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Biogeosciences
Discussions

Interactive comment on "North Pacific-wide spreading of isotopically heavy nitrogen from intensifieddenitrification during the Bølling/Allerød and post-youngerdryas periods: evidence from the Western Pacific" *by* S. J. Kao et al.

S. J. Kao et al.

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1-A: General comments Kao and coauthors present a new record of bulk sedimentary d15N from an intermediate-depth core in the Okinawa Trough, on the western margin of the northern subtropical Pacific gyre. The record shows little variability overall, but displays two clear millennial-timescale excursions during the latter half of the deglaciation that appear similar to many other sites in the North Pacific. They follow the existing paradigm, interpreting the deglacial excursions as reflecting changes in denitrification

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rates in the astern tropical North Pacific shadow zone. These new results contrast with previous results from the nearby South China Sea, which did not show similar variability through the deglaciation. The new data are a useful addition to the North Pacific d15N literature, and could provide an important constraint on the cross-basin gradient. However, the record does not stand very sturdily on its own, and would greatly benefit from ancillary measurements, as I suggest below. The discussion makes a general summary of the existing literature, but does not arrive at any firm or novel conclusions. In addition, there are some poorly-supported reasonings that need to be addressed. As such, I would recommend that the authors do a considerable amount of additional work, including gathering more data and making a more thorough analysis, if the article is to be published.

We appreciate the reviewer's recognition of our contribution to the study of nitrogen cycle in the North Pacific Ocean. We are grateful for the many useful suggestions provided by the reviewer. In response to the suggestions, we revised the manuscript thoroughly by adding newly obtained data, reorganizing the structure and having more discussions. We feel that the additional data and discussion have significantly strengthened our story.

1-B: Specific comments 1-B-1. Regarding the data: The d15N record appears to be of good quality, but contains a relatively small number of measurements from only a single core. Thus the interpretation is not very robust, and would greatly benefit from additional measurements. I would suggest two ways in which the interpretation could be strengthened. First, make additional measurements in the nearby MD12403 core, to verify the regionality of the signal and its magnitude. Second, measure d15N -NO3 in the water column. I believe d15N -NO3 measurements techniques have improved since the Liu (1996) data were published, and it would be very interesting to explore the suggestion that the South China Sea nitrogen supply is isotopically distinct from the OT nitrogen supply with modern data. I recognize that this would be contingent on the authors being able to obtain appropriate water samples, and that this may not be

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possible; however, given the potential these measurements would have to strengthen the interpretation of the sediment data, I would strongly encourage them to pursue it.

In this version we added d15N data from core 403 in the southern trough as suggested. The d15N signal in core 403 was affected by terrestrial input; however, it shows a similar pattern as that observed in core 404. Regarding the d15N -NO3, we are now at collecting data stage not able to provide sufficient data to constrain possibilities. In this paper, we referred to two reported d15N -NO3 profiles in water column in the Okinawa Trough and South China Sea, respectively.

1-B-2. Regarding the interpretation: Contrary to the authors' contention, the d15N record does not need local N2 fixation in order to explain it. The average sedimentary value is 4.8 +/- 0.2, with two excursions to higher d15N during the deglaciation. The value for NPIW is not an appropriate indicator of sub-euphotic zone nitrate; the data of Liu et al. (1996) show large gradients in d15N -NO3 through the upper 400m of the water column, with values $<$2 and $>$5 permil. The sedimentary values are, generally, higher than the shallow d15N -NO3, which is what would be expected to supply the surface nutrient pool. What's more, diagenetic alteration of sinking and sedimenting nitrogen has been shown to alter d15N in many environments, preventing the use of the absolute d15N value as a precise measure of organic matter d15N. Although there is ultimately a large contribution of nitrogen fixation to the d15N here - all nitrogen was fixed at some point - the evidence here does not require significant local N2 fixation.

The reviewer is correct in pointing out the difficulty in direct comparison between d15N values of nitrate in the water column and those of sedimentary nitrogen. However, we wish to emphasize that N2 fixation has been suggested to be an important N-cycle pathway in the western North Pacific based on isotopic analyses of both PON (Minagawa and Wada, 1986) and DIN (Liu et al., 1996). Hence, the contribution of N2 fixation to the sedimentary d15N record should be taken into consideration. In this version, we discussed the conditions with and without N2-fixation as suggested.

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1-B-3. Although a deglacial denitrification peak has been widely proposed in the literature, I think it is still worth acknowledging that the deglacial d15N signal could arise from other effects - not necessarily from a peak in water column denitrification rates. Kao et al. state that "it's highly probable that these are due to intensified denitrification in the ETNP" - based on what? The overall temporal coincidence? Many things were happening in the North Pacific through the deglaciation, and a number of things occurred within 1ky of the start of the Bolling, including large changes in temperature, ocean circulation, and sea level rise.

1. We separated results and discussion. In Results, we indicated the synchroniety of the d15N peaks in the Okinawa Trough cores with respect to those in the eastern North Pacific. 2. In Discussion, we discussed various mechanisms that may cause the peaks. The relative intensity of water column denitrification in the eastern North Pacific is certainly a very probable cause. First of all, it has been observed by Liu et al., (1996) that discernable isotopically heavy nitrate occurs in the Intermediate Water near Taiwan at the same isopycnal layer as that in the denitrifying zone of the Eastern North Pacific and may have the same origin. Secondly, other mechanisms of enriching 15N lack any supporting evidence. 3. Although the reviewer mentioned several possible changes in marine environmental conditions during deglaciation, it is unlikely that they may cause the kind of 15N enrichment through local processes (See Discussion). 4. One possible mechanism, which may have something to do with enhanced deep water ventilation, was proposed though we do not have any direct evidence.

1-B-4. I also think it is potentially significant that there is no glacial-interglacial change at this site - the Holocene and LGM d15N are identical, within measurement error. This agrees with the primary conclusions of Kienast (2000), and is markedly different from Western American margin records. I would like to see this more thoroughly commented upon.

It is noted in our manuscript that the overall changes in d15N values (4.3-5.8) observed in our cores are relatively small. As compared to the deglaciation peak values, the

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d15N values during the LGM and the late Holocene are rather close, but not identical. The late Holocene values showed a decreasing trend. The overall pattern appeared to resemble the model simulation of d15N of oceanic nitrate of Deutsch et al., (2004) that all feedback processes are included in major N-cycle pathways, namely, N2 fixation and water column and benthic denitrification. This resemblance suggests our data supportive of the notion that the marine N-cycle is regulated by closely coupled feedback processes. Under closely coupled N-cycle pathways, it is conceivable that the d15N variation observed in the Okinawa Trough differs significantly from those found in the eastern North Pacific, where major water column denitrification occurs. In the western North Pacific, the isotopically heavy nitrate originating from the eastern North Pacific is significantly diluted by the large nitrate reserve in the deep ocean. In addition, N2 fixation probably also help making up for the nitrogen loss in the eastern North Pacific and reducing the d15N enrichment.

1-B-5. The theoretical framework of Deutsch (2004) should be very useful in this respect, and comparisons to South Pacific d15N records might also prove illuminating. The placing of this record within the global context is tricky, but important.

As mentioned above, the theoretical framework of Deutsch et al. (2004) was added for discussion as suggested. The record off Chilean margin shows interesting temporal pattern in d15N, which is quite different from our records. Since the intensification of denitrification off the Chilean margin after the LGM was mainly due to reduced ventilation caused by large fresh water input from melting of the Patagonian ice sheet, there is little commonality between the Chilean system and ours. We feel that discussion on the Chilean case will shed little light on our results.

1-B-6. I find the references to the earlier works by Kao somewhat confused. Given that they are presented as a central part of the discussion, I would like to see the reasoning and referencing clarified. It seems to me that, in combination with their earlier works, these authors are on the verge of assembling a useful picture of historical variability of physical circulation and biogeochemical cycling in the vicinity of the Okinawa trough.

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However, it seems to me that there are - at present - an unacceptable number of inconsistencies between their various observations, that must be reconciled if this is to prove a useful contribution: in reading over the earlier work, I was worried to encounter what appear to be unexplained inconsistencies between their interpretations. For example, in one paper Corg is taken as a paleoproductivity proxy, while in another, Ba/Al is the chosen paleoproductivity proxy - however, the two quantities show an opposite sense of glacial-interglacial change!

Several issues are raised here. Our responses are as follows. 1. We have separated Results and Discussion in the revised version in order to clarify our arguments. 2. In the original manuscript, we referred to Kao et al. (2005) and Kao et al. (2006a) briefly to illustrate the influence of changes in hydrodynamic conditions in the Okinawa Trough on the biogeochemical properties of the sediments. Apparently, the brief referral caused some confusion. In fact, there is no inconsistency in our arguments in the current paper. We have provided more discussion to illuminate our point. However, some of the issues raised by the reviewer are not mentioned in our manuscript and beyond the scope of this paper. Hence, we do not include those in our added discussion. Instead, we provide some brief accounts as responses to the reviewer's quest. 3. The apparent inconsistency referred to by the reviewer is probably the higher Ba/Al ratio, which indicates higher productivity, in the Holocene observed in Core 403 in the southern Okinawa Trough by Kao et al. (2005). As shown in Kao et al. (2005) as well as in this paper, the Corg content in sediment corresponding to the period of high Ba/Al ratio was not unusually high. We believe that the Holocene paleoproductivity as recorded in core 403 (southern trough) was indeed high, but the lack of elevated Corg content mainly reflects the enhanced ventilation in the Okinawa Trough in Holocene and, consequently, enhanced organic matter degradation or reduced organic carbon preservation.

1-B-7. I am not familiar with the use of TS as a proxy for bottom water oxygenation. I would appreciate some references that explain how it is related to bottom water oxygen

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concentrations in recent marine sediments.

Because most, if not all, sedimentary sulfur is reduced sulfur, it represents the end product of sulfate reduction in the underlying sediments and its content is closely related to sulfate reduction rate in the sediment column. More TS indicates stronger sulfate reduction, which is favored by poorer supply of oxygen to the sediment column and vice versa, provided sufficient organic matter is available to fuel sulfate reduction. Kao et al. (2006a) demonstrated using a 3-D circulation model that sluggish ventilation in the Okinawa Trough enhances sulfate reduction. In the revised version, we calculated the flux of organic carbon consumed stoichiometrically during sulfate reduction corresponding to the TS accumulated in the sediment. We believe this value is a good indicator of the degree of oxygenation in the bottom water.

1-B-8. Finally, there are some parts of the discussion that seem out-of-place, or do not serve a clear purpose. The discussion of TS and the 3-D model (paragraph starting "However, sea-level change") seems somewhat aimless. Is this intended to provide an argument against local denitrification? Why is this important? The d13C discussion which follows is also confused and unconvincing.

In the revised version, we have discussed different mechanisms that may cause variation in d15N of sedimentary nitrogen. It has been demonstrated that local water column denitrification is an important process in controlling d15N of nitrate in the upper water column. Therefore, we tried to explore its possible significance in the Okinawa Trough. Our analysis indicated that the periods with relatively poorer oxygen supply did not match the d15N peaks, precluding local water column denitrification as an important mechanism for enriching 15N in nitrate. Another possible mechanism is local upwelling, which may provide excess nitrate to the upper water column resulting in a drop in d15N (e.g., Holmes et al., 1997). The discussion on d13C is to explore the possible significance of upwelling induced isotopic changes, which appears to be not very important in the Okinawa Trough.

1-B-9. Last, I don't understand why the precession cycle at 30N is shown. If this is to be included, there should be a discussion of a mechanism linking it to the d15N.

Reviewer is correct. We have eliminated the curve of solar insolation indicated by the precession cycle at 30 degree N, because it has little relevancy to our discussion.

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