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A mid-Holocene transition in the nitrogen dynamics of the western equatorial Pacific: Evidence of a deepening thermocline?

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[1] Sedimentary δ^{15}N records from the oligotrophic western equatorial Pacific (WEP) off Mindanao show that late Holocene sedimentary δ^{15}N is substantially lower than that of the early Holocene, following a gradual >3 ‰ decrease that occurred between 7 and 3 kyrs ago. Analyses of modern day nitrate isotope profiles from the same region indicate the sensitivity of the WEP N pools towards (1) the advection of ^{15}N-enriched nitrate from the Eastern Equatorial Pacific (EEP) by the North Equatorial Current (NEC) and the Mindanao Current in subsurface waters and, (2) at shallow depths, the input of new and ^{15}N-depleted nitrate through N_2 fixation. We suggest that the Holocene decrease in sedimentary δ^{15}N reflects a diminished relative input of ^{15}N-enriched nitrate to the surface biota, either through an increase of regional nitrogen fixation, a change in nitrate consumption along the advective path of nitrate supply, or a decrease in the vertical supply of ^{15}N-enriched nitrate from the NEC. The latter mechanism is consistent with a Holocene deepening of the WEP nitracline/thermocline.

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1. Introduction

[2] The equatorial Pacific is fundamental to Earth's climate system, and has been purported to be instigator, amplifier and mediator of past global climate change on timescales ranging from inter-annual to orbital. Today, the El Niño Southern Oscillation (ENSO) dominates the interannual variability of the tropical Pacific, with teleconnections affecting climate conditions worldwide, and it has thus been proposed that past global climate change on centennial to orbital time-scales could have been modulated by "ENSO-like" variations in the mean state of the tropical Pacific. However, many reconstructions of past ENSO variability rely exclusively on sea surface temperature (SST) or sea surface salinity (SSS) reconstructions in the eastern and western equatorial Pacific. This is problematic due to the inherent uncertainty of SSS reconstructions, multiple causes of SST variability, and, not least, to the potential non-stationarity of ENSO-related climatic teleconnections [*Bush*, 2007].

[3] The western equatorial Pacific (WEP) is considered to be an oligotrophic part of the ocean, where much of the nitrate is advected laterally from the eastern equatorial Pacific (EEP) [*Peña et al.*, 1994; *Turk et al.*, 2001], and interannual changes in vertical nitrate supply to the WEP euphotic zone are determined by the depth of the nitracline/thermocline, which itself is intimately linked with ENSO [*Turk et al.*, 2001]. Reconstructions of temporal variations in nitrate availability and supply in the WEP could thus provide a means of elucidating past variations in WEP thermocline depth and equatorial circulation, critical descriptors of the mean state of the tropical Pacific Ocean. Here we use modern water column profiles of the nitrogen and oxygen isotopic composition of nitrate in the WEP to provide calibration of the biogeochemical processes that drive N-transformations in the WEP today. High-resolution sedimentary δ^{15}N records from the WEP off Mindanao are presented in an attempt to elucidate Holocene variations in the WEP water column structure and circulation.

2. Materials and Methods

[4] Sediment cores MD98-2181 (6°18′N/125°49′E, 2114 m water depth), MD06-3067 (6°31′N/126°30′E, 1574 m water depth), and MD06-3075 (6°29′N/125°52′E, 1878 m water depth) were recovered during IMAGES expeditions aboard the *Marion Dufresne* in 1998 and 2006, respectively, at three different sites in the WEP off Mindanao, SE Philippines (see Figure S1 of the auxiliary material).[1] Sites 81 and 75 are located inside the Bay of Davao, whereas site 67 is more open oceanic. The water column samples were retrieved close to the two core sites (station 1, outside the bay at 6°28′N/126°28′E, and station 2, inside the bay at 6°28′N/125°52′E) by hydrocast during the 2006 expedition. Water samples were immediately filtered (0.2 μm) and frozen at $-80°$ C on board.

[5] Natural abundance nitrate N and O isotope ratio measurements (denoted as δ^{15}N and δ^{18}O, with $\delta = (R_{sample}/R_{standard})-1] \times 1000$, where R represents the ratio

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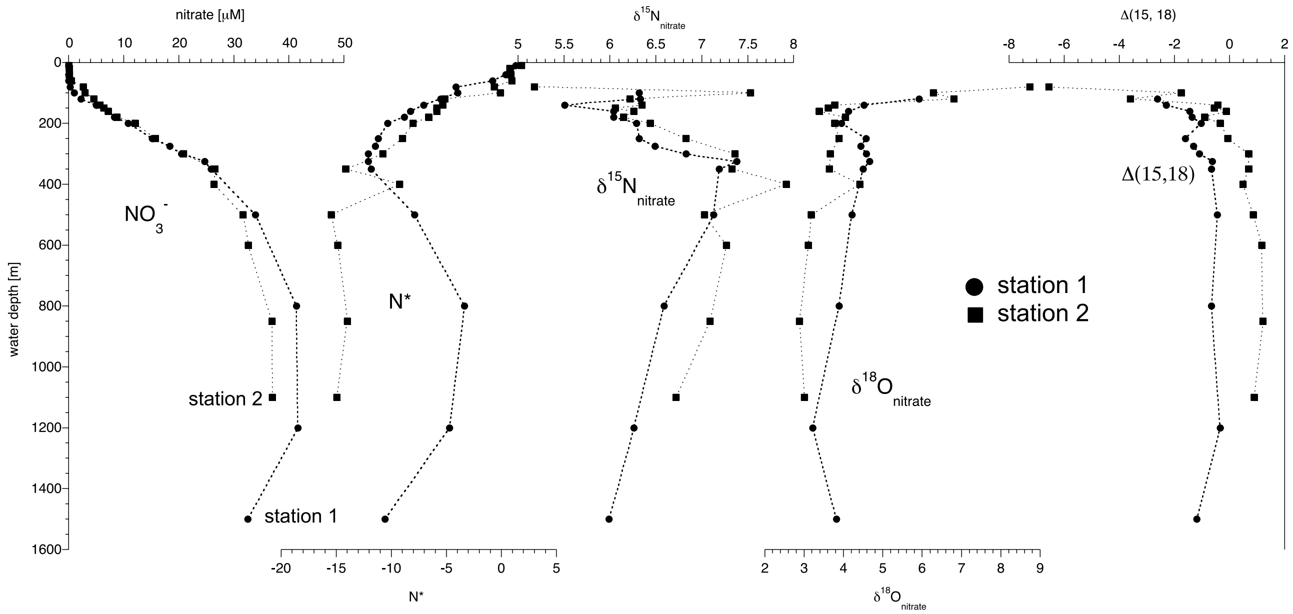

Figure 1. Water column profiles of nitrate concentration, N* and the isotopic composition of nitrate (see text for definitions) at two sites off Mindanao. Station 1 (circle) is located close to site MD06-3067, station 2 (square) close to MD06-3075 inside the bay of Davao (see Figure S1).

of ^{15}N to ^{14}N or ^{18}O to ^{16}O, respectively) were performed using the "denitrifier method" [*Casciotti et al.*, 2002; *Sigman et al.*, 2001] (see auxiliary material for details). N and O isotope ratios are reported in ‰ relative to atmospheric N_2 for N and V-SMOW for O isotopes, respectively. The reproducibility of the method (based on duplicate measurements of standards and samples) is generally better than ±0.2‰ for $\delta^{15}N$ and ±0.5‰ for $\delta^{18}O$.

[6] Sedimentary $\delta^{15}N$ was analyzed on dried, homogenized bulk sediment samples on an elemental analyzer coupled to a Finnigan Delta plus mass spectrometer at the Pacific Centre for Isotopic and Geochemical Research, UBC Vancouver, following standard procedures. Analytical precision of this method is better than ±0.2‰.

[7] The age model for core MD98-2181 is adopted from *Stott et al.* [2004]. For cores MD06-3067 and MD06-3075, the age models are based on aligning the planktonic foraminiferal oxygen isotope records (T. Bolliet et al., unpublished material, 2008) with the record from site 81, independently corroborated by ^{14}C dates. For site 67, the thus derived age model reveals that the top-most 3.5–4 kyrs were not recovered during coring operation. According to these age models, average sedimentation rates during the last 10 kyrs are ca. 14 cm/kyr at site 67, ca. 65 cm/kyr at site 75, and ca. 85 cm/kyr at site 81.

3. Results and Discussion

3.1. Water Column

[8] Both nitrate $\delta^{15}N$ and $\delta^{18}O$ below the mixed layer depth display values (Figure 1; 6–8 ‰ for $\delta^{15}N$ and 3–5 ‰ $\delta^{18}O$) that are higher than the values for "mean" oceanic nitrate (5.5‰ and 2.5‰, respectively [e.g., *Casciotti et al.*, 2002]) (see auxiliary materials for further details). The elevation above these mean oceanic nitrate isotope values, along with the comparatively low N* ($N^* = N - 16 \times P + 2.9$ mmol m^{-3}; Figure 1), can be explained by the advection of water masses that carry the geochemical signatures of water column denitrification, and originate in the Eastern Tropical Pacific [e.g., *Sigman et al.*, 2005]. This interpretation is consistent with previous studies suggesting import of ^{15}N enriched nitrate from the EEP to the WEP [*Yoshikawa et al.*, 2006].

[9] Consistent with a higher degree of partial denitrification (i.e., lower N* values), the ^{15}N-enrichment is more pronounced at Station 2 (Figure 1). Towards the surface, in association with the decrease in nitrate to concentrations below the detection limit, nitrate $\delta^{18}O$ increases at both stations, in agreement with isotope fractionation (i.e., the preferential consumption of nitrate containing the lighter isotope ^{16}O) associated with nitrate uptake by phytoplankton [*Casciotti et al.*, 2002]. The $\delta^{15}N$, on the other hand, decreases as we approach nitrate-free surface waters. Based on previous studies [e.g., *Casciotti et al.*, 2002; *Lehmann et al.*, 2005], we would expect a parallel evolution of nitrate $\delta^{15}N$ and $\delta^{18}O$ during algal nitrate uptake, with a ratio of ^{15}N to ^{18}O close to unity (see also auxiliary material). The decoupling of nitrate N and O isotope gradients (Figures 1 and S2) thus suggests the importance of additional N-transforming processes at the surface besides nitrate assimilation. The degree of the N-to O-isotope decoupling (i.e., the N-to-O isotope anomaly, or deviation from a 1:1 variation in $\delta^{15}N$ and $\delta^{18}O$) can be quantified using the approach of *Sigman et al.* [2005]:

$$\Delta(15,18) = \left(\delta^{15}N - \delta^{15}N_m\right) - \left(\delta^{18}O - \delta^{18}O_m\right), \qquad (1)$$

where $\delta^{15}N_m$ and $\delta^{18}O_m$ are the mean $\delta^{15}N$ and $\delta^{18}O$ of deep water, respectively. We assigned values of 5.5 ‰ for $\delta^{15}N_m$ and 2.5 ‰ for $\delta^{18}O_m$. A negative Δ(15,18) indicates a decrease (or lesser increase) in nitrate $\delta^{15}N$ relative to the $\delta^{18}O$. At both stations, surface waters are characterized by a clear negative nitrate isotope anomaly, with a decreasing trend in Δ(15,18) towards the ocean surface (Figure 1).

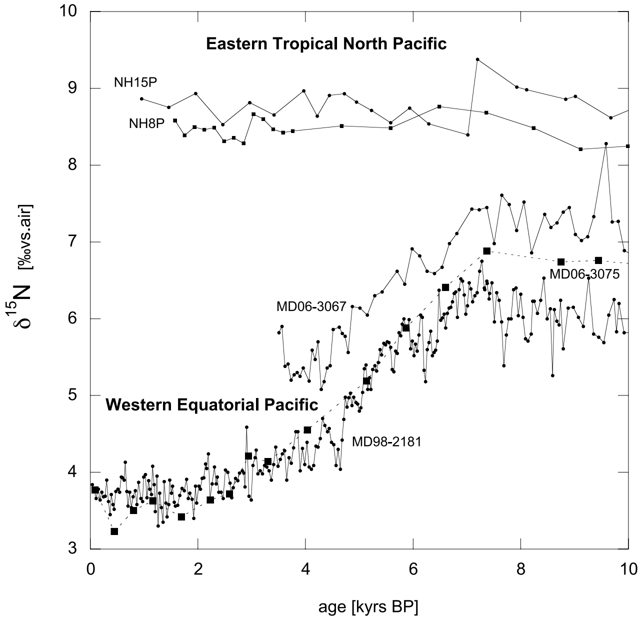

Figure 2. Holocene δ^{15}N records from sites MD98-2181, MD06-3067, and MD06-3075 in the western equatorial Pacific compared to two records from the eastern equatorial Pacific off Mexico (NH8P and NH15P) [*Ganeshram et al.*, 2000].

[10] Here we do not attempt a quantitative interpretation of the observed depth distribution of $\Delta(15,18)$ in the study area, because it would go beyond the scope of this paper. Several processes can theoretically lead to negative nitrate $\Delta(15,18)$ anomalies (e.g., nitrite re-oxidation [see *Sigman et al.*, 2005; *Casciotti and McIlvin*, 2007]). Yet, details aside, the negative $\Delta(15,18)$ anomaly towards the surface is most consistent with the remineralization and accumulation of newly fixed nitrogen (see also auxiliary material). This regeneration of fixed N is expected to decrease the nitrate δ^{15}N while it does not significantly affect its δ^{18}O (relative to deep water nitrate) [*Sigman et al.*, 2005; *Bourbonnais et al.*, 2008]. Following arguments made by *Bourbonnais et al.* [2008], the discrepancy in magnitude of the surface nitrate isotope anomaly between Stations 1 and 2 may indicate spatial variations in the relative importance of N_2 fixation, with higher relative N_2 fixation rates at Station 2, where more negative $\Delta(15,18)$ values were measured. However, it needs to be noted that the interstation-variation in $\Delta(15,18)$ in the upper water column is partially due to the fact that the source waters from the thermocline already display different $\Delta(15,18)$. Thus, the spatial variation in the N_2 fixation rates may be less pronounced than is indicated by the upper water column $\Delta(15,18)$ at the two stations. The fact that we do not observe strongly positive N* values in the surface waters (as has been observed, for example, in the eastern subtropical N-Atlantic, e.g., *Bourbonnais et al.* [2008]) does not a priori argue against a strong contribution of N_2 fixation to total N export in the WEP, since the whole region is impacted by advective import of low-N* water masses from the EEP. Indeed, most probably, residual geochemical denitrification signatures imported from the east are partially offset by the input of newly fixed N. The δ^{15}N of the sedimentary organic matter reflects the relative contribution of either nitrate source (^{15}N-enriched nitrate from the thermocline versus ^{15}N-depleted N from N fixation).

3.2. Sediment Record

[11] The bulk sedimentary δ^{15}N records from the western equatorial Pacific show stable values around 7 and 6 ‰, respectively, during the early Holocene (10-7 ka), followed by a > 3 ‰ decrease between 7 and 3 kyrs BP (Figure 2). During the latest Holocene (not recovered at site 67, see above), δ^{15}N values are more or less constant around 3.5–4 ‰. Present-day N limitation (at comparatively low δ^{15}N values) suggests that local N utilization has little impact on the bulk sedimentary δ^{15}N signal, and the lack of significant Holocene changes in chemical tracers of past productivity (TOC, biogenic opal, alkenone conc.; not shown) strongly suggests that variable local nitrate utilization is indeed an unlikely cause for the Holocene decrease. The similarity between the δ^{15}N records at the three sites (Figure 2) with very different sedimentation rates (see above) and proximity to terrigenous input also suggests that diagenetic overprint and/or variable inputs of organic or inorganic terrestrial N are not the prime cause of the Holocene decrease. The lower δ^{15}N values at sites 75 and 81 inside the bay are consistent with the dual nitrate isotopic signature suggesting a greater importance of N_2 fixation there (see above). Thus, sedimentary δ^{15}N in the study area is, indeed, reflective of the isotopic composition of the nitrate fueling phytoplankton production throughout the region.

[12] Water column concentrations of O_2 today [e.g., *Kashino et al.*, 1996] are well above the limit for local denitrification, and there is no evidence to indicate that this changed throughout the Holocene. While there are suggestions that the whole ocean nitrate ^{15}N/^{14}N may have reached a maximum value (1–2 ‰ greater than today) prior to 10 kyrs BP [e.g., *Deutsch et al.*, 2004], this contrasts with the >3 ‰ decrease in sedimentary δ^{15}N off Mindanao that clearly occurs only after 7 kyrs BP. More importantly, sedimentary records from the Eastern Tropical North Pacific (ETNP), the most proximal water-column denitrification zone, are interpreted to reflect largely unchanged rates and extent of denitrification during the Holocene [cf. *Ganeshram et al.*, 2000; *Thunell and Kepple*, 2004] (Figure 2), which, in turn, implies a more or less constant N isotopic signature of the source of subsurface nitrate at the core sites in the WEP, in particular during the 7-3 kyrs interval.

[13] Four non-exclusive scenarios are entertained to explain the higher sedimentary δ^{15}N during the early Holocene: reduced rates of N_2 fixation; increased cumulative N utilization along the advective supply route of nitrate; intensified lateral advection of ^{15}N-enriched nitrate from the ETNP to the WEP; and increased vertical supply of ^{15}N-enriched nitrate to the photic zone. We elaborate upon, and evaluate, each of these hypotheses in turn.

[14] 1. Given the importance of N_2 fixation implied by the modern water column profiles (see above), higher δ^{15}N during the early Holocene could be indicative of reduced N_2 fixation rates then. Because there is presently no independent proxy to quantify past N_2 fixation rates, and forcing mechanisms of variations in N_2 fixation remain elusive, we cannot assess the importance of this factor in determining temporal variations in sedimentary δ^{15}N off Mindanao. It is

worth noting, however, that a reduced physical supply of nitrate to the photic zone between 7 and 3 kyrs BP, suggested by hypotheses (3) and (4), would have given N_2-fixers an ecological advantage and may have shifted the balance of nitrogen nutrition towards newly fixed nitrogen.

[15] 2. The second scenario to explain the Holocene decrease in sedimentary δ^{15}N off Mindanao calls on a Holocene decrease in the "cumulative" N utilization along the advective nitrate supply route. The enrichment of δ^{18}O-nitrate within near-surface waters shows that the nitrate supplied to phytoplankton here is a residuum of nitrate upwelled elsewhere, and circulated and partially consumed within the equatorial and subtropical circulation to some degree prior to delivery at the surface off Mindanao. Although the corresponding enrichment of δ^{15}N-nitrate caused by this process is masked by modern N_2 fixation near the surface, past variations in the advective supply route could have modulated the cumulative N utilization component. A more pronounced utilization signature during the early Holocene could have been caused by variations in the ratio of macro- and micro-nutrients supplied to the photic zone, i.e., in the chemistry of the source waters [cf. *Altabet*, 2001], or by a change in the advective regime.

[16] Assuming that the substantial Holocene decrease in δ^{15}N off Mindanao is not solely caused by these two biogeochemical factors, the higher early Holocene δ^{15}N values could be interpreted to reflect increased supply of ^{15}N-enriched NO_3- from the EEP to the photic zone in the WEP, either laterally or vertically. 3. An increased lateral advection of ^{15}N-enriched nitrate across the Pacific could be explained by a stronger North Equatorial Current (NEC) during the early Holocene, a change in the bifurcation of the NEC into the Mindanao Current (MC) and Kuroshio, or a deeper NEC in the EEP tapping into a zone of more intense denitrification there. While we have no means to assess the latter scenario, interannual variations observed today are used as analog to qualitatively discuss changes in the NEC transport and bifurcation and their effect on the MC transport. The NEC bifurcation latitude has only little effect on the MC transport [*Kim et al.*, 2004], whereas the MC transport is highly correlated with NEC transport. The NEC transport, in turn, is driven in part by the northeasterly trade winds. To the extent that the easterly trade winds weaken with a more southerly position of the Intertropical Convergence Zone (ITCZ), the Holocene southward migration of the ITCZ [e.g., *Haug et al.*, 2001] would thus point to a Holocene decrease in NEC, and thus MC, transport, in line with the Holocene decrease in δ^{15}N. This scenario is also consistent with the inferred Holocene decrease in sea surface salinities at site 81 [*Stott et al.*, 2004] and in the WEP [*de Garidel-Thoron et al.*, 2007]. Reduced equatorial easterlies, however, would tend to shoal the WEP thermocline, which would increase the vertical supply of ^{15}N-enriched N (see below), and would thus counteract the effect of decreasing NEC transport.

[17] 4. Alternatively, the higher sedimentary δ^{15}N in the WEP during the early Holocene is reflective of an increase in the vertical supply of ^{15}N-enriched nitrate to the photic zone. This could have been caused by an overall shallower thermocline, an increase in upwelling associated with the Mindanao Dome (MD), or enhanced mixing through changes of the activity of westerly wind events and synoptic activity. Today, interannual variations in the strength of the Mindanao Dome appear to be caused by variations in the local upwelling due to positive curl associated with the Asian winter monsoon [*Masumoto and Yamagata*, 1991]. Reconstructions of a Holocene increase in winter monsoon activity [e.g., *Yancheva et al.*, 2007] thus render the latter cause of a Holocene decrease in the vertical supply of ^{15}N-enriched nitrate to the photic zone unlikely. We note, however, that ocean general circulation model analyses suggest a stronger MD upwelling associated with El Niño events [*Christian et al.*, 2004]. At the same time, the NEC transport also tends to intensify during El Niño [*Qiu and Lukas*, 1996], which would further increase the supply of ^{15}N-enriched N to the WEP photic zone (see above). A shallower early Holocene WEP thermocline as well as the link of the MD upwelling and NEC transport with ENSO events could thus be construed to reflect an intensification of the EEP cold tongue during the Holocene. A deeper late Holocene WEP thermocline inferred here is consistent with foraminiferal evidence from the WEP north of Papua New Guinea [*de Garidel-Thoron et al.*, 2007] and with upper water column δ^{13}C and temperature gradients in core MD06-3067 (surface and thermocline dwelling foraminiferal stable isotope and Mg/Ca data (T. Bolliet et al., unpublished material, 2008)). However, this scenario is incongruous with foraminiferal Mg/Ca SST reconstructions near the Galapagos Islands [*Koutavas et al.*, 2002], and we note that analysis of present-day and mid-Holocene coupled general circulation model simulations conducted as part of the Paleomodel Intercomparison Project [*Zheng et al.*, 2008] does neither reveal substantial changes of the mean thermocline in the Mindanao region, nor does it support the notion of a NEC weakening throughout the Holocene. It is tempting to speculate, however, that a Holocene deepening of the WEP thermocline, suggested by the δ^{15}N records presented here, provided a necessary condition for ENSO variations to occur at all.

4. Summary and Conclusion

[18] Bulk sedimentary δ^{15}N records from the western equatorial Pacific display a >3 ‰ decrease between 7 and 3 kyrs BP, evidencing a significant decrease in the δ^{15}N of nitrate utilized during primary production. At the same time, water column nitrate isotope profiles at the coring sites show the presence of ^{15}N-enriched nitrate advected from the EEP at depth. While the data in hand do not provide conclusive evidence for the exact mechanisms responsible for the observed isotopic shift in the WEP, we argue here that they reflect variations in the balance between new production (N_2 fixation) and the production from advective and diffusive preformed nitrate sources. These two N-cycle processes may in turn be mechanistically linked in that a higher nitrate availability will attenuate N_2 fixation, generating positive feedbacks with regards to changes in the sedimentary δ^{15}N. The Holocene decrease in sedimentary δ^{15}N off Mindanao could thus reflect an increased importance of local/regional N_2 fixation and/or a decrease in the cumulative impact of N utilization along the advective nitrate supply route. Alternatively, the sedimentary δ^{15}N records presented here could indicate a Holocene decrease

in the vertical supply of ^{15}N-enriched nitrate to the photic zone, caused by a deepening of the thermocline in the WEP.

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