

1 **Interactive comment on “Spatio-temporal variations of nitrogen**
2 **isotopic records in the Arabian Sea” by S.-J. Kao et al.**

3 **S.-J. Kao et al.**

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5 Received and published: 29 September 2014

6

7 **Reply to Anonymous Referee #1**

8 *1) Abstract: should be revised, and specifically talking about the modern-day first,*
9 *and then the downcore record and its implications.*

10 Reply: Modified as suggested.

11 “Available reports of dissolved oxygen, $\delta^{15}\text{N}$ of nitrate ($\delta^{15}\text{N}_{\text{NO}_3}$) and $\delta^{15}\text{N}$ of total
12 nitrogen ($\delta^{15}\text{N}_{\text{bulk}}$) for trap material and surface/downcore sediments from the Arabian
13 Sea (AS) were synthesized to explore its past nitrogen dynamics. Based on $25 \mu\text{mol}$
14 kg^{-1} dissolved oxygen isopleth at 150 m deep, we classified all reported data into
15 northern and southern groups. By using $\delta^{15}\text{N}_{\text{bulk}}$ of the sediments, we obtained
16 geographically distinctive bottom-depth effects for the northern and southern AS at
17 different climate stages. After eliminating the bias caused by bottom depth, the
18 modern day sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ values largely reflect the $\delta^{15}\text{N}_{\text{NO}_3}$ supply from the
19 bottom of the euphotic zone. For an addition to the dataset, nitrogen and carbon
20 contents versus their isotopic compositions of a sediment core (SK177/11) collected
21 from the most southeastern part of the AS were measured for comparison. We found a
22 one-step increase in $\delta^{15}\text{N}_{\text{bulk}}$ starting at the deglaciation with a corresponding decrease
23 in $\delta^{13}\text{C}_{\text{TOC}}$ similar to reports elsewhere revealing a global coherence. By synthesizing
24 and re-analyzing all reported down core $\delta^{15}\text{N}_{\text{bulk}}$, we derived bottom-depth correction
25 factors at different climate stages respectively for the northern and southern AS. The
26 diffusive sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ values in compiled cores became confined after bias
27 correction revealing a more consistent pattern except recent 6 ka. Such high similarity
28 to the global temporal pattern indicates that the nitrogen cycle in the entire AS had

1 responded to open-ocean changes until 6 ka BP. Since 6ka BP further enhanced
2 denitrification (i.e., increase in $\delta^{15}\text{N}_{\text{bulk}}$) in the northern AS had occurred and likely
3 driven by monsoon; while in the southern we observed a synchronous reduction in
4 $\delta^{15}\text{N}_{\text{bulk}}$ implying that nitrogen fixation was promoted correspondingly as the
5 intensification of local denitrification at the northern AS basin.”

6

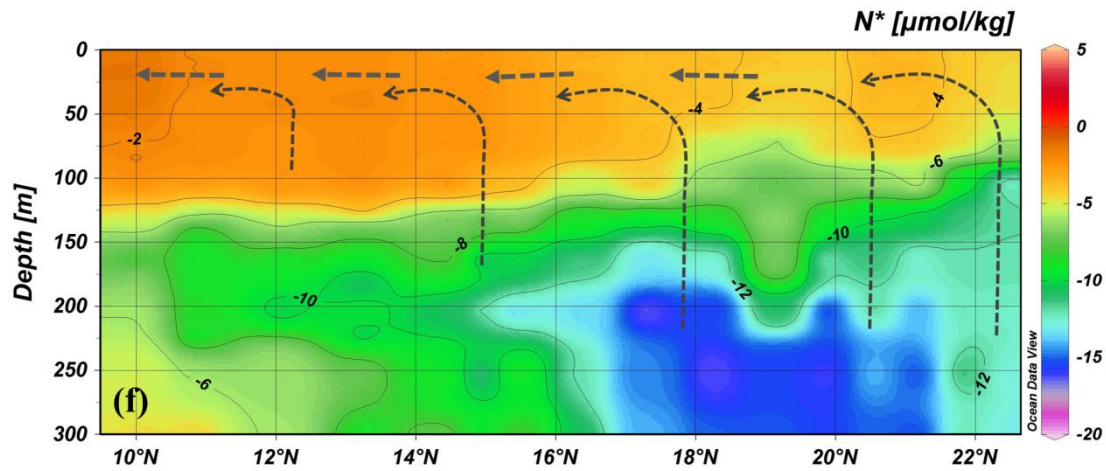
7 *2) Introduction: The second paragraph deals with how the $\delta^{15}\text{N}$ signal might be*
8 *altered. It is an important paragraph, though, I would put it at the beginning of the*
9 *"results" paragraph, somewhere in paragraph 4.2 or 5.1, where it is useful to*
10 *understand how the $\delta^{15}\text{N}$ signal might be altered. In the introduction it just alters the*
11 *flow of the manuscript.*

12 Reply: Thanks for this suggestion. We agree with the reviewer. This part had been
13 moved to the second graph in Section 5.1.

14

15 *3) Study area: A rapid sketch explaining how intermediate-depth water mass ventilate*
16 *the AS would be useful to figure out how the OMZ erodes from below, especially since*
17 *the core depth might be sensitive to that as well (see e.g. the Pichevin paper). For*
18 *example, it is unclear what is meant by in the last sentence of the paragraph. Arrows*
19 *on the transects, and their expansion, should help envision what you write.*

20 Reply: We added a new N^* transect specifically for the upper 300 m (Fig. 1f), in
21 which arrows were added to reveal the flow direction and the reference line of N^* of
22 -4 mentioned in text can be seen clearly.



1

2

3 *4) Material and methods: Second sentence: why pushing this? It is a useless sentence*
 4 *that alters the flow of the text - and it's probably wrong (check core MD77-191 in*
 5 *Bassinot et al., 2012, Climate of the Past).*

6 Reply: The sentence is only correct in terms of documenting $\delta^{15}\text{N}$. The sentence is
 7 now “Although the core MD77-191 locates further south in the AS (Bassinot et al.,
 8 2012), SK177/11 is so far the southernmost core with reference to $\delta^{15}\text{N}$ record.” This
 9 sentence is kept to emphasize we add one more core at the southern boundary (i.e.,
 10 more open-ocean type) into the dataset.

11

12 *5) Results: In paragraph 4.2, you can't say the $\delta^{15}\text{N}$ excursion at 13 ka occurs in the*
 13 *Younger Dryas chronozone given the uncertainties associated with your age model.*

14 Reply: Reviewer is right under considering the age uncertainties. The sentence is
 15 now: “The $\delta^{15}\text{N}$ values increased rapidly since ~19 ka BP, with a peak at ~15 ka BP
 16 and then started to decrease gradually toward modern day except the low $\delta^{15}\text{N}$
 17 excursion at ~14 ka BP.”

18

19 *6) Also, in the C and N increase seen in the first meter of sediments, could it be the*
 20 *signature of syn-sedimentary degradation of organic matter?*

21 Reply: We added more descriptions to the changing patterns of TOC and TN in the
 22 first meter. We also add the temporal variation of C/N into Figure 3 for discussion. In

1 this version, syn-sedimentary degradation was addressed; however, increased
2 sedimentation rate in Holocene should create higher preservation efficiency. Since
3 $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ did not show concomitant variations with C/N in first meter, we believe
4 the influence of organic degradation on isotope signal was insignificant, thus, no
5 influence on our original story. According to this comment, we added more
6 illustrations for the patterns in first meter in paragraph 4.2: “The upward increasing
7 TOC and TN patterns since Holocene were consistent with the increasing pattern of
8 sedimentation rate, suggesting a higher organic burial flux induced by enhance
9 productivity, which had been reported elsewhere in the AS (Altabet et al., 2002).

10 As for TOC/TN ratio, higher values appeared during deglacial transition and glacial
11 period (Fig. 3e). The highest value coincides with the $\delta^{13}\text{C}_{\text{TOC}}$ drop indicating there is
12 still some influence from terrestrial organics. However, the terrestrial organics
13 contains less nitrogen (C/N of 20; Meyers, 1997) thus the $\delta^{15}\text{N}$ did not drop
14 correspondingly. In the first meter, the downward decreasing pattern of TOC and TN
15 can also be attributed to syn-sedimentary degradation, if so a downward increasing in
16 TOC/TN should be evident. However, TOC/TN values varied in a narrow range
17 though an insignificant increasing trend can be observed if any. Higher preservation
18 efficiency in the Holocene is likely due to higher sedimentation rates. Nevertheless,
19 $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ did not show concomitant variations with C/N in the first meter or
20 throughout the core, the influence from either organic degradation or changes in
21 terrestrial organic input on isotopic signals is thus limited.”

22

23 *7) Discussion: Paragraph 5.1: please clarify the sentence "This implies that the*
24 *degree of addition processes, most likely the N₂-fixation, varied in concert with the*
25 *intensity of denitrification underneath." by mentioning the key results inferred in*
26 *Deutsch et al. (2007) cited just after. It would prepare the reader to get the mechanism*
27 *presented in paragraph 5.4.*

28 Reply: Thanks for this comment. We elaborated more about the spatial coupling
29 between N₂-fixation and denitrification following the mentioned sentence. “This

1 implies that certain degree of addition processes, most likely the N₂-fixation, varied in
2 concert with the intensity of denitrification underneath. Since the upwelling zones
3 distribute at the very north and the west of the AS and the upwelled water travels
4 southward (or outward) on the surface as shown in Fig. 1e, it is reasonable to see the
5 phenomenon of denitrification-induced N₂-fixation to compensate the nitrogen
6 deficiency. Consistent to this notion, Deutsch et al. (2007) discovered the spatial
7 coupling between denitrification in eastern tropical Pacific (upstream) and N₂-fixation
8 in western equatorial Pacific (downstream). Such horizontal nitrogen addition process
9 can also be seen clearly in our background information of N* (Fig. 1f).”

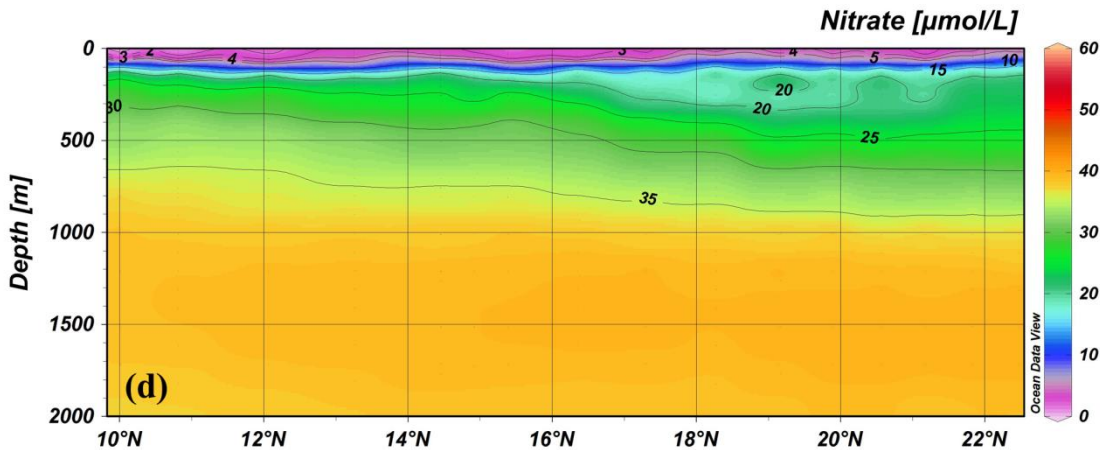
10

11 8) *Figure 1: why not expanding the panels b and c to the latitude where the core was*
12 *collected?*

13

Reply: We wish to have the data also, unfortunately, no available hydrographical data
14 extending to 8 degree. Nevertheless, we added nitrate transect (Fig. 1d) for
15 background introduction.

16



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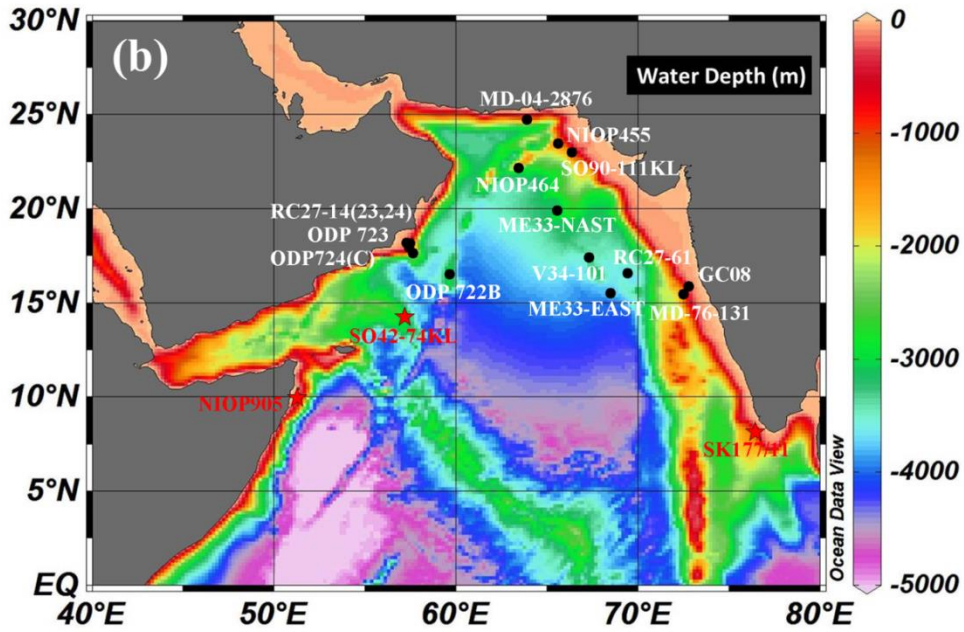
18

19 9) *You should try also to plot at depth the cores you deal with later, with appropriate*
20 *markers and colors, so that an easy comparison will help the reader checking where*
21 *the downcode records come from. It's really uneasy to figure out where the cores*
22 *mentioned are given the figure caption.*

23

Reply: We added a bathymetric map superimposed by core locations as Fig. 1b.

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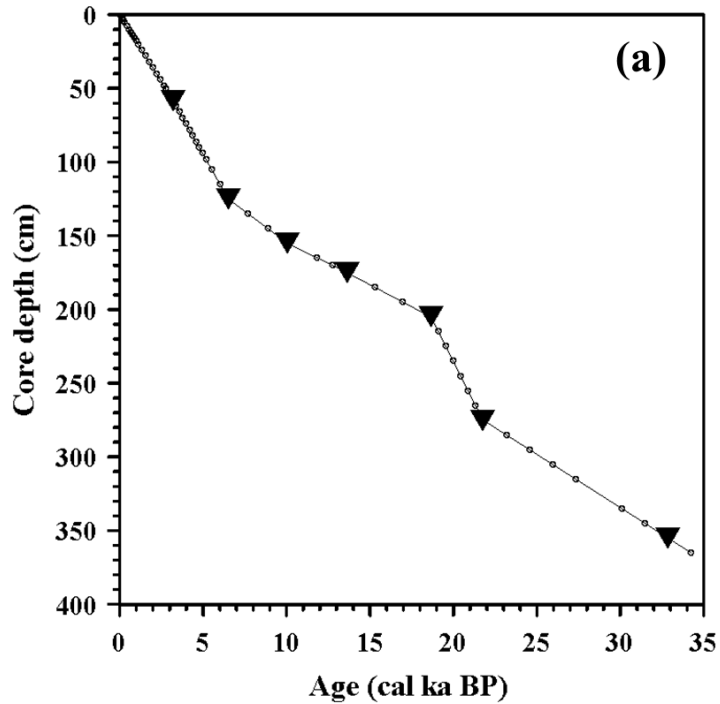
3

4 10) Figure 2: one radiocarbon date seems to be missing on panel A. Please provide
5 the calibration equation used.

6

7 Reply: The missing radiocarbon date has been added into Fig. 2a. The information
8 about calibration can be referred to Table 1.

8



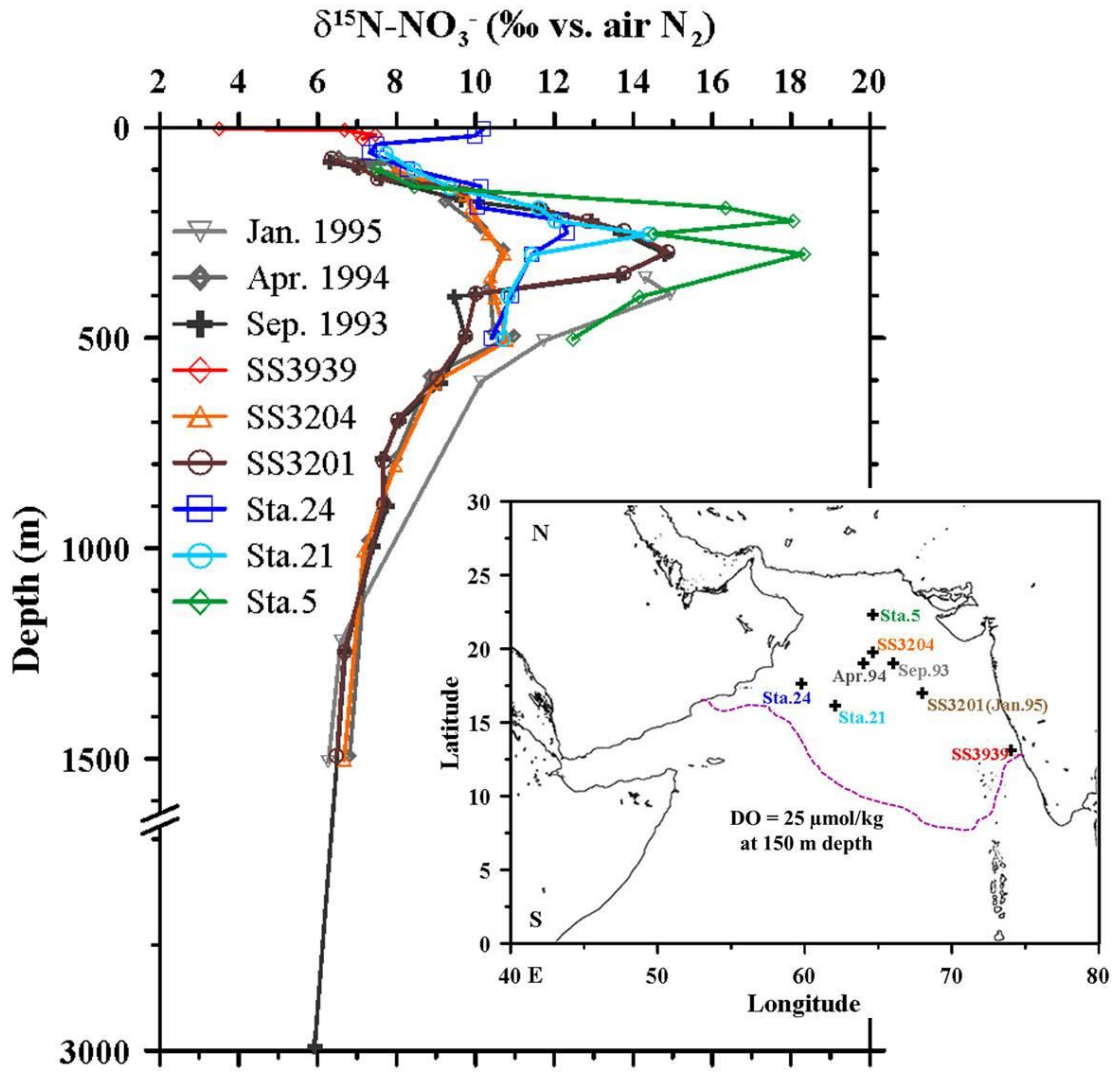
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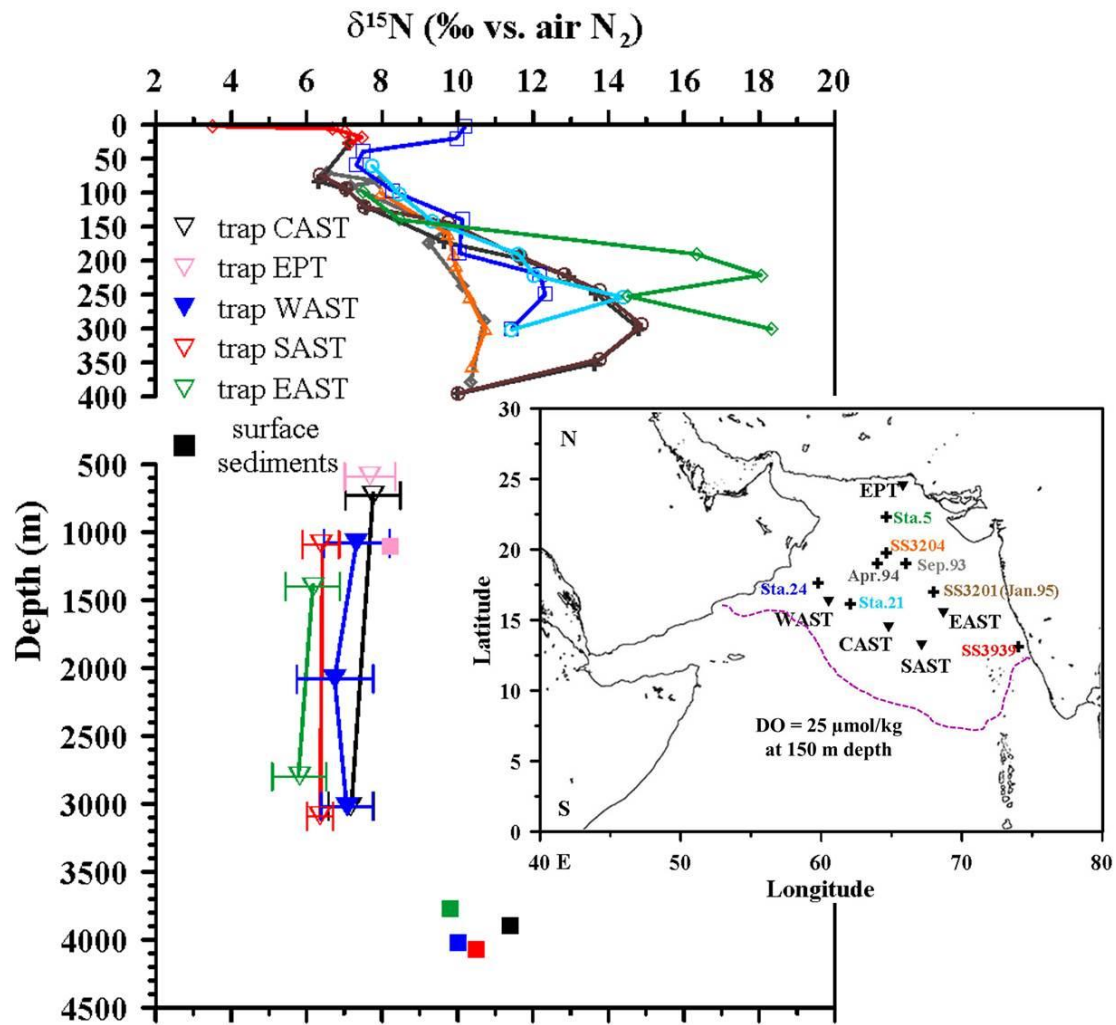
2 11) Figure 5: please enlarge the map and use colors on the map

3 Reply: Done. The new plots are shown below.

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12) Other details:
-choose between ODZ and OMZ (OMZ is more used)

Reply: We choose ODZ.

-in general, there are many English mistakes. A native English speaker should get a read over the manuscript.

Reply: We have our manuscript corrected by a native speaker.

Reply to Anonymous Referee #2

1
2 *1) Even the stratigraphy of core SK177/11 is well constrained by 7 AMS ^{14}C , the $\delta^{15}\text{N}$*
3 *record is very different from the two other records from the southern part of the*
4 *Arabian Sea (Fig. 8a; cores NIOP 905 and SO42-74KL). Is this difference only the*
5 *result of an age offset due to different methods of chronology or does it reflect a*
6 *peculiar dynamics off SW India?*

7 Reply: The foraminifera are absent in our core, thus, we have our dates by organic
8 carbon. This may introduce age uncertainties. Due to insufficiently high time
9 resolution, we cannot prove whether the differences during transition period were
10 caused by peculiar dynamics off SW India. Since the geographic and
11 glacial-interglacial differences in bottom-depth effect is one of the key points of this
12 paper, thus, we focus on the comparison between Holocene and glacial period when
13 water depth and climate condition were relatively stable.

14
15 *2) You cannot say that the $\delta^{15}\text{N}$ low at 13 ka occurs during the YD event which is*
16 *younger (Fig. 3 and text page 8720, lines 15). Anyway, this low should be in phase*
17 *with those centered during the YD of cores NIOP 905 and SO42-74KL (Fig. 8a).*
18 *Please clarify.*

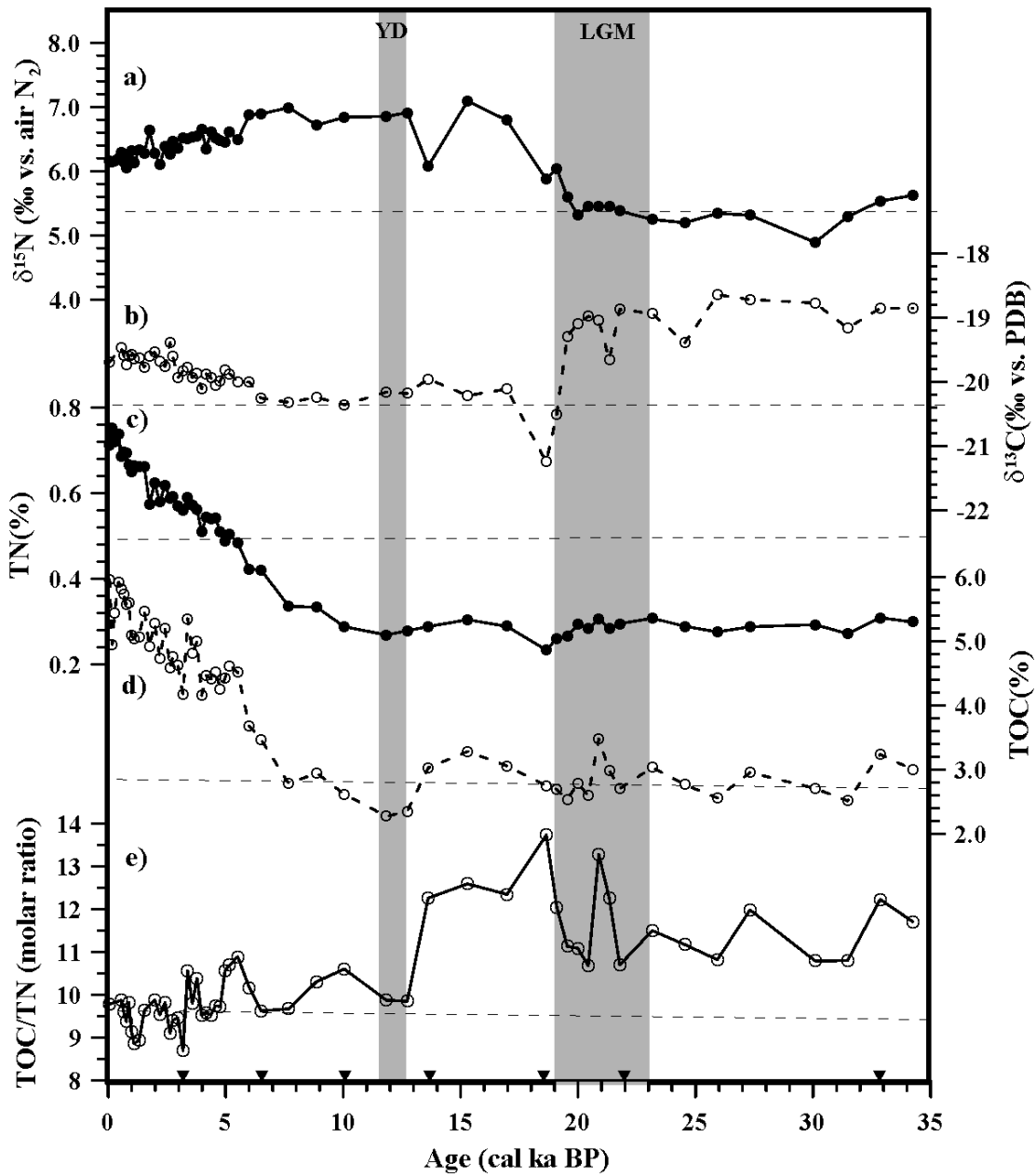
19 Reply: We do not mention YD in this version. And of course, this is the main reason
20 we exclude the transition period in our comparison.

21
22 *3) More details concerning especially the oceanography and climatology (nutrients,*
23 *production, water masses, and currents) of this region would be then helpful to better*
24 *constrain the dynamics of the region. For instance, are the $\delta^{15}\text{N}$ variations just a*
25 *matter of denitrification versus nitrogen fixation? Maps showing nitrate dynamics off*
26 *SW India (concentration, utilization) would be helpful.*

27 Reply: We added nitrate and shallow water N* transects as Figs. 1d and 1e.

4) C/N ratio and $\delta^{13}\text{C}_{\text{org}}$ (Fig 3 and 4) are clear indications that organic matter is pristine autochthonous (planktonic) material irrespectively of the climatic period. However, I would suggest the authors to plot the C/N profiles in Fig. 3.

Reply: We added the temporal variation of C/N into Figure 3. We also elaborate more about the temporal variation and the scatter plot (Fig. 4) according to the comment by Reviewer #1.



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1 *5) Moreover, the authors noticed that “An abrupt decrease in $\delta^{13}\text{C}$ was observed in*
2 *concert with the dramatic increase in $\delta^{15}\text{N}_{\text{bulk}}$ at the start of deglaciation”, and that*
3 *“A sharp decrease of $\delta^{13}\text{C}_{\text{TOC}}$ in SK177/11 at the start of deglaciation (Fig. 3b) may*
4 *indicate a rapid change of physical circulation had occurred in characteristics of the*
5 *intermediate water flowing into the AS”. They should also notice that the $\delta^{15}\text{N}$ and*
6 *$\delta^{13}\text{C}_{\text{Corg}}$ profiles mirror each other. It might be important and interesting to discuss*
7 *these observations in more details.*

8 Reply: This suggestion is well taken. We rewrote this paragraph and added more
9 illustrations to associated paragraphs. The latter one is now “In fact, the AAIW
10 penetrate further northward over 5°N in present day and even during the late
11 Holocene (You, 1998; Pichevin et al., 2007). Since the $\delta^{13}\text{C}$ of autochthonous
12 particulate organic carbon is negatively correlated to $[\text{CO}_2(\text{aq})]$ in euphotic zone (Rau
13 et al., 1991), the sharp decrease of $\delta^{13}\text{C}_{\text{TOC}}$ in SK177/11 at the start of deglaciation
14 (Fig. 3b) may infer the timing of a rapid accumulation of dissolved inorganic carbon
15 driven by the shrinking of oxygenated intermediate water (Pichevin et al., 2007) or
16 enhanced monsoon-driven upwelling (Ganeshram et al., 2000); both facilitate the
17 promotion of denitrification. Nevertheless, the mirror image between $\delta^{15}\text{N}$ and
18 $\delta^{13}\text{C}_{\text{TOC}}$ profiles revealed their intimate relation; of which, the variability was
19 attributable to the change of physical processes.”

21 *6) What do the authors mean by a rapid change of physical circulation in*
22 *characteristics of the intermediate water flowing into the AS?*

23 Reply: See reply above. This sentence is not clear and had been expanded to a
24 paragraph. “Since the $\delta^{13}\text{C}$ of autochthonous particulate organic carbon is negatively
25 correlated to $[\text{CO}_2(\text{aq})]$ in euphotic zone (Rau et al., 1991), the sharp decrease of
26 $\delta^{13}\text{C}_{\text{TOC}}$ in SK177/11 at the start of deglaciation (Fig. 3b) may infer the timing of a
27 rapid accumulation of dissolved inorganic carbon driven by the shrinking of
28 oxygenated intermediate water (Pichevin et al., 2007) or enhanced monsoon-driven
29 upwelling (Ganeshram et al., 2000); both facilitate the promotion of denitrification.

1 Nevertheless, the mirror image between $\delta^{15}\text{N}$ and $\delta^{13}\text{C}_{\text{TOC}}$ profiles revealed their
2 intimate relation; of which, the variability was attributable to the change of physical
3 processes.”

4

5 *7) In the core of the ms, the way the authors made to remove the bias due to water*
6 *depth is not clear. Please improve.*

7 Reply: We added an equation to make this clearer. “We applied the correction factor to
8 be equal to ((bottom depth – 100 m) × slope), ignoring the sea level changes during
9 the different climate stages.”

10

11 *8) My last comment concerns the choice of the authors to reject in their compilation*
12 *the record of Pichevin et al. (GBC, 2007) from the NE Arabian Sea (Kao et al., page*
13 *8725, lines 14-15), arguing that it might be influenced by terrigenous input. This*
14 *assumption contradicts the interpretations of Pichevin et al (2007). The authors*
15 *should integrate the record of Pichevin in their comparison.*

16 Reply: Thanks for reviewer’s correction. We have considered the MD-04-2876 in this
17 study for comparison. We revised the Table 2 as well accordingly. Since Pichevin’s
18 core was taken from shallower water depth, we suspected inorganic nitrogen
19 (clay-fixed) might have influence to deviate their $\delta^{15}\text{N}$ values from that of other cores
20 in northern basin. In this version, we put this core into estimation and follow their
21 explanation for the relatively low values.

22

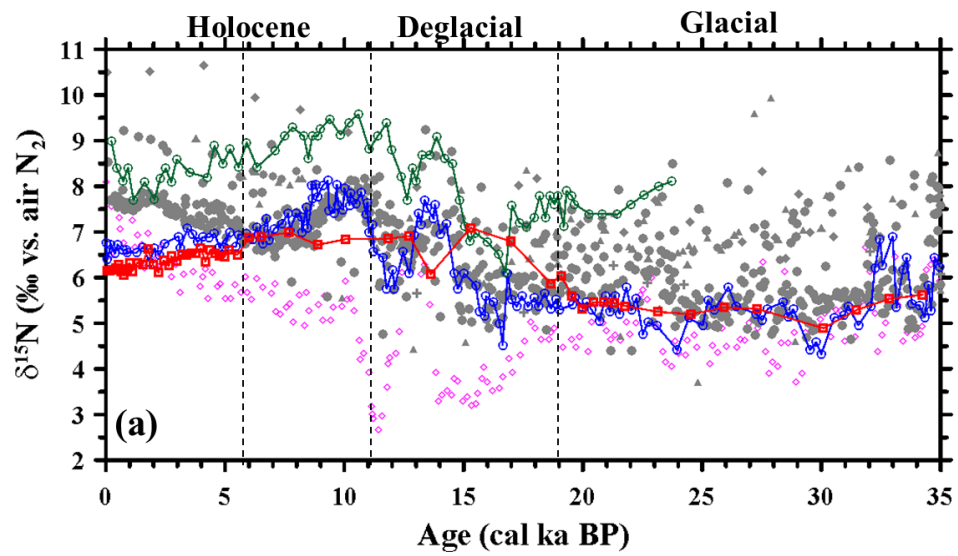
23 *9) Minor comments : Refs : Mollenhauer et al. instead of Mullenhauer et al.*

24 Reply: Corrected.

25

26 *10) Fig. 8a: I would suggest the authors to separate in two different graphs the 3*
27 *cores from the southern part of the Arabian Sea from the northern cores (including*
28 *Pichevin’s core). The figure would be then more readable.*

- 1 Reply: We used a lighter color for cores from the northern AS. Pichevin's core is also
- 2 included in this version.



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List of changes

We did a lot of changes in the manuscript because we took the referees' comments seriously.

1. As suggested by a native English-speaker, we would like to use the word “spatiotemporal” instead of “spatio-temporal” in the title to give a concise look.
2. One of our co-author Xianhui Sean Wan suggested adding his English name.
3. One of our co-author Chen-Tung Arthur Chen changed the name of his institution as “Department of Oceanography”
4. Abstract: we modified abstract as suggested by one referee.
5. In the text part, any changes or inserted are marked in blue and red.
6. In figures, most changes are supposed to support the related changes in the text. Figures with red crosses represent for the ones need to be deleted, while new ones of each can be seen its next page.

Spatio-temporal variations of nitrogen isotopic records in the Arabian Sea

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Abstract

~~Nitrogen and carbon contents versus their isotopic compositions for past 35 ka for a sediment core (SK177/11) collected from the most southeastern part of the Arabian Sea (AS) were presented. A one step increase in $\delta^{15}\text{N}_{\text{bulk}}$ starting at the deglaciation with a corresponding decrease in $\delta^{13}\text{C}_{\text{TOC}}$ was found similar to documentation elsewhere which showed a global coherence in general. We synthesized a~~ available reports ~~including~~ of dissolved oxygen, $\delta^{15}\text{N}$ of nitrate ($\delta^{15}\text{N}_{\text{NO}_3}$), ~~as well as~~ and $\delta^{15}\text{N}$ of total nitrogen ($\delta^{15}\text{N}_{\text{bulk}}$) for trap material and surface/downcore sediments ~~in from~~ the Arabian Sea (AS) were synthesized ~~in order~~ to explore ~~the its~~ past nitrogen dynamics ~~in the Arabian Sea~~. Based on ~~According to~~ 25 $\mu\text{mol kg}^{-1}$ dissolved oxygen isopleth at 150 m deep, we classified all reported data into northern and southern groups. By using $\delta^{15}\text{N}_{\text{bulk}}$ of the sediments, we ~~We~~ obtained geographically distinctive bottom-depth effects for the northern and southern AS at different climate stages. After ~~By~~ eliminating the bias caused by bottom depth, the modern day sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ values largely reflect the $\delta^{15}\text{N}_{\text{NO}_3}$ supply from the bottom of the euphotic zone. For an addition to the dataset, nitrogen and carbon contents versus their

1 isotopic compositions of a sediment core (SK177/11) collected from the most
2 southeastern part of the AS were measured for comparison. We found a one-step
3 increase in $\delta^{15}\text{N}_{\text{bulk}}$ starting at the deglaciation with a corresponding decrease in
4 $\delta^{13}\text{C}_{\text{TOC}}$ similar to reports elsewhere revealing a global coherence. By synthesizing
5 and re-analyzing all reported down core $\delta^{15}\text{N}_{\text{bulk}}$, we derived bottom-depth correction
6 factors at different climate stages respectively for the northern and southern AS.
7 ~~meanwhile,~~ The diffusive sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ values in long-compiled cores became
8 confined after bias correction revealing a more consistent pattern except recent 6 ka.
9 Such high similarity to the global temporal pattern indicates that ~~the~~ the nitrogen cycle
10 in the entire AS had apparently responded to open-ocean changes until 6 ka BP,
11 ~~during which~~ Since 6ka BP further enhanced denitrification (i.e., increase in $\delta^{15}\text{N}_{\text{bulk}}$)
12 in the northern AS had occurred and ~~was~~ likely ~~local and~~ driven by monsoon; while in
13 the southern we observed a synchronous reduction in $\delta^{15}\text{N}_{\text{bulk}}$ implying that AS either
14 nitrogen fixation was ~~enhanced~~ promoted correspondingly as the intensification of
15 local denitrification at the northern AS basin. ~~to the continuously reduced $\delta^{15}\text{N}_{\text{bulk}}$ for~~
16 ~~a compensation or the decreasing trend just followed the global pattern dominated by~~
17 ~~a longer term coupling of N_2 fixation and denitrification.~~

19 **1 Introduction**

20 Biogeochemical processes of nitrogen in the ocean are intimately related to various
21 elemental cycles synergistically modulate atmospheric CO_2 and N_2O concentrations,
22 thus feedback to climate on millennial time scale (Gruber, 2004; Falkowski and
23 Godfrey, 2008; Altabet et al., 2002). Though oxygen deficient zones (ODZs) occupy
24 only ~4% of ocean volume, the denitrification process therein contributes remarkably
25 to the losses of nitrate, leaving excess P in the remaining water mass to stimulate
26 N_2 -fixation while entering euphotic zone (Morrison et al., 1998; Deutsch et al., 2007)
27 and thus controlling the budget of bio-available nitrogen in ocean. Denitrification
28 leaves $^{15}\text{NO}_3^-$ in residual nitrate (Sigman et al., 2001); whereas, N_2 -fixation
29 introduces new bio-available nitrogen with low $\delta^{15}\text{N}$ values (Capone et al., 1997) into

1 ocean for compensation. The Arabian Sea (AS), as one of the three largest ODZs in
2 the world ocean with distinctive monsoon driven upwelling, accounts for at least one
3 third of the loss of marine fixed nitrogen (Codispoti and Christensen, 1985) playing
4 an important role in the past climate via regulating atmospheric N₂O concentration
5 (Agnihotri et al., 2006) or nitrogen inventory to modulate CO₂ sequestration through
6 biological pump (Altabet, 2006).

7 Sedimentary nitrogen isotope, measured as standard δ notation with respect to
8 standards of atmospheric nitrogen, is an important tool to study the past marine
9 nitrogen cycle. Nitrogen isotope compositions of sedimentary organic matter
10 potentially reflect biological processes in water column, such as denitrification
11 (Altabet et al., 1995; Ganeshram et al., 1995, 2000), nitrogen fixation (Haug et al.,
12 1998), and the degree of nitrate utilization by algae (Altabet and Francois, 1994;
13 Holmes et al., 1996; Robinson et al., 2004). However, alteration may occur (through
14 various ways or processes, e.g., diagenesis) before the signal of $\delta^{15}\text{N}$ of exported
15 production is buried. ~~Altabet and Francois (1994) reported little diagenetic alteration~~
16 ~~of the near-surface $\delta^{15}\text{N}$ in the equatorial Pacific, while an apparent +5‰ diagenetic~~
17 ~~enrichment relative to sinking particles in the southern ocean south of the polar front.~~
18 ~~In the Sargasso Sea, sedimentary $\delta^{15}\text{N}$ also enriched by 3–6‰ relative to sinking~~
19 ~~particles (Altabet et al., 2002; Gruber and Galloway, 2008). The degree of alteration~~
20 ~~was attributed to particle sinking rate and OM preservation (Altabet, 1988).~~
21 ~~Gaye-Haake et al. (2005) also suggested that low sedimentation rates benefit organic~~
22 ~~matter decomposition resulting in positive shift in bulk sedimentary $\delta^{15}\text{N}$ comparing~~
23 ~~to sinking particles in South China Sea. Finally, Robinson et al. (2012) concluded that~~
24 ~~oxygen exposure time at the seafloor is the dominant factor controlling the extent of N~~
25 ~~isotopic alteration.~~

26 Previous measurements of $\delta^{15}\text{N}_{\text{bulk}}$ in various cores and surface sediments in the AS
27 showed the following points: 1) near-surface NO₃⁻ in AS is completely utilized in an
28 annual cycle, resulting in small isotopic fractionation between $\delta^{15}\text{N}$ of exported
29 sinking particles and $\delta^{15}\text{N}$ of NO₃⁻ supplied to the euphotic zone (Altabet, 1988;

1 Thunell et al., 2004); 2) monsoon-driven surface productivity and associated oxidant
2 demand are regarded as the main control on water column denitrification in the past
3 (Ganeshram et al., 2000; Ivanochko et al., 2005); 3) sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ primarily
4 reflects the relative intensity of water column denitrification in this area (Altabet et al.,
5 1995; 1999) ; and 4) oxygen supply at intermediate depth by the Antarctic
6 Intermediate Waters (AAIW) can modulate the denitrification intensity in northern AS
7 (Schulte et al., 1999; Schmittner et al., 2007; Pichevin et al., 2007). Among previous
8 researches, the geographical features in sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ between north and south
9 basins of AS have not been discussed, particularly on the basis of bottom-depth effect
10 which might be different during glacial and interglacial periods.

11 In this study, a sediment core (SK177/11) collected from the slope of southeastern AS
12 was measured for organic C and N contents and their stable isotopes. We synthesized
13 previous hydrographical and isotopic data, such as dissolved oxygen (DO), N^* ($\text{N}^* =$
14 $\text{NO}_3^- - 16 \times \text{PO}_4^{3-} + 2.9$; Gruber and Sarmiento, 2002), and $\delta^{15}\text{N}$ of nitrate as well as
15 trapped material and surface/downcore sediments, among which surface and
16 downcore sediments may have experienced more intensified diagenetic alteration.
17 Based on the subsurface DO concentration of $25 \mu\text{mol kg}^{-1}$ isopleth at 150 m, the
18 datasets in the AS were separated into north and south basins by time span (glacial,
19 Holocene and modern) for comparison. We aim to (1) investigate the geographic and
20 glacial-interglacial differences in bottom-depth effect and to (2) retrieve extra
21 information from sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ by removing basin/climate stage specific
22 bottom-depth effects, thus, better decipher the environmental history of the Arabian
23 Sea.

24

25 **2 Study area**

26 The Arabian Sea is characterized by seasonal reversal of monsoon winds, resulting in
27 large seasonal physical/hydrographic/biological/chemical variations in water column
28 (Nair et al., 1989). Cold and dry northeasterly winds blow during winter from
29 high-pressure cell of the Tibetan Plateau, whereas heating of the Tibetan Plateau in

1 summer (June to September) reverses the pressure gradient leading to warm and moist
2 southwesterly winds and precipitation maximum. In present day, the SW monsoon is
3 much stronger than its northeastern counterpart. ~~Consistent seasonal oscillation in~~
4 ~~surface biological productivity was also revealed by satellite pictures in the entire~~
5 ~~Arabian Sea.~~

6 The spatial distribution of DO at 150 m deep for the AS is shown in Figure 1a (World
7 Ocean Atlas 2009, <http://www.nodc.noaa.gov/OC5/WOA09/woa09data.html>), which
8 shows a clear southward increasing pattern with DO increased from ~5 to >100 μmol
9 kg^{-1} and the lowest DO value appears at the northeast of the northern basin. As
10 denitrification, the dominant nitrate removal process, generally occurs in the water
11 column where DO concentration ranges 0.7~20 $\mu\text{mol kg}^{-1}$ (Paulmier et al., 2009), the
12 intensity of denitrification was reported to descend gradually, corresponding with the
13 DO spatial pattern from northern to southern parts of AS, and became unobvious at 11
14 or 12 $^{\circ}\text{N}$ (Naqvi et al., 1982). As indicated by upper 2000 m N-S transect of DO (Fig.
15 1c), a southward decreasing in ODZ thickness can be observed and the contour line of
16 5 $\mu\text{mol kg}^{-1}$ extends to around 13 $^{\circ}\text{N}$. Since the nitrate source is mainly from the
17 bottom of euphotic zone at around 150 m we postulate a geographically distinctive
18 sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ underneath ODZ. Thus, an isopleth of 25 $\mu\text{mol kg}^{-1}$ DO at 150 m
19 is applied as a geographic boundary to separate the northern from the southern part of
20 AS basin. The interface where DO concentration changed from 20 to 30 $\mu\text{mol kg}^{-1}$
21 was such a transition zone. On the other hand, the bottom layer of ODZ moves
22 shallower toward south as shown previously by Gouretski and Koltermann (2004).
23 Accordingly, the bottom oxygen content may also be a factor to influence the degree
24 of alteration in sedimentary $\delta^{15}\text{N}_{\text{bulk}}$.

25 As mentioned in Introduction, nitrate is removed via denitrification in ODZs resulting
26 in excess P to stimulate N_2 -fixation. In Figs. 1d, 1e and 1f, we presented the N-S
27 transect of nitrate and N^* (for both the upper 2000 m and 300 m) in January. Even
28 though there contains nitrate in the very surface water (Fig. 1d), as mentioned earlier
29 near-surface NO_3^- in AS is completely utilized in an annual cycle (Altabet, 1988;

1 Thunell et al., 2004). Furthermore, negative N* (P-excess) throughout the water
2 column represents a nitrate deficit and the lowest N* value appears at ~300 m at
3 18-20 °N, where DO is <1 μmol kg⁻¹. Fig. 1c shows the N-S transect of N*, which
4 represents nitrate deficit (Gruber and Darmieto, 2002), for the upper 2000 m. Overall,
5 the entire water column is negative in N* (P-excess) and the lowest N* value appears
6 at ~300 m at 19-20 °N, where DO is <1 μmol kg⁻¹. Meanwhile, a gradually southward
7 increase in N* can be observed for upper 100 m and the isopleth of N* of -4 deepens
8 southward with the highest N* (-2) appearing at ~10-12 °N. The volume expansion of
9 high N* water as well as a simultaneous increase in N* strongly indicate an addition
10 of bio-available nitrogen when surface water traveling southward.

12 **3 Material and method**

13 A sediment gravity core, SK177/11 (8.2 °N and 76.47 °E), was collected at water
14 depths of 776 m on the continental slope off southwest coast of India (Kerala) during
15 the 177th cruise of *ORV SagarKanya* on October 2002. Although the core MD77-191
16 locates further south in the AS (Bassinot et al., 2012), SK177/11 is so far the
17 southernmost core with reference to δ¹⁵N record. This is so far the reported sediment
18 core locates at the most southern part of the AS. The 3.65 m long core was
19 sub-sampled at intervals of 2 cm from top to 100 cm, and 5 cm from 100 cm to the
20 bottom of the core (open circles in Fig. 2a). At 1.70 m we see an obvious boundary,
21 above which sediments possessed by brownish grey clayey and greenish black clayey
22 sediments occupied the lower part. There is a distinct boundary at ~1.7 m, above
23 which the core contains mainly of brownish grey clayey sediments. Neither distinct
24 laminations nor turbidities can be observed by visual contact immediately after
25 collection or at the time during sub-sampling (Pandarinath et al., 2007). All
26 sub-samples were freeze-dried and ground into powder in an agate mortar with pestle.
27 Sand was almost absent (<1 wt.%) throughout the core.

28 The calendar chronology for core SK177/11 was based on 7 accelerator mass
29 spectrometry (AMS) radiocarbon (¹⁴C) dates of bulk organic matter (Fig. 2a).

1 Calendar years were calculated using calibration CALIB 6.0 with a reservoir age
2 correction of 402 years (Stuiver et al., 1998; Reimer et al., 2009). Details on the ^{14}C
3 age controlling points were presented in Table 1. Given that the AMS ^{14}C dates of
4 SK177/11 were obtained on total organic carbon, we may not be able to avoid the
5 mixture of organics of different ages during transport (Mollenhauer et al., 2005) or
6 interference by pre-aged organics sourced from land (Kao et al., 2008). However,
7 besides the reservoir age correction, higher TOC contents (Range: 2.2~5.5%) of
8 sediments and their marine-sourced organic carbon, as confirmed by stable C isotope
9 data and C/N ratio shown in Figs. 3b and 3e, we are confident that our age model is
10 reliable and less likely affected by age heterogeneity.

11 Bulk sedimentary nitrogen content and $\delta^{15}\text{N}$ analyses were carried out using a
12 Carlo-Erba EA 2100 elemental analyzer connected to a Thermo Finnigan Delta V
13 Advantage isotope ratio mass spectrometer (EA-IRMS). Sediments for total organic
14 carbon (TOC) analyses were acid-treated with 1N HCl for 16 hr, and then centrifuged
15 to remove carbonate. The acid-treated sediments were further dried at 60 °C for TOC
16 content and $\delta^{13}\text{C}$. The nitrogen isotopic compositions of acidified samples were
17 obtained in the same time for comparison. Carbon and nitrogen isotopic data were
18 presented by standard δ notation with respect to PDB carbon and atmospheric
19 nitrogen. USGS 40, which has certified $\delta^{13}\text{C}$ of -26.24‰ and $\delta^{15}\text{N}$ of -4.52‰ and
20 acetanilide (Merck) with $\delta^{13}\text{C}$ of -29.76‰ and $\delta^{15}\text{N}$ of -1.52‰ were used as working
21 standards. The reproducibility of carbon and nitrogen isotopic measurements is better
22 than 0.15‰. The precision of nitrogen and carbon content measurements were better
23 than 0.02% and 0.05%, respectively. Meanwhile, the acidified and non-acidified
24 samples exhibited identical patterns in $\delta^{15}\text{N}$ (not shown) with mean deviation of
25 0.3‰.

26

27 **4 Results**

28 **4.1 Sedimentation rate**

1 The age-depth curve was shown in Fig. 2a, in which age dates were evenly distributed
2 throughout the core though not high resolution. In Mollenhauer et al. (2005), the
3 largest age offset between total organic carbon and co-occurring foraminifera is ~3000
4 years and mostly <2000 years. Meanwhile, the offset remains more or less constant
5 throughout past 20 ka regardless of the deglacial transition. The youngest date in our
6 core is 3180 cal ka BP at 58 cm. We may expect younger age on the surface. Thus, if
7 our TOC samples contain any pre-aged organics as indicated by Mollenhauer et al.
8 (2005), the offset should not be too large to alter our interpretation for the comparison
9 between glacial and Holocene periods. The linear sedimentation rates derived from 7
10 date intervals range from 6 to 20 cm ka⁻¹ (Fig. 2b), with relatively constant value (~6
11 cm ka⁻¹) prior to Holocene except the excursion around the last glacial maximum. The
12 linear sedimentation rates started to increase since Holocene and reached 18~20 cm
13 ka⁻¹ when the sea level reached modern day level.

14 **4.2 Nitrogen and carbon contents and their isotopes**

15 Values of $\delta^{15}\text{N}_{\text{bulk}}$ ranged from 4.7‰ to 7.1‰ with significantly lower values during
16 glacial period (Fig. 3a). The $\delta^{15}\text{N}$ values increased rapidly since ~19 ka BP, with a
17 peak at ~15 ka BP and then started to decrease gradually toward modern day except
18 the low $\delta^{15}\text{N}$ excursion at ~14.3 ka BP ~~during the Younger Dryas event~~. Figure 3b
19 shows that values of $\delta^{13}\text{C}_{\text{TOC}}$ (-21.5~-18.5‰) were consistent with the $\delta^{13}\text{C}$ of
20 typical marine organic matter end-member (-22~-18‰; Meyers, 1997). An abrupt
21 decrease in $\delta^{13}\text{C}$ was observed in concert with the dramatic ~~decrease~~ increase in
22 $\delta^{15}\text{N}_{\text{bulk}}$ at the start of deglaciation.

23 Bulk nitrogen content (TN) had a range of 0.23~0.75% (Fig. 3c) and the total organic
24 carbon (TOC) content ranged from 2.2% to 5.5% (Fig. 3d). Both TN and TOC
25 showed similar trend over the last 35 ka BP with relatively constant values prior to
26 Holocene and an afterward elevation till modern day. The upward increasing TOC and
27 TN patterns since Holocene were consistent with the increasing pattern of
28 sedimentation rate, suggesting a higher organic burial flux induced by enhance

1 productivity, which had been reported elsewhere in the AS (Altabet et al., 2002).
2 As for TOC/TN ratio, higher values appeared during deglacial transition and glacial
3 period (Fig. 3e). The highest value coincides with the $\delta^{13}\text{C}_{\text{TOC}}$ drop indicating there is
4 still some influence from terrestrial organics. However, the terrestrial organics
5 contains less nitrogen (C/N of 20; Meyers, 1997) thus the $\delta^{15}\text{N}$ did not drop
6 correspondingly. In the first meter (since ~5 ka), the downward decreasing pattern of
7 TOC and TN can also be attributed to syn-sedimentary degradation, if so a downward
8 increasing in TOC/TN should be evident. However, TOC/TN values varied in a
9 narrow range not revealing a significant increasing trend can. Nevertheless, $\delta^{15}\text{N}$ and
10 $\delta^{13}\text{C}$ did not show concomitant variations with C/N in the first meter or throughout the
11 core, the influence from either organic degradation or changes in terrestrial organic
12 input on isotopic signals is thus limited.

13 Figure 4 shows the scatter plot of TOC against TN. The slope of the linear regression
14 line for TOC against TN ($\text{TOC} = (6.67 \pm 0.22) \times \text{TN} + (0.99 \pm 0.11)$, $R^2 = 0.94$, $n = 57$,
15 $p < 0.0001$) is 6.67 again indicating that organic matter is mainly marine-sourced.
16 Though this slope is slightly higher than the Redfield ratio of 5.68 (wt./wt.), it is
17 lower than that observed on the East China Sea shelf (7.46; Kao et al., 2003).
18 Meanwhile, the intercept of TN is negative when TOC downs to zero implying that
19 inorganic nitrogen can be ignored in our core. Obviously, if we force the regression
20 through the origin point, TOC/TN values for samples during the Holocene will have
21 the lower ratios reflecting even less contribution from terrestrial organics.

22 23 **5 Discussion**

24 **5.1 Downward transfer and transformation of N isotopic signal**

25 As mentioned, the signal of sedimentary $\delta^{15}\text{N}$ may be altered under different burial
26 conditions. Altabet and Francois (1994) reported little diagenetic alteration of the
27 near-surface $\delta^{15}\text{N}$ in the equatorial Pacific, while apparent +5% enrichment relative to
28 sinking particles in the Southern Ocean, south of the polar front. In the Sargasso Sea,

1 sedimentary $\delta^{15}\text{N}$ also enriched by 3~6‰ relative to sinking particles (Altabet et al.,
2 2002; Gruber and Galloway, 2008). The degree of alteration was attributed to particle
3 sinking rate and OM preservation (Altabet, 1988). Gaye-Haake et al. (2005) also
4 suggested that low sedimentation rates benefit organic matter decomposition resulting
5 in positive shift in bulk sedimentary $\delta^{15}\text{N}$ comparing to sinking particles in South
6 China Sea. Finally, Robinson et al. (2012) concluded that oxygen exposure time at the
7 seafloor is the dominant factor controlling the extent of N isotopic alteration. Thus, it
8 is necessary to follow the track of $\delta^{15}\text{N}$ signal to clarify the occurrence of deviation
9 during transfer.

10 The reported depth profiles of $\delta^{15}\text{N}_{\text{NO}_3}$ in the AS were shown in Fig. 5, in which
11 $\delta^{15}\text{N}_{\text{NO}_3}$ values of water depth deeper than 1200 m range narrowly around 6~7‰,
12 which is slightly higher than the global average of the deep oceans ((4.8 ± 0.2)‰ for >
13 2500 m, Sigman et al., 2000; (5.7 ± 0.7)‰ for >1500 m, Liu and Kaplan, 1989).
14 Below the euphotic layer, $\delta^{15}\text{N}_{\text{NO}_3}$ increases rapidly peaking at around 200~400 m.
15 The preferential removal of $^{14}\text{NO}_3$ by water column denitrification accounts for these
16 subsurface $\delta^{15}\text{N}_{\text{NO}_3}$ highs (Brandes et al., 1998; Altabet et al., 1999; Naqvi et al.,
17 2006). The subsurface $\delta^{15}\text{N}_{\text{NO}_3}$ maximum ranges from 10 to 18‰ for different stations
18 implying a great spatial heterogeneity in water column denitrification intensity. It is
19 worth mentioning that, higher values in general appear in the northeastern AS
20 (15~18‰; Fig. 5) highlighting that the focal area of water column denitrification is
21 prone to northeastern Arabian Sea (Naqvi et al., 1994; Pichevin et al., 2007), also
22 revealed by the DO spatial distribution (Fig. 1a). Contrary to higher denitrification in
23 the northeastern AS, the export production is always higher in the northwestern AS
24 throughout a year (Rixen et al., 1996). Such decoupling between productivity and
25 denitrification was attributed to the oxygen supply by intermediate water exchange
26 besides primary productivity oxygen demand (Pichevin et al., 2007). Note that, the
27 $\delta^{15}\text{N}_{\text{NO}_3}$ values at water depth of 100~150 m, which corresponds to the bottom depth
28 of euphotic zone (Olson et al., 1993), from different stations fall within a narrow
29 range of 7~9‰ despite of wide denitrification intensity underneath. The rapid addition

1 of new nitrogen as mentioned earlier might account for the relatively uniform $\delta^{15}\text{N}_{\text{NO}_3}$
2 at the bottom of euphotic layer. Unfortunately, there no either $\delta^{15}\text{N}_{\text{NO}_3}$ profiles or
3 sediment trap data from the southern basin for comparison.

4 Interestingly, reported $\delta^{15}\text{N}$ of sinking particles ($\delta^{15}\text{N}_{\text{SP}}$) collected by five
5 sedimentation traps deployed from 500 m throughout 3200 m deep ranged narrowly
6 from 5.1~8.5‰ (Fig. 6), which is slightly lower but overlaps largely with $\delta^{15}\text{N}_{\text{NO}_3}$
7 values at 100~150 m. Such similarity in $\delta^{15}\text{N}_{\text{NO}_3}$ at 100~150 m and sinking particles
8 strongly indicated that 1) NO_3^- source for sinking particles was coming from the
9 depth around 100~150 m instead of 200~400 m, the oxygen deficient zones (ODZs)
10 where the maximum $\delta^{15}\text{N}_{\text{NO}_3}$ value occurred (Schäfer and Ittekkot, 1993; Altabet et al.,
11 1999) and 2) little alteration had occurred in $\delta^{15}\text{N}_{\text{SP}}$ along sinking in the water column
12 as indicated by Altabet (2006). There were only these five trap stations with nitrogen
13 isotope information available in the AS (Gaye-Haaake et al., 2005). The trap locations
14 were in the same area but little south comparing with $\delta^{15}\text{N}_{\text{NO}_3}$ stations (insert map in
15 Fig. 6). The slightly lower $\delta^{15}\text{N}$ in sinking particle is attributable to their geographic
16 locations (see below) since incomplete relative utilization of surface nitrate has been
17 documented to have a very limited imprint on the $\delta^{15}\text{N}$ signal in the AS (e.g., Schäfer
18 and Ittekkot, 1993).

19 The uniformly low values of $\delta^{15}\text{N}_{\text{NO}_3}$ at the bottom of euphotic zone should be a
20 consequence resulted from various processes in the euphotic zone, such as
21 remineralization, nitrification and N_2 -fixation. Nevertheless, the distribution pattern of
22 N^* (Figs. 1e and 1f) illustrates that there must be an addition of $^{14}\text{NO}_3$ into the system
23 to cancel out the isotopic enrichment caused by denitrification. Note that the positive
24 offset in $\delta^{15}\text{N}_{\text{NO}_3}$ ($\Delta\delta^{15}\text{N}_{\text{NO}_3}$, 6~12‰) in ODZs caused by various degree of
25 denitrification were narrowed down significantly while nitrate transports upward. This
26 implies that certain degree of addition processes, most likely the N_2 -fixation, varied in
27 concert with the intensity of denitrification underneath. Since the upwelling zones
28 distribute at the very north and the west of the AS and the upwelled water travels
29 southward (or outward) on the surface as shown in Fig. 1e, it is reasonable to see the

1 phenomenon of denitrification-induced N₂-fixation to compensate the nitrogen
2 deficiency. Consistent to this notion, Deutsch et al. (2007) discovered the spatial
3 coupling between denitrification in eastern tropical Pacific (upstream) and N₂-fixation
4 in western equatorial Pacific (downstream). Such horizontal nitrogen addition process
5 can also be seen clearly in our background information of N* (Fig. 1f). ~~The spatial~~
6 ~~coupling of denitrification and N₂-fixation by Deutsch et al. (2007) is supportive of~~
7 ~~this notion.~~ In fact, fixed N had been proved to account for a significant part of
8 surface nitrate in modern day AS where denitrification is exceptionally intense
9 (Brandes et al., 1998; Capone et al., 1998; Parab et al., 2012).

10 Comparing with reported $\delta^{15}\text{N}$ of surface sediments retrieved from trap locations, a
11 significant positive shift in $\delta^{15}\text{N}$ can be seen at the seafloor (Fig. 6). Such positive
12 deviation can be seen elsewhere in previous reports (Altabet, 1988; Brummer et al.,
13 2002; Kienast et al., 2005) due to prolonged oxygen exposure after deposition
14 (Robinson et al., 2012) associated with sedimentation rate (Pichevin et al., 2007).
15 Although Cowie et al. (2009) found ambiguous relation between contents of
16 sedimentary organic carbon and oxygen in deep water, they also noticed the
17 appearance of maximum organic carbon contents at the lower boundary of ODZ,
18 where oxygen contents were relatively higher. Accordingly, they believed that there
19 existed other factors controlling the preservation of organic carbon, such as the
20 chemical characteristics of organic matter, the interaction between organic matters and
21 minerals, the enrichment and activity of benthic organism, or the physical factor
22 including the screening and water dynamic effect.

23 **5.2 Geographically-distinctive bottom depth effects in modern day**

24 As classified by oxygen content of $25 \mu\text{mol kg}^{-1}$ at 150 m, documented surface
25 sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ (Gaye-Haake et al., 2005) were separated into northern and
26 southern groups to examine the geographic difference in bottom-depth effect. Both
27 groups exhibit positive linear relationships between $\delta^{15}\text{N}_{\text{bulk}}$ and bottom depth (deeper
28 than 200 m) (Fig. 7a). The regression equations were shown in Table 2. Interestingly,

1 the regression differs statistically from each other in general in term of slope and
2 intercept. The slope represents the degree of positive shift of sedimentary $\delta^{15}\text{N}$ due to
3 bottom-depth effect. For the southern AS, the slope is $(0.76 (\pm 0.14) \times 10^{-3} \text{ km}^{-1})$,
4 which is close to the correction factor $(0.75 \times 10^{-3} \text{ km}^{-1})$ for the world ocean proposed
5 by Robinson et al. (2012) and further applied by Galbraith et al. (2012). By contrast,
6 the slope for the northern AS is significantly lower $(0.55 (\pm 0.08) \times 10^{-3} \text{ km}^{-1})$,
7 implying that the depth-associated alteration in the northern AS is smaller. The
8 correction factor for bottom-depth effect was suggested to vary in different regions
9 such as that in the South China Sea (Gaye et al., 2009). Since the magnitude of
10 oxygen exposure is the primary control of depth effect (Gaye-Haake et al., 2005;
11 Mobius et al., 2011; Robinson et al., 2012), we attributed this lower slope in the
12 northern AS to relatively higher sedimentation rates (not shown) and lower oxygen
13 contents as indicated by previous researches (Olson et al., 1993; Morrison et al., 1999;
14 Brummer et al., 2002).

15 On the other hand, the intercept for the northern AS regression (8.1 ± 0.2) is
16 significantly higher than that for the southern AS (6.0 ± 0.3) . As mentioned above,
17 $\delta^{15}\text{N}$ values of sinking particle resembled the $\delta^{15}\text{N}$ of nitrate sourced from 100~150 m
18 deep. According to the depth-dependent correction factor we may convert sedimentary
19 $\delta^{15}\text{N}_{\text{bulk}}$ values at various water depths into their initial condition when the diagenetic
20 alteration is minimal to represent the $\delta^{15}\text{N}$ of source nitrate. Higher intercept suggests
21 a stronger denitrification had occurred in northern AS surface sediments. The 2.1‰
22 lower intercept in the southern AS likely reflects the addition of N_2 -fixation in the
23 upper water column while it travels southward. The progressive increase of N^* toward
24 southern AS supports our speculation although none $\delta^{15}\text{N}_{\text{NO}_3}$ profiles had been
25 published in the southern basin. Future works about $\delta^{15}\text{N}_{\text{NO}_3}$ and $\delta^{15}\text{N}_{\text{SP}}$ in the
26 southern AS are needed.

27 In Fig. 7b, we presented corrected $\delta^{15}\text{N}_{\text{bulk}}$ values along with bottom depth for
28 northern and southern AS surface sediments for comparison. After removing
29 site-specific bias caused by bottom depth effect, the values and distribution ranges of

1 $\delta^{15}\text{N}_{\text{bulk}}$ for both northern and southern AS became smaller and narrower. For the
2 northern AS, the distribution pattern skewed negatively giving a standard deviation of
3 0.88‰, exactly falling in the range of 7~9‰ for $\delta^{15}\text{N}_{\text{NO}_3}$ (7~9‰) at the bottom of
4 euphotic zone. As a result, the corrected nitrogen isotopic signals in sediments more
5 truthfully represent the $\delta^{15}\text{N}_{\text{NO}_3}$ value at the bottom depth of euphotic zone.
6 Meanwhile, the statistically significant difference in $\delta^{15}\text{N}_{\text{bulk}}$ distribution between the
7 northern and southern AS further confirms the feasibility of our classification by using
8 DO isopleth of $25 \mu\text{mol kg}^{-1}$ at 150 m.

9 **5.3 Bottom-depth effect during different climate stages**

10 In order to better decipher the history of $\delta^{15}\text{N}_{\text{NO}_3}$ in the bottom euphotic zone of the
11 water column, we synthesized almost all available $\delta^{15}\text{N}_{\text{bulk}}$ of sediment cores reported
12 for the AS (see Figs. 1a and 1b for locations). Similar to modern surface sediments,
13 northern and southern groups were defined by the contour line of $25 \mu\text{mol kg}^{-1}$ DO.
14 ~~The data from core MD-04-2876 is abandoned since the relatively low $\delta^{15}\text{N}_{\text{bulk}}$ might~~
15 ~~be influenced by terrigenous inputs (Pichevin et al., 2007).~~ To keep data consistency
16 in temporal scale, we focused on the last 35 ka (Fig. 8a). Unfortunately, data points
17 were less in 0~6 ka and there were only three sediment cores in southern AS,
18 SK177/11 in this study and NIOP 905 and SO42-74KL in previous studies.

19 As shown in Fig. 8a, the original $\delta^{15}\text{N}_{\text{bulk}}$ from the northern (~~purple-gray~~ dots) and
20 southern AS (green, blue and red curves) are scattering in a wide range from 4.5 to
21 10.5‰ over entire 35 ka. The pink dots are for the data from core MD-04-2876,
22 which is peculiar since the relatively low $\delta^{15}\text{N}_{\text{bulk}}$ values deviated from all other
23 reports in the northern AS. Pichevin et al. (2007) excluded the influences from
24 incomplete nitrate utilization and terrestrial input, thus, we still include this core in
25 our statistical analyses. As f
26 For the southern cores, the temporal variations of $\delta^{15}\text{N}_{\text{bulk}}$
27 in core SK177/11 and NIOP 905 (red and blue) had a very similar trend distributing at
28 the lower bound of whole dataset. The mean $\delta^{15}\text{N}_{\text{bulk}}$ values for SK177/11 and NIOP
905 during glacial period were almost identical, and the deviation in the Holocene

1 was as small as 0.7‰. By contrast, the temporal pattern for $\delta^{15}\text{N}_{\text{bulk}}$ of core
2 SO42-74KL (green) resembles that of NIOP 905 yet with an enrichment in ^{15}N by ~2‰
3 for the entire period. The core SO42-74KL is retrieved from depth of 3212 m, which
4 the deepest among the three cores in southern AS, the positive offset is apparently
5 caused by the bottom depth effect. Thus inference should be made with caution when
6 compare sediment cores from different depths.

7 Below we consider two time spans, 0~11 ka (Holocene) and 19~35 ka (glacial), to
8 examine the bottom-depth effect at different climate stages. We ignore transgression
9 period, which is shorter with more variable in $\delta^{15}\text{N}_{\text{bulk}}$, to avoid bias caused by dating
10 uncertainties in different studies. Also, we will discuss the peculiar patterns for 0~6 ka
11 later. The mean and standard deviation of reported $\delta^{15}\text{N}_{\text{bulk}}$ values for the specific time
12 span were plotted against the corresponding depth of the core. Accordingly, we
13 obtained the correction factors for glacial and early Holocene, respectively, for
14 northern and southern AS (Figs. 8b and 8c). Since only 35 ka was applied in this
15 practice, the long term alteration (Reichert et al., 1998; Altabet et al., 1999) is ignored.
16 The regression curves for modern day (dashed lines) were plotted for comparison.

17 The difference among regressions of three climate stages in northern AS (Table 2) is
18 not significant (~~0.550.41~~ $\times 10^{-3} \text{ km}^{-1}$ to ~~0.700.60~~ $\times 10^{-3} \text{ km}^{-1}$); however, the regression
19 slopes for northern AS are significantly lower compared with those obtained from the
20 southern AS for all climate states. This might indicate the oxygen content in the
21 northern AS is always lower resulting in a lower degree of alteration of $\delta^{15}\text{N}_{\text{bulk}}$. On
22 the other hand, we may not exclude the effect by sedimentation rate changes over
23 these two stages, which also affect the oxygen exposure time; unfortunately,
24 insufficient sedimentation rate data in the northern AS in previous reports prevents us
25 to implement further analysis.

26 As for the southern AS, correction factors are always higher than that in northern AS.
27 The overall spatial temporal patterns are consistent with the oxygen distribution in
28 the Arabian Sea (Olson et al., 1993; Morrison et al., 1999; Pichevin et al., 2007) and
29 agree with the view that DO concentration was the dominant factor for organic matter

1 preservation (Aller, 2001; Zonneveld et al., 2010). Meanwhile, the regression slopes
2 remained high from $0.76 \times 10^{-3} \text{ km}^{-1}$ to $1.01 \times 10^{-3} \text{ km}^{-1}$ over different climate stages in
3 the southern AS suggesting that environmental situations, thus the correction factor,
4 change less relative to that in the northern AS. For SK177/11, sedimentation rate in
5 Holocene is 2-fold higher comparing to that in glacial period; however, the influence
6 caused by sedimentation rate changes is likely not significant enough to alter the
7 regression slopes for the southern AS basing on the small changes in slope (0.93×10^{-3}
8 km^{-1} and $1.01 \times 10^{-3} \text{ km}^{-1}$).

9 **5.4 Insights from temporal changes in geographic $\delta^{15}\text{N}_{\text{bulk}}$ distribution**

10 Based on the earlier comparison among $\delta^{15}\text{N}_{\text{NO}_3}$, sinking particles and surface
11 sediments, we recognized the regression intercept is representative of the nitrogen
12 isotope of nitrate source at depth of 100 m. Therefore, the regression-derived
13 intercepts given in Table 2 can be used to infer the $\delta^{15}\text{N}_{\text{NO}_3}$ source at different climate
14 stages, while the slopes can be used as correction factors to eliminate the positive shift
15 in $\delta^{15}\text{N}_{\text{bulk}}$ caused by bottom depth; by doing this, we can get the original signal of
16 $\delta^{15}\text{N}_{\text{bulk}}$ prior to alteration. We applied the correction factor to be equal to ((bottom
17 depth – 100 m) \times slope), ignoring the sea level changes during the different climate
18 stages.

19 Noticeably, the regression intercepts for both northern and southern AS are higher in
20 the Holocene compared to that in glacial period indicating the intensified isotopic
21 enrichment in $\delta^{15}\text{N}_{\text{NO}_3}$ in entire AS in Holocene. Such increment is almost the same to
22 be $\sim 1.7\text{‰}$, which is similar to the increase in Eastern Tropical North Pacific but
23 slightly smaller than that in the Eastern Tropical South Pacific (Galbraith et al., 2012).
24 The 120 m sea level increase, which may induce only 0.1‰ offset, cannot be the
25 reason for such a significant increase of average $\delta^{15}\text{N}_{\text{bulk}}$ during the Holocene.
26 Moreover, deviations between northern and southern AS at respective climate stage
27 are almost identical (0.8‰ for Holocene and 1.0‰ for glacial) indicating a
28 synchronous shift in the relative intensity of denitrification and N_2 -fixation over the

1 basin to keep such constant latitudinal gradient of subsurface $\delta^{15}\text{N}_{\text{NO}_3}$.

2 The ~~pattern of water exchange~~intermediate water formation near the polar region
3 controls the oxygen supply ~~for to the~~ intermediate water and thus the extent of
4 denitrification on global scale and, thus, the stoichiometry of nutrient source to
5 euphotic zone, ~~resulting in the feedback of nitrogen fixation to denitrification in~~
6 ~~global scale~~ (Galbraith et al., 2004). Lower glacial-stage sea surface temperature may
7 increase oxygen solubility, while stronger winds in high-latitude regions enhance the
8 rate of thermocline ventilation. The resultant colder, rapidly flushed thermocline
9 lessened the spatial extent of denitrification and, consequently, N fixation (Galbraith
10 et al., 2004). Therefore, such a basin wide synchronous increase in $\delta^{15}\text{N}_{\text{bulk}}$ is likely a
11 global control. The lower intercepts in glacial time (4.3‰ for south and 5.3‰ for
12 north), which are similar to the global mean $\delta^{15}\text{N}_{\text{NO}_3}$ ~~in modern day~~ (4.5~5‰, Sigman
13 et al., 1997), illustrates a better ventilation of intermediate water during glacial time in
14 the Arabian Sea (Pichevin et al., 2007). ~~Moreover~~In fact, the AAIW ~~was prevented~~
15 ~~from penetrate further~~ northward at over 5°-N in present day ~~out of the southern part~~
16 ~~in modern AS~~ and even during the late Holocene (You, 1998; Pichevin et al., 2007).
17 Since the $\delta^{13}\text{C}$ of autochthonous particulate organic carbon is negatively correlated to
18 [CO₂ (aq)] in euphotic zone (Rau et al., 1991), the sharp decrease of $\delta^{13}\text{C}_{\text{TOC}}$ in
19 SK177/11 at the start of deglaciation (Fig. 3b) may infer the timing of a rapid
20 accumulation of dissolved inorganic carbon driven by the shrinking of oxygenated
21 intermediate water (Pichevin et al., 2007) or enhanced monsoon-driven upwelling
22 (Ganeshram et al., 2000); both facilitate the promotion of denitrification. Nevertheless,
23 the mirror image between $\delta^{15}\text{N}$ and $\delta^{13}\text{C}_{\text{TOC}}$ profiles revealed their intimate relation;
24 of which, the variability was attributable to the change of physical processes.
25 ~~indicate a rapid change of physical circulation had occurred in characteristics of the~~
26 ~~intermediate water flowing into the AS.~~

27 The intercepts of the northern AS increase continuously from 5.3 to 8.1 from glacial
28 through modern day indicating the strengthened intensity of denitrification relative to
29 nitrogen fixation in the northern AS (Altabet, 2007). When we take a close look at the

1 temporal pattern of corrected $\delta^{15}\text{N}_{\text{bulk}}$ for long cores (Fig. 9), we can see an amplified
2 deviation since 6 ka, during which $\delta^{15}\text{N}_{\text{bulk}}$ increases continuously in the northern AS,
3 whereas it decreases in the southern AS. (Note that the northern most core,
4 MD-04-2876, also followed the increasing trend in recent 6 ka even though its
5 $\delta^{15}\text{N}_{\text{bulk}}$ values deviated from all other cores.) Such opposite trends indicate that the
6 controlling factors on nitrogen cycle in northern AS were different from that in the
7 southern AS, which means localized enhancement in specific process had occurred.

8 Besides the oxygen supply to the intermediate water, the intensity of water column
9 denitrification varies with primary productivity (Altabet, 2006; Naqvi et al., 2006).
10 Strong summer monsoon and winter monsoon drive upwelling or convective mixing
11 to enhance the primary productivity, which in turn intensify denitrification (Altabet et
12 al., 2002; Ganeshram et al., 2002). However, it was reported also that primary
13 productivity did not correlate well with water column denitrification underneath
14 during the Holocene in some parts of the northern AS (Banakar et al., 2005 and
15 references therein). Regardless of the declining summer monsoon strength since 5500
16 ka (Hong et al., 2003), the primary productivity in northern AS seem to be increased.
17 Similar to the patterns observed for TOC and TN in this study, productivity indicators
18 (TOC and Ba/Al ratios) reported by Rao et al. (2010) in the core SK148/4 located
19 nearby our SK177/11 also increased gradually since the Holocene. Incomplete nitrate
20 consumption can hardly explain the decreasing pattern for all three cores in the
21 southern AS where upwelling intensity is much less relative to that in the north.
22 Moreover, lower TOC/TN ratios observed in Holocene in SK177/11 as mentioned
23 earlier rules out the influence from terrestrial organic input. Therefore, a spatial
24 coupling of denitrification-dependent N_2 fixation is the more plausible cause of the
25 decreasing $\delta^{15}\text{N}_{\text{bulk}}$ pattern (Deutsch et al., 2007).

26 We suggested that intensified supply of excess phosphorous (phosphorus in
27 stoichiometric excess of fixed nitrogen) toward the southern AS to stimulate N_2
28 fixation, subsequently responsible for the decreasing $\delta^{15}\text{N}_{\text{bulk}}$ pattern in the southern
29 basin. The intensification in excess phosphorous supply can be driven by enhanced

1 upwelling or intensified subsurface water column denitrification or both. According to
2 the increasing pattern in $\delta^{15}\text{N}_{\text{bulk}}$ and primary productivity in the northern AS,
3 synergetic processes are suggested. The upwelled water in northern AS basin brings
4 up low N/P water to surface for non-diazotrophs to uptake. If we assume complete
5 consumption, the remaining excess phosphorous after complete consumption will be
6 transported toward south by clockwise surface circulation and advection, therefore,
7 N_2 -fixation in the southern AS acts as feedback to balance denitrification changes in
8 the northern AS. This phenomenon is similar to the illustration for the spatial coupling
9 of nitrogen inputs and losses in the Pacific Ocean proposed by Deutsch et al. (2007).
10 Why such forcing to expand the N-S deviation had not occurred before 6 ka warrants
11 more studies.

12

13 **6 Conclusions**

14 The available data showed that values of $\delta^{15}\text{N}_{\text{NO}_3}$ at the bottom of euphotic zone
15 (~150 m) were similar to $\delta^{15}\text{N}_{\text{SP}}$ implying that the source of nutrients for sinking
16 particulate organic matter was largely derived from the depth at around 150 m. Values
17 of sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ were obviously higher than $\delta^{15}\text{N}_{\text{SP}}$ in surrounding areas
18 suggesting such shift of sedimentary $\delta^{15}\text{N}_{\text{bulk}}$ occurred after deposition. It is necessary
19 to remove site-specific bias of $\delta^{15}\text{N}_{\text{bulk}}$ values caused by bottom depth to retrieve the
20 original signal before alteration. As a result, the corrected nitrogen isotopic signal in
21 sediments could be representative of the value of $\delta^{15}\text{N}_{\text{NO}_3}$ at the bottom depth of
22 euphotic zone. The bottom-depth effects in the northern AS varies during different
23 climate stages, but the variation is always lower than such effect in the southern AS in
24 general. The modern surface $\delta^{15}\text{N}_{\text{bulk}}$ values can be separated statistically into northern
25 and southern AS groups reflecting a special coupling of denitrification to the north
26 and N_2 -fixation to the south. This phenomenon is supported by the reported modern
27 day N^* distribution. As for historical records, the offset in $\delta^{15}\text{N}_{\text{bulk}}$ between southern
28 and northern AS remained relatively constant (0.8‰ for early Holocene and 1.0‰ for
29 glacial) prior to 6 ka indicating a synchronous shift in the relative intensity of

1 denitrification and N₂-fixation over the basin to keep such constant latitudinal gradient
2 of subsurface $\delta^{15}\text{N}_{\text{NO}_3}$. However, this offset expanded gradually since 6 ka due likely
3 to more localized intensifications in denitrification and N₂-fixation had occurred,
4 respectively, in the northern and southern Arabian Seas. The spatial coupling of
5 nitrogen inputs and losses in the Arabian Sea was proposed; yet, why the driving force
6 did not expand the N-S deviation before 6 ka warrants more studies.

7

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17

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3

1 Table 1. AMS ^{14}C dates of sediment core SK177/11. Radiocarbon ages were
 2 calibrated using CALIB 6.0 program (<http://calib.qub.ac.uk/calib/calib.html>, Reimer
 3 *et al.*, 2009).

Lab code	Depth cm	Dating materials	pMC	Raw ^{14}C age (yr BP)	Calibrated age (yr BP) (1 σ)	$\delta^{13}\text{C}$ (‰)
KIA24386	58	OM	65.58 \pm 0.17	3390 \pm 20	3186 \pm 24	-18.55 \pm 0.04
KIA26327	125	OM	46.65 \pm 0.20	6125 \pm 35	6504 \pm 26	-20.02 \pm 0.10
KIA24387	155	OM	31.38 \pm 0.13	9310 \pm 30	10054 \pm 104	-19.50 \pm 0.08
KIA26328	175	OM	21.96 \pm 0.12	12180 \pm 45	13618 \pm 104	-17.71 \pm 0.18
KIA24388	205	OM	13.94 \pm 0.11	15830 \pm 60	18646 \pm 54	-21.65 \pm 0.15
KIA24389	275	OM	9.81 \pm 0.12	18650 \pm 100(-90)	21774 \pm 194	-18.02 \pm 0.10
KIA26329	355	OM	2.76 \pm 0.06	28830 \pm 180	32857 \pm 207	-19.23 \pm 0.17

4 OM-Organic matter; pMC-Percent modern

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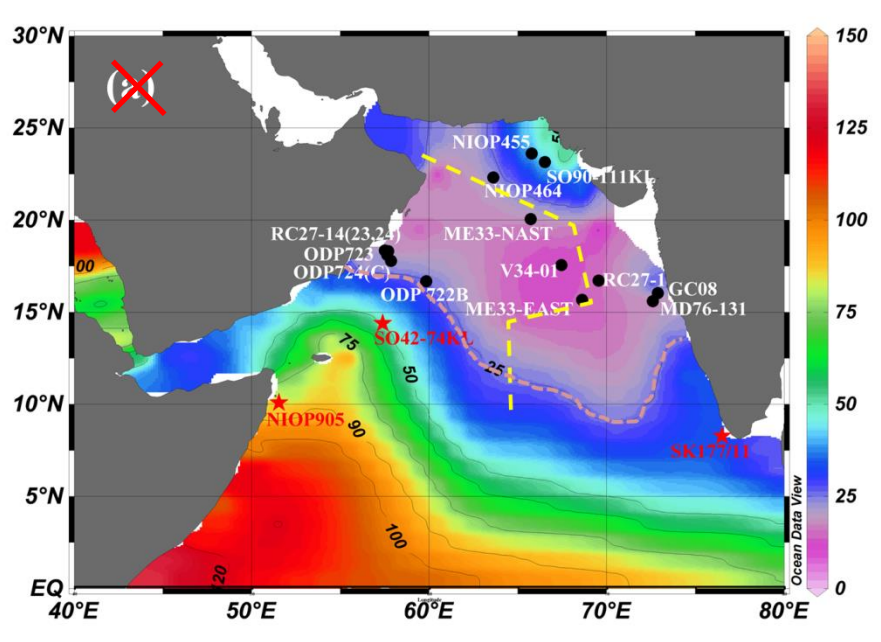
1 Table 2. Linear equations of bottom-depth effect during different climate stages

Location	Northern AS	Southern AS
Modern	$\delta^{15}\text{N} = 0.55 (\pm 0.08) \times 10^{-3} \times$ Depth + 8.1 (± 0.2) ($R^2 = 0.40$, n = 78, P < 0.0001)	$\delta^{15}\text{N} = 0.76 (\pm 0.14) \times 10^{-3} \times$ Depth + 6.0 (± 0.3) ($R^2 = 0.66$, n = 18, P < 0.0001)
Holocene	$\delta^{15}\text{N} = 0.7041 (\pm 0.20) \times 10^{-3} \times$ Depth + 6.79 (± 0.3) ($R^2 = 0.6132$, n = 165, P = 0.0067295)	$\delta^{15}\text{N} = 0.93 (\pm 0.06) \times 10^{-3} \times$ Depth + 5.761 (± 0.1) ($R^2 = 1.000999$, n = 3, P = 0.0152420)
Glacial	$\delta^{15}\text{N} = 0.640 (\pm 0.20) \times 10^{-3} \times$ Depth + 5.23 (± 0.3) ($R^2 = 0.6852$, n = 165, P = 0.001325)	$\delta^{15}\text{N} = 1.01 (\pm 0.31) \times 10^{-3} \times$ Depth + 4.3 (± 0.7) ($R^2 = 0.91$, n = 3, *P = 0.1899)

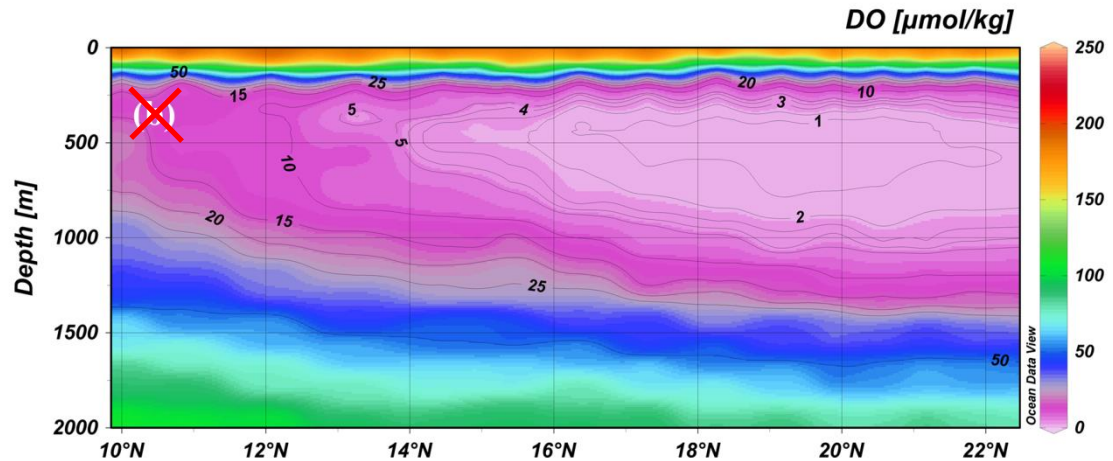
2 *insignificant by P value

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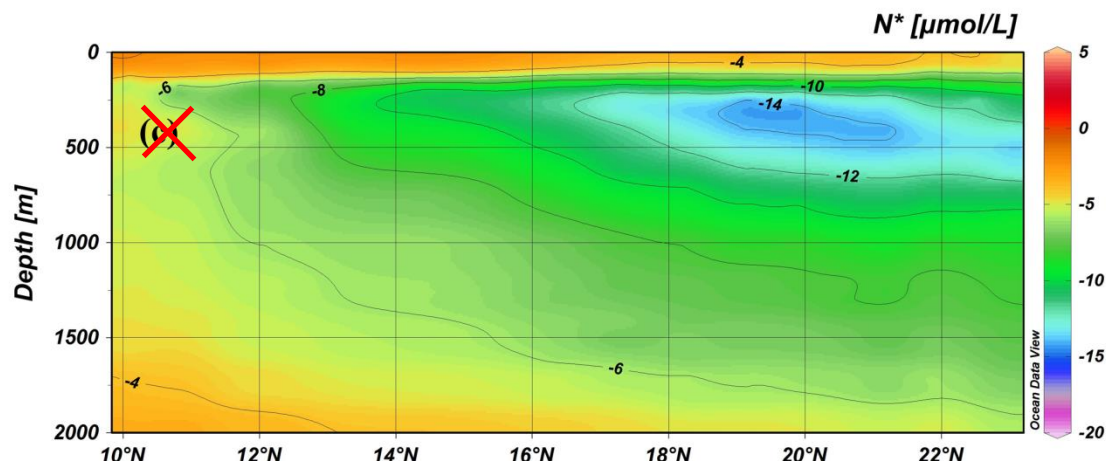
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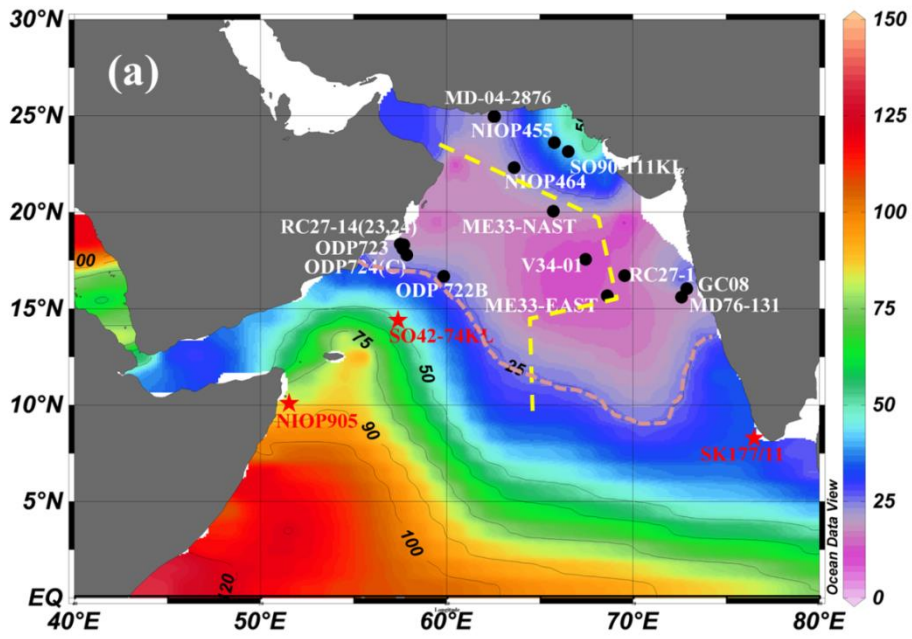


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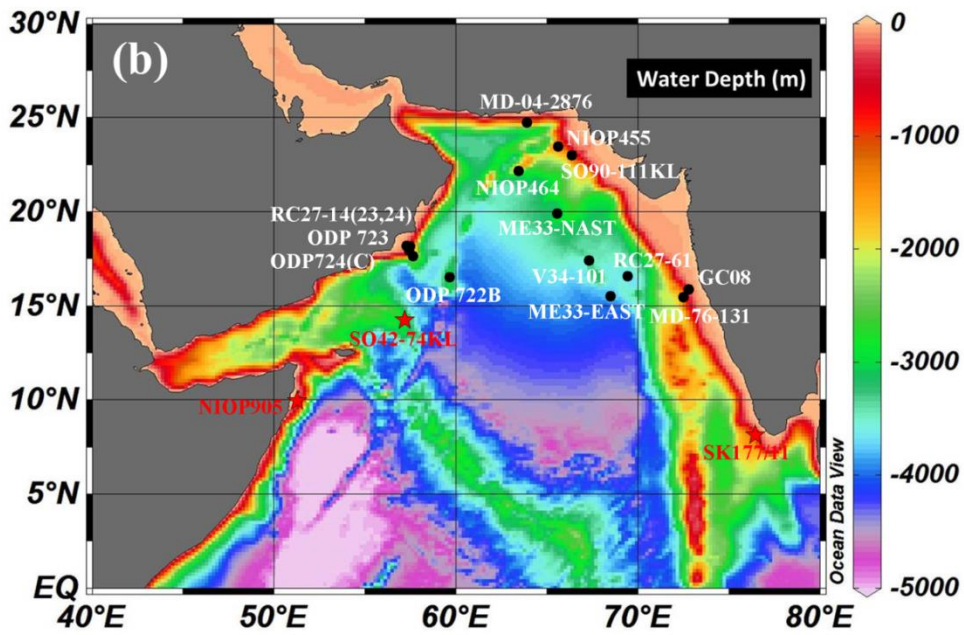


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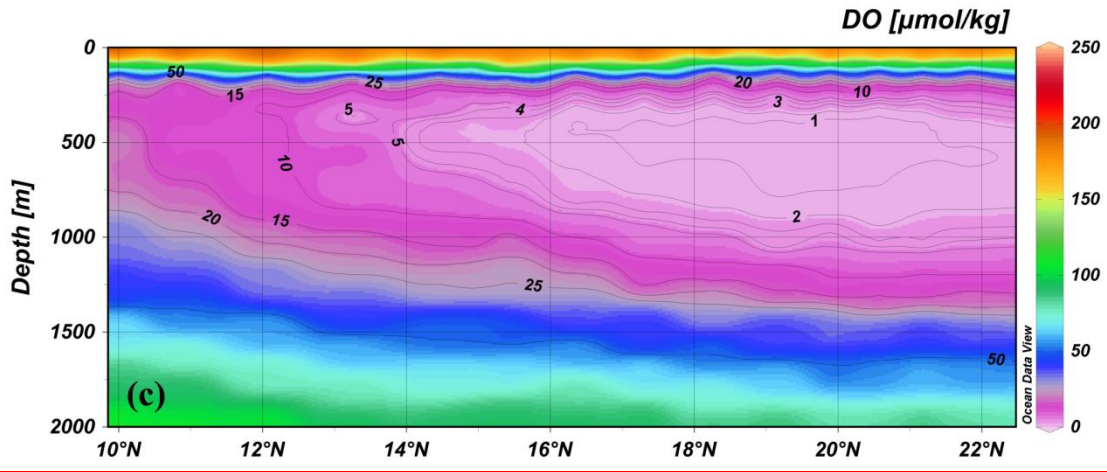


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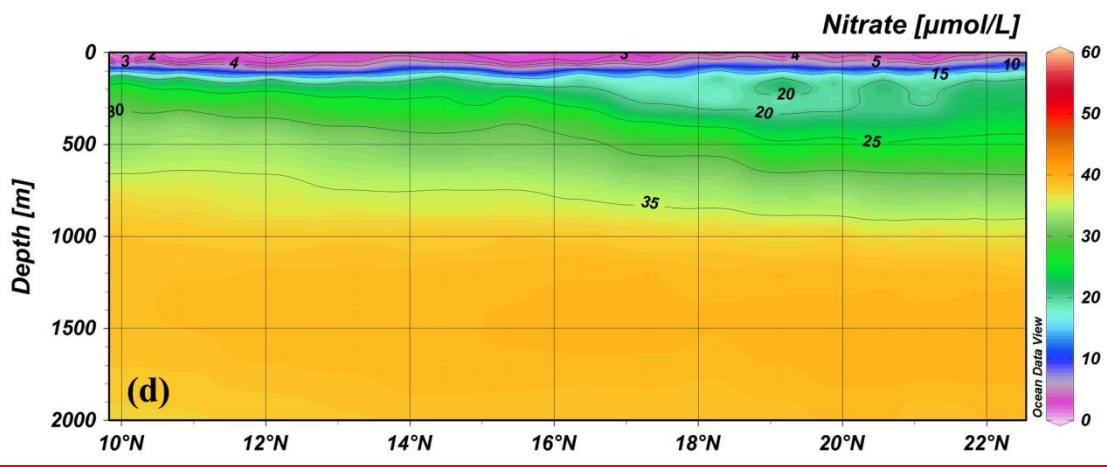


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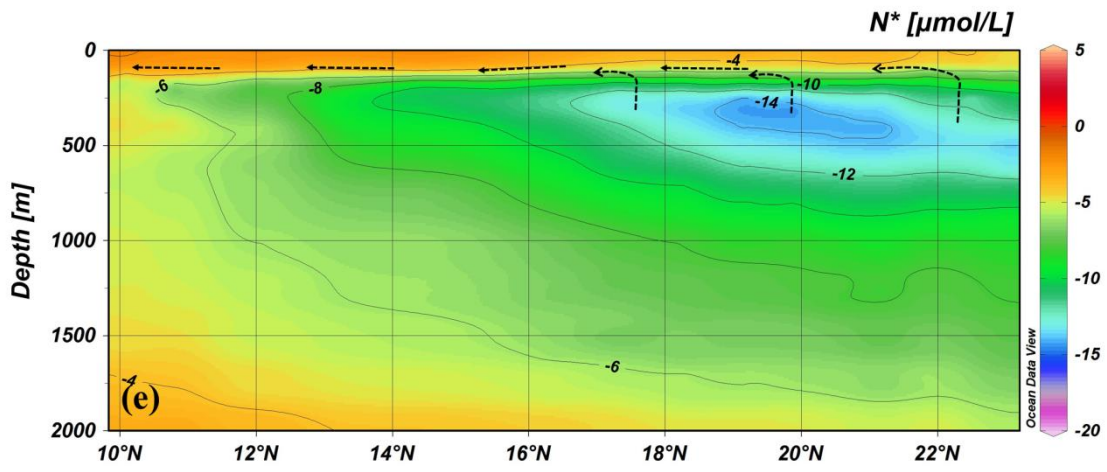
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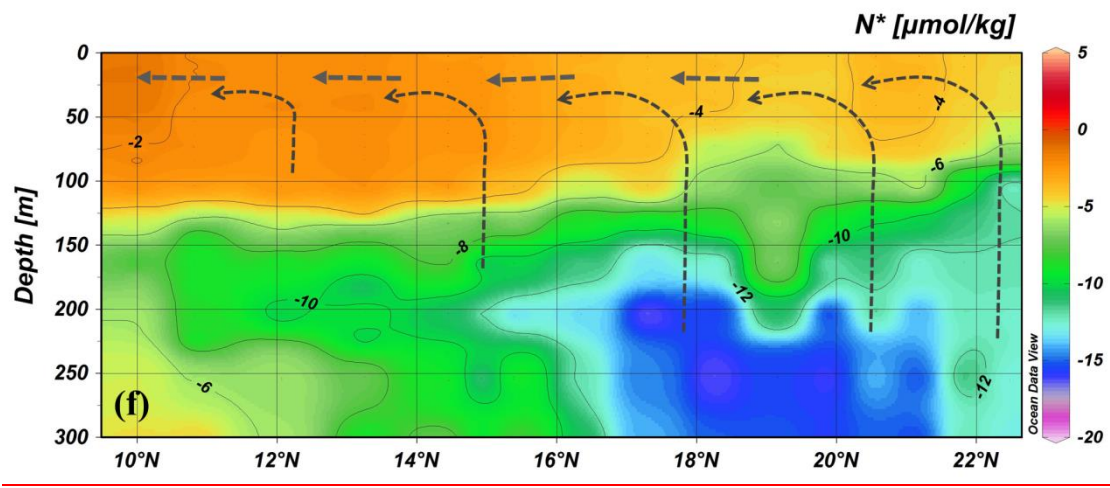


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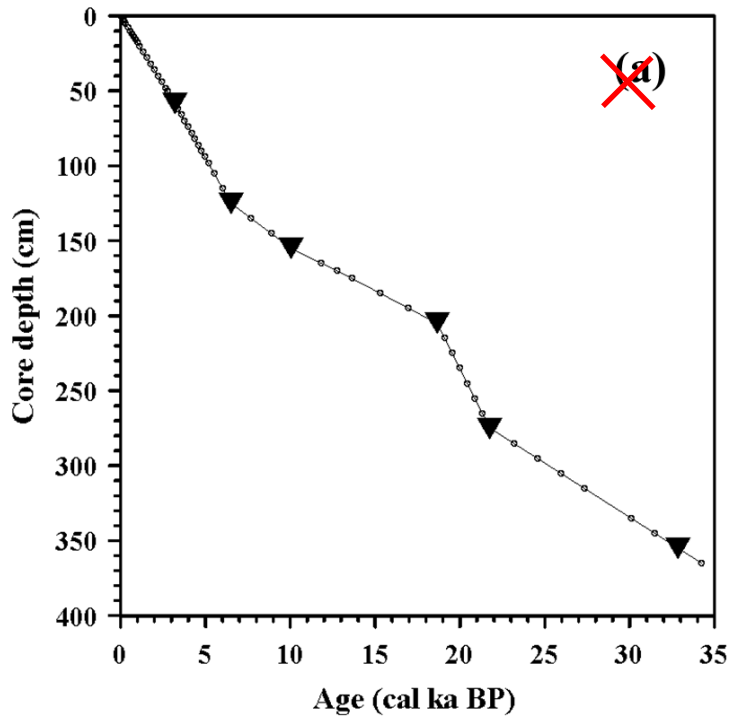




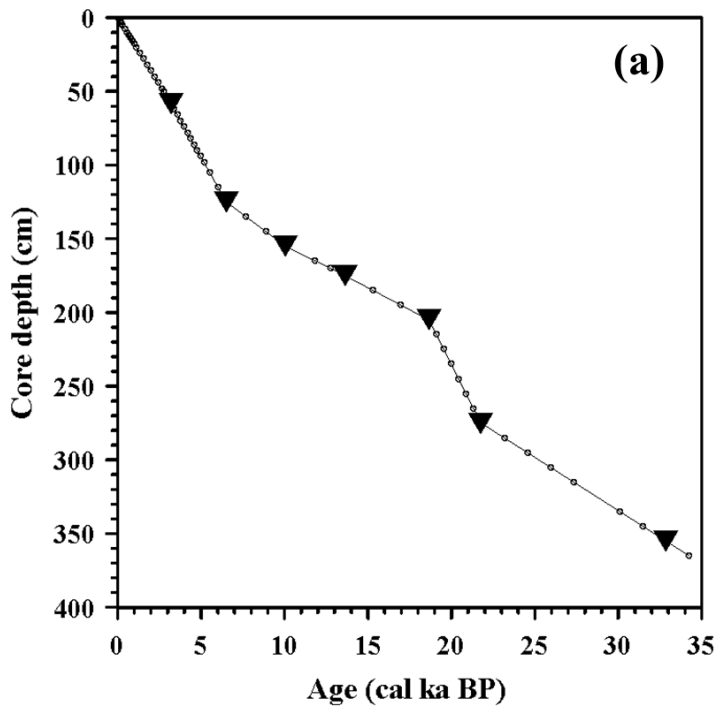
1 Figure 1. (a) Map of the Arabian Sea. Dissolved oxygen (DO) concentration at 150 m
2 (World Ocean Atlas 09) was shown in color contour. Southern (★) and northern (●)
3 categories of available cores and SK177/11 in this study were defined by DO of 25
4 $\mu\text{mol kg}^{-1}$ (see text, purple dash curve). (b) Bathymetric map superimposed by core
5 locations; and (c), (d) and (e) are DO, nitrate and N* transects (yellow dashed line in
6 (a), online data was originated from cruises of JGOFS in 1995), respectively, for
7 upper 2000 m. (f) N* transect for the upper 300m with arrows revealing the flow
8 direction. In (a), the northern cores include core MD-04-2876 (828 m, Pichevin et al.,
9 2007), core NIOP455 vs. NIOP464 (1002 m vs. 1470 m, Reichart et al., 1998),
10 SO90-111KL vs. ME33-NAST(775 m vs. 3170 m, Suthhof et al., 2001), ODP724C vs.
11 ME33-EAST (603 m vs. 3820 m, Möbius et al., 2011), RC27-24 vs. RC27-61 (1416
12 m vs. 1893 m, Altabet et al., 1995), ODP723, ODP722(B) vs. V34-101 (808 m, 2028
13 m vs. 3038m, Altabet et al., 1999), RC27-14 vs. RC27-23 (596 m vs. 820 m, Altabet
14 et al., 2002), GC08 (2500 m, Banakar et al., 2005), MD-76-131 (1230 m, Ganeshram
15 et al., 2000); the Southern cores include core SO42-74KL (3212 m, Suthhof et al.,
16 2001), NIOP905 (1586 m, Ivanochko et al., 2005) and SK177/11 (776 m, this study).

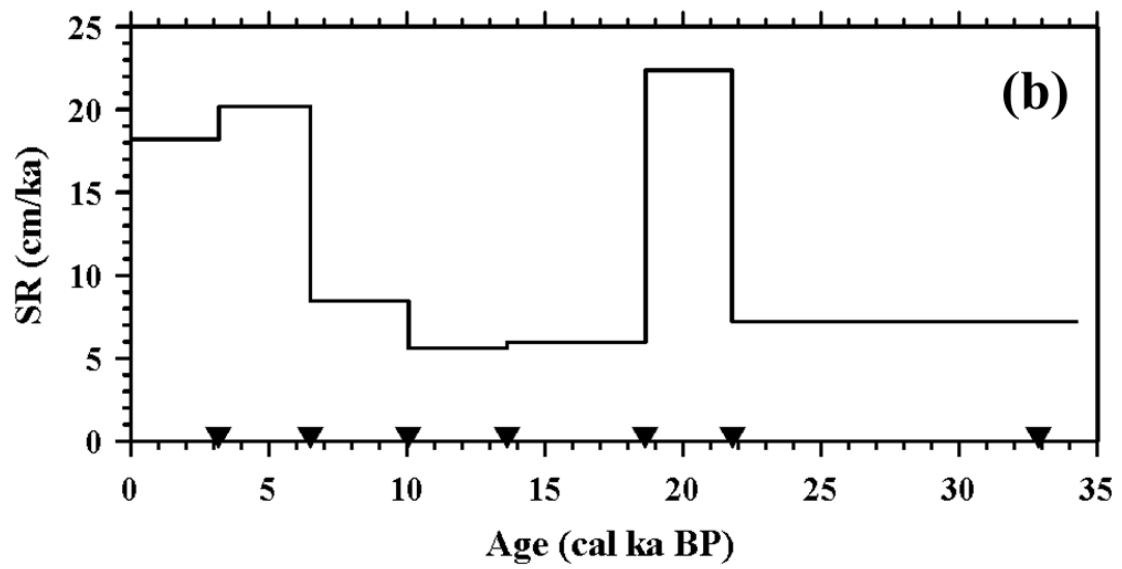
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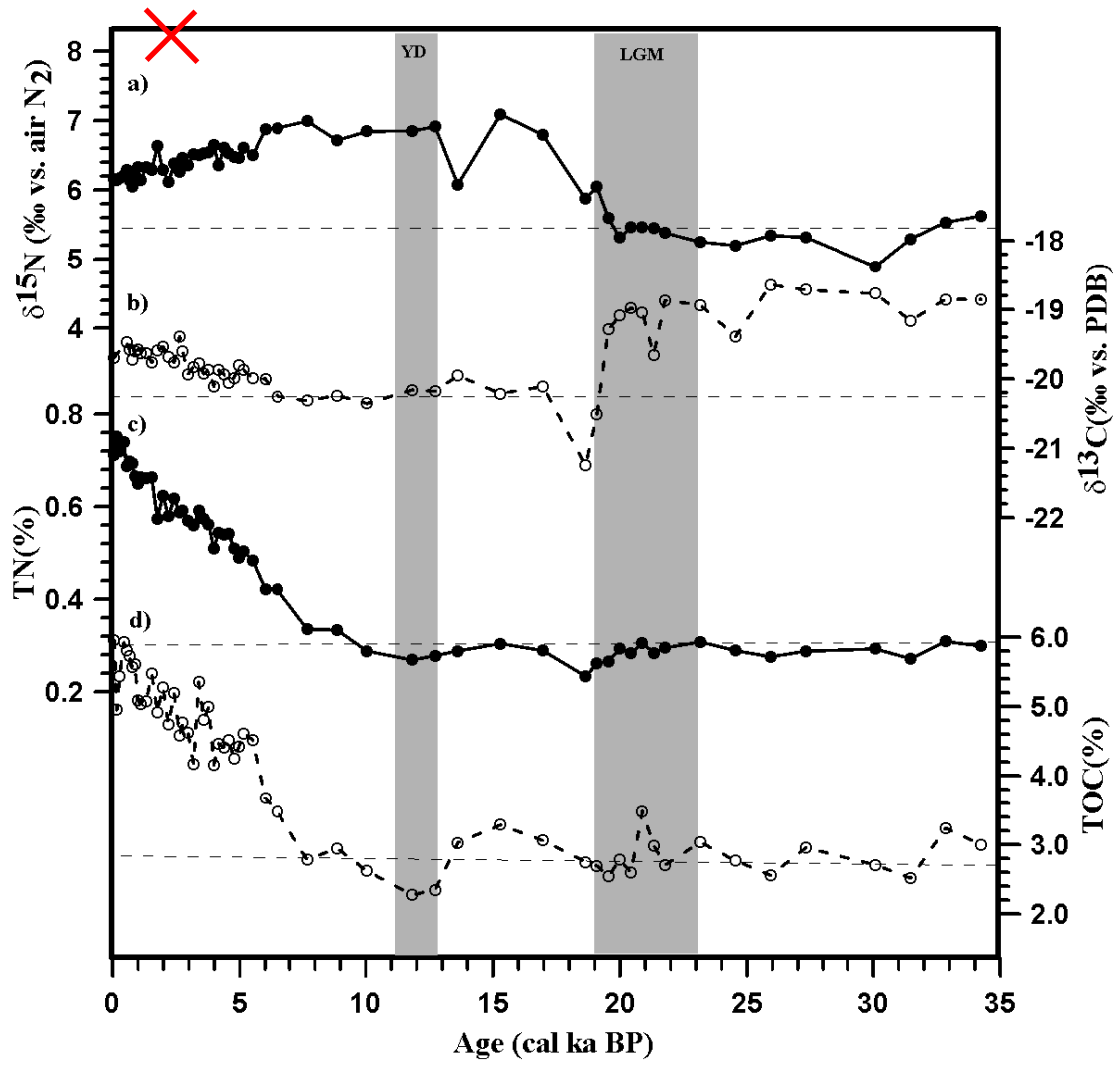


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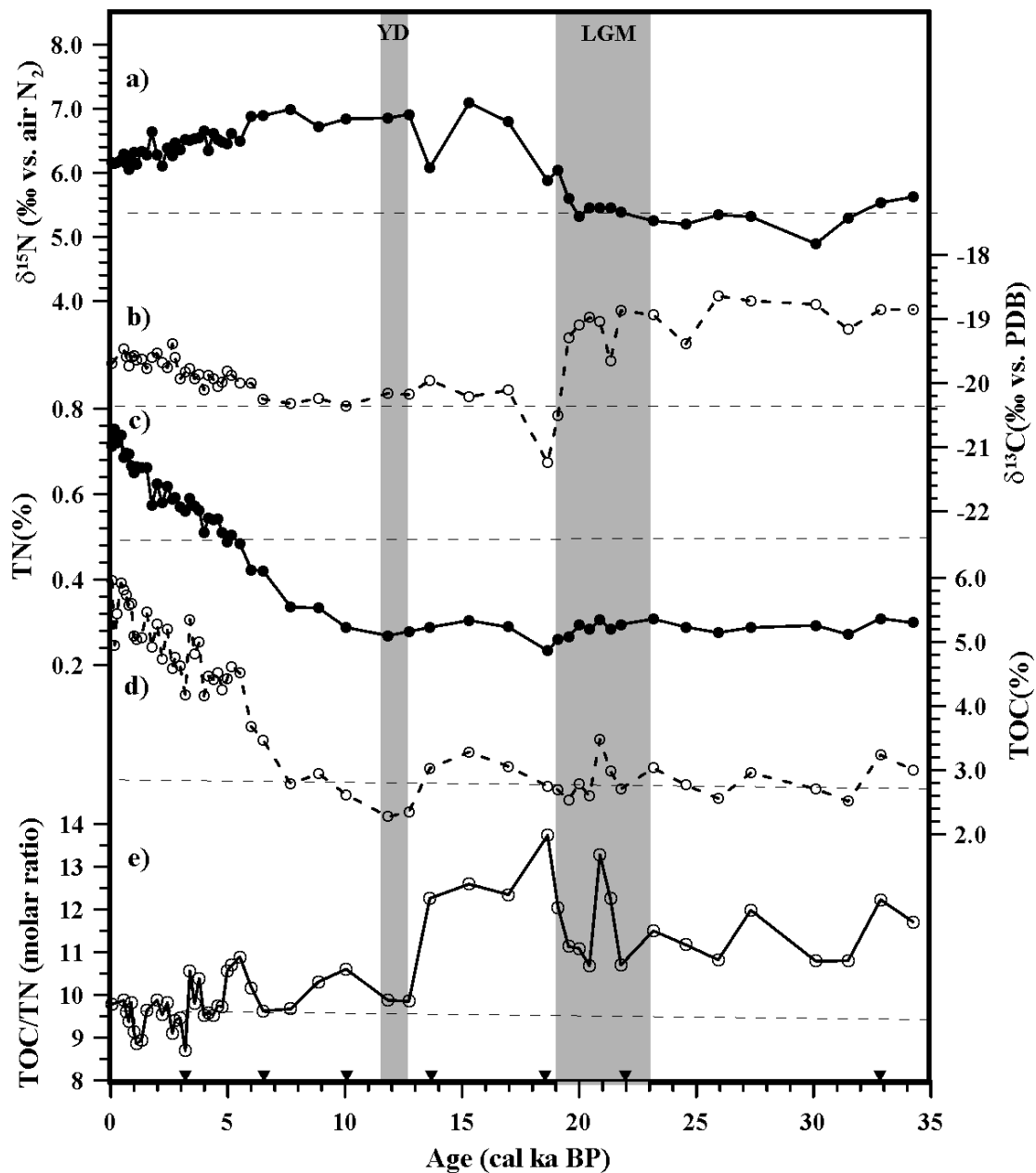
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3 Figure 2. (a) Plot of calendar age against depth; (b) Linear sedimentation rate (▼
 4 indicates the ¹⁴C age controlling points).

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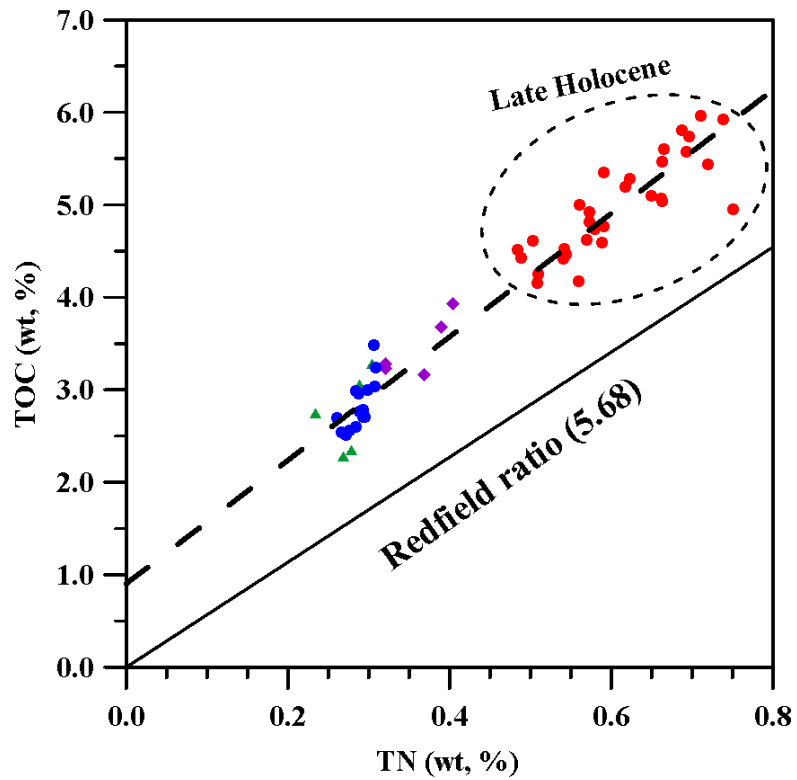
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3 Figure 3. Temporal variations of (a) stable isotopic compositions of bulk nitrogen
 4 ($\delta^{15}\text{N}$), (b) stable isotopic compositions of TOC ($\delta^{13}\text{C}$), (c) contents of total nitrogen
 5 and, (d) total organic carbon and (e) TOC/TN ratio. Horizontal dashed lines are
 6 references for low value periods.



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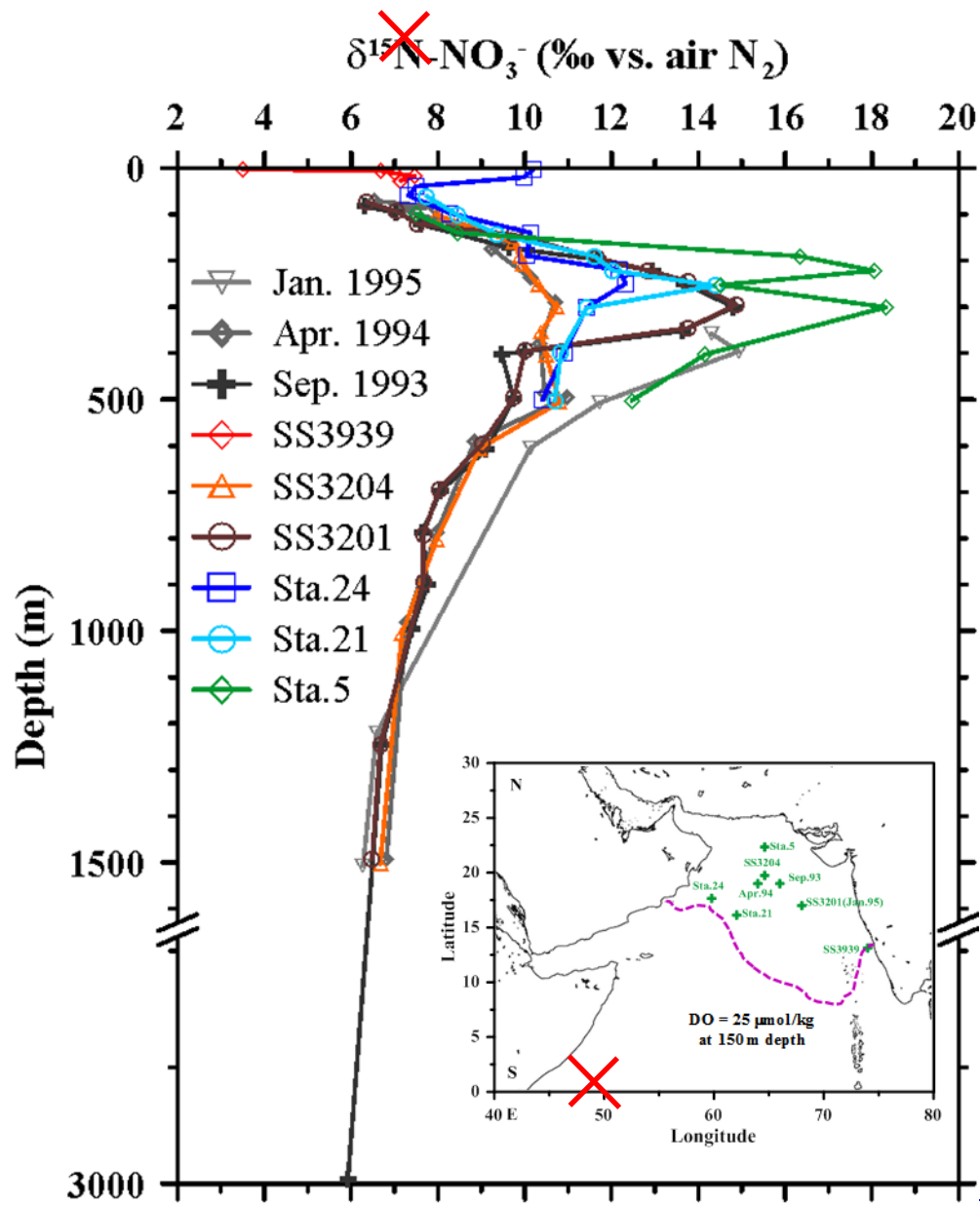
3 Figure 4. Scatter plot of the total organic carbon content against total nitrogen.

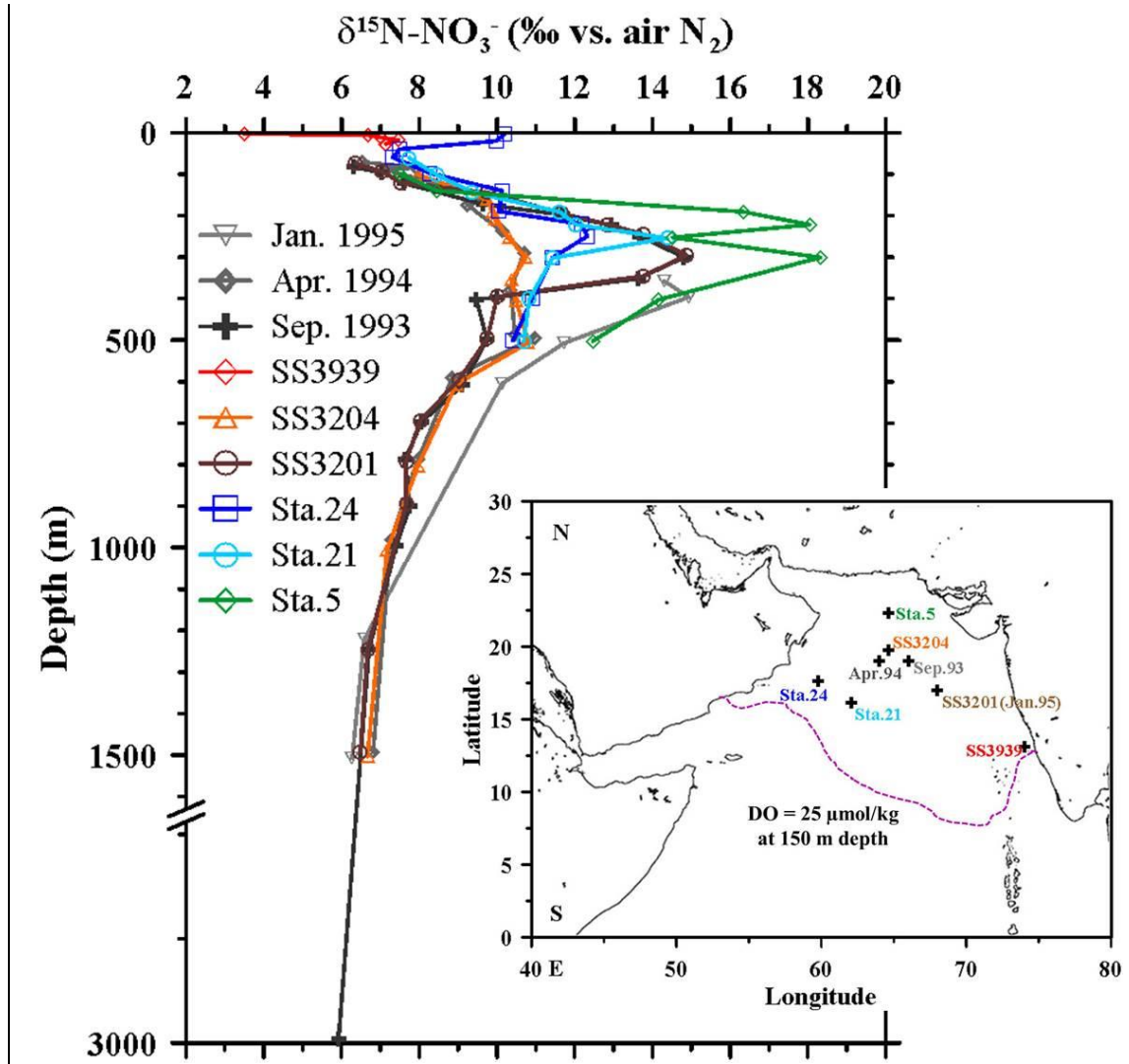
4 Redfield field ratio of 5.68 is shown in line. Bold dashed line stands for regression.

5 Red, purple, green and blue dots represent the late Holocene, early Holocene,

6 deglacial and glacial periods, respectively.

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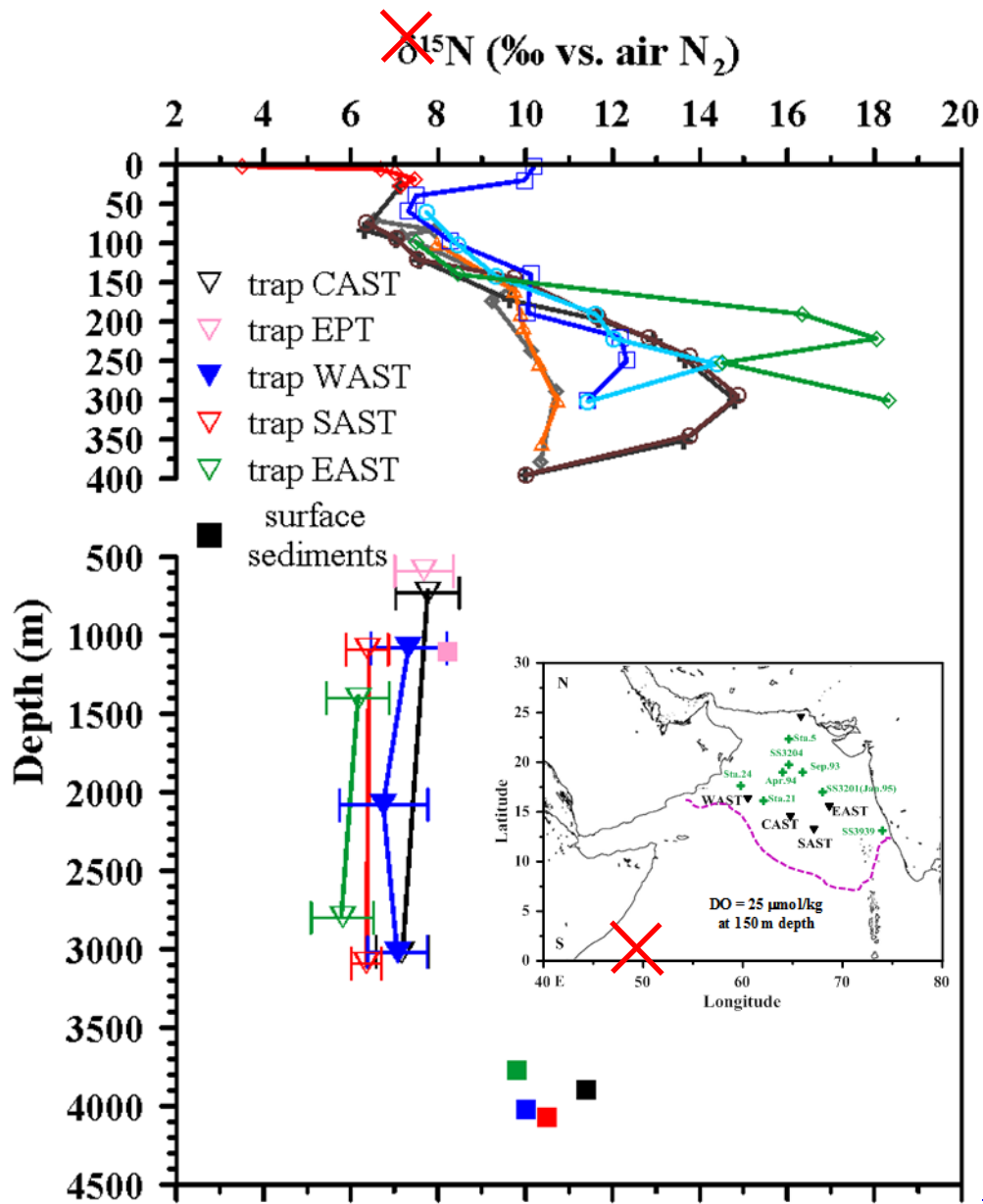
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3 Figure 5. Depth profiles of nitrogen isotope of nitrate ($\delta^{15}\text{N}_{\text{NO}_3}$) in water column (data

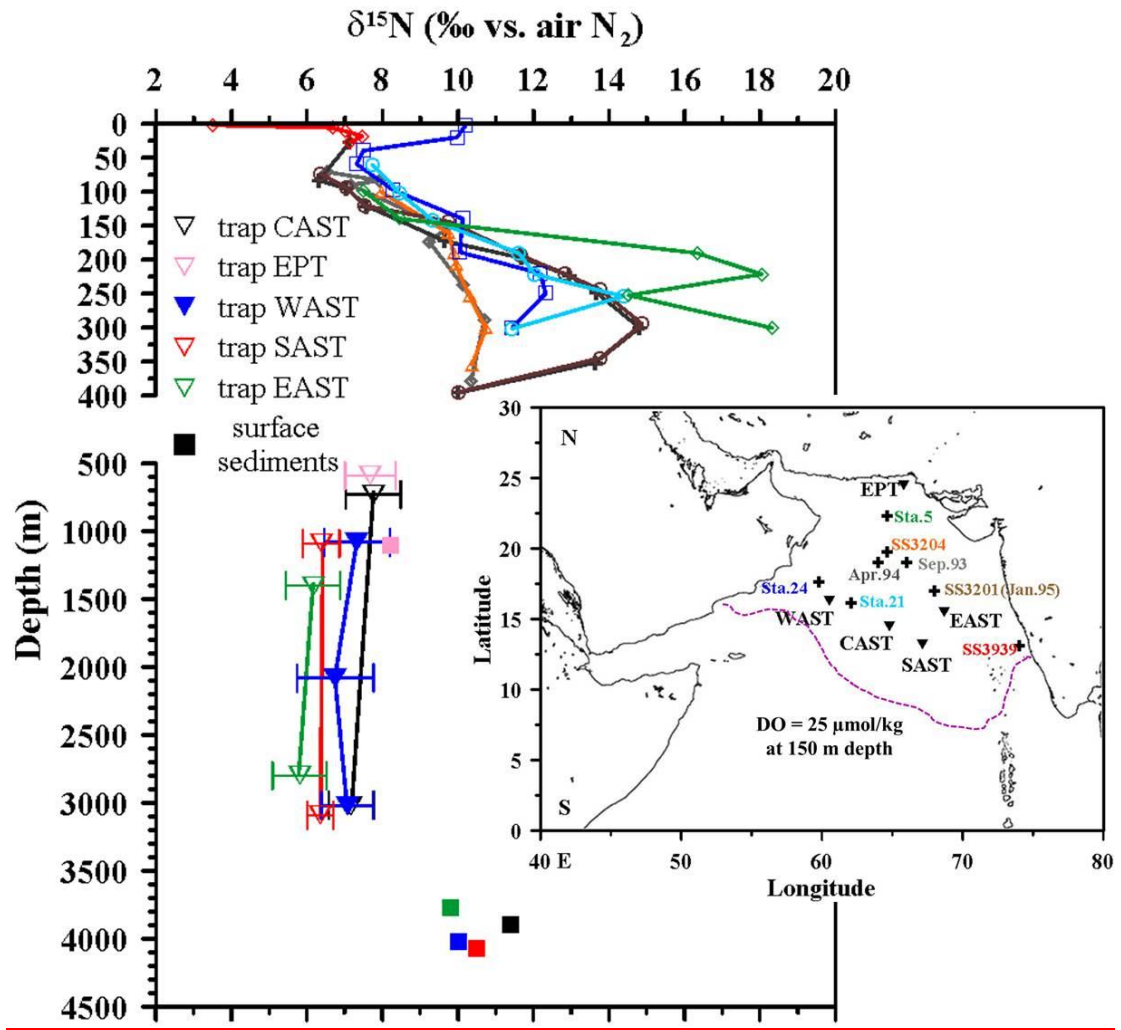
4 without months in mark are all from August and Sta. Jan. 1995 overlaps with Sta.

5 SS3201) (Data digitized from Brandes et al., 1998; Altabet et al., 1999; Naqvi et al.,

6 2006).

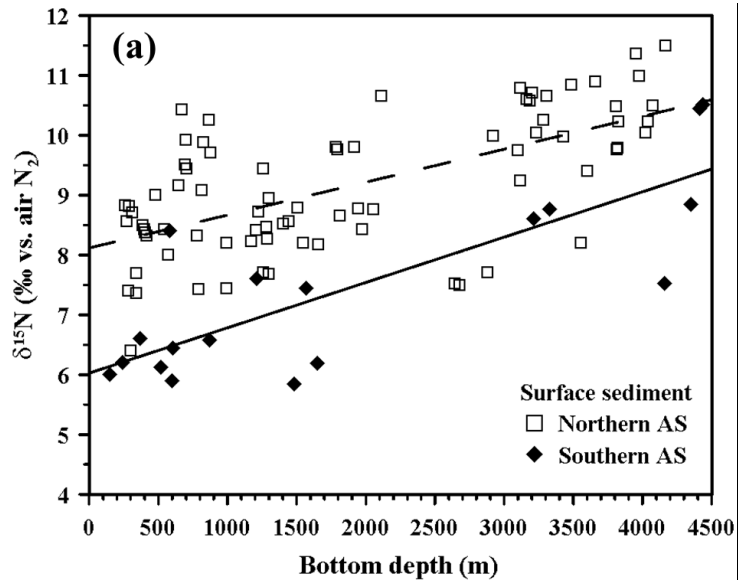


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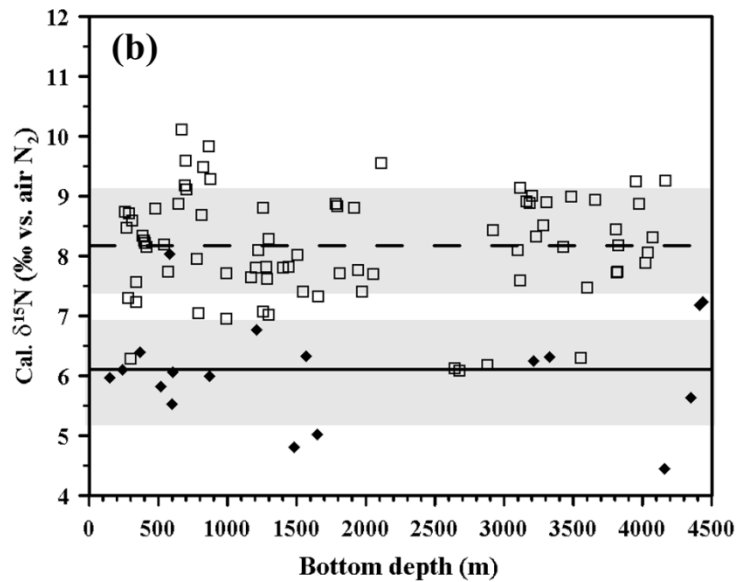


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Figure 6. Vertical profiles for nitrogen isotope of nitrate (green crosses in inserted map), sinking particles (inverse triangles in map) and trap-corresponding surface sediments. Data for sediment traps and surface sediments are from Gaye-Haake et al. (2005). Depth profile of $\delta^{15}\text{N}_{\text{NO}_3}$ follows that in Figure 5.—



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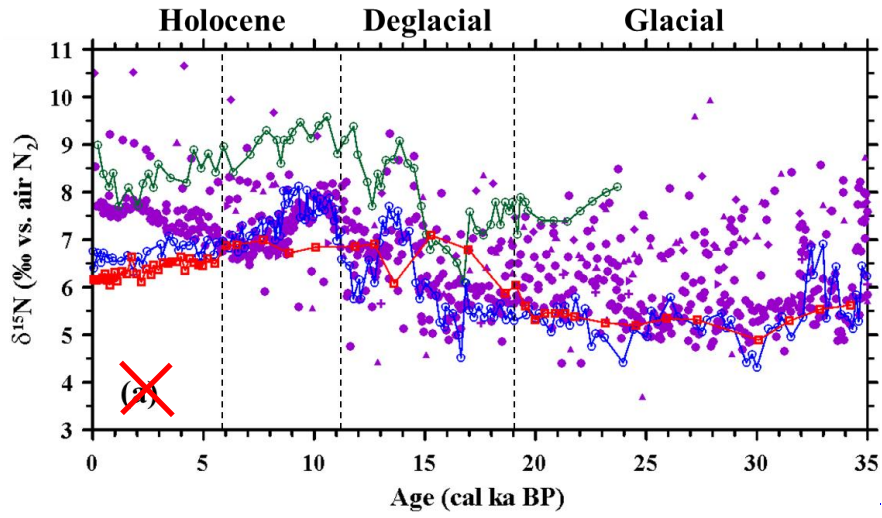


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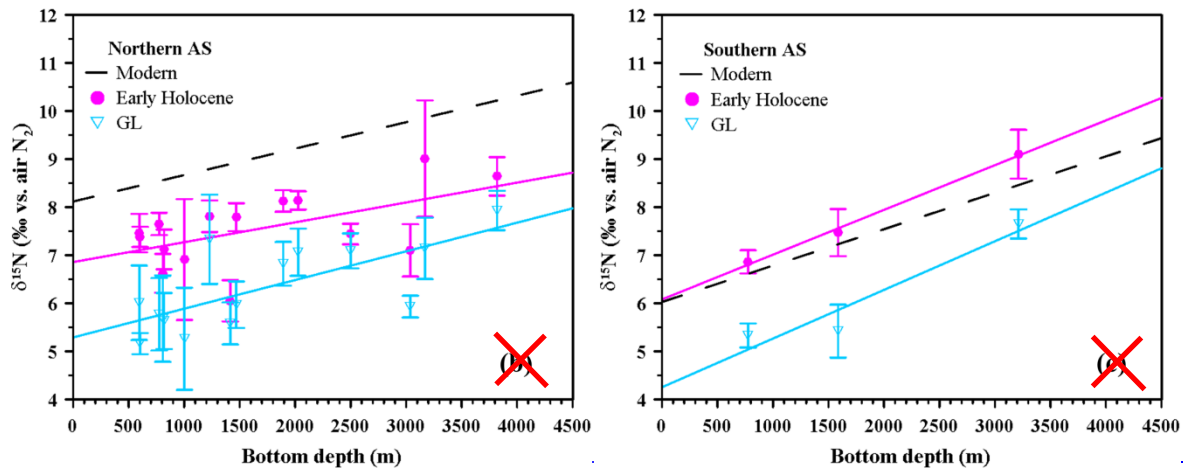
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4 Figure 7. (a) Non-corrected $\delta^{15}\text{N}$ values of modern surface sediments against
 5 corresponding bottom depth in northern and southern Arabian Sea (see text for N-S
 6 boundary). Regression lines were shown in dashed and solid lines, respectively, for
 7 northern and southern AS. (b) Corrected surface sedimentary $\delta^{15}\text{N}$ values against
 8 water depth.

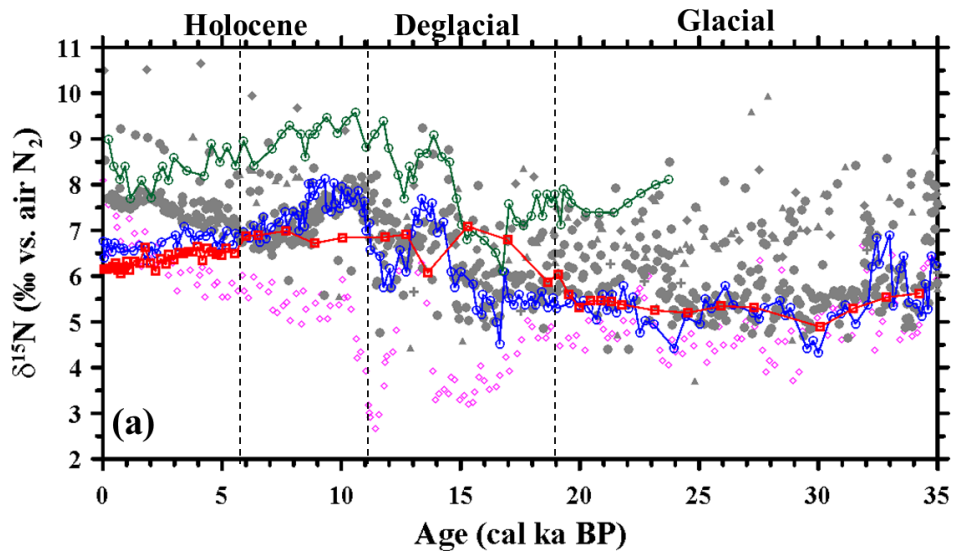
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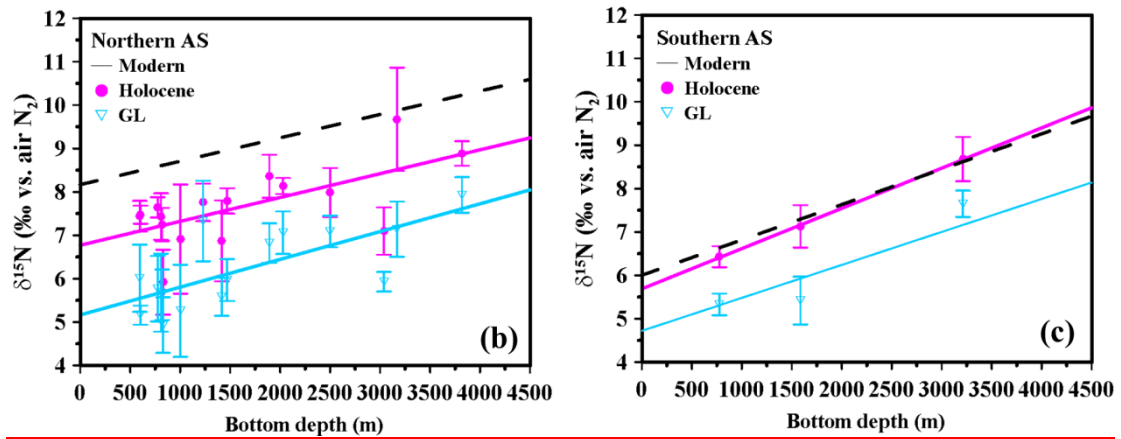


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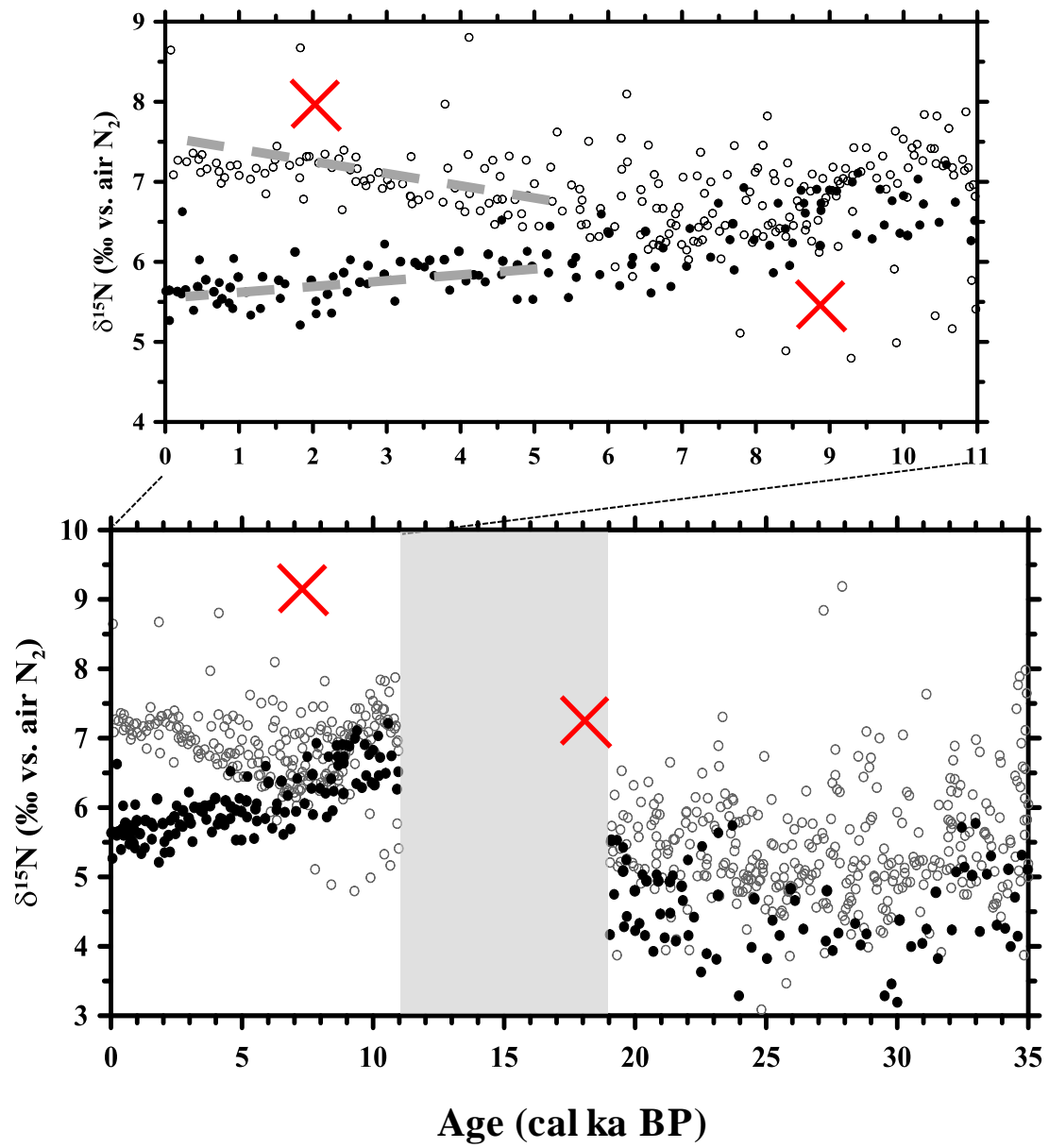




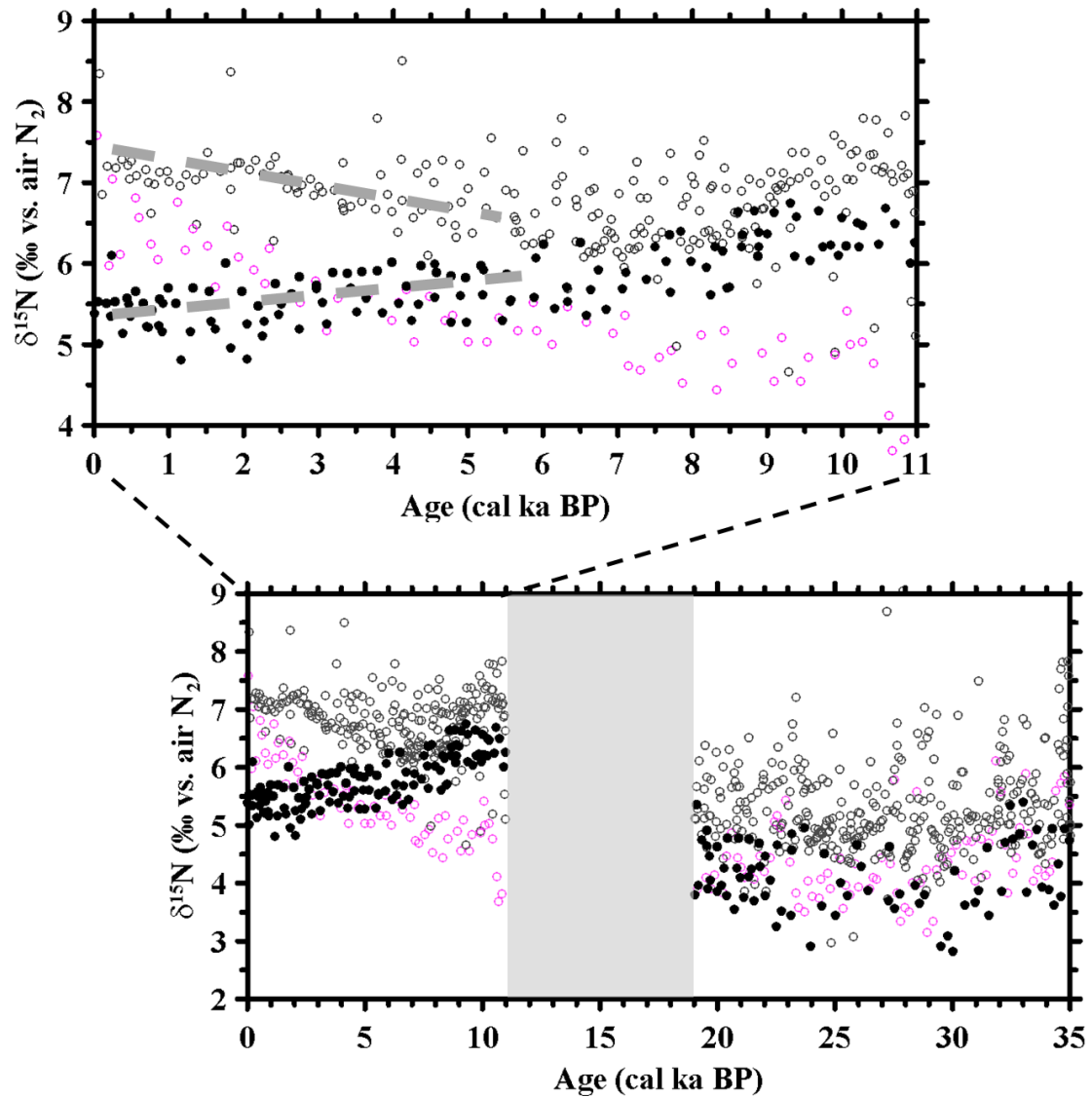
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3 Figure 8. (a) Temporal variations of non-corrected $\delta^{15}\text{N}_{\text{bulk}}$ values of all reported cores
 4 in the AS. Data shown in curves are for cores in the southern Arabian Sea (red for
 5 SK177/11, blue for NIOP 905 and green for SO42-74KL), dots in ~~purple-grey~~
 6 the northern part (pink dots are for core MD-04-2876). Mean values of $\delta^{15}\text{N}$ for fixed
 7 periods against corresponding water depths for cores in (b) northern and (c) southern
 8 Arabian Sea. Pink and indigo blue are for Holocene and glacial periods, respectively.
 9 Error bars represent the standard deviation for mean $\delta^{15}\text{N}_{\text{bulk}}$. The dashed regression
 10 lines for modern surface sediments are shown for reference.—



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Figure 9. Temporal variations of corrected $\delta^{15}\text{N}_{\text{bulk}}$ values of all reported cores in the AS. Gray and black dots are for northern and southern AS, respectively. Pink dots are for core MD-04-2876. The deglacial period is in shadow because non proper equations for bottom-depth effect correction. The upper panel is the blow-up for the Holocene period. The intensified deviation trends since 6 ka were marked by bold dashed lines.