Organic carbon and nitrogen contents and their isotopic compositions in surficial sediments from the East China Sea shelf and the southern Okinawa Trough

S.J. Kao^{a,*}, F.J. Lin^b, K.K. Liu^{b,c}

^aEnvironmental Change Research Center, Academia Sinica, P.O. Box 1-55, Nankang, Taipei, Taiwan

^bInstitute of Oceanography, National Taiwan University, Taipei, Taiwan

^cNational Center for Ocean Research, Taipei, Taiwan

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Abstract

The southern Okinawa Trough has been noted as an important depocenter for the particulate organic matter from the East China Sea shelf. In order to determine the major sources of sedimentary organic matter in the southern Okinawa Trough, we analyzed surficial sediments from the East China Sea shelf and the southern Okinawa Trough for organic carbon and nitrogen contents and their isotopic compositions in the decarbonated fraction. Distributions of total organic carbon content, $\delta^{13}\text{C}_{\text{org}}$ and nitrogen content all show a similar spatial pattern, resembling the distribution of fine-grained sediments. The coastal belt of elevated organic carbon content extends southward from the Changjiang mouth and veers offshore towards the southern Okinawa Trough just north of Taiwan, implying a pathway for channeling fine-grained sediments from the inner shelf to the depocenter. This distribution pattern is consistent with the shelf circulation. The isotope compositions ($\delta^{13}\text{C}_{\text{org}}$, $\delta^{15}\text{N}$) of sediments from the southern Okinawa Trough fall between those of riverine particulate organic matter and the mid-outer shelf sediments, but overlap with those of the inner shelf sediments. In addition, the previously reported $\delta^{13}\text{C}_{\text{org}}$ values of sinking particles collected by sediment traps in the southern Okinawa Trough are also close to those of the inner shelf sediments, especially during high-flux conditions. Therefore, the isotopic evidence strongly supports the notion that a major fraction of the sedimentary organic matter in the southern Okinawa Trough may originate from the inner shelf.

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1. Introduction

The East China Sea (ECS; Fig. 1) is one of the largest marginal seas in the world, which has moderately high primary production

(108–997 $\text{mg m}^{-2} \text{d}^{-1}$ with a mean value of 425 $\text{mg m}^{-2} \text{d}^{-1}$; Gong et al., 2003). It has been estimated that ECS may adsorb 20–30 Mt $\text{CO}_2\text{-C y}^{-1}$ mostly in the inner and middle shelves adjacent to the Changjiang River mouth (Peng et al., 1999; Tsunogai et al., 1999; Wang et al., 2000), which is considerably larger than the estimated flux of organic carbon burial in the shelf sediments (10 Mt C y^{-1} ; Chen and Wang,

*Corresponding author. Tel.: +886-2-27839910 × 809; fax: +886-2-2783-9871.

E-mail address: sjkao@earth.sinica.edu.tw (S.J. Kao).

Fig. 1. Flow pattern and sediment grain size distribution in the East China Sea shelf (after Liu et al., 2000). Arrows along the China coast represent the Changjiang runoff during summer (open arrow) and winter (solid arrows). The main stream of the Kuroshio Current is shown in solid curves. A dashed circle represents the upwelling center induced by a cyclonic eddy located at the northeast off Taiwan. The grain size distribution pattern is after Zhao and Yan (1994). Iso-contour lines of water depth of 200 and 1000 m are in dashed curves.

1999). Apparently, a major fraction of absorbed carbon must be exported out of the ECS shelf to balance the carbon budget. One of the possible mechanisms is the cross-shelf export of particulate organic matter (POM). The most likely site for deposition of exported POC from the ECS is the Okinawa Trough, which is located at the seaward edge of the shelf (Fig. 1). However, sediment deposition rates, carbon accumulation rates and POC fluxes in the Okinawa Trough are highly uneven spatially. Plenty of evidence indicates that the southern Okinawa Trough (SOT) is an important site for POC export from the shelf.

In the SOT region, sedimentation rates (Chung and Chang, 1995; Chen, 1995) reach $0.10\text{--}0.95\text{ cm y}^{-1}$; which are more than 10 times higher than those observed in the northern and central trough ($<0.01\text{--}0.08\text{ cm y}^{-1}$; Ikehara, 1995). The observed POC fluxes from the trap deployed in the SOT area (Stations marked by T in Fig. 2b) range from 10 to $750\text{ mg Cm}^{-2}\text{ d}^{-1}$ (Chung and Chang, 1995; Sheu et al., 1999; Chung and Hung, 2000), which are significantly higher than those ($< 40\text{ mg}$

Fig. 2. Map of sediment sample locations (dots): (a) in the East China Sea and (b) in the southern part of the Okinawa Trough. Also marked are the two sediment trap stations, SST₁ and SST₂ reported by Minagawa and Kadota (1994) and those (T₃–T₁₀) reported by Sheu et al. (1999). Bold dashed-line stands for the inner-shelf area defined in this study. Water depth of 200 m is marked by solid curve.

$\text{Cm}^{-2}\text{ d}^{-1}$; Iseki et al., 1995) obtained by traps of the Marginal Sea Flux Experiment (MASFLEX) deployed in the central and northern trough (Stations marked by SST₁ and SST₂ in Fig. 2a). For a longer term, the organic carbon accumulation rate has been estimated in the range of $5\text{--}56\text{ g Cm}^{-2}\text{ y}^{-1}$ (Chen, 1995) in the SOT. Apparently, the SOT receives more particulate organic matter

than other parts of the Okinawa Trough (Sheu et al., 1999; Hung et al., 1999; Liu et al., 2000).

Regarding the process responsible for feeding sediments to the SOT from the ECS shelf along the China coast, Liu et al. (2000) proposed a conceptual model illustrating the possible route of the sediment transport (Fig. 1). The fine-grained sediments from Changjiang tend to deposit along the south coast of the river mouth (McKee et al., 1983). During the northeast monsoon in winter, the China Coastal Current forms a southward jet flow, which moves the resuspended sediments southward along the coast (Beardsley et al., 1985; Chao, 1991). The sediments may be transported across the shelf in the northern Taiwan Strait due to the blocking of a topographic high in the middle section of the Strait (Jan et al., 1998) and a steady northward bottom current along the western coast of Taiwan (Chuang, 1985; see Fig. 1). As the main stream of the Kuroshio encounters the shelf break of the ECS, a cyclonic eddy forms off northeastern Taiwan, while the Kuroshio turns northeastward. It is the cyclonic eddy northeast of Taiwan, which may send the sediments offshore to the SOT.

In addition to sediments from the ECS shelf, sediments from Taiwan (Hsu et al., 1998) as well as primary production in the water column (Liu et al., 1995) are also potential sources of sedimentary organic matter in the SOT. The purpose of this study is to determine which of the potential sources is most important. For this purpose, we use dual isotope compositions ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) as tracers (Peters et al., 1978; Williams et al., 1992; Thornton and McManus, 1994; Middelburg and Nieuwenhuize, 1998) to elucidate the origins of sedimentary organic matter in the SOT. The total organic carbon content (TOC%), nitrogen content (TN%) and their isotopic compositions were analyzed for samples from a wide region in the ECS shelf and the SOT covering depths from 50 to 1500 m (Fig. 2). We also drew data from previous studies of sedimentary organic carbon (Lin et al., 1992; Chen, 1992) and settling particulate organic carbon (Sheu et al., 1999) in the ECS shelf and adjacent areas for the discussion.

2. Materials and methods

In this study we analyzed 71 surficial sediment samples from the ECS shelf (Fig. 2a i Table 1) and 14 samples from the SOT region (Fig. 2b), which were collected with a box corer on R/V Researcher-1 cruises between 1992 and 1996 and with a Van-Veen type grab on the R/V Vinogradov cruise KEEP-MASS in 1992. Sub-samples from the box cores were obtained with plastic tubings with an inner diameter of 5.3 cm and a length of 60 cm. Sealed samples were then preserved in a freezer. After taken to the laboratory, surficial sediments were scraped from the top 2 cm of the sealed samples with a stainless steel spatula, stored in plastic vials, and then placed in a freeze-drier for one week. The dried sediments were later ground to powder with pestle and mortar prior to analysis.

Carbon and nitrogen isotope samples were prepared using sealed-tube combustion method (Wedeking et al., 1983). About 0.2 g powdered sediments was acid-treated with 1N HCl for 3 days in pre-combusted (500°C) glass test-tubes to remove carbonate. They were then centrifuged for removal of solution and the acid-treated sediments were further dried at 60°C . Acidified samples were mixed with 1 g each of CuO, Cu pellets and several inches of silver wire, and then sealed in evacuated 9-mm quartz tubes. Quartz tubes were then heated at 900°C for 3 h, kept at 650°C for over 12 h, and cooled to room temperature. N_2 and CO_2 gases were extracted and purified in a vacuum line and collected in sealed 6-mm glass tubes. The abundances of carbon and nitrogen were measured manometrically in the vacuum line and presented in percentage. This method had been crosschecked previously by LECO Carbon Analyzer and gave consistent results ($\text{RSD} < 5\%$; Chang et al., 1991). The adsorbed NH_4^+ was generally eliminated during our pretreatment; yet the NH_4^+ fixed in the clay could not be removed. Therefore, the measured nitrogen may have included both organic and inorganic forms and is referred to as total nitrogen (TN) in this study. Carbon and nitrogen isotopic compositions were determined in a Micromass VG602E mass spectrometer. Results are presented in the standard δ notation with

Table 1
Sampling location, overlying water depth, total organic carbon, total nitrogen and isotopic compositions

No.	Water depth	Lat.	Long.	TOC%	TN%	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$
<i>Inner-shelf sediments</i>							
1	42	31.33	122.67	0.48	0.065	4.1	−22.4
2	38	31.17	122.67	1.31	0.170	3.6	−22.2
3	32	31.00	122.83	0.52	0.067	3.8	−21.7
4	48	30.01	122.84	0.67	0.099	4.5	−22.2
5	40	29.49	122.51	0.67	0.089	4.3	−22.3
6	45	29.01	122.35	0.63	0.086	4.4	−22.3
7	45	28.53	122.18	0.96	0.130	4.0	−22.4
8	48	28.00	122.01	0.73	0.100	4.1	−22.4
9	55	27.50	121.76	0.78	0.104	4.3	−22.1
10	70	26.51	121.01	0.88	0.130	3.9	−22.1
11	72	26.50	121.17	0.48	0.070	3.5	−22.3
12	78	26.50	121.33	0.46	0.065	3.9	−21.9
13	135	25.67	121.68	0.42	0.062	4.1	−22.1
<i>Middle-outer shelf sediments</i>							
14	49	30.67	122.95	0.53	0.082	4.2	−21.2
15	36	32.00	124.50	0.37	0.051	4.5	−20.0
16	51	32.00	125.31	0.57	0.082	5.1	−21.1
17	37	32.00	123.51	0.13	0.018	3.9	−20.3
18	60	31.63	125.78	0.78	0.116	5.0	−21.1
19	39	31.50	123.99	0.19	0.027	4.5	−19.8
20	44	31.50	125.00	0.40	0.059	4.5	−21.0
21	67	31.43	126.06	0.64	0.099	4.5	−21.2
22	86	31.06	126.60	0.52	0.078	5.1	−20.9
23	47	31.00	124.50	0.29	0.045	5.2	−20.1
24	56	31.00	123.33	0.22	0.030	4.2	−19.9
25	55	31.00	125.00	0.44	0.065	4.8	−20.7
26	96	30.71	127.01	0.47	0.062	4.6	−20.7
27	53	30.66	124.71	0.23	0.036	4.2	−20.6
28	54	30.51	123.50	0.30	0.042	3.8	−20.1
29	50	30.50	124.00	0.28	0.042	4.6	−20.1
30	128	30.29	127.55	0.25	0.035	4.8	−20.5
31	69	30.24	125.40	0.25	0.049	4.0	−20.8
32	62	30.00	124.50	0.29	0.044	4.3	−20.5
33	64	30.00	123.50	0.40	0.057	4.3	−20.4
34	77	29.87	125.91	0.31	—	—	−20.5
35	60	29.73	124.12	0.24	0.037	4.6	−20.2
36	70	29.58	124.38	0.27	0.040	4.9	−20.6
37	100	29.53	126.44	0.24	0.039	4.9	−20.6
38	85	29.50	125.00	0.48	0.068	4.5	−20.5
39	65	29.50	123.50	0.40	0.059	—	−20.0
40	73	29.49	124.00	0.37	0.056	4.7	−20.4
41	74	29.41	124.65	0.30	0.042	4.2	−20.5
42	78	29.05	125.15	0.23	0.034	4.7	−20.0
43	390	29.03	127.20	0.39	0.057	5.0	−21.0
44	67	29.00	123.51	0.36	0.053	4.1	−20.4
45	81	29.00	124.50	0.25	0.037	4.8	−20.2
46	85	28.89	125.44	0.36	0.050	4.2	−20.9
47	104	28.72	125.68	0.20	0.028	3.8	−20.3
48	67	28.51	123.50	0.25	0.036	4.0	−20.6
49	114	28.38	126.20	0.20	0.036	4.0	−20.4
50	84	28.28	124.17	0.25	0.037	4.9	−20.6

Table 1 (continued)

No.	Water depth	Lat.	Long.	TOC%	TN%	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$
51	104	28.01	124.74	0.29	0.048	4.9	−20.5
52	101	27.73	125.30	0.16	0.032	4.9	−20.2
53	82	27.68	122.83	0.25	0.039	4.5	−20.8
54	123	27.45	125.85	0.07	0.014	6.1	−19.7
55	330	27.38	126.09	0.12	0.021	5.3	−19.6
56	104	27.35	123.38	0.22	0.035	5.2	−20.4
57	106	27.17	122.83	0.21	0.031	—	−21.1
58	121	27.00	124.01	0.17	0.026	5.1	−20.1
59	116	26.92	123.58	0.19	0.032	—	−21.4
60	97	26.83	122.17	0.18	0.026	—	−21.0
61	118	26.58	122.92	0.22	0.034	—	−21.3
62	134	26.58	123.55	0.26	0.042	—	−20.3
63	100	26.25	122.25	0.10	0.017	—	−20.9
64	126	26.25	123.25	0.14	0.019	—	−20.4
65	108	26.17	122.83	0.10	0.018	—	−20.2
66	136	26.17	123.83	0.15	0.027	—	−20.5
67	107	26.00	122.50	0.08	0.015	6.3	−19.7
68	133	25.92	123.58	0.14	0.021	—	—
69	100	25.83	123.18	0.14	0.024	—	−20.4
70	194	25.83	124.17	0.15	0.022	—	—
71	104	25.75	122.75	0.14	0.025	—	−20.9
<i>SOT sediments</i>							
*72	52	24.71	121.91	0.52	0.070	4.1	−23.4
*73	115	24.58	121.93	0.49	0.069	3.9	−24.1
74	137	25.32	122.52	0.18	0.028	3.7	−21.1
75	380	25.35	122.17	0.31	0.045	4.0	−21.4
76	574	25.46	122.85	0.71	0.094	3.6	−21.2
77	786	25.53	123.01	0.75	0.110	3.8	−21.2
78	814	25.66	123.34	0.76	0.103	4.1	−21.3
*79	824	25.00	122.35	0.56	0.089	4.2	−23.5
80	831	25.25	122.85	0.72	0.100	3.6	−21.6
81	846	25.54	123.34	0.63	0.087	4.0	−22.5
82	886	25.38	122.90	0.75	0.110	3.8	−21.7
83	974	25.37	122.36	0.68	0.091	3.9	−22.4
84	1303	25.17	122.56	0.68	0.085	3.5	−21.1
85	1420	24.66	123.01	0.67	0.090	3.8	−21.5
86	1630	25.35	123.03	0.61	0.091	3.9	−21.3

Note: Symbol — stands for those samples influenced by Lanyang-Hsi river.

respect to standards of atmospheric nitrogen and PDB carbon. The precision of nitrogen and carbon isotopic determination for sediment samples is better than $\pm 0.2\%$.

In the presentation of distributions of sedimentary organic carbon and its isotopic composition, we include previously reported data of TOC% (from Lin et al., 1992) and $\delta^{13}\text{C}_{\text{org}}$ (from Chen, 1992) for surface sediments in the SOT area to make the contour map more complete. Lin et al.

(1992) used 1N HCl to remove carbonate, dried the acidified samples in the oven at 50°C , and then measured TOC with a LECO Carbon Analyzer. The results should be consistent with the vacuum extraction method according to the comparison study of Chang et al. (1991). Chen (1992) used 2.4 N HCl to remove carbonate and then dried the acidified samples without removing the solution. It has been demonstrated that pre-treatments for decarbonation with or without removing the acid

solution produced $\delta^{13}\text{C}_{\text{org}}$ values within 0.2‰ from each other (Kao et al., 2001). Similar results have also been reported by Schubert and Nielsen (2000).

Reported isotopic compositions of sinking particles collected at SST₁ and SST₂ from March 1993 to February 1994 (Fig. 2a) and at T₃, T₄, T₆, T₉ and T₁₀ from July 1993 to December 1995 (Fig. 2b) are included for discussion. The bottom depths at SST₁ and SST₂ were 1100 m and the trap depths were between 600 and 1050 m (Iseki et al., 1995). The bottom depths at Stations T₃, T₄, T₆, T₉ and T₁₀ range between 578 and 1440 m, and the trap depths range between 378 and 1340 m. Samples from sites SST₁ and SST₂ have dual isotope compositions (Minagawa and Kadota, 1994), while samples from the other sites were preserved with sodium azide and, therefore, were analyzed for $\delta^{13}\text{C}_{\text{org}}$ values but not $\delta^{15}\text{N}$ values (Sheu et al., 1999).

3. Results

The distribution of TOC in surficial sediments on the ECS shelf from the Changjiang River mouth to the SOT (Fig. 3) shows that most of the shelf area has less than 0.4%. TOC a strong seaward gradient appears in the inner shelf with higher TOC values (>0.50%; Fig. 3a) near the coast; and the maximum value (>0.9%) occurs near the Changjiang mouth. Outside the inner shelf TOC percentage decreases seaward in general and drops to less than 0.2% near the shelf break. Two areas with distinctively high TOC stand out against the generally low TOC background away from the coastal zone (Fig. 3a). One is located at the northeastern corner (around 32°N; 126°E) of the contour map and the other one is in the SOT.

The areas with TOC > 0.50% matches well the mud patches (Fig. 1) (Zhao and Yan, 1994; Qu and Hu, 1993; Hu, 1984; Hu et al., 1998). The coastal belt with elevated TOC content extends southward from the Changjiang mouth. It is noteworthy that this belt veers offshore and extends toward the SOT as it approaches the northern end of the Taiwan Strait. Lin and Chen (1996) also observed a similar spatial distribution

Fig. 3. Distribution of the organic carbon content (TOC%) in sediments in (a) the East China Sea shelf and (b) the southern Okinawa Trough. Scales are the same for the two panels. Filled circles represent data from this study and squares represent data from Lin et al. (1992). The Lanyang-Hsi River, located at the northeastern Taiwan, is also shown. Three triangles locate the three samples affected by riverine sediments (see text).

for fine-grained sediments. It is likely that the seaward extension of this belt is connected with the high TOC zone in the SOT (Fig. 3b).

The Lanyang-Hsi River (Fig. 3b), which has a sediment discharge rate of 6–9 Mt y⁻¹ (Kao and Liu, 2000), and other high sediment-yielding Taiwanese watersheds have been suggested as the main contributors of lithogenic materials to the SOT (Hsu et al., 1998). However, contrary to the high TOC% found at the Changjiang mouth, the TOC% off the Lanyang-Hsi River

Fig. 4. The scatter plot of total organic carbon and nitrogen content in sediments. Redfield ratio of 5.7 (wt/wt) is shown in solid line. The dashed line represents the regression function.

mouth was rather low (Fig. 3b). The low TOC% indicates a strong mineral dilution effect, which is typical for the mountainous watersheds (Kao and Liu, 1996). Aside from the differences in TOC%, it is shown later that the isotopic characteristics of the riverine POC also differ considerably from the sedimentary organic carbon in the SOT.

The spatial distribution pattern of total nitrogen (TN%) in the decarbonated sediments (not shown) is quite similar to that of TOC%. The TN values range from 0.02% to 0.19% with the highest values occurring in the mud patches. The TN% shows a strong linear correlation with TOC% (Fig. 4) with a regression equation as follows:

$$\text{TOC}\% = 7.46 \text{ TN}\% - 0.03$$

$$R^2 = 0.98, p < 0.001, n = 85.$$

The regression line almost passes through the origin, suggesting that most of the nitrogen measured by our method is related to sedimentary organic carbon and, therefore, probably in the organic form (Hedges et al., 1986). Fig. 4 also shows that sediments with high TOC contents have C/N ratios higher than the Redfield ratio of 5.7 (wt/wt), while those with lower TOC 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 > 12; Kukal, 1971; Aitkenhead and McDowell, 2000).

The $\delta^{13}\text{C}_{\text{org}}$ values in surficial sediments show a rather large range, from -25.0‰ to -19.6‰ . Most of the lower values (-21.5‰ or less) occur in the coastal mud belt within about 100 km from the coast, and higher values in the middle and outer shelves (Fig. 5). The results are consistent with the findings of Tan et al. (1991) for Changjiang delta sediments and of Sheu et al. (1995) for middle and outer ECS shelf sediments. However, the data presented here over the entire shelf reveal an isotopic distribution pattern of sedimentary organic carbon has not previously shown. A belt with relatively low $\delta^{13}\text{C}_{\text{org}}$ extends from the Changjiang River mouth southward along the coast and goes offshore near Taiwan. This isotopic distribution pattern is quite similar to that of TOC with only minor differences.

The lower $\delta^{13}\text{C}_{\text{org}}$ values ($< -21.5\text{‰}$) in the inner shelf suggest a stronger influence of terrigenous organics, which has a typical endmember value of -26.5‰ (e.g., Peters et al., 1978, Wada et al., 1987). The higher $\delta^{13}\text{C}_{\text{org}}$ values more offshore generally suggest a higher fraction of marine organic matter, which has a typical endmember value of -20.0‰ (e.g., Goericke and Fry, 1994). On the other hand, the relatively high $\delta^{13}\text{C}_{\text{org}}$ in sedimentary organic matter could be attributed to C_4 plants and its degraded residuals (Goni et al., 1998; Onstad et al., 2000). However, the $\delta^{15}\text{N}$ values of the same samples (to be shown later) indicate the sedimentary organic matter in the mid-outer shelf more akin to marine origin than to C_4 plants.

The surface sediments in the SOT showed relatively low $\delta^{13}\text{C}_{\text{org}}$ values (Fig. 5b), consistent with the trend on the shelf that lower $\delta^{13}\text{C}_{\text{org}}$ values associate with higher TOC%. It is noted that a different relationship between $\delta^{13}\text{C}_{\text{org}}$ and TOC was found off the Lanyang-Hsi River mouth, where the lowest $\delta^{13}\text{C}_{\text{org}}$ values ($< -23\text{‰}$, Fig. 5b) did not correspond to high TOC% values but to relatively low values ($< 0.5\%$, Fig. 3b). This is

Fig. 5. The same as Fig. 3 except for $\delta^{13}\text{C}_{\text{org}}$ in sediments. Solid squares in the lower panel represent data from Chen (1992).

understandable that the sedimentary organic carbon in this region is predominantly terrigenous and, therefore, has a low $\delta^{13}\text{C}_{\text{org}}$ value; the low TOC content is due to mineral dilution effect, which is common in sediments from mountainous watersheds (Kao and Liu, 1996).

The $\delta^{15}\text{N}$ values of surficial sediments range from 3.4‰ to 6.3‰, with most values lower than 5‰. The $\delta^{15}\text{N}$ values greater than 5.0‰ exist mainly in a narrow band at the southeastern edge of the ECS shelf (Fig. 6a), where Kuroshio intrudes onto the shelf (Chao, 1991; Tang et al., 1998, 2000; Liang et al., 2003). The slightly elevated $\delta^{15}\text{N}$ value in the outer shelf is a clear indication of marine origin (Peters et al., 1978). Against rather uniform $\delta^{15}\text{N}$ values around 4.5‰

Fig. 6. The same as Fig. 3 except for $\delta^{15}\text{N}$ of nitrogen in sediments.

over the middle and outer shelves, the inner shelf shows relatively low $\delta^{15}\text{N}$ values; so does the SOT (Fig. 5b).

4. Discussion

In order to determine the most important one among potential sources for the sedimentary organic matter in the SOT, we compare the $^{13}\text{C}_{\text{org}}-^{15}\text{N}$ compositions of sediments from the ECS shelf and the SOT, the suspended particulate matter in the Lanyang-Hsi River and coastal sediments off its mouth. The dual isotope composition plot (Fig. 7) shows distinctive isotopic composition fields for the inner shelf and for the

Fig. 7. The $\delta^{13}\text{C}_{\text{org}}-\delta^{15}\text{N}$ plot of the potential sources and sedimentary organic matter in the southern Okinawa Trough. Rectangle—(1), (2), (3) and (4) represents the isotopic composition field of marine endmember, riverine POM, SOT sediments, and trapped material collected at SST₁ and SST₂, respectively.

mid-outer shelf. The inner shelf, whose sediments are characterized by higher TOC% ($>0.5\%$) and lower $\delta^{13}\text{C}_{\text{org}}$ values ($<-21.5\%$), has a width of roughly 100 km and is marked out in Fig. 2a. Although the inner shelf sediments mainly originate from the Changjiang (Mckee et al., 1983), their $\delta^{13}\text{C}_{\text{org}}$ values are not characteristic of typical terrigenous organic matter, indicating the reworking and addition of marine organic matter. In the following, we refer to the inner-shelf sediments as a distinctive source of sedimentary organic matter rather than an alias of the Changjiang-discharged materials. The $\delta^{13}\text{C}_{\text{org}}$ values (-21.4% to -19.5%) found in the mid-outer shelf sediments agree with the range of POM observed for the latitude of $20-25^\circ\text{N}$ (-21.0 to -19.5% ; Goericke and Fry, 1994). The $\delta^{15}\text{N}$ values found in the mid-outer shelf have a mean value of $4.6 \pm 0.5\%$, which is similar to that of the upwelled nitrate ($\delta^{15}\text{N} = 4.7 \pm 0.6\%$) observed in the southern ECS (Liu et al., 1996). As mentioned earlier, the slightly elevated $\delta^{13}\text{C}_{\text{org}}$ values in the mid-outer shelf may be attributable to organic matter derived from C₄ plants. However, the associated $\delta^{15}\text{N}$ values are typical of marine origin rather than

terrestrial characteristics; the observed $\delta^{15}\text{N}$ values for C₄ plants on Taiwan were -4.1 to -2.6% as reported by Kao and Liu (2000). Therefore, the dual isotope compositions strongly suggest that sedimentary organic matter in the mid-outer shelf is mainly of marine origin. Since the outcropping area of the nitrate-laden water is quite limited (Hsu et al., 2000), we assume 100% uptake (i.e. no biological fractionation) of the nitrate reaching the euphotic zone along the shelf break, and, therefore, there is little net isotopic shift in the POM pool as a whole. Based on this assumption, $\delta^{15}\text{N}$ by Liu et al. (1996) and $\delta^{13}\text{C}$ by Goericke and Fry (1994), we derived a composition field for marine endmember (rectangle — (1) in Fig. 7).

For the source sediments from Taiwan (rectangle — (2) in Fig. 7), we plotted the dual isotope compositions (flux weighted mean) of particulate organic carbon and nitrogen ($\delta^{13}\text{C}_{\text{org}} = -25.5 \pm 0.3\%$ and $\delta^{15}\text{N} = 3.6 \pm 0.3\%$) exported from the Lanyang-Hsi watershed (Kao and Liu, 2000). The isotope compositions of riverine POM from other major rivers ($\delta^{13}\text{C}_{\text{org}} = -25.0 \pm 0.3\%$ and $\delta^{15}\text{N} = 3.8 \pm 0.3\%$) on Taiwan are quite similar to those from the Lanyang-Hsi watershed (S. J. Kao, unpublished data).

The composition field for sediments from the SOT (rectangle — (3) in Fig. 7) is plotted from compiled data in this study, which have rather uniform $\delta^{15}\text{N}$ values ranging from 3.5% to 4.2% (this study), and more variable $\delta^{13}\text{C}_{\text{org}}$, ranging from -21.0% to -23.0% (this study and Chen, 1992). Also plotted are three samples (represented by triangles in Figs. 5b, 6b and 7) near the Lanyang-Hsi River mouth, where the local river loading probably dominates the sediment supply.

The isotope composition of SOT sediments falls between the terrestrial POM from Taiwan on one side of the composition field and sedimentary organic matter of marine origin on the other side (Fig. 7), but it overlaps with those observed for the inner shelf sediments. Although the organic matter deposited in SOT could be a mixture of different fractions of these potential sources, it is conceivable that the inner-shelf sediments is most likely the dominant source for SOT sediments as suggested by the high degree of isotopic similarity. To investigate this possibility, we draw the

Fig. 8. (a) $\delta^{13}\text{C}_{\text{org}}$ values of different kind of samples. (b) Scatter plots of $\delta^{13}\text{C}_{\text{org}}$ values vs. corresponding organic carbon fluxes (OC-flux) for sinking particles collected in the southern Okinawa Trough (data from Sheu et al. 1999). Note the scale changes in the y-axis.

reported sediment trap data of Sheu et al. (1999) for discussion.

For the convenience of comparison, we plot the $\delta^{13}\text{C}_{\text{org}}$ ranges of the SOT sediments and the settling particles collected in the SOT in Fig. 8a. The particulate organic carbon fluxes varied in a wide range, from 10 to 750 $\text{mg C m}^{-2} \text{d}^{-1}$ (Fig. 8b). The trapped materials increased significantly with increasing trap depth (Sheu et al., 1999), indicating a strong particle supply from lateral transport as observed in the mid-Atlantic Bight during the SEEP projects (Biscaye et al., 1988, Biscaye and Anderson, 1994). The $\delta^{13}\text{C}_{\text{org}}$ values of the sediment trap samples range from -24.8‰ to -21.5‰ , overlapping with the range of the SOT sediments and inner-shelf sediments. Some of the trap materials had $\delta^{13}\text{C}_{\text{org}}$ values (Fig. 8a) very close to that of the POC from the Taiwanese rivers ($-25.5 \pm 0.3\text{‰}$; Fig. 7a). However, these samples all corresponded to rather low organic carbon fluxes (Fig. 8b), suggesting a limited contribution

from the Taiwanese rivers as far as POC is concerned.

As the flux increased (e.g. $> 150 \text{ mg C m}^{-2} \text{d}^{-1}$ in Fig. 8b), the $\delta^{13}\text{C}_{\text{org}}$ values of trapped particles approached a uniform $\delta^{13}\text{C}_{\text{org}}$ value (-23.0‰ to -22.2‰). The narrow range overlaps with those of the sedimentary organic matter from the inner-shelf and the SOT. Because the enhanced fluxes are higher than the background flux ($< 50 \text{ mg C m}^{-2} \text{d}^{-1}$) by factors of 3–15, they must represent a major fraction of the total depositional flux, and very likely originate from the inner shelf. Heussner et al. (1999) reported similar phenomenon observed in the Cap-Ferret Canyon in the ECO-MARGE study. They found that the chemical composition in trapped materials approached the composition of surficial sediments, when total flux increased. They attributed the compositional uniformity to a pre-mixed source material. In our case, this pre-mixed source could be the inner shelf sediments transported across the shelf by the shelf circulation.

On the other hand, the trapped POM could be a mixture of the river-derived POM and sediments from mid-outer shelf as indicated by the compositional field in Fig. 7. If this is indeed the case, it would be necessary to keep the mixing ratio nearly constant for all high flux occasions in order to maintain a rather uniform composition. There seems to be no plausible mechanism to maintain constant mixing ratios between these source materials, whenever the carbon flux is enhanced. Therefore, it is unlikely that the isotopic similarity between the enhanced carbon fluxes and the inner shelf sediments was only fortuitous. Instead, this clearly suggests that the enhance fluxes originate from a premixed single source with rather uniform isotopic composition. The inner-shelf sediment is most likely a major source for the sinking POC in the SOT region.

For further comparison with trapped POM in the adjacent region, we also plotted the dual isotope compositions of sediment trap samples collected at Station SST₁ and SST₂ (Fig. 2a) reported by Minagawa and Kadota (1994). The $\delta^{13}\text{C}_{\text{org}}$ values obtained from the northern sites match with those from the southern sites surprisingly well (rectangle — (4) in Fig. 7). Moreover,

the rather uniform $\delta^{15}\text{N}$ values of surficial sediments from the SOT match almost perfectly with the median value of the $\delta^{15}\text{N}$ of the northern trap samples. It appears that the northern and the southern parts of the Okinawa Trough may receive similar materials from the shelf despite of their rather large differences in flux magnitude.

The isotopic signal presented above is not the only evidence supporting the inner-shelf sediments as a major source for sediments deposited in the SOT. Elemental analyses of lithogenic materials indicate that the geochemical characteristics of trapped materials in the SOT are much like those of the inner shelf sediments (Hung et al., 1999). An elevated inventory of ^{210}Pb (300–900 dpm cm^{-2}) on the seafloor in the SOT demands a supply of ^{210}Pb from about 10% of the ECS shelf area (Chung and Chang, 1995), which roughly matches the area of the fine-grained sediments in the inner shelf.

The rather long distance, over which the POM must travel to get from the inner shelf to the SOT, deserves comment. It is the fine grain size associated with POM that makes the long-range transport possible. As mentioned earlier, the distributions of TOC and $^{13}\text{C}_{\text{org}}$ resemble the mud distribution in the ECS shelf. Lin and Chen (1996) found a strong positive correlation between fine-grained fraction ($<64\mu\text{m}$) and TOC in sediments in the southern ECS shelf. This correlation can be explained by the high specific surface area of fine-grained sediments, which have a large capacity for adsorption of organic matter (Mayer, 1994; Keil et al., 1994; Hedges and Keil, 1995). Alternatively, this close correlation also can be attributed to the hydrodynamic equivalence between organic particle and fine-grained sediments (Pedersen and Calvert, 1990). In either case, the distribution pattern of sediments with relatively high TOC content is consistent with the proposed route of sediment transport on the ECS shelf (Liu et al., 2000).

A recent study of circulation in the ECS by a three-dimensional numerical model (Lee and Chao, 2003) supports the notion that the China Coastal Current veers offshore in the northern Taiwan Strait in winter. The modeled flow fields

reveal the cyclonic turn of the winter coastal jet in the northern Taiwan Strait and also the northward flowing current in the western Taiwan Strait as shown in Fig. 1. Their numerical experiment of drifters has successfully demonstrated the proposed across-shelf path of particle transport. Similar cross-shelf transport also has been observed near the depocenter off Cape Hatteras on the east coast of the US (DeMaster et al., 1994), where the circulation pattern known as the “Hatteras funnel” is responsible for the seaward export of particulate matter (Rhoads and Hecker, 1994).

It is interesting to note that the $\delta^{13}\text{C}_{\text{org}}$ range of the SOT sediments is slightly higher than the upper limit of the trap samples. Although sediment traps have been widely used to collect materials transported from the shelf to the slope (e.g., Biscaye and Anderson, 1994; Heussner et al., 1999; Chung and Hung, 2000), they are known for their problems with trapping bias and efficiencies, especially in the slope region (Gardner, 1997; Liu et al., 2000). For example, sediment traps may not be able to catch fluid mud in the bottom boundary layer, which is only recently recognized as one of the major forms of cross shelf sediment export (e.g. Trakovski et al., 2000; Ogston et al., 2000). Therefore, the ^{13}C -enriched fraction may represent an unaccounted density-driven transport of the ^{13}C -enriched outer-shelf sediments within the bottom boundary layer. On the other hand, it also may represent input of organic matter produced in the overlying water column. The slight isotopic offset between the inner-shelf sediments and the trapped POC at high flux (Fig. 8) may be attributed to selective decomposition and/or the addition of POM from Taiwanese rivers during cross-shelf transportation.

Finally, we will discuss the significance of such a pathway to the export of organic carbon in the continental margin. The total primary production in the East China Sea shelf is estimated to be 115 Mt C y^{-1} based on the average given by Gong et al. (2003) and an area of 740000 km^2 . If 15% of the fixed carbon is exported from the shelf as organic carbon, the flux is 17 Mt C y^{-1} , which is on the same order of magnitude as the estimated air

to sea CO₂ flux. How much of the export may be accomplished by the pathway for POC demonstrated here is difficult to assess. The burial flux of organic carbon in the SOT is only on the order of 1 Mt C y⁻¹ based on a mean burial rate of 2.2 g C m⁻² y⁻¹ (Chen, 1995) and area of 4500 km². However, it is not clear how much organic carbon gets oxidized before burial or how much fine organic matter gets swept away by the strong current of the Kuroshio flowing in the Okinawa Trough. Further study is warranted to investigate these processes.

5. Summary and conclusions

Elemental and isotopic analyses of decarbonated surficial sediments from the East China Sea shelf and the southern Okinawa Trough reveal a similar spatial pattern in the distributions of total organic carbon content, its isotopic composition and nitrogen content. The coastal belt of relatively high total organic carbon contents in sediments and low $\delta^{13}\text{C}_{\text{org}}$ values extends southward from the Changjiang mouth along the coast and veers offshore towards the southern Okinawa Trough in the southern East China Sea near Taiwan. This spatial pattern resembles the mud distribution, indicating the close association between sedimentary organic matter and fine-grained sediments.

The $\delta^{13}\text{C}_{\text{org}}-\delta^{15}\text{N}$ compositions of surficial sediments in the southern Okinawa Trough ($-22.0 \pm 1\text{‰}$ and $3.9 \pm 0.4\text{‰}$, which represent compositional ranges) falls between the compositional fields of the riverine particulate organic matter from Taiwan ($-25.5 \pm 0.3\text{‰}$ and $3.6 \pm 0.3\text{‰}$) and sediments from the mid-outer shelf of the East China Sea ($-20.5 \pm 0.9\text{‰}$ and $5.1 \pm 1.3\text{‰}$), but overlaps with that of the inner shelf sediments ($-22.0 \pm 0.4\text{‰}$ and $4.0 \pm 0.5\text{‰}$), which is probably the major source of sedimentary organic matter in the southern Okinawa Trough. This notion is further supported by isotopic data of sediment trap samples collected in the southern Okinawa Trough that the $\delta^{13}\text{C}_{\text{org}}$ values ($-22.8 \pm 0.2\text{‰}$) of sinking particles at high flux are close to those found in the inner shelf. The long

range transport of fine-grained sediments facilitated by the shelf circulation is probably responsible for the delivery of sedimentary organic matter from the inner shelf to the southern Okinawa Trough. It is difficult to assess how significant this pathway can contribute to the carbon export in the continental margin, because little is known of the fraction of organic carbon oxidized or swept away before burial in the SOT.

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Appendix

Sampling location, overlying water depth, total organic carbon, total nitrogen and isotopic compositions are given in Table 1.

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