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# North Pacific-wide spreading of isotopically heavy nitrogen during the last deglaciation: Evidence from the western Pacific

**S. J. Kao[1,4], K. K. Liu[2], S. C. Hsu[1], Y. P. Chang[3], and M. H. Dai[4]**

[1]Research Center for Environmental Changes, Academia Sinica, Taiwan
[2]Institute of Hydrological Science, National Central University, Jung-Li, Taiwan
[3]Institute of Applied Geosciences, National Taiwan Ocean University, Keelung, Taiwan
[4]State Key Laboratory of Marine Environmental Science, Xiamen University, Xiamen, China

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**Abstract.** Sedimentary $\delta^{15}$N records in two IMAGES cores (MD012404 and MD012403) retrieved from the Okinawa Trough (OT) in the western North Pacific reveal deglacial increases with two peaks occurring during the Bølling/Allerød and the Preboreal/early Holocene periods. These peaks are synchronous with previously reported $\delta^{15}$N peaks in the Eastern Tropical North Pacific, although the amplitudes (from 3.8 to 5.8‰) are much smaller in the OT. Similar $\delta^{15}$N values for the last glacial maximum and the late-Holocene observed by us at a site far from the present-day zones of water-column denitrification (WCD) indicate that the mean $^{15}$N/$^{14}$N of nitrate in the upper ocean did not differ much between the two climate states. The accumulation rate of organic carbon and total sulfur content are used as indices of the local WCD potential. The results suggest that enhancement of global WCD rather than local denitrification should be responsible for the deglacial maxima of sedimentary $\delta^{15}$N in the Okinawa Trough. Our data could provide additional constraints to better understand changes in nitrogen budget during the glacial to interglacial transition.

## 1 Introduction

As an essential nutrient, changes in the oceanic inventory of biologically available N (or "fixed nitrogen", which is dominated by nitrate) would be expected to impact the biological carbon pump over large regions of the ocean through glacial-interglacial cycles (Falkowski, 1997; Broecker and Henderson, 1998; Archer et al., 2000; Naqvi et al., 2008). Thus, there is a growing interest in the interaction between climate and N biogeochemistry. Evidence for strong links between climate and key N cycling processes – $N_2$ fixation and denitrification – has been accumulating (Altabet et al., 1995; Ganeshram et al., 1995; Falkowski, 1997), and several researchers have hypothesized indirect influences of the marine fixed N inventory on paleo-climate (McElroy, 1983; Ganeshram et al., 1995, 2000; Pedersen and Bertrand, 2000; Suthhof et al., 2001; Altabet et al., 2002). However, considerable uncertainty still remains regarding changes in global ocean nitrate inventory, particularly during the last glacial period (Deutsch et al., 2004; Altabet, 2007).

Sedimentary nitrogen isotope ($\delta^{15}$N) records from oligotrophic regions distant from zones of vigorous water column denitrification (WCD) may help in assessing past changes in the magnitude of global WCD, which along with sedimentary denitrification is the main pathway of losses of fixed N from the ocean (Brandes and Devol, 2002; Altabet, 2007; Naqvi et al., 2008). One of the unsettled issues is the intensity of WCD in the North Pacific during the last deglaciation and its effect on global oceanic N inventory. Evidence from the South China Sea (SCS), a *cul-de-sac* (Fig. 1a) of North Pacific Intermediate Water (NPIW) (You et al., 2005), believed to be well-suited to determine the extent of basin-wide influence of WCD in the Eastern Tropical North Pacific (ETNP), shows insignificant responses in sedimentary $\delta^{15}$N during the last deglaciation when WCD peaked in the ETNP (Kienast, 2000; Higginson et al., 2003). However, in order to adequately assess the effect of intensified WCD on marine N inventory during the last deglaciation more sedimentary $\delta^{15}$N records are needed from oligotrophic regions of the ocean, particularly the western Pacific.

*Correspondence to:* S. J. Kao
(sjkao@gate.sinica.edu.tw)

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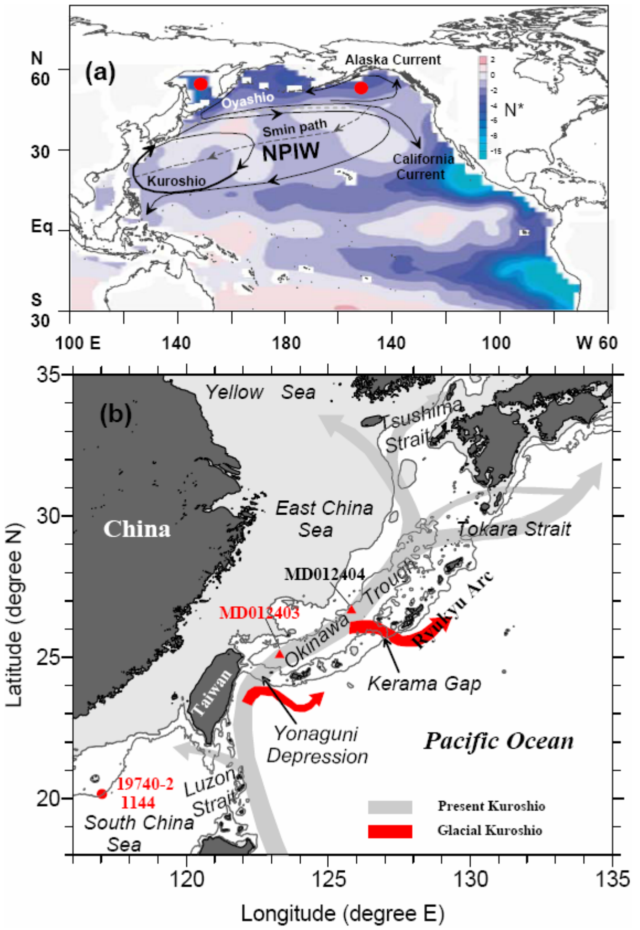


**Fig. 1.** **(a)** Geographic setting of the North Pacific. Circulation, path of salinity minimum ($S_{min}$, see You et al., 2006) and two sources (black circles) of intermediate water are indicated. **(b)** Location map for IMAGES core MD012404 and MD012403, and Cores 17940 and 1144 used in previous reports are also shown. The land (deep gray), shelf of $<-100$ m (light gray) and $-1000$ m isobaths are also shown. The flow path of present Kuroshio and glacial Kuroshio are shown in gray and red arrows.

We present sedimentary $\delta^{15}$N records in the Okinawa Trough (OT) showing changes that are synchronous with global climate events during the last deglaciation. The synchroneity and similarity of $\delta^{15}$N records from the western and eastern North Pacific allow us to infer enhanced supply of the isotopically heavy nitrogen to the upper ocean during the last deglaciation when WCD had intensified in the ETNP. The amplitude of the isotopic variation may shed new light on the potential changes in the fixed N inventory in the entire North Pacific.

## 2 Study area

The OT is located in the western Pacific (Fig. 1a). The Kuroshio Current (KC) enters the OT through the Yonaguni Depression (Fig. 1b) at its southern end. While the OT is around 2000m deep, the depth of Yonaguni Depression ranges from 300 to 800 m, deep enough to allow the Kuroshio Intermediate Water (KIW) to enter the OT.

Recent studies suggest that nitrate influx by the KC is presently 1.5–3.4 times the river loading (Li, 1994; Chen and Wang, 1999; Liu et al., 2000). Thus the KIW is the main supplier of nutrients to the ECS (Chen, 1996), of which the OT serves as a boundary and a sink for the shelf production. It is conceivable that the primary nutrient source for the ECS in the pre-Anthropocene period was dominated by input through the shelf intrusion of the KIW (Liu et al., 2000). In addition to the Yonaguni Depression, the Kerama Gap in the middle Ryukyu Arc (Fig. 1b), although narrow, is sufficiently deep (2000 m) to afford some intermediate and deep water exchange between the OT and the northwestern Pacific.

The NPIW spreads throughout the North Pacific (Fig. 1a) with a subtropical salinity minimum (about 34.0–34.3) within the depth range of 300–800 m, confined to the subtropical North Pacific (see You et al. (2005) and references therein). It has been demonstrated by $\delta^{15}$N values of nitrate that the KIW carries isotopically heavy nitrate originating from the Eastern North Pacific, presumably transported by the NPIW (Liu et al., 1996). Following Brandes et al. (2007), we present distribution of N* (N* = [nitrate] – 16[phosphate] + 2.9) along the surface of constant density of 1026.6 kg m$^{-3}$ ($\sim$600 m depth) in Fig. 1a for reference. The N* distribution clearly illustrates that the denitrification signal (negative N*) in ETNP intermediate water may be transmitted to the western North Pacific. On the other hand, the N* distribution in the surface ocean (Brandes et al., 2007) suggest that nitrogen input from $N_2$-fixation (positive N*, not shown) is higher in the western North Pacific, which is broadly consistent with the biogeography of *Trichodesmium* observed from ships and satellites. Liu et al., (1996) also found isotopically light nitrate within the 200–400 m depth range in the KC. The isotopically light nitrate was attributed to the regeneration of regional $N_2$-fixation since isotopically light (negative $\delta^{15}$ N) particulate nitrogen (PN) has been repeatedly observed in the surface waters of the Philippine Sea off eastern Taiwan (Wada and Hattori, 1976; Saino and Hattori, 1987).

Besides circulation and biological factors, sea-level change in the past (see sea level curve in Fig. 2a) might also significantly affect KC volumetric transport, surface and bottom circulations in the OT. Hydrological changes and subsequent effects on the sedimentary sulfur, organic carbon biogeochemistry (Kao et al., 2005, 2006a) and water column nutrient dynamics are discussed below.

## 3 Materials and methods

Data are being reported for two giant piston core (Fig. 1b), MD012404 (125.81° E, 26.65° N, water depth 1397, core length 43 m) and MD012403 (123.28° E, 25.07° N, water depth 1420, core length 20 m), that were recovered by R/V Marion Dufresne during the cruise IMAGES VII, WEPAMA (Bassinot et al., 2002). The coring site of MD012404 is located in a small topographic low near the western edge of the OT, which is ideal for trapping downward settling biogenic particles in the water column, as well as suspended sediments transported from the shelf of the East China Sea (ECS, see Fig. 1b). Sediments in MD012404 are mainly composed of nearly homogenous nanno-fossil ooze or diatom-bearing nanno-fossil ooze and no visible turbidite or tephra layer was found in the core (Chang et al., 2005). Compared to MD012404 located in the middle trough, MD012403 came from the southern OT that receives significant terrestrial inputs from Taiwan resulting in 4× higher mean sedimentation rate in the past 30 000 years (Kao et al., 2008). Comparison of the data between MD012404 and MD012403 allows us to examine the influence of terrestrial input on carbon and nitrogen isotopes.

Sediment cores were sliced into 1-cm-thick segments during the cruise and preserved in freezer. A preliminary age model of core MD012404 based on 5 AMS$^{14}$C dates over the last 30 ka has been published previously (Chang et al., 2005). Chang et al. (2008a, b) recently published a new fine-tuned age model for this core with 14 additional $^{14}$C ages over the last 40 000 years (blue triangles in Fig. 2b). The AMS $^{14}$C measurements were made using ∼20 mg of the planktonic foraminifers *G. ruber* and *G. sacculifer* (>250 $\mu$m) at the Micro Analysis Laboratory, Tandem Accelerator (MALT), University of Tokyo. All AMS $^{14}$C ages were adjusted for a mean Pacific reservoir age of 400 years, and then calibrated according to Fairbanks et al. (2005). No age reversal was observed between any adjacent $^{14}$C dating horizons. We also found a layer of low carbonate and high magnetic susceptibility (measured on board ship) (Bassinot et al., 2002) that coincides with the timing (∼7.3 kyr BP, see Fig. 2a) of the eruption of the Japanese volcano Kikai-Ah (Machida, 2002). More information on the age model including the AMS data can be found in Chang et al. (2008b). Details of age model and related information for MD012403 have been reported in Kao et al. (2005).

For the $\delta^{13}C_{TOC}$ and $\delta^{15}N$ analyses, samples were treated with 1 N HCl for 16 h to remove carbonate; the residue was centrifuged and freeze-dried (Kao and Liu, 2003). Details of the sample preservation and pretreatments have been reported in Kao et al. (2006a). Carbon and nitrogen isotope analyses were carried out using a Carlo-Erba EA 2100 elemental analyzer connected to a Thermo Finnigan Delta$^{plus}$ Advantage isotope ratio mass spectrometer (IRMS). Carbon and nitrogen isotopic compositions are presented in the standard $\delta$ notation with respect to PDB carbon and atmospheric nitrogen. USGS 40, which has certified $\delta^{13}$C of −26.24‰ and $\delta^{15}$N of −4.52‰ and acetanilide (Merck) with $\delta^{13}$C of −29.76‰ and $\delta^{15}$N of −1.52‰ were used as working standards. The reproducibility of carbon and nitrogen isotopic measurements is better than 0.15‰İn order to check if decarbonation process affected the $\delta^{15}$N results we randomly selected 11 samples for measuring $\delta^{15}$N in untreated sediment. The results show similar values of $\delta^{15}$N for acidified and non-acidified samples (blue circles in Fig. 2e).

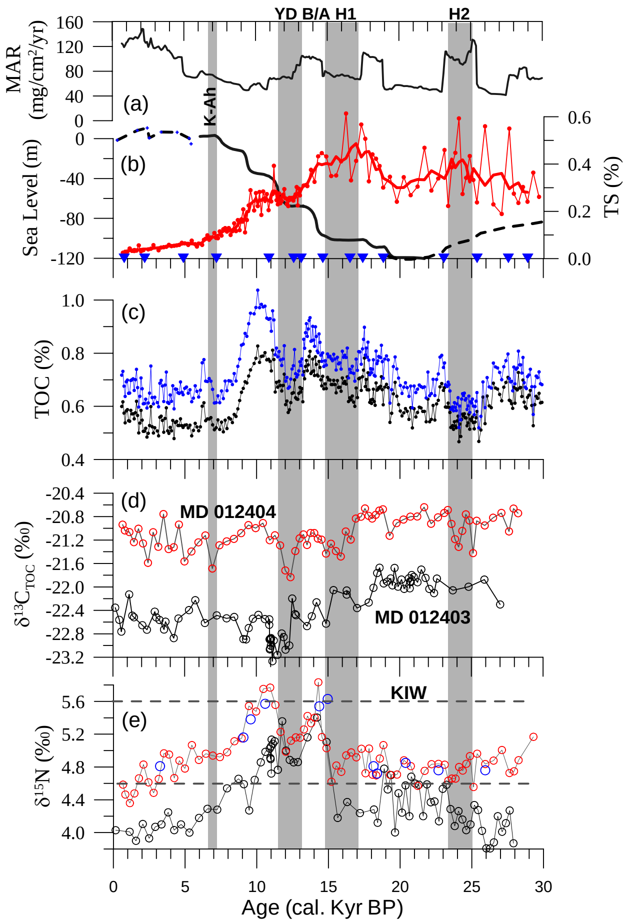


**Fig. 2.** **(a)** Accumulation rate for sediment (MAR). **(b)** Sea level curve. Blue dotted-line and black curve represent data digitized, respectively, from Saito et al. (1998) and Liu et al. (2006). Dates (▼) and total sulfur content (TS, ●) are plotted against age. The 5-point running average for TS is shown in red curve. **(c)** Temporal variation of total organic carbon (TOC, black dots). Carbonate-free TOC is shown in blue. **(d)** Temporal variations of $\delta^{13}$C of total organic carbon for MD012404 (○) and MD012403 (∘). **(e)** Sedimentary $\delta^{15}$N for MD012404 (○) and MD012403 (∘). Blue circles represent $\delta^{15}$N values for selected non-acidified samples in MD012404. Upper and lower horizontal dashed lines are for mean $\delta^{15}$N values of Kuroshio Intermediate Water (KIW) and the upper 400 m of Kuroshio, respectively. YD, H1 and H2 mark the periods of Younger-Dryas, Heinrich 1 and Heinrich 2, respectively.

Total organic carbon content (TOC) and total sulfur content (TS, in lower time resolution) for both cores have been reported previously (Kao et al., 2005, 2006b). Following their method for TS, we obtained a higher resolution TS record in this study. By using porosity, wet bulk density (Bassinot et al., 2002) and linear sedimentation rate derived from $^{14}$C dates we computed the mass accumulation rate (MAR, in mg cm$^{-2}$ yr$^{-1}$).

Organic carbon accumulation rate (CAR, in mgC cm$^{-2}$ yr$^{-1}$) was obtained by multiplying MAR by the TOC content. We also calculated the accumulation rate of remineralized organic carbon. The metabolized organic carbon corresponding to sulfur deposition was estimated with reasonable assumptions. Due to sufficient reactive iron for iron sulfide formation in seas surrounding Taiwan (Kao et al., 2004a, b, 2005, 2006b), it is assumed that all reduced sulfur has been fixed as sulfide minerals. Based on this assumption, we back calculated the amount of organic carbon metabolized via sulfate reduction by using molar stoichiometric ratio following Morse and Berner (1995). Multiplying this value by MAR, we compute the extent of the organic carbon metabolization (EOCM).

This paper largely focuses on the central trough, using some published supporting data from previous studies (Kao et al., 2006a, 2008) for discussion.

## 4 Results

Results for the two cores are presented in Figs. 2 and 3 along with relevant ancillary data. The plotted parameters include MAR, sea level curve, TS, TOC, $\delta^{13}C_{TOC}$ and $\delta^{15}N$ for Core MD012404. Also plotted are TOC, $\delta^{13}C_{TOC}$ and $\delta^{15}N$ from Core MD 012403.

The MAR ranges from $\sim$40 to 150 mg cm$^{-2}$ yr$^{-1}$ (black curve in Fig. 2a). The MAR peaked to about 100 mg cm$^{-2}$ yr$^{-1}$ during the Heinrich 2 cold period, the early stage of sea level rise (see sea level curve in Fig. 2b), and the Bølling/Allerød warm period; during other periods it remained near the background level, around 45 mg cm$^{-2}$ y$^{-1}$, except the late Holocene, when it gradually rose to 150 mg cm$^{-2}$ yr$^{-1}$ around 2 cal. ka BP.

The TS and TOC contents show different temporal patterns. TS was high (>0.2%, red curve in Fig. 2b) with peak values reaching 0.6% when the sea level was low. Since 17 cal. ka BP, the TS content decreased continually in association with increasing sea level, without corresponding fluctuations in the TOC content (Fig. 2c, black dot-curve), which varied from 0.48% to 0.80% peaking around 10 cal. ka BP. Note that dilution by carbonate had little effect on the TOC pattern, as carbonate-free TOC content shows the same variation pattern as does the original curve (see blue curve in Fig. 2c).

The $\delta^{13}C_{TOC}$ values range from −21.8 to −20.6‰(red circles in Fig. 2d) in MD012404. Core MD012403 in the southern trough shows a similar trend (open circles in Fig. 2d) but with consistently lower $\delta^{13}C_{TOC}$ (by $\sim$1‰, ranging from −23.2 to −21.8‰), apparently caused by inputs of terrestrial organics due to the proximity of the site to Taiwan. More information about sedimentary organic carbon supply to MD012403 can be found in Kao et al. (2008).

As for the N isotopes, the $\delta^{15}N$ in core MD012404 ranges from 4.4 to 5.8‰with a temporal pattern resembling that of MD012403 but with consistently higher values (Fig. 2e). This offset is similar to that found in $\delta^{13}C_{TOC}$ records. The lower bound of $\delta^{15}N$ is very close to that (3–4‰) of the terrestrial end-member of particulate matter in the Taiwanese rivers (Kao and Liu, 2000). This is consistent with the notion that sediments at the MD012403 site received significant terrestrial inputs. Because of the probable terrestrial influence at this site, we focus the discussion on records from Core MD012404, which should be more representative of the marine environment in the western North Pacific Ocean.

## 5 Discussion

### 5.1 Sediment-upper water column coupling

The sedimentary $\delta^{15}N$ records may be affected by the origin of nitrogen and diagenetic alteration. In a dominantly marine environment, biogenic particulate organic nitrogen (PON) is the main source of sedimentary nitrogen. The isotopic composition of biogenic PON is, in turn, controlled by the nitrogen source and the degree of utilization. In the oligotrophic Kuroshio water, the nutrient-rich subsurface water (e.g., Gong et al., 1996; Liu et al., 2000) and the surface dwelling nitrogen fixers (Minagawa and Wada, 1986; Liu et al., 1996) are the two major sources of fixed nitrogen.

According to Liu et al. (1996), the weighted mean $\delta^{15}N$ value of nitrate for the upper 400 m of the Kuroshio water is 3.3–4.7‰. The upper limit of this range is very close to the $\delta^{15}N$ values (4.4–4.6‰) for the surface sediment of MD012404 and previous reported values in the southern Okinawa Trough (Kao and Liu, 2003). Since the Kuroshio surface water (the upper 100 m) is nutrient-depleted with nanomolar nitrate concentrations (Chiang et al., 1997), the upwelled nitrate gets completely utilized. Consequently, the sinking PON should have isotopic composition very close to that of the subsurface nitrate without significant fractionation occurring during phytoplankton uptake (Altabet et al., 2001). The closeness of the nitrogen isotopic composition of the surface sediments with that of the subsurface nitrate reservoir suggests that the diagenetic alteration of the nitrogen isotopic composition during sediment deposition was not significant. Thus, it is reasonable to assume that the sedimentary $\delta^{15}N$ record of MD012404 tracked nitrogen isotopic changes in the overlying upper water column in the past as well.

It is noted that the nitrogen isotopic composition of nitrate in the subsurface Kuroshio water is significantly lighter than that in the KIW below 500 m. The KIW has a mean $\delta^{15}N_{NO_3}$ value of 5.6‰, which is close to the global mean (Sigman et al., 1999). As the KIW upwells towards the surface, nitrate uptake in the euphotic zone is expected to remove isotopically light nitrate leaving behind the $^{15}$N-enriched nitrate in the upper water column (Altabet et al., 2001). The trend observed in the upper water column in the KC contradicts this expected pattern, indicating that other processes dictate the nitrogen isotopic variation. It has been suggested that lowering of the $\delta^{15}N_{NO_3}$ value in the Kuroshio subsurface water may be due to remineralization of isotopically light PON from $N_2$-fixers (Liu et al., 1996).

## 5.2 Local environmental variation or signal from ETNP?

The temporal variation of sedimentary $\delta^{15}$N records reflects the variation of $\delta^{15}N_{NO_3}$ in the upper water column, which may be controlled by remote as well as local processes. We compare the MD012404 record with the sedimentary $\delta^{15}$N records (Fig. 3a) from the Eastern North Pacific (ENP) during last deglaciation (JPC-56 from the Gluf of California,Pride et al., 1999; and ODP-893A from the Santa Barbara Basin, Emmer and Thunell, 2000). Record of $\delta^{18}$O in GISP 2 ice core is also plotted (Fig. 3b) for comparison (Grootes et al., 1993). Despite the much reduced amplitude, fluctuations of the sedimentary $\delta^{15}$N in OT follow those in the Gulf of California quite closely. The $\delta^{15}$N fluctuations also correspond to fluctuations in $\delta^{18}$O in the GISP 2 ice core during the pre-Holocene period, suggesting close relationship with climate events. The two major $\delta^{15}$N peaks occurred during two warm periods: Bølling/Allerød and the period after Younger Dryas during transgression (Fig. 3b) resembling the trends observed not only in the ENP but also in the Arabian Sea (Ganeshram et al., 1995, 2000; Deutsch et al., 2004 and references therein).

A decreasing trend in $\delta^{15}$N since the beginning of the Holocene has been reported previously (Higginson et al., 2003; Meckler et al., 2007; Altabet, 2007; and references therein). It is attributed to decreasing WCD and increasing $N_2$-fixation. Synchronous changes in $\delta^{15}$N (though of much smaller amplitude) in the OT and their close correlation with the warm and cold events and with the temporal patterns observed in ETNP and Arabian Sea suggest that these brief events found in the OT were global in nature and climate-related.

However, changes in local environmental conditions, which have been quite large, might also contribute to $\delta^{15}$N variation in the past 25 kyr (e.g., Kao et al., 2005, 2006a). The past environmental variation can be constructed from geochemical properties of the sediment core. It has been proposed that local WCD could be an important process in providing $^{15}$N-enriched nitrate to the upper water column (e.g., Meckler et al., 2007). However, conditions conducive

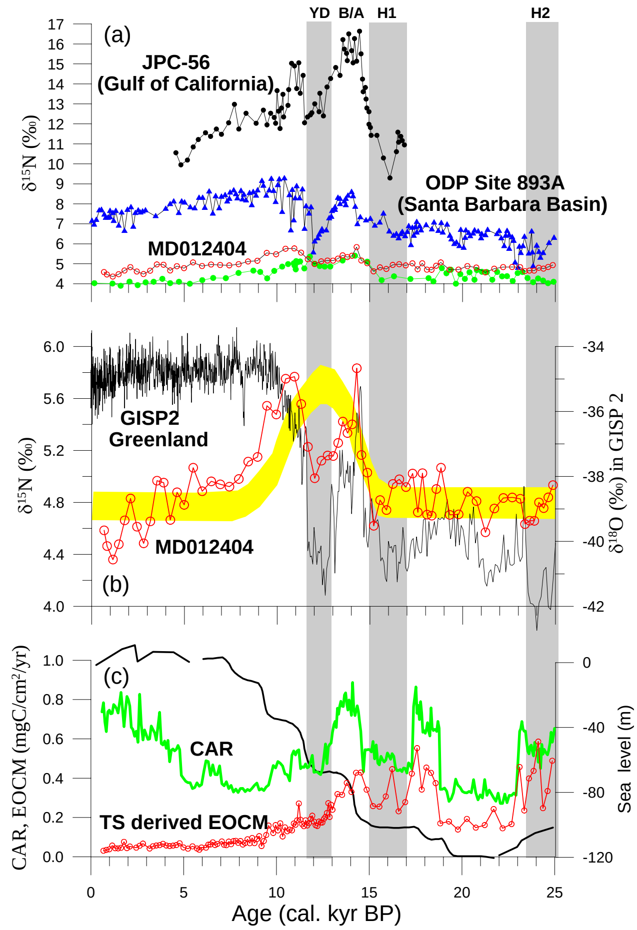


**Fig. 3.** **(a)** Temporal trends of sedimentary $\delta^{15}$N for MD012404 (○), MD012403 (●), JPC-56 in the Gulf of California (•) and ODP 893A in the Santa Barbara Basin (▲). **(b)** Sedimentary $\delta^{15}$N for MD012404 (○) in enlarged scale. Record of $\delta^{18}$O for GISP2 ice core (black curves) is shown for comparison. Yellow zone represents the pattern of mean ocean $\delta^{15}$N of nitrate (Deustch et al., 2004). **(c)** Enlarged plots of accumulation rates for organic carbon (CAR, in green) and the extent of organic carbon metabolization (EOCM, in red) derived from total sulfur (see text).

for occurrence of WCD –complete oxygen consumption – develop only in selected areas of the oceans, mostly along the oceans' eastern boundaries, where subsurface water renewal is sluggish and oxygen demand is high due to high surface productivity. It is debatable whether such conditions could have occurred in OT in the past. For the sake of argument, we will discuss at what time the local environment might favor the occurrences of such conditions, if they had ever occurred.

Higher TOC burial in sediments generally reflects higher productivity (e.g., Calvert et al., 1995) and/or lower oxygen content of the overlying water column. However, organic carbon supply and the redox condition in the water column are closely related, because high organic carbon supply from the surface may result in rapid consumption of dissolved oxygen in the underlying water column and thus create condition conducive for WCD, such as in ETNP. It is, however, hard to determine the relative importance of the two factors. Since WCD is mainly affected by organic carbon supply and the redox condition in the water column, past record of $\delta^{15}$N may also be regarded as index of WCD potential in the OT.

In fact, the TOC in sediments represents only the fraction of organic matter that escaped degradation during transportation and burial processes. It has been shown that the OT is not a steady depositional environment, where the sedimentation rate has fluctuated substantially in the past. The TOC content may be diluted (see the Holocene period in Fig. 2c) to different extents by the lithogenic materials. Consequently, the CAR (green curve in Fig. 3c) is a better indicator that is not compromised by mineral dilution during sedimentation. On the other hand, a significant fraction of organic matter gets oxidized during burial and a major fraction of the oxidation is attributed to sulfate reduction (Berner, 1984). Therefore, CAR together with the EOCM should provide a good indication of the organic supply down the water column and the potential of WCD as well.

Here we apply CAR together with the EOCM (Fig. 3c) to evaluate the magnitudes of organic supply and its remineralization. Except the late Holocene, the temporal pattern of EOCM resembled that of the CAR, which showed three peaks occurring, respectively, during the Heinrich 2 (H2) period, the very early stage of sea rise prior to Heinrich 1 (H1) (Hemming, 2004), and the Bølling/Allerød period. Based on high CAR and EOCM, high WCD potential is inferred for the three periods. For the late Holocene since 7 ka, faster mass accumulation (comprising mainly lithogenic materials) resulted in a dilution of TOC content, yet high CAR values.

Had the local water column denitrification been responsible for producing the $^{15}$N-enriched nitrate, the two peaks of sedimentary $\delta^{15}$N in MD012404 (Fig. 3b), it should have happened during the three periods of high WCD potential. However, only one $\delta^{15}$N peak (around 14 kyr cal. B.P.) was associated with a maximum in CAR. During the two other periods of higher WCD potential, no elevation of sedimentary $\delta^{15}$N was detected. One of the $\delta^{15}$N peaks occurred around 10 kyr cal. B.P. corresponding to a peak in the TOC record, which deserves attention. The MAR values around this time were lower thus resulting in a modest CAR. Usually fresh marine organics are the best quality organic matter and discernible by their carbon isotopic composition. A close examination of the $\delta^{13}C_{TOC}$ record of MD012404 (red circles in Fig. 2d) shows relatively higher values during the sea level lowstand, whereas, slightly lower $\delta^{13}C_{TOC}$ values appear during transgression and the Holocene. However, the variability of $\delta^{13}C_{TOC}$ in entire MD 012404 is small ($-20.6$ to $-21.8$‰) with a mean value of $-21.2$+0.3‰reflecting mainly the marine source of the organics (Goericke and Fry, 1994; Meyers, 1997). The consistently marine-like $\delta^{13}C_{TOC}$ and such a small variability of $\delta^{13}C_{TOC}$ during the entire 30 ka period indicate an unimportant effect of terrestrial inputs to the central OT. Based on $\delta^{13}C_{TOC}$ data, we suggest that the high TOC during the Holocene is due to lower mineral dilution and higher portion of reworked marine organics that previously resided on the shelf.

It is noted that high organic supply alone does not necessarily lead to the suboxic condition necessary for WCD to occur. Sluggish water exchange is also required for the suboxic condition to develop. If the suboxic condition was to develop in the lower water column in the OT in the past, the bottom sediments would experience greater anoxia, enhancing sulfur deposition through sulfate reduction.

The TS content was considerably higher during the glacial period and early deglaciation stage, suggesting existence of more intense reducing condition in the sediments of the OT then, which has been attributed to weaker ventilation (Kao et al., 2006a, b). The relationship between sea level and deep water ventilation was investigated using a 3-D model by Kao et al. (2006b). The model revealed a reduction of KC throughflow due to the bifurcation of the flow before entering the Suao-Yonaguni Pass (with ∼30% of the flow getting diverted away from OT; see Fig. 1b) during the glacial period. This would lead to weaker ventilation in deepwater; at the same time, KC outlet switched from Tokara Strait to Kerama Gap (see Fig. 1b). A similar trend in TS content (in terms of age) was found in core MD012403 in the southern OT (not shown) despite a ∼4 times higher sedimentation rate (Kao et al., 2005). This indicates that bottom water circulation change was a trough-wide phenomenon. There is also evidence for a transformation from foliation to anomalous sedimentary magnetic fabric (dynamic depositional environment) in the southern OT when sea level reached −40 m during the early Holocene (Kao et al., 2005). Thus, it appears that sea level change affected the KC throughflow resulting in changes in deepwater circulation, and consequently oxygen supply to the deepwater, depositional environment and sedimentary biogeochemistry. Down-core diagenetic sulfate reduction might have contributed to the TS profile to some degree; however, the main temporal pattern was probably caused by circulation changes (Kao et al., 2006b). In addition, fossil records from the sediment core show that relatively high abundance of benthic fauna (unpublished data) before 17 cal. ka BP during low sea level stand. This implies higher food (organic detritus) supply to seafloor when the sea level was low, again supporting higher WCD potential.

From the above discussion, it follows that, if at all, conditions suitable for WCD to occur could have developed only when the sea level was lower, especially during the three periods of high supply rate of organic carbon. Yet, only one of the two sedimentary $\delta^{15}$N peaks occurred during the period of relatively high TS, when the bottom water ventilation in the OT was limited and deepwater oxygen levels are expected to have been lower. Significantly, no increase in $\delta^{15}$N occurred prior to the Bølling/Allerød warm period, when the TS was in its maximum. In contrast, the other sedimentary

$\delta^{15}$N peak occurred when both the CAR and the TS were much lower. We therefore consider it very unlikely that local WCD was responsible for the elevated sedimentary $\delta^{15}$N values. Similarly decoupled nitrogen isotopic compositions of nitrate in the upper water column and in the bottom water have also been observed in the sea off southern California (Liu and Kaplan, 1989).

It may thus be concluded that the most likely origin of the heavy $\delta^{15}$N signals during the deglaciation was in the ETNP, and if local WCD ever occurred in the OT in the past, its effect on the sedimentary $\delta^{15}$N records is not detectable.

### 5.3 $\delta^{15}$N records in the South China Sea

The range of $\delta^{15}$N values in the two cores examined by us is similar to those observed in 17940-2 (Kienast, 2000) and 1144 (Higginson et al., 2003) from the northern SCS (see Fig. 1b for locations). Unlike the $\delta^{15}$N records from the OT, those from the SCS did not show any recognizable pattern despite the coring sites being close to each other.

It has been reported that $N_2$-fixation could be important in the SCS (Wong et al., 2002). Moreover, basin-wide deep ventilation down to 2000 m occurs in the SCS (Chao et al., 1996). Both processes might have contributed to attenuation of the ETNP denitrification signal in the SCS. Variations in surface and subsurface circulations and exchanges, such as Kuroshio intrusion, are known to occur due to climate fluctuations (Qu et al., 2004). Similar or more pronounced changes very likely occurred in the past due to climate and sea level changes. Consequently, the inflows of water masses at different density levels, which probably had nitrate with different $\delta^{15}$N values, probably varied considerably in the past. These complicated processes that may have led to the unrecognizable patterns of temporal variations of $\delta^{15}$N during the last deglaciation have been described previously (Higginson et al., 2003). On the other hand, the terrestrial input from different sources might also affect the $\delta^{15}$N record in the SCS (Kienast et al., 2005). By comparison, the relatively simple circulation in the open North Pacific Ocean allows the transmission of the signal across the entire basin from the eastern tropical Pacific (Fig. 1a) without losing its integrity, resulting in a clear pattern of the $\delta^{15}$N variability in the OT.

### 5.4 Implications

There is now sufficient evidence to show that the oxygen minimum zone (OMZ) in northern and eastern Pacific was more intense than it is today during the deglaciation, interrupted by the Younger Dryas event (Behl and Kennett, 1996; Cannariato and Kennett, 1999; Zheng et al., 2000; Ivanochko and Pederson, 2004; Mckay et al., 2005). Whether these events were caused by increased export production or suppressed ventilation remains debatable, though. Recently, Galbraith et al. (2004) proposed that $O_2$ supply from high latitudes to the tropical intermediate waters might exert a key control on denitrification and the coupled nitrogen fixation, with both processes intensifying (weakening) during interglacial (glacial) periods. The conceptual model explains well most temporal variations of $\delta^{15}$N in various oceanic settings.

In our study area in the western North Pacific, synchronous increases in $\delta^{15}$N during the two warm periods indicate that the signal of enriched $^{15}$N due to enhanced denitrification in ETNP might have been transmitted to the western North Pacific during the last deglaciation. The small amplitude of sedimentary $\delta^{15}$N changes in the OT was likely caused by reservoir dilution (Deutsch et al., 2004) or by counteraction of regional $N_2$-fixation (Deutsch et al., 2007). The upper bound of $\delta^{15}$N value in MD012404 is close to that in the North Pacific Deep Water (Fig. 2e) and the lower bound is close to the integrated mean $\delta^{15}$N of nitrate in the upper 400 m of KC (3.3–4.7‰). Except the transgression, no distinctive shift in sedimentary $\delta^{15}$N has been found between glacial and interglacial periods. Thus, it would be reasonable to conclude that the isotopic composition of nitrate in the upper ocean was the same during the two climate states as suggested by Kienast (2000). On the other hand, it is conceivable that the nitrate reservoir in the upper water column had an elevated mean $\delta^{15}$N value during these warm periods reflecting a decrease in the nitrogen inventory during the transgression period.

The low frequency variation of $\delta^{15}$N follows the changing trend of mean ocean $\delta^{15}$N in nitrate (yellow zone in Fig. 3b) as suggested by Duetsch et al. (2004). We conclude that such substantial $\delta^{15}$N changes in a region very distant from the intensive denitrifying zones in the ETNP lend support to the notion of significant changes in N inventory in the North Pacific during deglaciation. Our results can provide valuable constraints for model simulations of the nitrogen cycle during the last glacial-deglacial cycle.

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