Composition and sources of organic matter and its solvent extractable components in surface sediments of a bay under serious anthropogenic influences: Daya Bay, China

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ABSTRACT

Organic matter of nine surface sediments from the Daya Bay was Soxhlet-extracted with a mixture of 2:1 (v/v) dichloromethane–methanol and separated into five fractions: non-aromatic hydrocarbons, aromatic hydrocarbons, ketones, alcohols, and fatty acids and asphaltenes, and analyzed to determine their bulk and biomarker composition. Marine autogenic input appears to be a major source of organic matter. Generally, non-aromatic hydrocarbons are the most dominant fraction of solvent-extractable organic matter (EOM) followed by the other four fractions in decreasing amounts: fatty acids and asphaltenes, ketones, alcohols and aromatic hydrocarbons. On average, both non-aromatic hydrocarbon fraction and fatty acid and asphaltene fraction account for ∼40% of EOM. The sources of acyclic methyl ketones, alkanols and fatty acids were examined. For n-alkan-2-ones, allochthonous input is a more important source than marine autogenetic input; the reverse is true for n-fatty acids; for n-alkanols, allochthonous and autogenetic inputs seem comparable. Both n-alkan-2-ones and n-fatty acids in the surface sediments of different areas appear to be derived from common sources.

1. Introduction

Organic matter and its lipid components in different sedimentary environments have received extensive study because they can provide much information for interpreting sedimentary records regarding the sources, transport pathways and diagenetic processes of organic matter (e.g., Duan, 2000; Duan and Ma, 2001; Meyers, 2003; Tuo and Li, 2005; Bai et al., 2006; Lü and Zhai, 2006). Organic matter and its lipid components are naturally occurring materials, but their composition may be influenced by many human activities. Marine coastal areas are subject to a great deal of stress from human activities as modern civilization spreads, as more than half of the world population now lives within 200 km of the coast and that number is still increasing (De Souza et al., 2003). Abundant natural and anthropogenic organic matter derived from land and carried by rivers and the atmosphere are imported to and buried in marine coastal areas (e.g., Gagosian et al., 1987; Ludwig et al., 1996; Raymond and Bauer, 2001; de Haas et al., 2002), which inevitably has an influence on the original characteristics of the lipid composition of organic material in these areas.

Daya Bay is a busy region greatly affected by human activities. The surrounding area is an important economic development district in Guangdong Province. Petrochemical, plastic, printing and other industries as well as harbors are present (Song et al., 2004). Two nuclear plants, Daya Bay Nuclear Power Station and Ling'ao Nuclear Power Station, which came into operation in 1994 and 2002, respectively, are situated on its western coast. Fish, shrimp and shellfish aquaculture are well developed in Daya Bay and profit from its good water quality. Owing to its special status on both environmental and economic levels, Daya Bay has become one of the few bays that have been extensively studied in China. Especially since the 1980s, it has received much attention from researchers in different fields and a national research station with an emphasis on the marine biology of Daya Bay was established in 1984 for regular monitoring (e.g., Pan and Cai, 1996; Pan and Wang, 1998; Zhou et al., 2001; Tang et al., 2003; Zhou and Maskaoui, 2003; Song et al., 2004; Wang et al., 2006). The long-term dataset indicates that the ecosystem and environment of Daya Bay has deteriorated rapidly over the past 20 years (Wang et al., 2004). However, reports related to solvent extractable organic matter (EOM) and lipids in Daya Bay are very scarce. For the first time, this research presents the composition, origin and distribution of EOM and its lipid components including acyclic methyl ketones, alkanols and fatty acids in Daya Bay surface sediments. The information derived from bulk analysis and multi-biomarker data

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is combined to obtain insights into the sources of target lipids in this area and provide a background reference for future research.

2. Materials and methods

2.1. Regional setting

Daya Bay is a subtropical drowned valley bay of the northern South China Sea located on the eastern coast of Guangdong Province, southern China (Fig. 1). It is one of a series of large embayments along the southern coast of China and covers an area of ~600 km^2 with a width of about 20 km and a north–south length of about 30 km. The water depth in Daya Bay averages 11 m, and generally ranges from 6 to 16 m, with the western part being deeper than the eastern part (Xu, 1989; Han, 1995). Although there are no large rivers discharging into the bay, there are over 10 seasonal streams that lead to the bay within a short distance of each other along the coast (Han, 1995).

The average water temperature in the coastal region of Daya Bay is 29.3 °C in summer (July to September) and 17.3 °C in winter (December to February); the weighted mean wind velocity is 4.8 m s^{-1} in summer and 4.6 m s^{-1} in winter; the daily average salinity is about 28.0‰; the tidal current in Daya Bay is dominated by an irregular semidiurnal tide with an average tidal day of about 24.7 h; the mean tidal range is 1.01 m and the maximum is 2.57 m; the current velocities are lower than 0.5 m s^{-1} and currents form a clockwise gyre system in spring and autumn (Zeng et al., 2002). The surface water has an average resident time of about 3.2 days (Wang et al., 1996). The annual mean precipitation in Daya Bay is 1827 mm; dry and rainy seasons can be distinguished (Han, 1995). In recent decades, the mean sedimentary rate of the bay is 0.9 cm year^{-1} (Han, 1995).

2.2. Sampling

Sediment samples were collected from nine sites in Daya Bay using a box grab sampler on January 12 and 13, 2006 (Fig. 1). The details of the sampling sites are given in Table 1. Sediments from the undisturbed top 5 cm were collected and placed in pre-cleaned glass jars and stored at −20 °C in the dark until further analysis. Water depth and temperature of each sampling station are listed in Table 1.

2.3. Chemical analyses

After being freeze-dried, sediment samples were finely ground and homogenized with an agate pestle and mortar. Then, about 80 g of those pretreated samples were Soxhlet-extracted with a
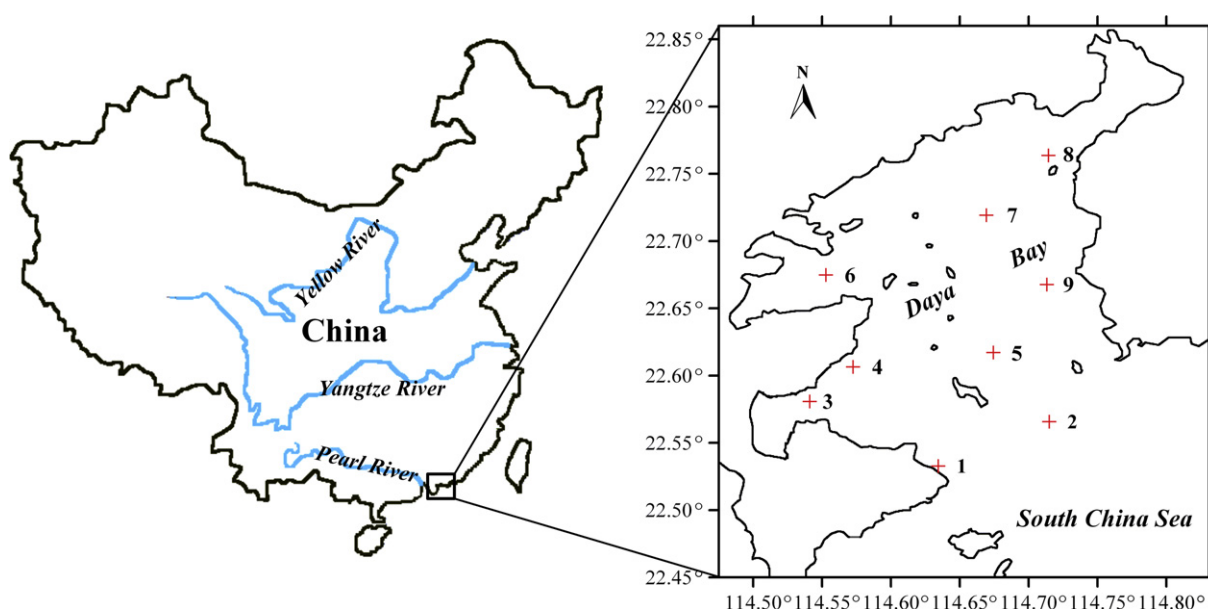

Fig. 1. Location of sampling stations in Daya Bay, South China Sea.

Table 1
The location, water depth, and temperature of each sampling site and bulk geochemical parameters of the sediments sampled

Site	Longitude (E)	Latitude (N)	Water depth (m)	Temperature (°C)	Clay (%)	Silt (%)	Sand (%)	TOC (%)	TN (%)	C/N	EOM$_1$ (μg g^{-1})	EOM$_2$ (%)	F1 (μg g^{-1})	F2 (μg g^{-1})	F3 (μg g^{-1})	F4 (μg g^{-1})	F5 (μg g^{-1})
1	114°38.06′	22°31.97′	15.1	16.5	4.3	93.8	1.9	0.86	0.105	9.6	130.0	1.52	54.7	5.2	16.3	6.2	47.6
2	114°42.89′	22°33.95′	16.2	16.7	5.7	86.9	7.4	1.08	0.118	10.7	163.9	1.51	51.8	5.0	21.0	11.6	74.5
3	114°32.46′	22°34.85′	12.2	17.1	5.8	89.1	5.1	1.40	0.166	9.8	284.7	2.04	182.4	5.8	23.3	1.0	72.2
4	114°34.35′	22°36.39′	11.5	17.4	15.6	73.5	10.9	0.87	0.096	10.6	197.8	2.28	123.5	8.7	17.8	9.9	37.9
5	114°40.46′	22°37.03′	10.9	16.8	28.9	55.6	15.5	1.06	0.109	11.3	88.1	0.83	31.9	5.4	13.6	6.4	30.8
6	114°33.17′	22°40.49′	7.4	16.5	34.6	65.3	0.1	1.60	0.180	10.4	436.1	2.72	275.8	11.7	34.6	21.2	92.8
7	114°40.15′	22°43.16′	10.2	16.8	31.7	66.3	2.0	1.42	0.138	12.0	1637.4	11.53	95.2	4.7	16.5	7.2	1513.8
8	114°42.86′	22°45.82′	7.0	16.2	36.8	61.8	1.4	1.53	0.206	8.7	202.4	1.32	52.3	5.6	27.2	13.1	104.2
9	114°42.79′	22°40.06′	9.9	16.0	34.5	63.4	2.1	1.46	0.196	8.7	168.3	1.15	72.5	5.3	24.4	8.9	57.2

Temperature was measured at a depth of 2 m above the seabed; TOC: total organic carbon; TN: total nitrogen; C/N: atomic carbon/nitrogen ratio; EOM$_1$: extractable organic matter concentration in dry sediment samples evaluated by gravimetry; EOM$_2$: extractable organic matter relative to TOC; F1–F5: the concentration of different EOM fractions in dry sediment samples evaluated by gravimetry.

mixture of 2:1 (v/v) dichloromethane–methanol for 72 h (Duan, 2000; Duan and Ma, 2001). Total extracted portions were treated with activated copper to remove sulfur, weighed to determine EOM content and fractionated by column chromatography on alumina over silica gel with different solvents sequentially. A series of fractions, including non-aromatic hydrocarbons (fraction F1), aromatic hydrocarbons (fraction F2), ketones (fraction F3), alcohols (fraction F4), and fatty acids and asphaltenes (fraction F5), were obtained by successive elution with petroleum-ether, benzene, 10% ethyl acetate in hexane, 20% ethyl acetate in hexane and methanol (Lü and Zhai, 2006; Lü et al., 2006). Each fraction was taken to dryness and then gravimetrically quantified. Prior to instrumental analysis, the F4 fraction of each sample was derivatized with bis(trimethylsilyl)trifluoroacetamide (BSTFA) in pyridine to convert alcohols into their trimethylsilyl ethers. The F5 fraction was saponified with 0.5 mol l^{-1} KOH in methanol and subsequently acidified to pH 2, and the released fatty acids were extracted in hexane and then methylated with 10% BF$_3$–methanol solution to yield fatty acid methyl esters (FAME) (Muri et al., 2004; Lü and Zhai, 2006; Lü et al., 2006).

Lipid fractions were analyzed by gas chromatography–mass spectrometry (GC–MS), using a HP 6890N GC equipped with an HP-5 fused silica capillary column (30 m, 0.25 mm inner diameter, 0.25 μm film thickness) and interfaced to a HP 5973N MS. The GC–MS system was operated in the full scan mode. The GC oven temperature was programmed to increase from 80 to 290 °C at 4 °C min^{-1}, and then kept constant for 30 min. Helium was used as the carrier gas. MS operating conditions were: EI ionization, ion source 230 °C, electron energy 70 eV, interface temperature 280 °C. Identification of the target compounds was based on the NIST (US National Institute of Standards and Technology) mass spectral library supplied with the GC–MS instrument.

The sample granulometry was analyzed using a Malvern Mastersizer 2000 laser diffractometer capable of analyzing particle sizes between 0.02 and 2000 μm. The percentages of the following three grain size groups were determined: <4 (clay), 4–63 (silt), and >63 μm (sand). Crushed and homogenized sediment samples were acidified with 10% HCl overnight to remove carbonates, rinsed with distilled water and dried before the analysis of total organic carbon (TOC) and total nitrogen (TN) using a Perkin–Elmer PE 2400 Series II elemental analyzer (Hedges and Stern, 1984).

3. Results and discussion

3.1. Bulk geochemical parameters

3.1.1. Grain size

The sand content in five of the nine sampling sites were <5% (Table 1), indicating that clay and silt are the main components of the surface sediments in Daya Bay. There was clearly higher clay content in the northern part of the bay than in the southern part.

The relatively high and stable amount of clay in the northern part of the bay probably indicates that the hydraulic conditions in this area are relatively weak.

3.1.2. TOC and TN

TOC content varied in the range of 0.86–1.60% of the dry sediment weight with an average of 1.25% (Table 1). The highest and lowest TOC values were recorded at the sites located at the most extensive aquacultural region in the northwest part of Daya Bay—Aotou Cove (site 6) and the western nearshore part of the bay mouth (site 1), respectively. The total nitrogen content varied in the range of 0.096–0.206% of the dry sediment weight with an average of 0.146% (Table 1). In the surface sediments of Daya Bay, the average TOC content is higher than that of the Yangtze River and Pearl River estuaries and their adjacent shelves; the average TN content is higher than that of the Yangtze River surrounding area and is comparable to that of Pearl River estuaries and their adjacent shelf (Hu et al., 2006a; Lü et al., 2006). The percentages of TOC and TN are significantly correlated (Table 2).

Generally, the hydraulic conditions of an estuary with significant fresh water discharge are relatively stronger than those of a semi-closed bay like Daya Bay; this makes it more difficult for fine suspended particles to be deposited in the sediments of an estuary. Perhaps for this reason, the grain sizes of Daya Bay sediments are finer than those of the Yangtze River and Pearl River estuaries (Zhou et al., 2004; Dai et al., 2005). Previous research indicates that fine-grained sediments easily accumulate and preserve organic matter (e.g., Guo et al., 1999; Lin et al., 2002), and the results of this study, obtained by analyzing the correlations between TOC and the percentage of different grain size fractions, generally agree with these previous reports. As shown in Table 2, a positive correlation exists between TOC and the finest fraction of the sediment, and negative correlations exist between TOC and the other two coarser fractions of the sediment. Similar results can be achieved by analyzing the correlations between TN and the percentage of different grain size fractions. Besides the influence of sediment grain size composition, the difference in organic supply as well as variation in sedimentary inorganic materials (dilution effect) from one area to another can also lead to the fluctuation of TOC and TN concentrations.

Although influenced by extensive early diagenesis, the atomic ratio of TOC to TN (hereafter abbreviated as C/N ratio) is one of the parameters frequently used to identify changes in the proportions of sedimentary organic matter originating from marine autogenic and terrigenous inputs (e.g., Verardo and McIntyre, 1994; Meyers, 1997; Twichell et al., 2002; Boot et al., 2006). Compared to cellulose- and lignin-rich vascular land plants, marine algae are rich in proteins (Twichell et al., 2002). Therefore, fresh marine autogenic organic matter shows C/N ratios typically between 5 and 8, whereas organic matter from terrestrial sources is characterized by C/N ratios of 20 or greater (Emerson and Hedges, 1988; Meyers,

Table 2
The correlation coefficients between different fractions of EOM, EOM, TOC, TN, C/N ratio and granulometric parameters

	F2	F3	F4	F5	EOM	TOC	TN	C/N	Clay (%)	Silt (%)	Sand (%)
F1	**0.821**	**0.681**	0.416	−0.025	0.156	0.461	0.301	0.066	0.085	0.045	−0.349
F2		0.621	**0.709**	−0.260	−0.104	0.197	0.115	0.039	0.220	−0.179	−0.119
F3			**0.703**	−0.252	−0.115	**0.746**	**0.779**	−0.450	0.379	−0.152	−0.618
F4				−0.126	−0.034	0.363	0.320	−0.056	0.492	−0.375	−0.332
F5					**0.983**	0.256	−0.034	0.594	0.275	−0.181	−0.263
EOM						0.344	0.030	0.594	0.299	−0.180	−0.331
TOC							**0.911**	−0.216	0.662	−0.442	−0.612
TN								−0.592	0.595	−0.361	−0.648
C/N									−0.037	−0.126	0.433

Numbers in boldface indicate that the correlation is significant at $p < 0.05$.

1994). The C/N ratios in this study varied in a narrow range of 8.7–12.0 with an average of 10.2, indicating a weak spatial difference (Table 1). Most of the C/N ratios were slightly higher than the typical high end of marine autogenic organic matter (5–8), suggesting a predominance of marine-derived organic matter in Daya Bay.

Hydrodynamic sorting of sediment by grain size can affect the C/N ratios. In general, the C/N ratios of organic matter in fine-sized sediments are lower than those in coarse sediments, partially because fine sediment fractions contain larger proportions of clay minerals, which have both large surface areas and negative surface charges and therefore adsorb ammonia well (Meyers, 1997). No correlation is found between C/N ratio and the different components of sediment, indicating the weak influence of sediment grain size on C/N ratios. Combined with the narrow variation range of C/N ratios, it seems that organic matter in the surface sediments of Daya Bay is from similar non-point sources.

3.1.3. EOM

The concentrations of EOM in the dry sediments varied within a wide range of 88–1637 µg g^{-1} dry sediment; the maximum value appeared at site 7, and was much higher than those at the other sites (Table 1). Extractable organic matter is a small part of TOC

and only accounts for <3% of the TOC in the dry sediments except at site 7 (Table 1). The percentages of EOM to TOC in the surface sediments of Daya Bay are higher than those of the northern South China Sea outer continental shelf (∼0.42% on average) and the area around the Yangtze River estuary (∼0.45% on average) (Lü et al., 2006; Gao et al., 2007). The correlation between TOC and EOM was not significant (Table 2), which seems to provide evidence that organic matter, if it was from the same source, might experience different degrees of degradation in sediments from different areas of Daya Bay. Like those of TOC, a positive correlation exists between EOM and the finest fraction of sediment, and negative correlations exist between EOM and the other two coarser fractions of sediment. The correlation coefficients between them are low and indicate weak relationships (Table 2).

The concentration and percentage of each fraction of EOM are shown in Table 1 and Fig. 2, respectively. Taken as a whole, fraction F1, namely non-aromatic hydrocarbons (NAH), is the most abundant component of EOM in the surface sediments of Daya Bay. Its concentration varies in the range of 32–276 µg g^{-1} with an average of 104 µg g^{-1} dry sediment and accounts for 5.8–64.1% of the EOM with an average of 41.6%. Fraction F5 ranks as the second most prevalent component of EOM with a few exceptions and was
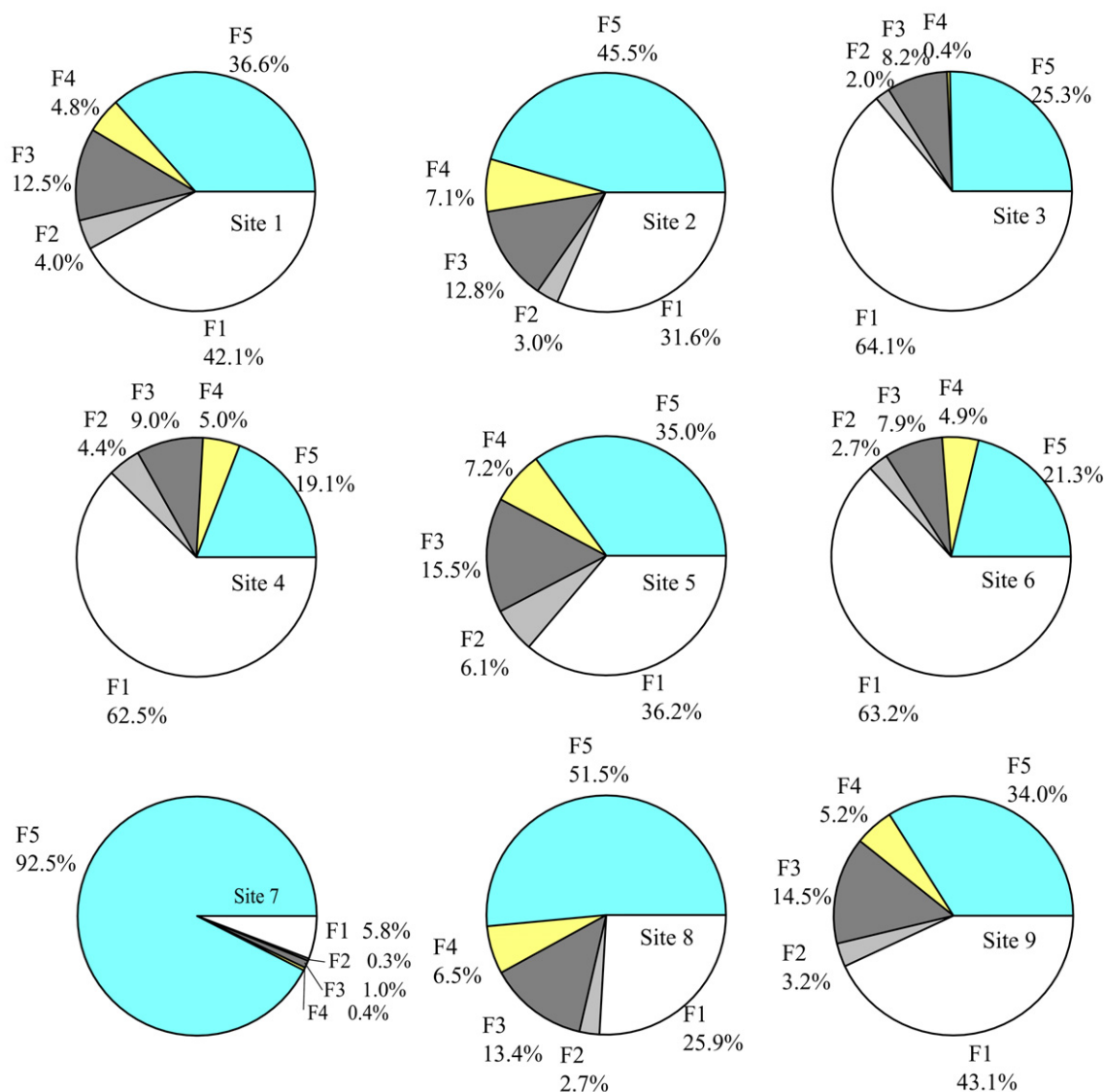

Fig. 2. The percentage of each fraction of solvent-extractable organic matter (EOM).

226 μg g^{-1} dry sediment on average. Its contribution to the EOM concentration was comparable to that of fraction F1 (Fig. 2), averaging about 40.1%. At site 7, the extremely high concentration of fraction F5 caused its EOM concentration to be much higher than that of the other sites. There was a significant correlation between fraction F5 and EOM (Table 2). Fractions F3 and F4, on average accounted for 10.5% and 4.6% of EOM, respectively, and were the third and fourth most prevalent components of EOM. The mean concentration of aromatic hydrocarbons (fraction F2) was 6.4 μg g^{-1} dry sediment. Its contribution to EOM was the smallest among all five fractions, with only about 3.2% on average.

The concentration of NAH in the surface sediments of Daya Bay is much higher at the present time than those of the northern South China Sea outer continental shelf (<10 μg g^{-1} dry sediment) (Gao et al., 2007) and comparable with other marine areas that receive important anthropogenic inputs such as Montevideo Harbor (Muniz et al., 2004), western north and south Mediterranean (Zaghden et al., 2005) and the Bay of Fort de France (Mille et al., 2006). The aromatic hydrocarbons in the surface sediments of Daya Bay are much lower than those of the Bay of Fort de France (Mille et al., 2006). Their average value in the surface sediments of the former is only about one-ninth of the value of the latter. Research carried out in August 1999 showed that in Daya Bay, the total concentration of 16 polycyclic aromatic hydrocarbons (PAHs) varied from 4228 to 29,325 ng l^{-1} in water, and from 115 to 1134 ng g^{-1} dry weight in sediments (Zhou and Maskaoui, 2003). The PAH composition patterns in sediments suggest dominance by medium to high molecular weight compounds, and the ratios of certain related PAHs indicate important pyrolytic and petrogenic sources (Zhou and Maskaoui, 2003).

The influence of sediment grain size on the composition of EOM seems to be weak, because there is no significant correlation between fractions F1–F5 and the granulometric parameters (Table 2). The contents of the EOM fractions were also statistically compared to test common sources. As seen in Table 2, fractions F2 and F3 are significantly correlated with fractions F1 and F4, suggesting that at least some of them are from common sources.

3.2. Biomarkers

3.2.1. Acyclic methyl ketones

The distribution of acyclic methyl ketones in recent sediments is not widely used as an indicator of the origin and fate of organic matter (Pancost and Boot, 2004; Lü and Zhai, 2006). Compared with other straight chain ketones, saturated n-alkan-2-ones are quite common and have been found in higher plant and phytoplankton biomass and a wide variety of depositional environments such as soils, peats, lacustrine and marine sediments, glacier ice, stalagmites, and aerosols (e.g., Hernandez et al., 2001; Tuo and Li, 2005; Bai et al., 2006 and references therein). The n-ketones occurring in higher plants and phytoplankton are generally C_{23}–C_{33} mid-chain ketones with a high predominance of odd numbered carbon chain lengths maximizing at C_{25}, C_{27}, C_{29}, or C_{31} (e.g., Baker, 1982; Hernandez et al., 2001). The distribution of n-ketones in different geological samples typically showed similar distributions with a high predominance of odd-over-even carbon number homologues with maxima at C_{27} or C_{29} (Hernandez et al., 2001; Tuo and Li, 2005). Some researchers proposed that the process of microbial oxidation of n-alkanes and β-oxidation of fatty acids followed by decarboxylation could produce n-alkane-2-ones (e.g., Allen et al., 1971; Volkman et al., 1981; Lehtonen and Ketola, 1990). Apart from natural sources, the anthropogenic discharge of cooking smoke, wood burning smoke, and automobile exhaust are also sources of n-alkan-2-ones (e.g., Simoneit, 1985; Standley and Simoneit, 1987; Rogge et al., 1991).

In the surface sediments of Daya Bay, n-alkan-2-one homologues in the range of C_{15}–C_{36} were identified and they exhibited very similar monomodal distributions (Fig. 3). The high molecular
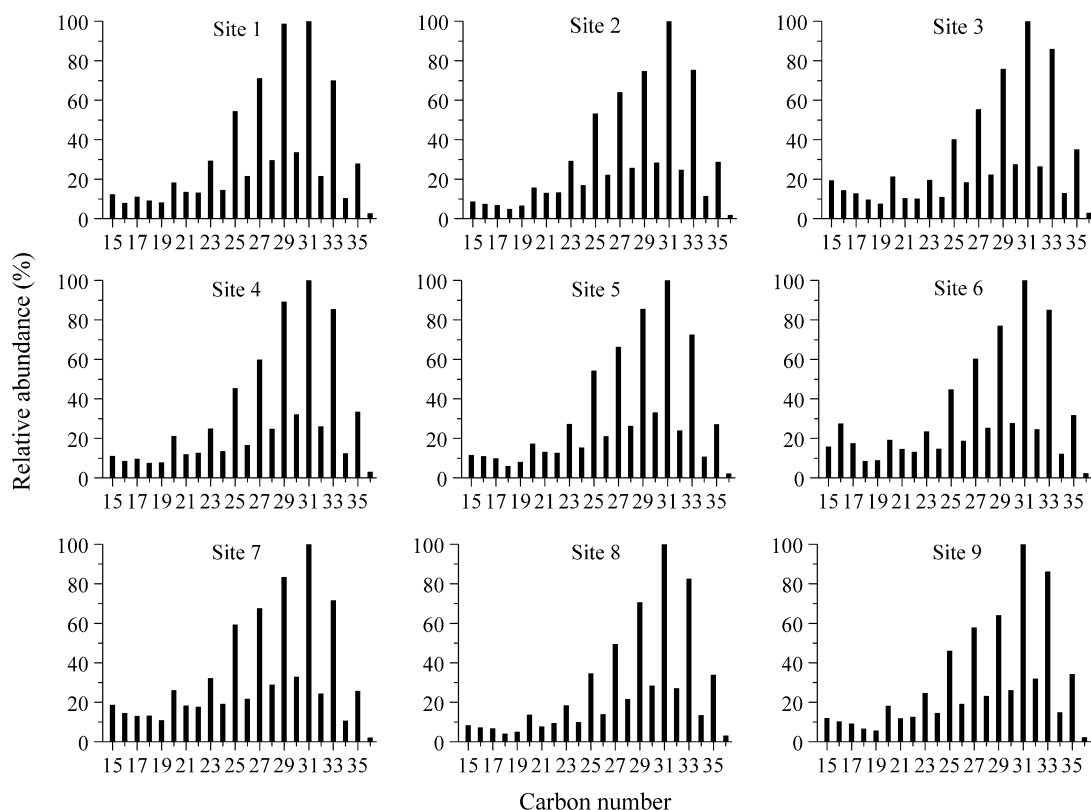

Fig. 3. Distribution patterns of n-alkan-2-ones from the surface sediment samples of Daya Bay, normalized to the most abundant components.

Table 3
Analytical data from acyclic methyl ketones

Index	Site								
	1	2	3	4	5	6	7	8	9
Major peaks	C_{31}	C_{31}	C_{31}	C_{31}	C_{31}	C_{31}	C_{31}	C_{31}	C_{31}
CPI	3.4	3.3	3.4	3.4	3.3	3.4	3.2	3.3	3.1
ACL	29.2	29.4	29.7	29.5	29.3	29.6	29.2	29.7	29.6
LMW/HMW	0.13	0.11	0.17	0.13	0.13	0.20	0.19	0.10	0.13
%Isoprenoid	14.6	10.4	20.5	15.9	15.3	17.4	20.4	8.6	11.7

$CPI = 2\sum n\text{-}C_{25-35}(\text{odd})/(\sum n\text{-}C_{24-34}(\text{even}) + \sum n\text{-}C_{26-36}(\text{even})).$
$ACL = (23(n\text{-}C_{23}) + 24(n\text{-}C_{24}) + 25(n\text{-}C_{25}) + \cdots + 35(n\text{-}C_{35}))/(n\text{-}C_{23} + n\text{-}C_{24} + n\text{-}C_{25} + \cdots + n\text{-}C_{35}).$
$LMW/HMW = \sum n\text{-}C_{15-21}/\sum n\text{-}C_{22-36}.$
%Isoprenoid = the percentage of 6,10,14-trimethylpentadecan-2-one to total n-alkan-2-ones.

weight (HMW) n-alkan-2-ones ($\geqslant C_{22}$) show odd carbon numbers predominating with CPI values that varied in a narrow range of 3.1–3.4 (Table 3). The low molecular weight (LMW) n-alkan-2-ones ($<C_{22}$) show no carbon number predominance (Fig. 3). The dominant peak occurred at C_{31} in all samples (Fig. 3 and Table 3). The LMW/HMW ratios were all very low and no greater than 0.20, showing that the homologues with relatively longer chains are the predominant members of the n-alkan-2-ones (Table 3). Fig. 3 and the CPI values of HMW n-alkan-2-ones clearly indicate that the dominance of odd numbered homologues in the surface sediments of Daya Bay is not as remarkable as those in typical biomass. Based on all evidence, although there is no doubt that a majority of HMW n-alkan-2-ones are from terrestrial higher plants, there is likely a sizeable proportion of n-alkan-2-ones, both LMW and HMW, derived from anthropogenic input and/or degradation products of other lipid precursors.

The average chain length (ACL) is a parameter that can be used to identify n-alkyl lipids from different vegetation (Ternois et al., 2001; Boot et al., 2006; Jeng, 2006). Vegetation types are the main influence on chain length of terrigenous leaf lipids. For example, leaf lipids derived from grasslands may have longer chain lengths on average than leaf lipids from plants in forests (Cranwell, 1973). The n-alkan-2-one ACL data varied in a narrow range of 29.2–29.7 indicating that n-alkan-2-ones in the surface sediments of Daya Bay may be from similar non-point sources.

Similar to what was found in the surface sediments of Changjiang Estuary by Lü and Zhai (2006), 6,10,14-trimethylpentadecan-2-one (6,10,14-TMPD-2-one) was the only isoprenoidal methyl ketone identified in the surface sediments of Daya Bay. Isoprenoid ketones may be produced in several processes including bacterial degradation and photosensitized oxidation of free phytol, photodegradation of chlorophyll phytyl side chains, and photosensitized oxidation of some isoprenoid hydrocarbons such as pristane and phytane (Rontani et al., 1992). The two former processes have been proposed as the potential main sources of 6,10,14-TMPD-2-one in the marine environment (Rontani and Bonin, 2000). It has also been reported that in oxygenated surface sediments of temperate marine areas, the hydrolysis of chlorophyll photoproducts is probably another major source for sedimentary 6,10,14-TMPD-2-one (Rontani and Bonin, 2000). In the surface sediments of Daya Bay, 6,10,14-TMPD-2-one relative abundance exceeds the most abundant n-alkan-2-ones except for at sites 2, 8, and 9. Its abundance alone accounted for 8.6–20.5% of the total acyclic methyl ketones with an average of 15.0% (Table 3), which was higher than that of pristane versus total n-alkanes (7.3% on average, Gao et al., unpublished data). This suggests multiple causes for the observed distributions of isoprenoid ketones (Tuo and Li, 2005).

3.2.2. Alkanols

In coastal environments, n-alkanols from fresh leaf waxes exhibit a pronounced even-over-odd carbon number predominance of longer chain components, which mainly comprise the n-C_{20} to

n-C_{34} homologues (Baker, 1982; Bianchi, 1995). Shorter chain n-alkanols are considered to originate from aquatic organisms such as bacteria or plankton (Ogura et al., 1990; Rielley et al., 1991). However, it has been argued that shorter chain n-alkanols are not accurate indicators of biotic sources due to their ubiquitous nature (Meyers et al., 1984).

The n-alkanols with carbon numbers ranging from 14 to 34 were detected in the surface sediments of Daya Bay. The distributions of n-alkanols were bimodal and characterized by an even carbon predominance (Fig. 4). In all samples, n-C_{16} was the most dominant component of short chain (C_{14}–C_{20}) n-alkanols; n-C_{22} or n-C_{28} was the most dominant component of long chain (C_{21}–C_{34}) n-alkanols (Fig. 4 and Table 4). The abundant mid-chain length C_{22} and C_{24} n-alkanols may be attributed to aquatic macrophytes (Ficken et al., 1998). The CPI values of n-alkanols ranged from 4.2 to 6.5 (Table 4). The strong even carbon number predominance and the molecular distributions suggest that n-alkanols are mainly from fresh biogenic sources and have not been deeply degraded. The ratios of n-alkanols with carbon numbers $\leqslant 20$ to those with >20 carbons (LMW/HMW) show clear spatial differences with the highest and lowest values appearing at the northeast (site 8) and middle (site 5) parts of the bay, respectively (Table 4). The mean LMW/HMW ratio is 0.94 suggesting that the contribution of marine and terrigenous inputs seem comparable. The n-alkanol ACL data varied in the range of 25.4–27.6, which was wider than that for n-alkan-2-ones (Table 4). This may be attributable to either source or preservation differences. Combined with the data of the C/N ratio and EOM, the preservation difference between sampling stations seems to be a major factor.

3.2.3. Fatty acids

As shown in Fig. 5, saturated n-fatty acids (SFAs) with carbon numbers ranging from 14 to 32 were recorded in all samples and exhibited bimodal distributions with maxima at C_{16} and C_{24} for short chain (C_{14}–C_{21}) and long chain (C_{22}–C_{32}) homologues, respectively (Table 5). CPI values ranged from 4.2 to 5.5 (Table 5), clearly indicating that SFAs in the samples are dominated by the components with even carbon numbers. The samples from the two sites located at the bay's mouth have higher CPI values than the other sites. Such a distribution pattern of SFAs has been widely recognized in marine sediments (e.g., Simoneit, 1978). The relative abundance of individual SFAs shows significant differences among the sediments. For example, the most abundant saturated n-fatty acid, namely n-$C_{16:0}$, accounted for about 31–84% of the total SFAs with the lowest observed at site 1 and the highest at site 7. Compared with n-$C_{16:0}$, n-$C_{24:0}$, the most abundant component of long chain SFAs was much lower and only occupied about 1–11% of the total SFAs with the lowest found at site 7 and the highest at site 1.

Generally, n-fatty acids from higher plant waxes show carbon numbers of >C_{22} with a maximum at C_{24} or C_{26}, whereas those from algae and bacteria range between C_{12} and C_{20} with a maximum at C_{16} (e.g., Eglinton and Calvin, 1967; Simoneit, 1978).
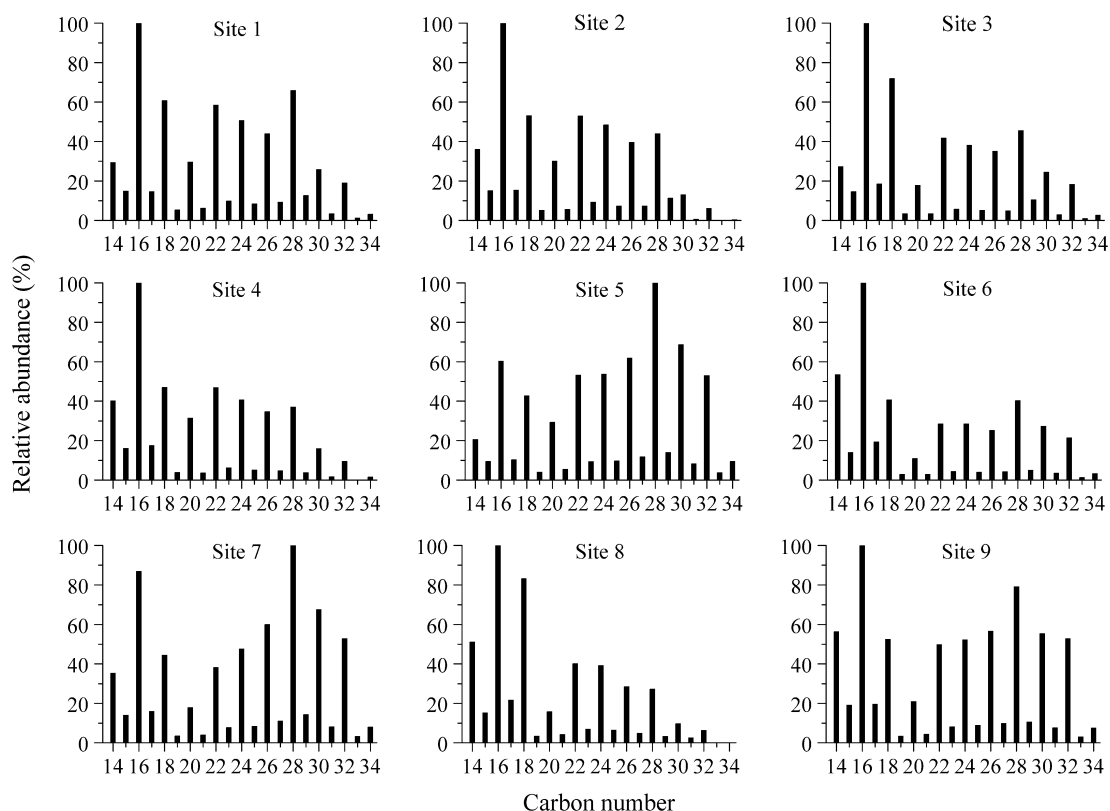
Fig. 4. Distribution patterns of *n*-alkanols from the surface sediment samples of Daya Bay, normalized to the most abundant components.

Table 4
Analytical data from alkanols

Index	Site								
	1	2	3	4	5	6	7	8	9
Major peaks	C_{16}, C_{28}	C_{16}, C_{22}	C_{16}, C_{28}	C_{16}, C_{22}	C_{16}, C_{28}	C_{16}, C_{28}	C_{16}, C_{28}	C_{16}, C_{22}	C_{16}, C_{28}
CPI	4.7	4.2	5.4	6.4	6.1	6.5	6.3	4.6	6.3
ACL	26.1	25.4	26.4	25.7	27.4	27.0	27.6	25.3	27.3
LMW/HMW	0.80	1.03	1.06	1.21	0.38	1.21	0.51	1.62	0.67

CPI = $\sum n\text{-}C_{24-34}(\text{even})/\sum n\text{-}C_{23-33}(\text{odd})$.
ACL = $(22(n\text{-}C_{22}) + 24(n\text{-}C_{24}) + 26(n\text{-}C_{26}) +\cdots+ 34(n\text{-}C_{34}))/(n\text{-}C_{22} + n\text{-}C_{24} + n\text{-}C_{26} +\cdots+ n\text{-}C_{34})$.
LMW/HMW = $\sum n\text{-}C_{14-20}/\sum n\text{-}C_{21-32}$.

Previous research has also shown that sewage discharge can bring fatty acids such as $n\text{-}C_{16:0}$ and $n\text{-}C_{18:0}$ to marine coastal areas and influence their original characteristics (Quemeneur and Marty, 1992). Daya Bay is a eutrophic bay with high phytoplankton abundance and primary productivity at most times of the year and receives little freshwater inputs (Song et al., 2004; Wang et al., 2006). The sewage-derived fatty acids are unlikely to be a major source of fatty acids in the sediments. In the surface sediments of Daya Bay, the LMW/HMW ratios of SFAs varied within a large range of 1.8–23.2 (Table 5), suggesting that the contribution of SFAs from algae and bacteria is greater than that from higher plant waxes, especially for site 7. Like those of *n*-alkan-2-ones, the ACL data of SFAs showed no clear spatial variation, which indicated similar sources (Table 5).

Branched (*iso-* and *anteiso-*) fatty acids are representative of those from bacterial origins (e.g., Budge and Parrish, 1998; Carrie et al., 1998). Branched fatty acids identified in this study were C_{15}, C_{16}, and C_{17} *iso*-fatty acids, and C_{15} and C_{17} *anteiso*-fatty acids. Their abundance was relatively low and only accounted for 2.3–12.2% of the total SFAs with an average of 7.9% (Table 5). These

values combined with the data of LMW/HMW ratios suggest that algae-derived fatty acids are the major source of total fatty acids in the surface sediments of Daya Bay. The percentage of branched fatty acids to total SFAs was much lower in the surface sediments of Daya Bay than in those of the Pearl River estuary and its adjacent marine area where this value was ∼40% on average (Hu et al., 2006b).

Few unsaturated fatty acid homologues, such as $C_{16:1}$, $C_{18:1}$, and $C_{18:2}$, were detected in some samples with relative lower amounts when compared to other fatty acid compounds. Unsaturated fatty acids are rich in marine organisms. The ratio of $C_{16:1}/C_{16:0}$ can be used to distinguish between diatom- and dinoflagellate-derived fatty acids, and values >1.6 are thought to be evidence of fatty acids originating from diatoms in different aquatic systems (Budge and Parrish, 1998). However, during the process of sedimentation, unsaturated fatty acids are more vulnerable to biological and chemical degradation than saturated ones; this results in the ratio of $C_{16:1}/C_{16:0}$ in surface sediments being well below 1 (Birgel et al., 2004). In this study, no value was found for the $C_{16:1}/C_{16:0}$ ratios that was >0.1, suggesting a deep degradation of fatty acids. The ab-
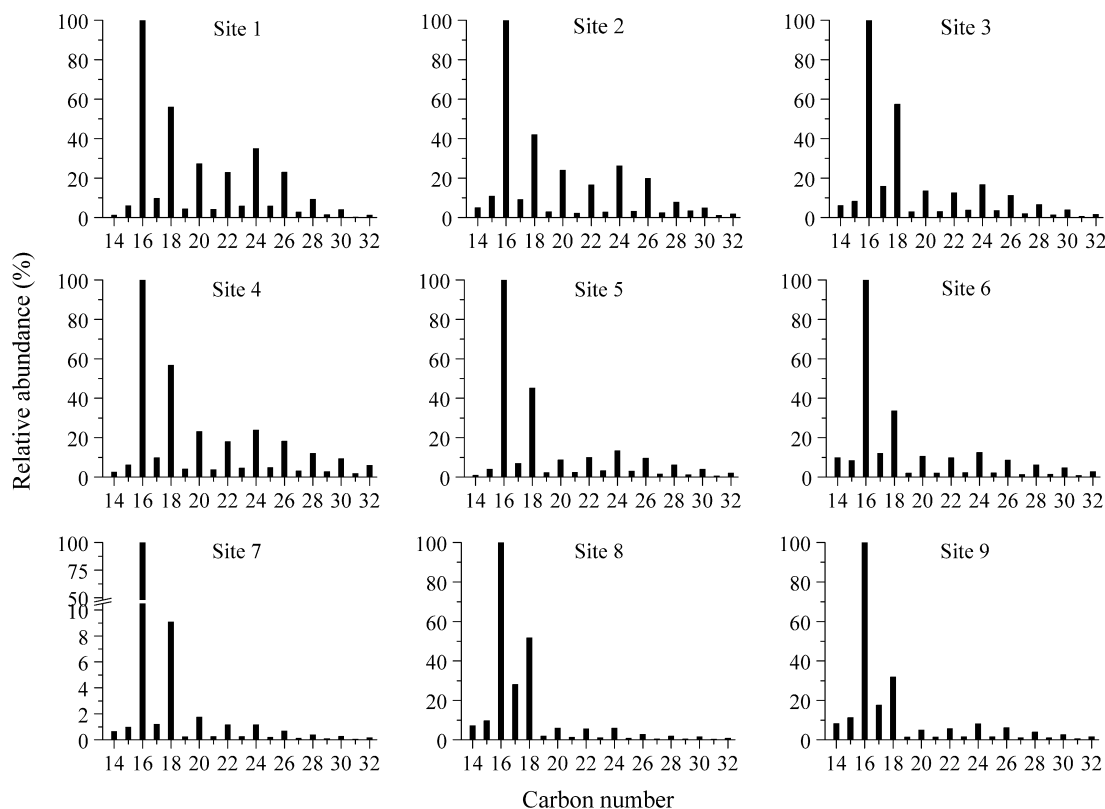
Fig. 5. Distribution patterns of saturated n-fatty acids from the surface sediment samples of Daya Bay, normalized to the most abundant components.

Table 5
Analytical data from fatty acids

Index	Site								
	1	2	3	4	5	6	7	8	9
Major peaks	C_{16}, C_{24}	C_{16}, C_{24}	C_{16}, C_{24}	C_{16}, C_{24}	C_{16}, C_{24}	C_{16}, C_{24}	C_{16}, C_{24}	C_{16}, C_{24}	C_{16}, C_{24}
CPI	5.5	5.3	4.2	4.6	4.2	4.8	4.3	4.6	4.3
ACL	25.0	25.1	25.2	25.5	25.3	25.4	25.0	25.0	25.4
LMW/HMW	1.8	2.1	3.0	1.9	2.9	3.2	23.2	8.7	4.9
%Branched	6.6	12.2	9.8	6.8	4.8	8.6	2.3	9.6	10.1

CPI $= 2\sum n\text{-}C_{24-32}(\text{even})/(\sum n\text{-}C_{23-31}(\text{odd}) + \sum n\text{-}C_{25-33}(\text{odd}))$.
ACL $= (22(n\text{-}C_{22}) + 24(n\text{-}C_{24}) + 26(n\text{-}C_{26}) +\cdots+ 32(n\text{-}C_{32}))/(n\text{-}C_{22} + n\text{-}C_{24} + n\text{-}C_{26} +\cdots+ n\text{-}C_{32})$.
LMW/HMW $= \sum n\text{-}C_{14-20}/\sum n\text{-}C_{21-32}$
%Branched = the percentage of branched fatty acids to the total saturated n-fatty acids.

sence of most polyunsaturated fatty acids, which are useful biomarkers for phytoplankton community differentiation (e.g., Colombo et al., 1996; Carrie et al., 1998; Meziane and Tsuchiya, 2000), further validates the deep degradation of fatty acids. These compounds are hard to preserve in their original amounts in marine sediments owing to their labile characteristics of rapid loss through bacterial degradation and/or zooplankton grazing (Carrie et al., 1998).

4. Conclusions

The surface sediments from Daya Bay were sampled and analyzed to investigate their bulk organic geochemical characteristics and the composition and origin of some lipid biomarkers including acyclic methyl ketones, alkanols and fatty acids.

The evidence of C/N ratios indicates that the organic matter at all sampling sites are derived from both marine autogenic and terrestrial origins in different proportions. The marine autogenic

input seems to be the major origin of organic matter. Spatial differences were not significant. The weak correlation between TOC and EOM seems to provide evidence that organic matter in sediments from different areas of Daya Bay has experienced different degrees of degradation.

On average, the relative abundance of different fractions of EOM followed the sequence of non-aromatic hydrocarbons > fatty acids and asphaltenes > ketones > alcohols > aromatic hydrocarbons. The contribution of non-aromatic hydrocarbons, and fatty acids and asphaltenes to EOM is comparable and much higher than the other three fractions. Together, they account for 71.2–98.3% of EOM with a mean of ~80%. Sediment grain sizes have a weak influence on the composition of EOM.

For n-alkan-2-ones, allochthonous input to the sediments is a more important source than marine autogenetic input. The reverse is true for n-fatty acids. The n-alkanols from allochthonous and autogenetic inputs seem comparable. Based on all evidence, it seems that point-source influences on the composition of

n-alkan-2-ones and *n*-fatty acids are weak. The *n*-alkan-2-ones in the surface sediments from different areas of Daya Bay seem to be derived from common sources, as do *n*-fatty acids.

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