

## NOTE

# Sedimentary organic carbon and nitrogen contents and their isotopic compositions along two transects on the East China Sea shelf

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Surficial sediment samples were collected from two transects on the shelf of the East China Sea: one across the shelf and the other along the coast, both starting near the Changjiang river mouth. They were analyzed for contents of total nitrogen (TN%) and total organic carbon (TOC%) and for their isotopic compositions ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}_{\text{TOC}}$ ). The TOC% and TN% values varied concomitantly in the ranges of 0.2 to 1.5% and 0.02 to 0.19%, respectively. The  $\delta^{13}\text{C}_{\text{TOC}}$  value ranged from  $-25.1$  to  $-19.6\text{‰}$ . The lower  $\delta^{13}\text{C}_{\text{TOC}}$  values ( $<-21.5\text{‰}$ ) all occurred in the inner shelf corresponding to higher TOC% ( $>0.5\%$ ). Away from the coastal zone, TOC% dropped sharply with corresponding increase in the  $\delta^{13}\text{C}_{\text{TOC}}$  value. The  $\delta^{15}\text{N}$  value, varying from 3.6 to 4.9‰, displayed a more complicated distribution pattern. Distinctively lower  $\delta^{15}\text{N}$  values ( $<4.1\text{‰}$ ) occurred near the Changjiang river mouth and near the shelf edge. Isotopically light riverine nitrogenous materials and preferential uptake of isotopically light nitrogen in the rich supply of nutrients in the Changjiang plume may have contributed to the observed low values near the river mouth, while nitrogen fixers in the Kuroshio waters may have contributed to those near the shelf break.

## INTRODUCTION

The East China Sea (ECS) receives large quantities of terrigenous materials from the northeast Asia. The riverine nutrient fluxes and the marine nutrient inputs from onwelling subsurface waters sustain an active biological productivity in the shelf sea (Gong *et al.*, 1996, 2000; Chen *et al.*, 1999). In order to understand the origins of the sedimentary organic matter in the ECS shelf, we use stable isotopes as tools to trace the sources of organic carbon and nitrogen in sediments.

Generally, the terrigenous materials are more depleted in  $^{15}\text{N}$  and  $^{13}\text{C}$ , while the autochthonous marine organic matter are usually more enriched in  $^{15}\text{N}$  and  $^{13}\text{C}$  (Peters *et al.*, 1978; Sweeney and

Kaplan, 1980; Wada *et al.*, 1987). In the surface ocean, the preferential uptake of isotopically light nitrogen by primary producers occurs, when availability of dissolved inorganic nitrogen is high (Mariotti *et al.*, 1981; Altabet and Francois, 1994). Therefore, nitrogen isotopic composition of sediments can be used to study nutrient dynamics in the overlying water (e.g., Calvert *et al.*, 1992; Nakatsuka *et al.*, 1995).

Previous studies of ECS sediments, which mostly focused on sedimentological and mineralogical parameters (e.g., DeMaster *et al.*, 1985; Milliman *et al.*, 1985; Saito and Yang, 1994), hold scant attention to other properties that might be helpful for discerning the carbon and nitrogen cycles. Sheu *et al.* (1995) studied the organic car-

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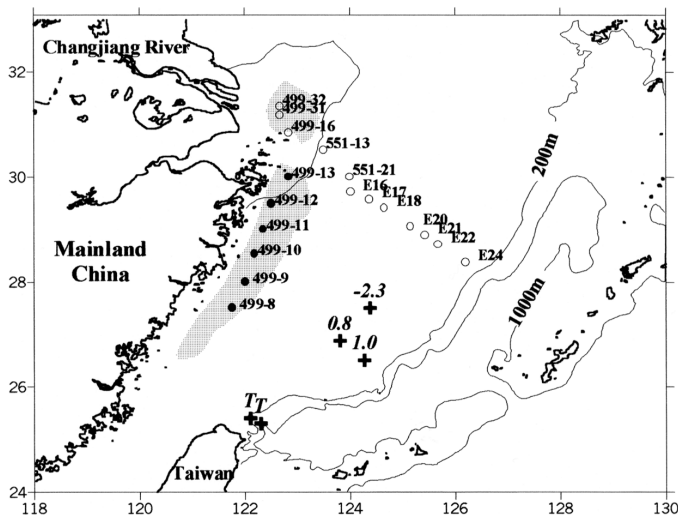


Fig. 1. Sites for the sediment coring on the East China Sea shelf. The open circles are for the cross-shelf transect and the solid circles for the coastal transect. The shaded areas indicate the mud patches (after Hu *et al.*, 1998). The solid curve are the seasonally averaged isohaline of 29 psu in summer (Chen, 1992), indicating the mean position of the Changjiang plume. The crosses indicate stations where abundant *Trichodesmium* (labeled by **T**) or isotopically light particulate nitrogen was found. Lines with numericals indicate bottom depths.

bon content and its isotopic composition in the ECS sediments, but none of their samples were collected in the coastal zone of China and no nitrogen properties were determined.

The Kuroshio Current (KC) flows northward along the shelf edge in the ECS (Beardsley *et al.*, 1985). The primary nutrient source for the ECS shelf is the persistent KC upwelling (Gong *et al.*, 1996; Liu *et al.*, 2000). Changjiang River runoff is the second important nutrient source. The Changjiang Diluted Water (CDW) spreads mainly to the northeast in summer (as shown in Fig. 1) and flows southward along the coast as a jet in winter (Chao, 1991). Other sources, including the northward flow through the Taiwan Strait (Chung *et al.*, 2001) and nitrogen fixation (Chang *et al.*, 2000), also contribute to the nitrogen supply to the ECS. In summer, the primary production showed distinctive high values (up to 1900 mgC/m<sup>2</sup>/d) within the Changjiang plume (Chen *et al.*, 1999; Gong *et al.*, 2000).

The sediments on the ECS shelf originate mainly from Changjiang discharge (~500 MT/y;

Milliman and Meade, 1983). Although the Yellow River discharges annually ~1000 MT sediments into the Gulf of Bohai, less than 2% of these modern sediments can be transported to the south of 32°N by the Yellow Sea Coastal Current (DeMaster *et al.*, 1985). The fine-grained sediments from the Changjiang runoff may disperse eastward or northeastward in summer following the Changjiang plume (Beardsley *et al.*, 1985; Sternberg *et al.*, 1985). The final destiny of sediments appears to be dominated by the China Coastal Current, which flows southward in winter (Chao, 1991) and distributes sediments along the coastal zone (Fig. 1), where deposition takes place (Saito and Yang, 1994).

In order to understand isotopic characteristics of surficial sediments deposited on the ECS shelf, we selected 18 samples (Fig. 1). Samples cover different regimes: the river mouth, and the inner, middle and outer shelves. We analyzed the TOC%, TN%,  $\delta^{13}\text{C}_{\text{TOC}}$  and  $^{15}\text{N}$  of these sediments to trace their possible origins.

Table 1. Sampling locations and the relative distance to Station 499-32

Station	Distance (km)	Latitude (°N)	Longitude (°E)
Cross-shelf transect			
499-32	0	31.333	122.668
499-31	16	31.167	122.667
499-16	60	30.833	122.833
551-13	121	30.508	123.497
551-21	170	30.001	124.001
E-16	225	29.716	124.013
E-17	254	29.581	124.377
E-18	286	29.416	124.654
E-20	348	29.053	125.153
E-21	380	28.893	125.437
E-22	411	28.717	125.683
E-24	473	28.375	126.196
Coastal transect			
499-13	148	30.003	122.834
499-12	205	29.492	122.509
499-11	260	29.007	122.351
499-10	314	28.534	122.182
499-9	375	28.003	122.011
499-8	434	27.502	121.764

## MATERIALS AND METHODS

The coordinates and water depth of the sampling stations are listed in Table 1. Eighteen samples were selected distributing along two transects (Fig. 1): one across the shelf and the other along the coast, both starting from the Changjiang river mouth. Samples were taken on board R/V *Researcher-I* on Cruise 499 (June 1999), Cruise 551 (August 1999) and the *KEEPMASS* Cruise (July 1992). Sediments were collected with box corer. Subsamples from the box cores were obtained with plastic liners of 5.3 cm in inner diameter and 60 cm in length. Sealed plastic liners were preserved in the freezer. Upon returning to the laboratory, we collected surficial sediments by scraping the surface 0–2 cm sediment with a stainless steel spatula. They were stored in plastic vials, and then placed in a freeze-drier for one week to dry. The dried sediments were later ground to powder with pestle and mortar.

About 0.2 g of powdered samples were acid-treated with 1N HCl for 3 days in pre-combusted (500°C) glass test-tube and then centrifuged for

the removal of solution. The adsorbed  $\text{NH}_4^+$  was generally eliminated during this pretreatment, yet the  $\text{NH}_4^+$  fixed in the clay is not removed. The acid-treated sediments were dried at 60°C.

A handful of box core samples from the same sites (but not the same subcores) were treated with desalting after decarbonation. For these samples, 5 g of non-pulverized bulk sample was treated with 2N HCl for 3 days followed by a twice rinse with deionized water. Since desalting after acid treatment of sediment samples has been recommended, a comparison was made between results obtained for samples treated with or without desalting.

Carbon and nitrogen isotope samples were prepared using sealed-tube combustion (Wedeking *et al.*, 1983). Decarbonated samples were transferred into a pre-combusted copper capsule, which is then put into the 9-mm quartz tube with 1 g each of CuO and Cu pellets and 10 cm of silver thread. The quartz tubes are evacuated, sealed and then heated at 900°C for 3 hours, kept at 650°C for over 12 hours, and then cooled to room temperature.  $\text{N}_2$  and  $\text{CO}_2$  gases were extracted and purified in a vacuum line and collected in sealed 6-

mm glass tubes. Abundances of carbon and nitrogen were measured manometrically. The precision for nitrogen and carbon content were  $\pm 0.005\%$  and  $\pm 0.01\%$ , respectively. Carbon isotopic compositions were determined in a Finnigan Delta-Plus mass spectrometer. Nitrogen isotopic compositions were determined in a Micromass 602E mass spectrometer. Results are presented in the standard  $\delta$  notation with respect to standards of PDB carbon and atmospheric nitrogen.

## RESULTS AND DISCUSSION

The results of analyses are presented in Fig. 2. The distances to Station 499-32 are used for data presentation for both transects. The values of TOC% ranged from 0.20 to 1.49% with higher values ( $>0.5\%$ ) found near the river mouth and in the coastal transect (Fig. 2(a)). The content of TOC increased slightly and then dropped sharply within 50 km from the river mouth along the cross-shelf transect. The distribution of TN% follows that of TOC% closely. The highest value of TOC% and TN% almost all occurred in the mud patches along the coast shown in Fig. 1 (Hu *et al.*, 1998). Values of TOC% and TN% (0.02–0.19%) are positively correlated. Figure 3 shows that sediments with high organic contents have C/N ratios higher than the Redfield ratio of 6.63, while those with lower organic contents have C/N ratios approaching the Redfield ratio. The elevated C/N ratios in the coastal zone are consistent with higher contributions of organic matter from terrestrial sources, which usually show higher C/N ratio (C/N atomic ratio  $\geq 12$ ; Kukal, 1971).

The  $\delta^{13}\text{C}_{\text{TOC}}$  values obtained for samples treated with two different pretreatment procedures (Table 2) are reasonably consistent, except samples from two sites which show much lower values obtained from desalted samples. Aside from these two sites, the values from the non-desalted samples are higher by 0.18‰ on average. It is not clear what caused such an offset. If some of the  $\text{CO}_2$  was retained by reacting with the calcium ion left in the non-desalted sample, the removed  $\text{CO}_2$  would be enriched in  $^{13}\text{C}$  under equilibrium con-

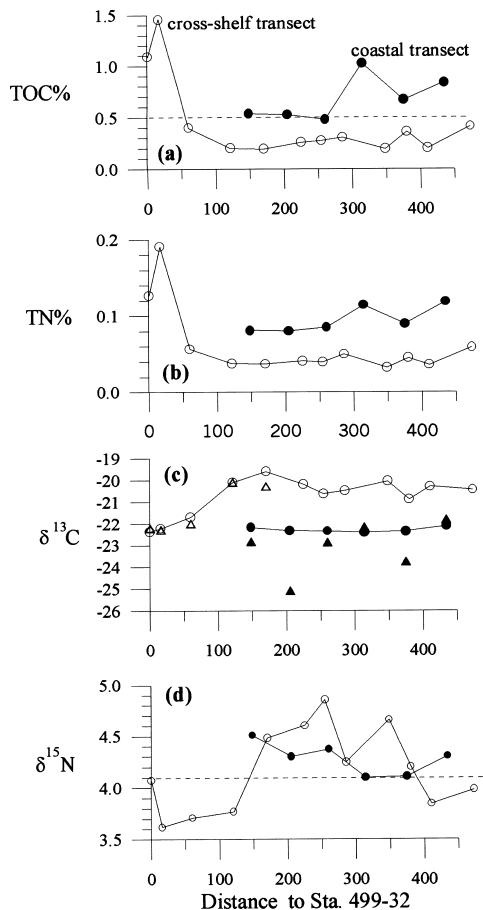


Fig. 2. The variations of (a) total organic carbon content, (b) total nitrogen content, (c)  $\delta^{13}\text{C}_{\text{TOC}}$  and (d)  $\delta^{15}\text{N}$  along the two transects. The distances are measured with respect to Site 499-32. Solid and open symbols are for coastal and cross-shelf transects, respectively. Triangles stand for desalted samples.

ditions. In other words, this would make the measured  $\delta^{13}\text{C}$  values lower, not higher. Therefore, the consistently higher  $\delta^{13}\text{C}$  values of the non-desalted samples suggest that the centrifuging process during the preparation of the non-desalted samples removed most of the salt generated by acidification. On the other hand, the consistently lower  $\delta^{13}\text{C}$  values of the desalted samples suggest that some isotopically heavier labile fraction of the organic carbon may have been lost in the preparation procedure (Schubert and Nielsen, 2000). It is cautioned that there are other processes (such as

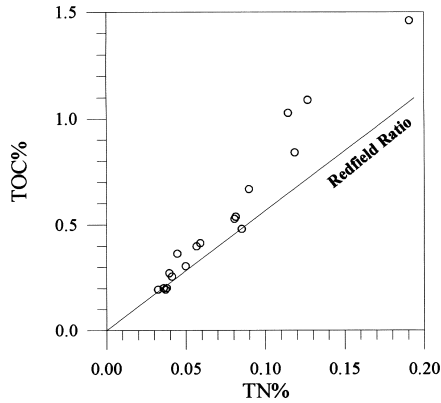


Fig. 3. Plot of total organic carbon content vs. total nitrogen content. The line indicates the Redfield ratio of C/N = 6.63.

reaction kinetics) that may cause alteration in the isotopic composition during sample preparation. The isotope effects associated with such procedures are not well understood and warrant further investigation. The two samples that show very low values ( $-25.1$  and  $-23.8$ ‰) obtained from desalting procedure were collected in the coastal zone, where high heterogeneity in sediments was found. We have found that clay and silt fractions from coastal sediments have  $\delta^{13}\text{C}_{\text{TOC}}$  values as different as  $-20.3$ ‰ vs.  $-26.7$ ‰. It is likely that the differences in  $\delta^{13}\text{C}_{\text{TOC}}$  values for the two coastal samples could have resulted from inhomogeneity of sediments.

In Fig. 2(c), it is interesting to note that all lower values of  $\delta^{13}\text{C}_{\text{TOC}}$  ( $-25.1$  to  $-22.1$ ‰) occurred at stations near the river mouth or along the coast, apparently due to a supply of large amount of organic matter from land. This is consistent with the notion of Saito and Yang (1994), that the coastal mud belt is enriched with terrigenous organic matter. Corresponding to the decrease of TOC seaward from the Changjiang river mouth, both sets of  $\delta^{13}\text{C}_{\text{TOC}}$  values showed a significant increase of about  $2.0$ ‰, suggesting a shift towards increasing contribution from marine sources of organic matter (Fig. 2(c)). This also indicates that deposition of terrigenous materials is confined to the inner shelf.

Table 2. The carbon isotope compositions obtained from two decarbonation pretreatments (see text)

Station	Centrifuged	Desalted
499-32	$-22.4$	$-22.2$
499-31	$-22.2$	$-22.3$
499-16	$-21.7$	$-22.0$
499-13	$-22.2$	$-22.8$
499-12	$-22.3$	$-25.1$
499-11	$-22.3$	$-22.9$
499-10	$-22.4$	$-22.2$
499-9	$-22.4$	$-23.8$
499-8	$-22.1$	$-21.8$
551-13	$-20.1$	$-20.1$
551-21	$-19.6$	$-20.3$

Although the nitrogen content of sediments was well correlated with the organic carbon content, the variation of  $\delta^{15}\text{N}$  value of sedimentary nitrogen did not follow the simple pattern of isotopic variation of sedimentary organic carbon (cf., Figs. 2(c) and (d)). The incongruent behavior suggests that processes other than simple mixing of isotopically uniform end-members, namely the terrigenous and the marine organic matter, controlled the isotopic composition of sedimentary nitrogen.

Figure 2(d) shows that samples from the along coast transect had  $\delta^{15}\text{N}$  values between  $4.1$  and  $4.5$ ‰ with a mean of  $4.3$ ‰. Most of these samples had  $\delta^{13}\text{C}_{\text{TOC}}$  values between  $-22.4$  and  $-22.1$ ‰ with a mean of  $-22.3$ ‰, from which we calculated the mean terrigenous fraction of  $\sim 40\%$  for the organic carbon under the assumption of typical carbon isotopic compositions for the terrestrial ( $-26$ ‰; Smith and Epstein, 1971; Newman *et al.*, 1973) and marine ( $-20$ ‰; Goericke and Fry, 1994) end members. For the  $\delta^{15}\text{N}$  values, we took that ( $4.7 \pm 0.6$ ‰) reported for the nitrate in the Kuroshio upwelling water (Liu *et al.*, 1996) for the marine end member, and  $+2.0$ ‰ (Wada *et al.*, 1987) for the terrestrial end member. For the C/N ratios we also assumed the typical values of 6.63 (Redfield *et al.*, 1963) for the marine end member and 20 for the terrestrial one, which is a compos-

ite of warm mixed woodlands and cool deciduous forests (Aitkenhead and McDowell, 2000). Then the calculated mean  $\delta^{15}\text{N}$  value for these samples was 4.2‰, which matched well with the observations.

The  $\delta^{15}\text{N}$  values along the cross-shelf transect showed more variability. The highest  $\delta^{15}\text{N}$  values (4.2–4.9‰) occurred in the middle shelf, where the mean  $\delta^{15}\text{N}$  value was  $4.5 \pm 0.2\text{‰}$ , which is close to the  $\delta^{15}\text{N}$  value of nitrate in the Kuroshio upwelling water (Liu *et al.*, 1996). The high  $\delta^{13}\text{C}$  values (–20.8 to –19.6‰) of these samples also indicate a high fraction (>85%) of marine organic materials. The calculated mean  $\delta^{15}\text{N}$  value was 4.6‰, which matched well with the observed mean.

It is noteworthy that the samples near the river mouth had  $\delta^{13}\text{C}$  values similar to those from the coastal transect but had distinctively lower  $\delta^{15}\text{N}$  values. One likely reason for it was due to additional sources of isotopically light nitrogen. The most important source of nutrients in this area is apparently the river runoff (Gong *et al.*, 1996). Unfortunately no isotopic data of the dissolved inorganic nitrogen (DIN), namely, ammonia and nitrate, in the Changjiang runoff are available. It is reasonable to assume that the DIN in the Changjiang runoff, which originates from the terrestrial nitrogen pool, is isotopically light, similar to the terrigenous organic nitrogen. If so, it is conceivable that the autochthonous marine organic nitrogen in the Changjiang plume would be isotopically lighter than typical marine organic nitrogen. Moreover, the Changjiang plume often has excess nitrate (Gong *et al.*, 1996; Wong *et al.*, 1998), which would allow isotope fractionation during uptake of nitrate by phytoplankton (Wada and Hattori, 1978; Mariotti *et al.*, 1981). This would also result in production of isotopically light organic nitrogen (Altabet and Francois, 1994). The isotopically light sedimentary nitrogen appeared to be confined to the area near the river mouth, suggesting that the autochthonous organic nitrogen produced in the Changjiang plume might be rapidly remineralized in the water column or diluted by other isotopically heavier

nitrogenous materials during its dispersion away from the river mouth.

Another group of samples with low  $\delta^{15}\text{N}$  values was from the shelf break region, dominated by the Kuroshio Water. The highest *Trichodesmium* abundance in the ECS shelf was observed in summer in the Kuroshio Water near the shelf break (see Fig. 1, Chang *et al.*, 2000). Isotopically light particulate nitrogen ( $\delta^{15}\text{N} = -2.3$  to  $1.0\text{‰}$ ) has been observed in the outer shelf of the East China Sea in August 2000 (K. L. Chen, unpublished data). Thus, the input of isotopically light nitrogen from  $\text{N}_2$ -fixation was probably responsible for these low  $\delta^{15}\text{N}$  values. Assuming the  $\delta^{15}\text{N}$  of the nitrogen fixer to be 0‰ (–2‰ to 1‰; Minagawa and Wada, 1986), we calculated the fraction of organic nitrogen in these samples derived from nitrogen fixers to be 15–28%.

Compared to the high  $\delta^{15}\text{N}$  values of 7–9‰ determined for the fine-grained sediments from Huanghe by Mayer *et al.* (1998), the  $\delta^{15}\text{N}$  values observed in this study were considerably lower. It is not clear what makes the particulate nitrogen from Changjiang and Huanghe so different isotopically. However, our observations are in good agreement with the notion by DeMaster *et al.* (1985) that little sediments from Huanghe can be transported to the south of 32°N.

It is cautioned that selective decomposition and diagenesis may cause isotopic variation of organic matter in the sediments (Cifuentes *et al.*, 1988), which certainly warrants careful study. Here we only present the interesting phenomenon of decoupled variation of carbon and nitrogen isotopic compositions in the sedimentary organic matter from the ECS shelf and make suggestions on the probable causes for it. Further study is necessary to validate the hypotheses posted here. Most important of all, the isotopic compositions of nitrogenous materials in the Changjiang runoff must be determined.

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## REFERENCES

- Aitkenhead, J. A. and McDowell, W. H. (2000) Soil C/N ratio as a predictor of annual riverine DOC flux at local and global scale. *Global Biogeochem. Cycles* **14**, 127–138.
- Altabet, M. A. and Francois, R. (1994) The use of nitrogen isotopic ratio for reconstruction of past changes in surface ocean nutrient utilization. *NATO ASI Series, Carbon Cycling in the Glacial Ocean: Constraints on the Oceans's Role in Global Change* (Zahn, R. et al., eds.), 281–306, New York-Heidelberg-Tokyo, I17.
- Beardsley, R. C., Limeburner, R., Yu, H. and Cannon, G. A. (1985) Discharge of the Changjiang (Yangtze River) into the East China Sea. *Cont. Shelf Res.* **4**, 57–76.
- Calvert, S. E., Nielsen, B. and Fontugne, M. R. (1992) Evidence from nitrogen isotope ratios for enhanced productivity during formation of eastern Mediterranean sapropels. *Nature* **359**, 223–225.
- Chang, J., Chiang, K. P. and Gong, G. C. (2000) Seasonal variation and cross-shelf distribution of the nitrogen-fixing cyanobacterium, *Trichodesmium*, in southern East China Sea. *Cont. Shelf Res.* **20**(4–5), 479–492.
- Chao, S.-Y. (1991) Circulation of the East China Sea, a numerical study. *J. Oceanogr. Soc. Japan* **46**, 273–295.
- Chen, G. (1992) *Marine Atlas of Bohai Sea, Yellow Sea and East China Sea, Hydrography*. China Ocean Press, Beijing.
- Chen, Y. L. L., Lu, H. B., Shiah, F. K., Gong, G. C., Liu, K. K. and Kanda, J. (1999) New production and f-ratio on the continental shelf of the East China Sea: comparisons between nitrate inputs from the subsurface Kuroshio current and the Changjiang River. *Estuar., Coast. Shelf Sci.* **48**, 59–75.
- Chung, S. W., Jan, S. and Liu, K. K. (2001) Nutrient fluxes through the Taiwan Strait in spring and summer 1999. *J. Oceanogr.* **57**, 47–53.
- Cifuentes, L. A., Sharp, J. H. and Fogel, M. L. (1988) Stable carbon and nitrogen isotope biogeochemistry in the Delaware estuary. *Limnol. Oceanogr.* **33**, 1102–1115.
- DeMaster, D. J., Mckee, B. A., Nittrouer, C. A., Qian, J. C. and Cheng, G. D. (1985) Rates of sediment accumulation and particle reworking based on radiochemical measurements from continental shelf deposits in the East China Sea. *Cont. Shelf Res.* **4**, 143–158.
- Goericke, R. and Fry, B. (1994) Variations of marine plankton  $\delta^{13}\text{C}$  with latitude, temperature, and dissolved  $\text{CO}_2$  in the world ocean. *J. Neurol.* **8**, 85–90.
- Gong, G. C., Chen, Y. L. L. and Liu, K. K. (1996) Chemical hydrography and chlorophyll-*a* distribution in the East China Sea: implications in nutrient dynamics. *Cont. Shelf Res.* **16**, 1561–1590.
- Gong, G. C., Shiah, F. K., Liu, K. K., Wen, Y. H. and Liang, M. H. (2000) Spatial and temporal variation of chlorophyll a, primary productivity and chemical hydrography on the south East China Sea. *Cont. Shelf Res.* **20**, 411–436.
- Hu, D., Saito, Y. and Kempe, S. (1998) Sediment and nutrient transport to the coastal zone. *Asia Change in the Context of Global Climate Change* (Galloway, J. N. and Melillo, J. M., eds.), IGBP book series, 245–270, Cambridge University Press.
- Kukul, Z. (1971) *Geology of Recent Sediments*. Academic Press, N.Y., 490 pp.
- Liu, K. K., Su, M. J., Hsueh, C. R. and Gong, G. C. (1996) The nitrogen isotopic composition of nitrate in the Kuroshio water northeast of Taiwan: evidence for nitrogen fixation as a source of isotopically light nitrate. *Mar. Chem.* **54**, 273–292.
- Liu, K. K., Tang, T. Y., Gong, G. C., Chen, L. Y. and Shiah, F. K. (2000) Cross-shelf and along-shelf nutrient fluxes derived from flow fields and chemical hydrography observed in the southern East China Sea off northern Taiwan. *Cont. Shelf Res.* **20**(4–5), 493–523.
- Mariotti, A., German, J. C., Hubert, P., Kaiser, P., Letolle, R., Tardieux, A. and Tardieux, P. (1981) Experimental determination of nitrogen kinetic isotope fractionation: some principles; illustrations for the denitrification and nitrification process. *Plant Soil* **62**, 413–430.
- Mayer, L. M., Keil, R. G., Macko, S. A., Joye, S. B., Ruttenger, K. C. and Aller, R. C. (1998) Importance of suspended particulates in riverine delivery of bioavailable nitrogen to coastal zones. *Global Biogeochem. Cycles*. **12**, 573–580.
- Milliman, J. D. and Meade, R. H. (1983) World-wide delivery of river sediments to the oceans. *J. Geol.* **91**, 1–21.
- Milliman J. D., Shen, H. T., Yang, Z. S. and Meade, R. H. (1985) Transport and deposition of river sediment in the Changjiang estuary and adjacent continental shelf. *Cont. Shelf Res.* **4**, 37–45.
- Minagawa, M. and Wada, E. (1986) Nitrogen isotope ratios of red tide organisms in the East China Sea: a characterization of biological nitrogen fixation. *Mar. Chem.* **19**, 245–259.

- Nakatsuka, T., Watanabe, K., Handa, N., Matsumoto, E. and Wada, E. (1995) Glacial to interglacial surface nutrient variations of Bering deep basins recorded by  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  of sedimentary organic matter. *Paleoceanography* **10**, 1047–1061.
- Newman, J. W., Parker, P. L. and Behrens, E. W. (1973) Organic carbon isotope ratios in Quaternary cores from the Gulf of Mexico. *Geochem. Cosmochim. Acta* **37**, 225–238.
- Peters, K. E., Sweeney, R. E. and Kaplan, I. R. (1978) Correlation of carbon and nitrogen stable isotope ratios in sedimentary organic matter. *Limnol. Oceanogr.* **23**, 598–604.
- Redfield, A. C., Ketchum, B. H. and Richards, F. A. (1963) The influence of organisms on the composition of sea water. *The Sea* (Hill, M. N. *et al.*, eds.), **2**, 26–49, Wiley, N.Y.
- Saito, Y. and Yang, Z. (1994) The Huanghe River: Its water discharge, sediment discharge and sediment budget. *J. Sed. Soc. Japan* **40**, 7–17.
- Schubert, C. J. and Nielsen, B. (2000) Effects of decarbonation treatment on  $\delta^{13}\text{C}$  in marine sediments. *Mar. Chem.* **72**, 55–59.
- Sheu, D. D., Jou, W. C., Chen, M. J., Lee, W. Y. and Lin, S. (1995) Variations of calcium carbonate, organic carbon and their isotopic compositions in surface sediments of the East China Sea. *Terrestrial, Atmospheric, Oceanic Science* **6**(1), 115–128.
- Smith, B. N. and Epstein, S. (1971) Two categories of  $^{13}\text{C}/^{12}\text{C}$  ratios for higher plants. *Plant Physiol.* **47**, 380–384.
- Sternberg, R. W., Larsen, L. H. and Miao, Y. T. (1985) Tidally driven sediment transport on the East China Sea continental shelf. *Cont. Shelf Res.* **4**(1/2), 105–120.
- Sweeney, R. E. and Kaplan, I. R. (1980) Natural abundances of  $^{15}\text{N}$  as a source indicator for near-shore marine sedimentary and dissolved nitrogen. *Mar. Chem.* **9**, 81–94.
- Wada, E. and Hattori, A. (1978) Nitrogen isotope effects in the assimilation of inorganic nitrogenous compounds by marine diatoms. *Geomicrobiol. J.* **1**, 85–101.
- Wada, E., Minagawa, M., Mizutani, H., Tsuji, T., Imaizumi, R. and Karasawa, K. (1987) Biogeochemical studies on the transport of organic matter along the Otsuchi River watershed, Japan. *Estuarine Coast. Shelf Sci.* **25**, 321–336.
- Wedeking, K. W., Hayes, J. M. and Matzigkeit, U. (1983) Procedure of organic geochemical analysis. *Earth's Earliest Biosphere: Its Origin and Evolution* (Schopf, J. W., ed.), 428–441, Princeton Univ. Press, Princeton, N.Y.
- Wong, G. T. F., Gong, G. C., Liu, K. K. and Pai, S. C. (1998) 'Excess Nitrate' in the East China Sea. *Estuarine Coast. Shelf Sci.* **46**, 411–418.