A comparison of sedimentary aliphatic hydrocarbon distribution between East China Sea and southern Okinawa Trough

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Abstract

In this study, 34 surface sediments were collected from the East China Sea (ECS) shelf, and their aliphatic hydrocarbon concentrations measured. The hydrocarbon distribution of the ECS was compared with that of the southern Okinawa Trough (SOT) to explore for possible relations between the two coastal marine areas. Results from principal component analysis performed on hydrocarbon data of ECS and SOT sediments indicated that the first two components explained 74% of the total variance and that the score plot of the two components showed three groups. The first group defined by the 12 SOT sediments contained three sediments on the ECS inner shelf. Eight sediments except one on the ECS inner shelf formed group II, and the remaining 21 sediments located away from the inner shelf (mainly on the mid-to-outer shelf) formed group III. Even though three inner shelf sediments cluster with group I, the separation between groups I and II suggests that the hydrocarbon relation between the inner shelf and the SOT is very weak. Groups II and III do not cluster together; their separation is supported by previous studies showing that Changjiang River sediment tends to remain in the coastal waters and is transported to the south by the coastal currents along the Chinese coast. Also, group I does not cluster with group III, signifying a great difference between them; this can be attributed to little material exchange between the ECS and the SOT because of the Kuroshio Current. In addition, the SOT and ECS inner shelf had carbon preference index values averaging 2.92 ± 0.79 ($n = 12$) and 4.56 ± 0.93 ($n = 8$), terrestrial/aquatic ratios averaging 9.64 ± 4.33 ($n = 12$) and 50.4 ± 58.0 ($n = 8$), and $(nC_{27} + nC_{29} + nC_{31})$/TOC ratios averaging $1731 \pm 482 \times 10^{-7} \, \mathrm{g(g\,C)}^{-1}$ ($n = 12$) and $3016 \pm 868 \times 10^{-7} \, \mathrm{g(g\,C)}^{-1}$ ($n = 8$), respectively; strongly indicating considerable differences between the two areas. Based on the present results, hydrocarbon export from the ECS inner shelf to the SOT appears to be unlikely.
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1. Introduction

The main circulation patterns of the Changjiang Estuary and adjacent East China Sea (ECS) can be characterized by the northward flow of warm (13 °C) and saline (34 psu) waters of the Taiwan Warm Current (TWC) and a southward flow of the colder (5 °C) and less saline (30 psu) waters of the Yellow Sea Coastal Current (YSCC) (Fig. 1). During high river runoff, one part of the Changjiang plume with the freshest water extends to the south along the coast, and the other part with low salinity extends offshore on average toward the northeast. However, during low river runoff, the surface plume spreads toward the south only, possibly because of the influence of the TWC and YSCC convergence (Beardsley et al., 1985).

The ECS has a very broad continental shelf. The Changjiang River annually discharges $\sim 5 \times 10^8$ tons of sediment directly into the ECS (Milliman and Meade, 1983). This sediment is confined to the coastal zones of the ECS and ultimately transported south and southwestward by the Changjiang Coastal Water (Milliman et al., 1985); offshore transport has been prevented by tidal currents and by the northward movement of the Taiwan Warm Water (Milliman et al., 1989). DeMaster et al. (1985) have reported that the accumulation rates of ECS shelf sediments are relatively high in muddy areas and low in

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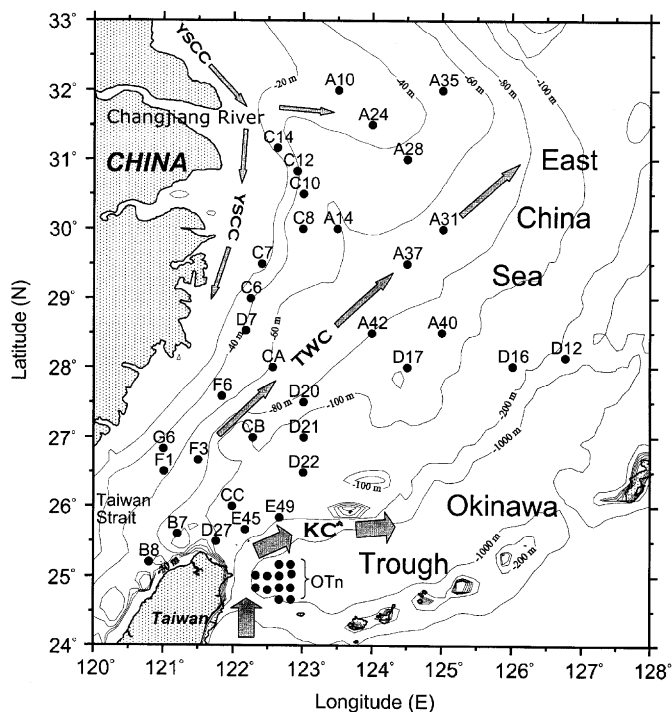*E-mail address:* wljeng@oc.ntu.edu.tw (W.-L. Jeng).

Fig. 1. Sample sites on the East China Sea shelf and in the Okinawa Trough with water circulation patterns. Three major currents are the Yellow Sea Coastal Current (YSCC), the Taiwan Warm Current (TWC) and the Kuroshio Current (KC). Sample codes for the Okinawa Trough (OTn) are top row (from left): stations 18 and 5; second row: stations 1, 3, 4 and 22; third row: stations 12, 23, 19 and 9; bottom row: stations 20 and 8.

results of field observations and diagnostic numerical experiments, Yanagi et al. (1996) have concluded that suspended matter is transported from the shelf edge to the inner shelf in summer and from the inner shelf to the shelf edge in autumn (which is maximum) and winter due to the vertical circulation mainly induced by the monsoon wind. Studying time-series sediment trap samples collected at SST-1 (29°21.6′N, 128°13.5′E, 1100 m water depth), Katayama and Watanabe (2003) have concluded that lateral transport through the bottom layer is important in the transport of terrigenous particles from the ECS shelf to the mid-OT and that Changjiang influence is relatively high during winter. As far as organic carbon is concerned, analyzing 11 sediment cores from the mid-ECS, Oguri et al. (2003) have reported that organic carbon accumulation rates range from 0.42 to 3.1 mg C cm^{-2} yr^{-1} and are higher in the inner shelf edge and the slope areas. Based on the balance of organic carbon budget in the shelf sediments, Kao et al. (2003) consider that a major fraction of adsorbed carbon from primary production must be exported out of the ECS shelf. They further propose that one possible mechanism is the cross-shelf export of particulate organic matter (POM) and that the most likely site for deposition of exported POM from the ECS is the SOT. Liu et al. (2000) propose a conceptual model illustrating the possible route of transporting sediment. From isotopic evidence, Kao et al. (2003) conclude that a major fraction of the sedimentary organic matter in the SOT may originate from the inner shelf of the ECS.

In the present study, 34 surface sediments from the ECS were analyzed for their aliphatic hydrocarbons. A comparison of hydrocarbon distribution in sediments between ECS and SOT (data from Jeng and Huh, 2006) was made in order to understand the significance of cross-shelf export of particulate organic carbon to the SOT. To find the possible relation of hydrocarbon distribution between the two areas, principal component analysis (PCA) was employed since it was a data reduction technique used to simplify data sets while retaining the underlying patterns within the data. This result of comparing hydrocarbon similarity between ECS and SOT may shed some light on the transport of sedimentary lipids between them, and might provide some information about the question "Does the ECS export organic carbon to the SOT?"

2. Experimental

In the present study, 34 surface sediments were collected from the ECS shelf on board R/V Ocean Researcher I using a box corer (Fig. 1). The top 3–4 cm of sediment was stored in stainless steel boxes (pre-cleaned with solvent) and kept frozen (−20 °C) until analyzed. Prefixes A, B, C, D, E, F and G of sample codes denote cruise numbers 551 (May 29–June 4, 1999), 542 (March 8–11, 1999), 525 (August 17–24, 1998), 493 (July 8–13, 1997), 460 (August 20–26, 1996), 456 (July 6–12, 1996) and 417 (April 24–May 1, 1995), respectively.

sandy areas. The sedimentation rates on the ECS shelf range from ~2 to 0.02 cm yr^{-1}, and generally decrease southward along the inner shelf and eastward offshore (Huh and Su, 1999). On the other hand, the southern Okinawa Trough (SOT) has high sedimentation rates located approximately in the lower trough, deeper than 1000 m (range from 0.25 to 0.52 cm yr^{-1} estimated from the excess ^{210}Pb profiles of cores), while low sedimentation rates are generally in the upper trough, shallower than 1000 m (Chung and Chang, 1995). Based on results from a site (24°48.24′N, 122°30.00′E) of ODP Leg 195 in the SOT, the sedimentation rate has always been high, reaching 325 cm kyr^{-1} (0.325 cm y^{-1}) since the late Holocene (ODP, 2001). Therefore, the SOT is apparently an area of focused sedimentation along the path of the Kuroshio Current (KC). Furthermore, the lower slope (>1000 m) sediments consist almost entirely of silty mud (Chen et al., 1995); for instance, mud accounts for over 97% (Jeng and Chen, 1995), while the upper slope (<1000 m) sediments are composed of sand with little mud (Chen et al., 1995).

Transport processes of particulate matter in the marginal seas were first investigated in the middle Atlantic Bight during the SEEP (Shelf Edge Exchange Processes I and II) program (Walsh et al., 1988; Biscaye et al., 1988) and later in several other seas in the world. For the ECS and the SOT, it is the KEEP (Kuroshio Edge Exchange Processes) program (Wong et al., 2000; Liu et al., 2003). Using the

In the lab, frozen sediments were freeze-dried. Following the addition of an internal standard (n-$C_{24}D_{50}$), the dried sediment was extracted with a mixture of dichloromethane and methanol (1:1; v/v) for 24 h in a Soxhlet apparatus. The lipid extract was then saponified by reflux for 3 h with 0.5 N KOH solution in methanol. The non-saponifiable lipids were isolated by hexane extraction four times and concentrated using N_2 gas. The aliphatic hydrocarbon fraction was isolated from the neutral lipids by silica gel (deactivated with 5% H_2O) column chromatography using hexanes.

For gas chromatography (GC) analysis, an HP 5890A gas chromatograph equipped with a split/splitless injector and a flame ionization detector (FID) was used. Separation of aliphatic hydrocarbons was achieved by an SPB-1 capillary column (30 m × 0.25 mm i.d. × 0.25 μm). Oven temperature programming was 45–90 °C at 15 °C min^{-1} and 90–270 °C at 3 °C min^{-1} for analyzing aliphatic hydrocarbons. Identification was made with co-injection of authentic standards and gas chromatography–mass spectrometry (GC–MS). The GC–MS analyses were performed with an HP 6890 GC (HP-1MS crosslinked methyl siloxane column, 30 m × 0.25 mm i.d. × 0.25 μm) interfaced directly to an HP 5973 quadrupole mass selective detector (electron ionization, electron energy 70 eV, scanned from 50 to 550 Da).

An SGE (Australia) OCI-5 cool on-column injector was also fitted in the gas chromatograph for obtaining the best quantification. GC peak areas of all hydrocarbons and the internal standard were obtained using an electronic integrator (Chromatopac C-R6A, Shimadzu, Japan). Each hydrocarbon concentration was determined using the internal standard. Based on eight replicate analyses, the analytical precision (expressed as the percent coefficient of variation) of hydrocarbon abundances was calculated to be 2–8%.

All sediment samples were determined for their total organic carbon by the dichromate–acid oxidation method (Gaudette et al., 1974) modified by addition of Ag_2SO_4 to H_2SO_4 at the rate of 15 g L^{-1}. Titration was carried out with an automatic titrator (Metrohm 702 SM Titrino, Switzerland). The relative standard deviation of TOC determination was generally <1%.

PCA was performed on GC data of aliphatic hydrocarbons. Each compound was used as a variable. Prior to PCA, all hydrocarbon data were standardized by subtracting each data from the mean and dividing by the standard deviation. PCA was performed using xlSTAT version 5.1 (Addinsoft, France).

3. Results and discussion

From the GC chromatograms of aliphatic hydrocarbons, the sediments showed a general picture of similar distribution patterns with n-alkanes predominant along with squalene, diploptene, UCM (unresolved complex mixtures), etc. (Fig. 2). In general, higher-molecular-weight n-alkanes of terrigenous sources—C_{25}, C_{27}, C_{29} and C_{31} (Rieley et al., 1991) dominated over lower-molecular-weight ones of marine sources such as phytoplankton—C_{15}, C_{17}, C_{19} and C_{21} (Winters et al., 1969; Blumer et al., 1971). This result can be attributed to n-alkanes derived from marine sources being degraded faster than those from terrigenous ones (Meyers et al., 1984; Gagosian and Peltzer, 1986). Also, land plants have proportionally larger amounts of hydrocarbons than do marine algae. Squalene exhibited a wide concentration range as compared to n-alkanes (Table 1) probably because it is a lipid constituent of most marine organisms and particularly of phytoplankton (Bieger et al., 1997; Salas et al., 2006). Also, it possesses relatively high stability like n-C_{18} and pristane (Jeng and Huh, 2004). Diploptene (hop-22(29)-ene) is derived from terrestrial higher plants and is also formed by bacteria (Rohmer et al., 1984). If higher plants are the sole source of diploptene, a strong correlation between diploptene and terrestrial higher plant n-alkanes has been demonstrated (Prahl et al., 1992). A positive correlation between diploptene and higher plant n-alkanes was found for 34 ECS sediments ($\sum C_{25-33} = -152 + 13.2$ Di, $r = 0.96$), strongly indicating a higher plant source to the sediments. UCM, consisting of cyclic and branched alkanes, is known to resist microbial degradation more effectively than n-alkanes and thus has a greater tendency to remain in the environment after n-alkanes have degraded (Gough and Rowland, 1990; Bouloubassi and Saliot, 1993). It has a linkage to degraded or weathered petroleum residues (Venkatesan et al., 1980; Readman et al., 1987). Most marine sediments analyzed generally contained a medium UCM (Fig. 2; Table 1). This does not necessarily reflect the presence of degraded petroleum in the sediments since in some cases concentrations of UCM < 10 μg g^{-1} are common in coastal marine environments far from petrogenic hydrocarbon sources (Matsumoto, 1983; Tolosa et al., 1996). In addition, the distribution of the lower molecular weight n-alkanes ($<C_{23}$) looked like a petrogenic origin (carbon preference index (CPI))close to 1) which could probably be attributed to recycled organic matter since the mid-to-outer shelf was composed mainly of relict sediments (Niino and Emery, 1961). Further, only five stations gave Pr/Ph ratios close to 1 (A14, 0.82; B7, 0.81; D16, 0.86; D27, 1.21; E49, 1.05; calculated from Table 1). Combining the UCM and Pr/Ph ratio results, it can be concluded that the petrogenic hydrocarbon contamination on the ECS shelf is minimal.

The sediment textures varied greatly as reflected by the TOC content of sediments ranging from 0.043 to 0.85 g C(100 g)$^{-1}$ (Table 2). To compensate for the grain size difference, all hydrocarbon data were normalized to TOC before performing multivariate analysis.

Prior to PCA, cluster analysis was performed on all variables (e.g. individual aliphatic hydrocarbons) of ECS and SOT sediments (Tables 1 and 3). The resulting dendrogram (not shown) illustrated that UCM, diploptene, and squalene merged with the clusters in the final stages
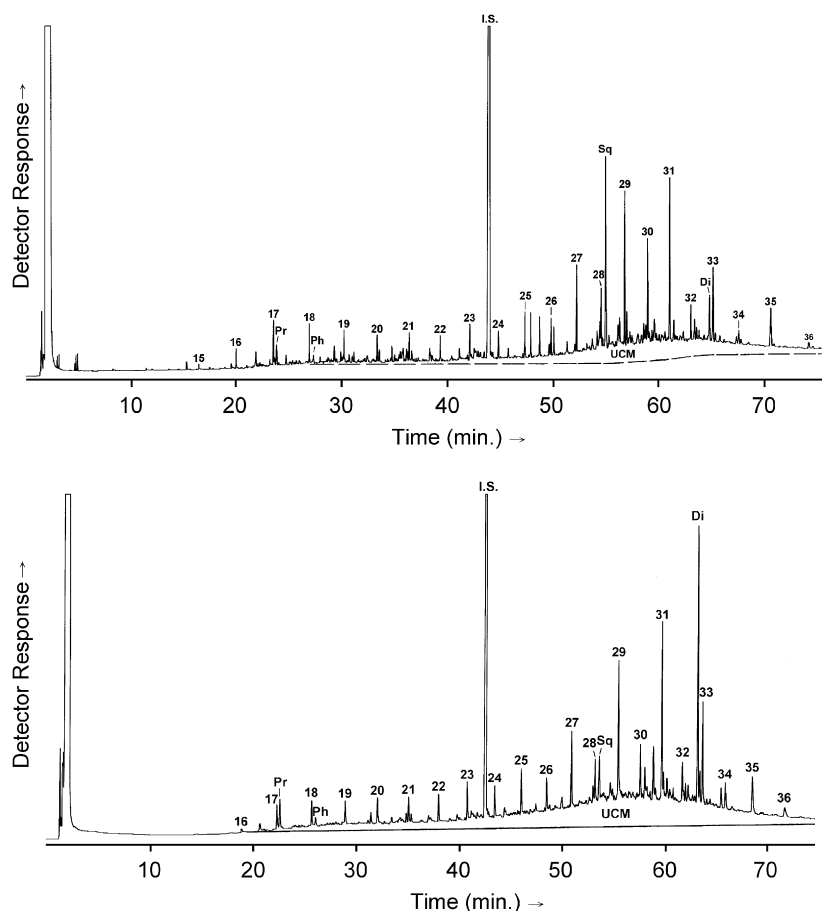
Fig. 2. Gas chromatograms of the aliphatic hydrocarbon fraction from station A40 (upper) and OT8 (lower). Numbers denote carbon numbers of *n*-alkanes. I.S., internal standard; Pr, pristane; Ph, phytane; Sq, squalene; Di, diploptene; and UCM, unresolved complex mixtures.

Table 1
Hydrocarbon concentrations (ng g^{-1}) for East China Sea (ECS) sediments

Sample	Aliphatic hydrocarbons[a]								
	C_{15}	C_{16}	C_{17}	Pr	C_{18}	Ph	C_{19}	C_{20}	C_{21}
A10	–	1.66	4.27	5.95	3.99	1.42	2.96	2.37	7.78
A14	–	–	6.80	3.13	12.2	3.83	13.5	12.6	20.3
A24	–	5.19	32.1	41.8	15.1	5.81	16.6	7.91	11.8
A28	0.372	0.521	0.397	–	2.97	0.818	3.15	3.07	3.72
A31	2.17	7.09	15.7	13.1	13.1	4.99	12.4	10.3	16.0
A35	–	–	3.67	2.60	8.64	–	10.1	10.5	16.4
A37	4.11	10.3	24.4	18.4	19.0	6.36	17.0	12.7	21.0
A40	0.789	4.90	12.7	10.3	11.7	2.33	9.34	8.97	10.1
A42	3.36	13.3	30.4	22.2	26.9	9.89	21.1	17.0	22.6
B7	–	–	2.10	1.68	5.39	2.07	5.26	5.60	7.42
B8	–	12.5	38.7	45.6	33.9	13.4	32.8	28.1	35.1
C6	–	–	–	–	11.2	–	25.8	24.8	41.7
C7	2.88	14.7	42.9	50.0	27.8	9.85	39.7	23.1	37.8
C8	–	4.34	17.2	11.8	17.5	6.36	16.8	13.7	19.7
C10	–	–	3.66	1.30	9.34	–	13.2	13.6	21.4
C12	–	–	–	–	–	–	10.3	18.4	34.4
C14	–	–	–	–	–	–	11.5	16.3	29.4
CA	–	–	2.29	–	8.74	3.01	13.2	13.9	19.8
CB	–	–	1.93	1.13	4.82	1.76	5.24	5.06	7.35
CC	–	–	–	–	1.72	–	2.37	2.91	3.21
D7	25.4	32.1	70.8	62.6	42.5	16.7	50.6	32.7	57.3
D12	–	3.11	12.1	8.05	13.4	5.26	10.2	9.46	11.1

Table 1 (*continued*)

Sample	Aliphatic hydrocarbons[a]								
	C_{15}	C_{16}	C_{17}	Pr	C_{18}	Ph	C_{19}	C_{20}	C_{21}
D16	0	0.918	5.33	2.40	6.84	2.78	4.86	4.50	4.77
D17	2.56	8.09	18.7	14.1	15.5	3.01	11.6	9.79	12.9
D20	–	3.48	11.4	8.08	11.8	4.60	8.14	7.21	8.33
D21	–	–	–	–	4.30	0.513	5.92	7.07	9.00
D22	–	4.64	10.8	6.88	10.3	3.93	7.18	6.39	7.66
D27	–	1.35	5.02	4.13	6.50	3.40	5.26	4.67	5.67
E45	–	–	–	–	–	–	0.958	1.83	2.95
E49	–	1.20	3.35	1.60	3.67	1.52	2.35	2.16	2.30
F1	44.6	48.6	94.8	120	66.6	27.8	62.2	60.8	71.4
F3	46.4	48.4	93.2	141	65.3	26.0	60.2	69.1	66.6
F6	40.3	41.7	116	91.0	59.3	23.1	60.7	44.1	65.0
G6	–	–	20.6	21.3	26.1	9.56	32.7	28.7	42.1

	C_{22}	C_{23}	C_{24}	C_{25}	C_{26}	C_{27}	C_{28}	Sq	C_{29}
A10	2.67	4.68	3.10	4.62	3.00	6.82	6.97	76.0	14.6
A14	17.8	31.3	21.0	39.6	28.6	65.0	32.9	134	136
A24	7.91	13.5	9.42	15.0	10.2	22.6	12.4	161	48.4
A28	2.95	4.91	3.17	4.21	4.21	7.86	6.27	41.6	12.7
A31	13.1	21.2	13.1	24.7	15.4	37.2	24.1	93.4	80.4
A35	14.6	26.0	16.6	37.8	19.8	63.3	27.5	125	144
A37	16.2	28.6	18.8	36.8	22.6	56.1	33.7	197	117
A40	9.38	14.3	9.67	17.4	13.8	30.7	22.2	83.2	60.2
A42	21.2	32.8	22.5	35.1	23.1	53.3	28.6	138	106
B7	7.54	10.5	7.42	9.01	6.39	10.2	7.95	92.5	17.1
B8	36.7	44.9	40.8	51.3	34.6	56.9	32.5	194	98.0
C6	47.4	89.6	58.3	144	66.4	237	104	100	565
C7	40.3	80.0	47.3	125	56.4	218	89.1	133	523
C8	21.4	35.9	27.0	42.0	22.9	55.6	30.7	51.8	127
C10	22.8	36.6	31.8	50.9	37.6	73.3	48.3	160	140
C12	45.3	89.0	69.1	156	90.2	262	135	141	587
C14	36.6	70.0	39.9	111	51.9	196	80.4	108	485
CA	22.2	37.4	32.7	52.2	40.9	71.1	45.8	65.1	118
CB	6.15	9.95	6.33	10.8	6.19	14.0	7.03	62.1	27.0
CC	3.79	5.79	3.88	5.49	3.00	6.35	3.53	24.7	11.3
D7	54.0	90.7	58.2	123	63.8	213	105	119	512
D12	12.0	19.1	13.9	30.0	20.7	55.5	45.0	57.2	109
D16	4.80	8.32	5.12	8.00	3.94	10.8	5.54	50.3	22.2
D17	12.0	18.5	13.1	23.7	14.8	34.7	22.3	87.1	76.0
D20	7.66	12.4	9.93	16.4	8.57	21.0	12.5	70.1	43.1
D21	8.88	14.6	10.1	21.4	10.5	25.9	11.2	145	53.5
D22	6.69	10.8	7.10	12.3	6.39	14.0	12.8	78.4	25.8
D27	5.36	9.08	7.37	10.7	9.75	20.0	24.7	64.0	38.6
E45	3.42	5.86	3.93	6.36	4.00	9.80	4.61	12.5	17.9
E49	2.28	4.09	2.43	3.64	2.05	5.11	2.14	6.89	8.85
F1	70.5	85.6	57.9	83.9	58.9	116	73.4	111	201
F3	69.3	83.2	56.2	84.5	62.3	118	75.0	116	205
F6	65.4	95.8	67.0	131	74.9	205	98.3	275	457
G6	41.3	70.5	46.7	90.7	54.8	147	97.9	161	312

	C_{30}	C_{31}	C_{32}	Di	C_{33}	C_{34}	C_{35}	UCM	CPI
A10	12.9	14.2	2.45	13.1	6.14	0.672	6.26	388	1.71
A14	60.1	167	21.4	57.7	79.6	9.46	47.8	2330	3.09
A24	42.9	51.0	8.64	28.8	19.9	2.28	24.5	1100	1.97
A28	12.6	11.1	3.84	6.92	4.98	1.12	6.25	–	1.41
A31	52.3	93.7	15.3	49.0	44.3	6.91	45.0	3410	2.40
A35	80.8	186	17.3	71.3	80.1	6.92	40.2	2180	3.26
A37	71.0	137	20.3	60.2	62.0	6.95	61.3	2980	2.56
A40	43.0	63.5	13.0	23.1	33.8	6.31	23.8	852	2.06
A42	41.6	126	19.0	42.3	60.3	9.26	44.2	3990	2.98
B7	11.2	17.4	4.68	10.7	9.59	1.78	10.9	658	1.83
B8	43.8	123	22.4	54.7	71.4	9.07	34.4	–	2.56

Table 1 (continued)

	C_{30}	C_{31}	C_{32}	Di	C_{33}	C_{34}	C_{35}	UCM	CPI
C6	102	778	68.0	156	357	24.4	108	9250	5.46
C7	129	747	77.1	141	330	20.8	101	6400	5.05
C8	23.0	174	23.4	42.3	75.9	9.16	32.0	6560	4.05
C10	43.5	168	29.1	48.2	83.0	14.2	53.1	5490	2.85
C12	120	796	82.3	142	340	31.6	128	7920	4.49
C14	77.0	648	57.4	98.2	280	18.8	85.2	5120	5.82
CA	43.1	138	28.3	39.9	69.2	13.8	49.5	2960	2.48
CB	8.93	31.3	5.38	24.5	14.0	1.69	14.2	1300	3.10
CC	4.05	13.9	2.05	6.19	6.33	1.19	5.98	–	2.89
D7	108	687	82.2	167	298	21.3	97.6	9420	4.61
D12	46.8	131	36.7	35.0	66.4	21.2	32.0	1300	2.35
D16	5.33	25.7	3.73	14.6	11.4	1.54	6.46	–	3.60
D17	27.0	90.9	18.9	42.6	44.9	8.30	29.3	1940	2.89
D20	20.7	49.7	9.11	33.1	22.4	4.36	13.7	1210	2.64
D21	20.4	62.0	12.6	41.1	29.6	7.94	18.3	1290	3.02
D22	6.88	29.9	4.56	26.0	12.0	0.972	11.0	911	2.73
D27	31.8	56.6	36.7	20.2	42.2	14.3	17.4	1250	1.48
E45	4.42	22.3	3.05	2.64	9.43	1.30	3.62	–	3.54
E49	2.12	11.0	1.18	2.30	4.60	0.455	2.55	–	3.77
F1	81.3	239	41.5	75.6	129	20.7	46.4	4580	2.63
F3	64.4	247	48.8	70.4	134	19.0	96.8	6660	2.75
F6	104	607	90.1	166	308	28.8	112	5820	4.12
G6	76.8	431	51.2	110	219	18.1	79.7	3530	3.84

[a]Pr, pristane; Ph, phytane; Sq, squalene; Di, diploptene; UCM, unresolved complex mixtures; CPI, carbon preference index $= (1/2)[(nC_{25} + nC_{27} + nC_{29} + nC_{31} + nC_{33})/(nC_{24} + nC_{26} + nC_{28} + nC_{30} + nC_{32}) + (nC_{25} + nC_{27} + nC_{29} + nC_{31} + nC_{33})/(nC_{26} + nC_{28} + nC_{30} + nC_{32} + nC_{34})]$.

Table 2
Total organic carbon (TOC in $g\,C\,(100\,g)^{-1}$) in East China Sea and southern Okinawa Trough sediments

East China Sea				Southern Okinawa Trough	
Sample	TOC	Sample	TOC	Sample	TOC
A10	0.059	CA	0.20	OT1	0.65
A14	0.34	CB	0.10	OT3	0.56
A24	0.18	CC	0.057	OT4	0.79
A28	0.046	D7	0.49	OT5	0.75
A31	0.45	D12	0.23	OT8	0.71
A35	0.21	D16	0.082	OT9	0.73
A37	0.31	D17	0.17	OT12	0.81
A40	0.15	D20	0.098	OT18	0.82
A42	0.26	D21	0.13	OT19	0.59
B7	0.063	D22	0.11	OT20	0.60
B8	0.31	D27	0.064	OT22	0.85
C6	0.42	E45	0.047	OT23	0.76
C7	0.42	E49	0.043		
C8	0.16	F1	0.46		
C10	0.24	F3	0.43		
C12	0.45	F6	0.52		
C14	0.33	G6	0.46		

with Euclidean distances of 6.246, 7.034 and 8.717, respectively, indicating that the three components had very weak relation with other hydrocarbons. As a consequence, these three components were excluded for performing PCA. The score plot of the first two components explaining 74% of the total variance is shown in Fig. 3. To facilitate discussion, peripheral samples of all

OT samples were connected by line segments, forming a polygon; those in positive PC1 and negative PC2 as well as those in negative PC1 were, respectively, connected in the same way. Therefore, three groups are visible in the score plot, those virtually with a positive score on PC1 and with low positive and low negative scores on PC2 (group I), those with a positive score on PC1 and a high negative score on PC2 (group II), and those with negative scores on PC1 (group III). Of the three groups, group I contained 12 OT samples and three inner shelf samples (C8, D7 and F6). With the exception of sample D27, group II comprised seven samples on the inner shelf. The majority of samples (21) on the ECS shelf located away from the inner shelf constituted group III. In addition, samples F1 and F3, situated in high positive scores on PC1 and high positive scores on PC2, were fairly peculiar and might be considered outliers.

It is of importance to note that group I defined by 12 OT samples includes three samples on the inner shelf; this is especially unreasonable because group I clusters with distant samples instead of nearby samples. This requires more discussion. The Lanyang River (nearest river to the SOT, not shown) has been reported to be not a major hydrocarbon source for OT sediments, but to a less extent it does influence two stations OT 1 and OT 12 closest to the river mouth based on the individual TAR (terrigenous/aquatic ratio) and CPI values (Jeng and Huh, 2006). This also reflects in OT1 in Fig. 3 that the sample is situated relatively away from the cluster of other OT samples. That is, sample OT1 has a low similarity to the other OT

Table 3

Hydrocarbon concentrations (ng g^{-1}) for Okinawa Trough (OT) sediments (from Jeng and Huh, 2006)

Sample	Aliphatic hydrocarbons[a]								
	C_{15}	C_{16}	C_{17}	Pr	C_{18}	Ph	C_{19}	C_{20}	C_{21}
OT1	1.94	29.3	97.6	170	110	45.9	116	85.1	87.5
OT3	–	–	23.8	28.1	61.4	23.2	55.7	50.3	49.2
OT4	–	–	42.7	49.4	81.1	26.2	96.1	75.4	82.4
OT5	–	–	20.1	17.9	56.7	17.9	66.6	70.7	82.1
OT8	–	32.7	90.4	118	88.7	28.7	95.1	71.5	77.8
OT9	–	–	18.3	17.1	55.7	15.8	89.5	83.1	95.9
OT12	80.3	63.8	103	147	89.1	32.0	102	65.5	64.7
OT18	10.7	42.9	103	101	103	36.3	79.5	73.8	73.1
OT19	11.2	32.2	67.2	89.8	65.5	20.5	52.2	47.7	56.5
OT20	10.8	28.2	52.7	34.1	52.0	15.7	40.2	28.6	36.0
OT22	–	14.5	80.6	113	101	37.9	104	78.8	84.5
OT23	–	–	14.9	15.6	49.4	17.4	54.6	56.5	60.7

	C_{22}	C_{23}	C_{24}	C_{25}	C_{26}	C_{27}	C_{28}	Sq	C_{29}
OT1	96.4	137	114	171	111	241	165	217	418
OT3	53.2	70.6	65.0	98.6	54.8	136	87.6	178	253
OT4	88.7	125	98.6	156	96.7	243	168	199	473
OT5	86.7	122	97.3	149	95.4	220	157	271	382
OT8	82.3	119	96.8	150	96.3	221	145	161	421
OT9	108	135	116	156	97.6	214	139	317	392
OT12	65.1	103	97.6	160	102	240	176	184	457
OT18	72.3	107	77.7	148	79.7	214	149	230	451
OT19	69.8	94.7	73.5	110	73.0	168	88.3	100	319
OT20	41.2	99.0	77.3	178	94.8	346	121	99.0	690
OT22	86.6	125	99.6	145	101	236	141	28.2	433
OT23	64.4	94.7	78.6	133	76.4	195	127	171	359

	C_{30}	C_{31}	C_{32}	Di	C_{33}	C_{34}	C_{35}	UCM	CPI
OT1	195	564	150	450	273	65.9	167	8930	2.35
OT3	102	378	77.9	874	167	25.9	121	3410	2.82
OT4	202	634	178	1130	295	76.3	186	8080	2.46
OT5	148	561	162	363	280	72.0	179	4450	2.46
OT8	168	573	153	917	263	69.8	182	8760	2.53
OT9	161	574	131	1310	260	55.8	165	8790	2.61
OT12	172	616	121	586	278	49.1	203	7420	2.72
OT18	106	606	127	626	265	44.4	206	–	3.23
OT19	97.9	456	80.0	915	206	26.4	94.9	–	3.25
OT20	114	885	91.1	79.9	371	26.4	101	–	5.24
OT22	195	618	167	716	316	76.9	206	10700	2.53
OT23	147	549	115	1210	230	34.8	177	3540	2.81

[a]Pr, pristine; Ph, phytane; Sq, squalene; Di, diploptene; UCM, unresolved complex mixtures; CPI, carbon preference index $= (1/2)[(nC_{25} + nC_{27} + nC_{29} + nC_{31} + nC_{33})/(nC_{24} + nC_{26} + nC_{28} + nC_{30} + nC_{32}) + (nC_{25} + nC_{27} + nC_{29} + nC_{31} + nC_{33})/(nC_{26} + nC_{28} + nC_{30} + nC_{32} + nC_{34})]$.

samples. If sample OT1 is excluded from Fig. 3, the area of group I is greatly reduced, and samples C8, D7 and F6 are not included in group I. In other words, the relation between these three samples and group I depends entirely on sample OT1, and it can be concluded that this relation is very weak, or even does not exist at all.

It is of great interest to examine the hydrocarbon distribution in the southwestern part of the ECS since it is an important area proposed for transporting POM from the ECS to the SOT by Liu et al. (2000). In their conceptual model, the possible route of transporting sediment is from the Changjiang River mouth, southward along the south-eastern coast of China, eastward across northern Taiwan Strait, along offshore northern Taiwan to the SOT. Considering the samples collected from the Changjiang River mouth to the SOT (22 samples on the shelf west of longitude 123° and 12 in the SOT), it was found that only three samples (C8, D7 and F6) clustered with group I (OT samples) and the other 19 samples do not. This suggests that the hydrocarbon relation between the inner shelf and the SOT is actually weak, implying that the possibility of transporting hydrocarbons along the route of the conceptual model to SOT is very low. This result can be quite reliable because *n*-alkanes are relatively unreactive and
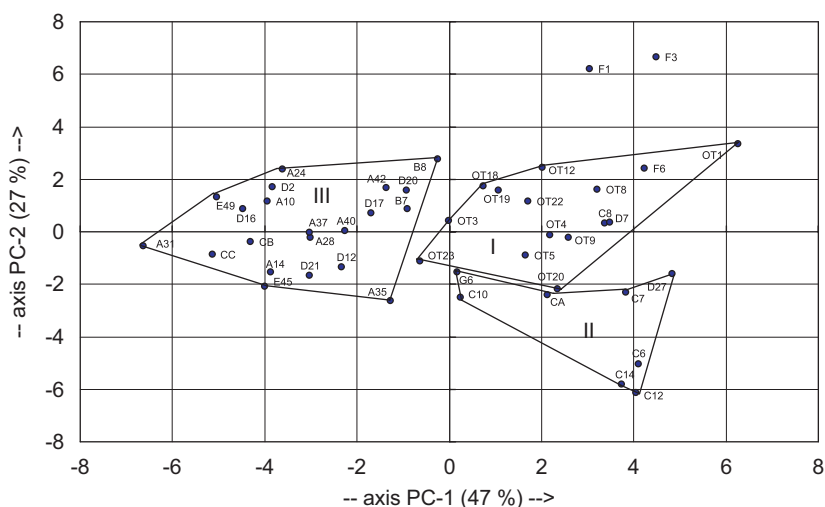
Fig. 3. Score plot of PCA for ECS and SOT sediments. Sample codes for ECS samples have prefixes of A–G, and those for SOT samples have a prefix of OT. Group I contains 12 SOT sediments and three sediments on the inner shelf, group II consists of eight but one sediments on the inner shelf, and group III is composed of 21 sediments mainly on the mid-to-outer shelf.

unlikely to be affected by any mechanism that would be selective for alternating homologs in the sequence (Hedges and Prahl, 1993).

One marked feature is that groups II and III are, respectively, located in positive and negative PC1, signifying their difference in hydrocarbon distribution. This is supported by the literature result that Changjiang sediments have been transported to the south via the coastal current along the Chinese coast and that offshore transport has been prevented by tidal currents and by the northward movement of the TWC (Milliman et al., 1989). Another reason may be that the inner shelf consists largely of muddy sediments, which are considered recent deposits, and the mid-to-outer shelf is composed chiefly of sandy sediments (Huh and Su, 1999), which are known as relict sediments (Niino and Emery, 1961). In addition, stations F1 and F3 located near the inner shelf have no relation with group II, which might be attributed to the influence of the TWC (Fig. 1). Furthermore, sample locations G6, F1 and F3 are very close to each other geographically (Fig. 1), but sample G6 separates clearly from samples F1 and F3 in the score plot (Fig. 3), suggesting that the deposition of Changjiang River sediments terminates at station G6.

As expected, groups I and III are separated from each other in Fig. 3, likely attributable to little material exchange between them. The KC flows northward and turns northeastward when approaching the ECS shelf although it intrudes onto the shelf in the winter and spring time (Chern and Wang, 1992; Hsueh et al., 1992).

Note that the SOT contains 12 stations within a relatively small area, but the hydrocarbon distribution of the samples varies widely. For example, the areas covered by groups I and III in Fig. 3 are comparable, but their respective geographical areas in the SOT and on the mid-to-outer shelf differ greatly (Fig. 1). The reason can be explained as follows. The lipid inputs by suspended matter/

sediment transport to the seas off northeastern Taiwan can derive from three sources (Fig. 1). From the northwest, a small alongshore flow just off northern Taiwan (south of station D27) along with the eddy flow (Tang et al., 1999) may carry materials from the southern ECS and Taiwan Strait to the SOT. The Lanyang River (from the west) exerts small influence on the SOT sediments as discussed earlier. Another source from south is other river runoffs from eastern Taiwan carried by the Kuroshio Current to the SOT.

Terrestrial higher plant wax contribution to sediment or soil is generally expressed by the CPI of n-alkanes. Higher CPI_{25-33} values indicate greater contribution from vascular plants (Rieley et al., 1991); CPI_{25-33} values close to one are thought to indicate greater input from marine microorganisms, recycled organic matter, and/or petroleum (Kennicutt et al., 1987). The present CPI_{25-33} values of the ECS ranged widely from 1.41 to 5.82 with an average of 3.12 (calculated from Table 1), suggesting non-uniform distribution of terrigenous hydrocarbons. In general, high CPI values were found on the inner shelf, and low values on the mid-to-outer shelf. This suggests that the Changjiang River contributes more to the former than to the latter, consistent with the literature result indicated earlier. On the other hand, the average CPI value (3.12, $n = 34$) of the ECS was comparable to that (average 2.92, $n = 12$) of the SOT. These two averages are considered to be relatively high, suggesting that fossil fuel contamination is minimal. However, it should be noted that CPI values would become smaller if there is any addition of 'old' hydrocarbons (e.g., CPI of petroleum close to 1; Bray and Evans, 1961) to the sediments from pollution. Some samples were found to contain different proportions of 'old' hydrocarbons as evidenced by the presence of $17(\alpha)H,21(\beta)H$-hopanes. This fossil component could be contributed from fossil fuels through contamination (Albaigés and Albrecht,

1979; Shaw et al., 1985; Readman et al., 1996) from boat traffic or a spill. The addition of 'old' hydrocarbons will lower some CPI values observed in ECS sediments. The purpose of this study is to compare hydrocarbon distribution between ECS and SOT and to explore if they are related. For comparing the two areas, eight samples (C14, C12, C10, C8, C7, C6, D7 and F6; Fig. 1) on the ECS inner shelf and 12 samples in the SOT were chosen; their average CPIs were, respectively, 4.56 ± 0.93 and 2.92 ± 0.79, suggesting that the hydrocarbon sources to the two areas were quite different. The result is quite dependable because the CPI is a diagenetically insensitive ratio (Hedges and Prahl, 1993), implying that the CPI would not change over time during transport.

The ratio of terrigenous-to-aquatic *n*-alkanes were estimated as

$$\mathrm{TAR} = (nC_{27} + nC_{29} + nC_{31})/(nC_{15} + nC_{17} + nC_{19}).$$

This ratio is valuable for determining changes in relative contributions of organic matter from land and aquatic flora although it may over-represent the absolute amounts from terrigenous sources (Meyers, 1997). This ratio was adopted in the present study to estimate the relative contribution of terrigenous and marine sources to ECS and SOT sediments. The average TAR for the ECS inner shelf was 50.4 ± 58.0 ($n = 8$), and that for the SOT was 9.64 ± 4.33 ($n = 12$) (calculated from Tables 1 to 3). This suggests that the two areas receive different hydrocarbon inputs. The TAR values are expected to be larger for SOT sediments than for ECS sediments (since the SOT is much deeper than the ECS and since aquatic components degrade faster) if they have the same source. The present result is reversed and suggests the ECS and SOT having different sources. Note that this ratio is one of the factors used for comparing the differences of the multiple study sites. Higher plant *n*-alkanes were the predominant components in the sediments from the ECS and SOT; to correct for the grain size effect, $(nC_{27} + nC_{29} + nC_{31})/\mathrm{TOC}$ was used to estimate the input of higher plant *n*-alkanes to the two areas (Prahl, 1985). The SOT and ECS inner shelf gave $(nC_{27} + nC_{29} + nC_{31})/\mathrm{TOC}$ averages of $1731 \pm 482 \times 10^{-7}\, \mathrm{g\,(g\,C)^{-1}}$ ($n = 12$) and $3016 \pm 868 \times 10^{-7}\, \mathrm{g\,(g\,C)^{-1}}$ ($n = 8$) (calculated from Tables 1 to 3), respectively, indicating different inputs of higher plant *n*-alkanes to the two areas. In the HMW region, the C_{27} and C_{29} *n*-alkanes are diagnostic of waxes from trees and shrubs, and the *n*-C_{31} alkane is diagnostic of grass waxes (Cranwell, 1973). The $nC_{31}/(nC_{27} + nC_{29} + nC_{31})$ ratio was employed to see if any difference existed between the two areas. The average ratios of $nC_{31}/(nC_{27} + nC_{29} + nC_{31})$ for the SOT and ECS inner shelf were, respectively, 0.478 ± 0.012 ($n = 12$) and 0.483 ± 0.018 ($n = 8$) (calculated from Tables 1 to 3), showing no difference between the two averages. This can be attributed to *n*-alkane distribution maximizing at C_{31}, which makes the change in C_{31} insensitive.

Seven stations (not shown) between Taiwan and 12 SOT sample sites were selected for collecting total suspended matter (2 m below sea surface) on July 23–26, 2000 (cruise #590). The average CPI of the seven samples was 1.01 ± 0.12. This low CPI average reflects the contribution from a nearby river (Lanyang River), which is known to contain "old" hydrocarbons in its suspended matter and sediments (Jeng and Kao, 2002; Jeng and Huh, 2006). In addition, one of our cruises (#525 on August 17–24, 1998) to the ECS happened to be the high runoff period of the Changjiang River. The runoff of the river had a very different color than the offshore seawater of the ECS. Two stations—C14 and C15 (31°19.96′N, 122°30.09′E, not shown) representing the runoff of the river were occupied, and one sample of total suspended matter (2 m below sea surface) for each station was collected. CPI values of the two stations were 1.55 and 1.32, which differed greatly from those of off northeast Taiwan. These results indicate that transport of suspended sediment from the Changjiang River mouth to the SOT is quite unlikely.

In conclusion, most sediments on the ECS inner shelf did not group with those in the SOT, indicating dissimilarity in hydrocarbon distribution between the two areas. Moreover, striking differences in CPI, TAR and $(nC_{27} + nC_{29} + nC_{31})/\mathrm{TOC}$ values were found between SOT and ECS inner shelf. In addition, hydrocarbon distribution differences in suspended matter from Changjiang River runoff and off northeastern Taiwan were also observed, suggesting that hydrocarbon sources to the two areas were different. Based on these results, it can be concluded that hydrocarbon contribution from the ECS inner shelf to the SOT appears to be minimal.

References

Albaigés, J., Albrecht, P., 1979. Fingerprinting marine pollutant hydrocarbons by computerized gas chromatography-mass spectrometry. International Journal of Environmental Analytical Chemistry 6, 171–190.

Beardsley, R.C., Limeburner, R., Yu, H., Cannon, G.A., 1985. Discharge of the Changjiang (Yangtze River) into the East China Sea. Continental Shelf Research 4, 57–76.

Bieger, T., Abrajano, T.A., Hellou, J., 1997. Generation of biogenic hydrocarbons during a spring bloom in Newfoundland coastal (NW Atlantic) waters. Organic Geochemistry 26, 207–218.

Biscaye, P.E., Anderson, R.F., Deck, B.L., 1988. Fluxes of particles and constituents to the eastern United States continental slope and rise: SEEP-I. Continental Shelf Research 8, 855–904.

Blumer, M., Guillard, R.R.L., Chase, T., 1971. Hydrocarbons of marine phytoplankton. Marine Biology 8, 183–189.

Bouloubassi, I., Saliot, A., 1993. Investigation of anthropogenic and natural organic inputs in estuarine sediments using hydrocarbon markers (NAH, LAB, PAH). Oceanologica Acta 16, 145–161.

Bray, E.E., Evans, E.D., 1961. Distribution of n-paraffins as a clue to recognition of source beds. Geochimica et Cosmochimica Acta 22, 2–15.

Chen, M.P., Lin, K.L., Huang, C.K., 1995. Sedimentary structure and texture of surficial sediment off northeastern Taiwan. Ti-Chih (ROC) 15, 15–47 (in Chinese with English abstract).

Chern, C.S., Wang, J., 1992. On the season variation of the Kuroshio intrusion onto the East China Sea. Acta Oceanographica Taiwanica 29, 1–17.

Chung, Y., Chang, W.C., 1995. Pb-210 fluxes and sedimentation rates on the lower continental slope between Taiwan and the South Okinawa Trough. Continental Shelf Research 15, 149–164.

Cranwell, P.A., 1973. Chain-length distribution of n-alkanes from lake sediments in relation to postglacial environment change. Freshwater Biology 3, 259–265.

DeMaster, D.J., McKee, B.A., Nittrouer, C.A., Qian, J.C., Cheng, G.D., 1985. Rates of sediment accumulation and particle reworking based on radiochemical measurements from continental shelf deposits in the East China Sea. Continental Shelf Research 4, 143–158.

Gagosian, R.B., Peltzer, E.T., 1986. The importance of atmospheric input of terrestrial organic material to deep sea sediments. Organic Geochemistry 10, 661–669.

Gaudette, H.E., Flight, W.R., Toner, L., Folger, D.W., 1974. An inexpensive titration method for the determination of organic carbon in recent sediments. Journal of Sedimentary Petrology 44, 249–253.

Gough, M.A., Rowland, S.J., 1990. Characterization of unresolved complex mixtures of hydrocarbons in petroleum. Nature 344, 648–650.

Hedges, J.I., Prahl, F.G., 1993. Early diagenesis: consequences for applications of molecular biomarkers. In: Engel, M.H., Macko, S.A. (Eds.), Organic Geochemistry: Principles and Applications. Plenum Press, New York, pp. 237–253.

Hsueh, Y., Wang, J., Chern, C.S., 1992. The intrusion of the Kuroshio across the continental shelf northeast of Taiwan. Journal of Geophysical Research 97 (C9), 14323–14330.

Huh, C.A., Su, C.C., 1999. Sedimentation dynamics in the East China Sea elucidated from ^{210}Pb, ^{137}Cs and ^{239,240}Pu. Marine Geology 160, 183–196.

Jeng, W.L., Chen, M.P., 1995. Grain size effect on bound lipids in sediments off northeastern Taiwan. Organic Geochemistry 23, 301–310.

Jeng, W.L., Huh, C.A., 2004. Lipids in suspended matter and sediments from the East China Sea Shelf. Organic Geochemistry 35, 647–660.

Jeng, W.L., Huh, C.A., 2006. A comparison of sedimentary aliphatic hydrocarbon distribution between the southern Okinawa Trough and a nearby river with high sediment discharge. Estuarine, Coastal and Shelf Science 66, 217–224.

Jeng, W.L., Kao, S.J., 2002. Lipids in suspended matter from the human-disturbed Lanyang River, northeastern Taiwan. Environmental Geology 43, 138–144.

Kao, S.J., Lin, F.J., Liu, K.K., 2003. Organic carbon and nitrogen contents and their isotopic compositions in surficial sediments from the East China Sea shelf and the southern Okinawa Trough. Deep-Sea Research II 50, 1203–1217.

Katayama, H., Watanabe, Y., 2003. The Huanghe and Changjiang contribution to seasonal variability in terrigenous particulate load to the Okinawa Trough. Deep-Sea Research II 50, 475–485.

Kennicutt II, M.C., Barker, C., Brooks, J.M., DeFreitas, D.A., Zhu, G.H., 1987. Selected organic matter source indicators in the Orinoco, Nile and Changjiang deltas. Organic Geochemistry 11, 41–51.

Liu, K.K., Iseki, K., Chao, S.Y., 2000. Continental margin carbon fluxes. In: Hanson, R.B. (Ed.), The Changing Ocean Carbon Cycle, IGBP Book Series. Cambridge University Press, Cambridge, pp. 187–239.

Liu, K.K., Peng, T.H., Shaw, P.T., Shiah, F.K., 2003. Circulation and biogeochemical processes in the East China Sea and the vicinity of

Taiwan: an overview and a brief synthesis. Deep-Sea Research II 50, 1055–1064.

Matsumoto, G., 1983. Comparative study on organic constituents in polluted and unpolluted inland aquatic environments—V. Organic carbons and hydrocarbons in sediments. Water Research 17, 823–830.

Meyers, P.A., 1997. Organic geochemical proxies of paleoceanographic, paleolimnologic, and paleoclimatic processes. Organic Geochemistry 27, 213–250.

Meyers, P.A., Leenheer, M.J., Eadie, B.J., Maule, S.J., 1984. Organic geochemistry of suspended and settling particulate matter in Lake Michigan. Geochimica et Cosmochimica Acta 48, 443–452.

Milliman, J.D., Meade, R.H., 1983. World-wide delivery of river sediment to the oceans. The Journal of Geology 91, 1–21.

Milliman, J.D., Shen, H.T., Yang, Z.S., Meade, R.H., 1985. Transport and deposition of river sediment in the Changjiang estuary and adjacent continental shelf. Continental Shelf Research 4, 37–45.

Milliman, J.D., Qin, Y.S., Park, Y.A., 1989. Sediments and sedimentary processes in the Yellow and East China Seas. In: Taira, A., Masuda, F. (Eds.), Sedimentary Facies in the Active Plate Margin. Terra Scientific Publishing Company, Tokyo, pp. 233–249.

Niino, H., Emery, K.O., 1961. Sediments of shallow portions of East China Sea and South China Sea. Geological Society of America Bulletin 72, 731–762.

ODP, 2001. Seafloor observatories and the Kuroshio Current. Ocean Drilling Program, Leg 195 Preliminary Report. Available at: ⟨http://www-odp.tamu.edu/publications/prelim/195_prel/195toc.html⟩.

Oguri, K., Matsumoto, E., Yamada, M., Saito, Y., Iseki, K., 2003. Sediment accumulation rates and budgets of depositing particles of the East China Sea. Deep-Sea Research II 50, 513–528.

Prahl, F.G., 1985. Chemical evidence of differential particle dispersal in the southern Washington coastal environment. Geochimica et Cosmochimica Acta 49, 2533–2539.

Prahl, F.G., Hayes, J.M., Xie, T.M., 1992. Diploptene: an indicator of terrigenous organic carbon in Washington coastal sediments. Limnology and Oceanography 37, 1290–1300.

Readman, J.W., Mantoura, R.F.C., Rhead, M.M., 1987. A record of polycyclic aromatic hydrocarbons (PAH) pollution obtained from accreting sediments of the Tamar Estuary, UK: evidence for non-equilibrium behavior of PAH. The Science of the Total Environment 66, 73–94.

Readman, J.W., Bartocci, J., Tolosa, I., Fowler, S.W., Oregioni, B., Abdulraheem, M.Y., 1996. Recovery of the coastal marine environment in the Gulf following the 1991 war-related oil spills. Marine Pollution Bulletin 32, 493–498.

Rieley, G., Collier, R.J., Jones, D.M., Eglinton, G., 1991. The biogeochemistry of Ellesmere Lake, UK—I: Source correlation of leaf wax inputs to the sedimentary lipid record. Organic Geochemistry 17, 901–912.

Rohmer, M., Bouvier-Nave, P., Ourisson, G., 1984. Distribution of hopanoid triterpenes in prokaryotes. Journal of General Microbiology 130, 1137–1150.

Salas, N., Ortiz, L., Gilcoto, M., Varela, M., Bayona, J.M., Groom, S., Álvarez-Salgado, X.A., Albaigés, J., 2006. Fingerprinting petroleum hydrocarbons in plankton and surface sediments during the spring and early summer blooms in the Galician coast (NW Spain) after the Prestige oil spill. Marine Environmental Research 62, 388–413.

Shaw, D.G., Hogan, T.E., McIntosh, D.J., 1985. Hydrocarbons in the sediments of Port Valdez, Alaska: consequences of five years' permitted discharge. Estuarine, Coastal and Shelf Science 21, 131–144.

Tang, T.Y., Hsueh, Y., Yang, Y.J., Ma, J.C., 1999. Continental slope flow northeast of Taiwan. Journal of Physical Oceanography 29, 1353–1362.

Tolosa, I., Bayona, J.M., Albaigés, J., 1996. Aliphatic and polycyclic aromatic hydrocarbons and sulfur/oxygen derivatives in northwestern Mediterranean sediments: spatial and temporal variability, fluxes and budgets. Environmental Science and Technology 30, 2495–2503.

Venkatesan, M.I., Brenner, S., Ruth, E., Bonilla, J., Kaplan, I.R., 1980. Hydrocarbons in age-dated sediment cores from two basins in the

Southern California Bight. Geochimica et Cosmochimica Acta 44, 789–802.

Walsh, J.J., Biscaye, P.E., Csanady, G.T., 1988. The 1983–1984 Shelf Edge Exchange Processes (SEEP)—I. Experiment: hypotheses and highlights. Continental Shelf Research 8, 435–456.

Winters, K., Parker, P.L., Van Baalen, C., 1969. Hydrocarbons of blue-green algae: geochemical significance. Science 158, 467–468.

Wong, G.T.F., Chao, S.Y., Li, Y.H., Shiah, F.K., 2000. The Kuroshio Edge Exchange Processes (KEEP) study—an introduction to hypotheses and highlights. Continental Shelf Research 20, 335–347.

Yanagi, T., Takahashi, S., Hoshika, A., Tanimoto, T., 1996. Seasonal variation in the transport of suspended matter in the East China Sea. Journal of Oceanography 52, 539–552.