Chemical Characteristics and Source Implications of Petroleum Hydrocarbon Contaminants in the Sediments near Major Drainage Outfalls along the Coastal of Laizhou Bay, Bohai Sea, China

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Abstract The associated industrial and urban developments are located to a large extent along the Laizhou Bay, Bohai Sea coastal and raw sewage is often discharged into near shore waters with little treatment. To find out chemical characteristics and pollution source of the petroleum related contaminations in sediments near the major drainage outfalls located in the coastal, in this study, 10 surface sediment samples were collected during June. Sediment samples were extracted by organic solvents, separated by silica gel column chromatography and the concentrations and the profiles of *n*-alkane, biomarker and PAH in sediments were analyzed by gas chromatography-mass selective detector (GC-MSD). The use of several molecular markers and related indexes derived for *n*-alkane and PAHs has been proposed for assessing the relative contributions to the environment of hydrocarbon sources. As a result, *n*-alkanes reflect that the sea area of paper mill (Station ZZ08) is dominated by vascular plant. DY petroleum oil field and outer shore of the paper mill (Station ZZ02) have some degrees of petroleum related hydrocarbon contamination. Whereas the contamination of the sea area of TH River may be ascribed to different sources such as territorial non-point pollution source, domestic sewages, and stormwater runoff. Judged by their PAH ratios, the sediments near the paper mill (Station ZZ02) and the outer station of the oil field (Staion TH2) were pyrolytic. The estuary of Tiao River including the inner Station THX, TH10 and TH05 are petrogenic. The marine sediment near DY drainage outfall may have a mixture source of PAH both pyrolytic and petrogenic.

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1 Introduction

Polynuclear aromatic hydrocarbons (PAHs) as ubiquitous environmental contaminants are of great concern because they are carcinogenic and resistant to biodegradation (Van Metre et al., 2000). In the aquatic environment, hydrocarbons such as PAHs and *n*-alkanes tend to associate with particulate material

(PM) because of their low water solubility and hydrophobic nature (P. J. Gearing et al., 1980). The continuous settling of PM matter along with flocs and fecal pellets favors the transport of hydrocarbons to the bottom sediments, which act as a long-term reservoir of these hydrophobic contaminants. Therefore, the chemical nature of the hydrocarbons in sediments can act as indicator of the interrelationship between natural and anthropogenic variables (Salomons, 1995). The characteristics of the concentration levels and distribution of *n*-alkanes, PAHs and biomarkers in sediments have been proven to be an efficient tool for petroleum related pollution source identification and impact assessment (Kowalewska et al., 1997; Witt, 1995), and A series of hydrocarbon indexes have been developed and successfully applied to characterize petroleum related contamination sources (Thomas et al., 1989; Zheng & Richardson, 1999).

Laizhou Bay make up of 10% of the total area of Bohai Sea in northern China. The coastal region surrounding Bohai Sea is one of the three most densely populated and industrialized areas in China, along with Yantze river delta in Shanghai in Mid-China, and Pearl River Delta near Hong Kong in the south. Industrial and urban developments in Laizhou Bay are located to a large extent along the coast, and raw sewage is often discharged into the near-shore waters with little treatment. Three groups of sediments near the major drainage outfalls in the coastal of Laizhou Bay, Bohai Sea have been studied and compared in this work. The first is located at the western shore of the Bay near DongYing (DY) oil field, which suspected of oil contamination. The second was collected at the estuary of the adjacent Tiao River west of the DY oil field. The river flows into the Bay with influx of terrestrial sewage as well as industrial wastewaters more than 5 miles upstream. The third sediment samples were collected at the eastern shore of the Bay, where locate a of a paper mill with a production of more than 2,500 tons paper pulp every year.

As a result of shipping, sewage and industrial discharges, many locations along the coastal waters of the Bay are no longer considered fit for their designated beneficial uses. The government and the public are accelerating their effort in recent years to control the situation, and our study is a part of a large research program dedicated to the development of pollution monitoring technology for applications in

marine environment. The objective of this study was to characterize and compare the hydrocarbon profiles of the surface sediment near the above three major drainage outfalls along the coastal of Laizhou Bay namely, DY oil field, TH River, and ZZ paper mill. The data was intended for the tracing and apportion of major sources of hydrocarbon pollution using *n*-alkane profiles, PAH distributions and petroleum biomarkers.

2 Experimental

2.1 Sediment collection

A total of 10 surface sediment samples were obtained at 10 stations near the major drainage outfalls including DY petroleum oil field, TH River and ZZ paper mill along the coastal of Laizhou Bay, Bohai Sea by a grab sampler in June, 2004 (Fig. 1 and Table 1). All samples were stored under frozen conditions and transported to the laboratory. Before chemical treatments, individual samples were dried to constant weigh using a freeze dry system (Labconco). The freeze dried sediment solids were then ground, sieved (60 meshes) and stored at −20°C until use.

2.2 Hydrocarbon extraction from sediments

Approximately 20 g (±0.001 g) dry sediment was placed in 125 ml solvent-rinsed Erlenmeyer flask, spiked with the appropriate aromatic hydrocarbon surrogate (*o*-terphenyl), and mixed with 1 g copper powder to desulfurize and 2 g sodium sulfate to dry the sample. The mixture was then serially extracted three times with 60 ml of 1:1 hexane/dichloromethane (DCM), DCM, and DCM for 40 min by sonication. The extracts were combined and then concentrated to appropriate volumes by rotary evaporation. The halogenated solvent was exchanged to hexane, and the hexane extract was further concentrated to a small volume (0.5–1 ml). All hexane and methylene chlorides were of pesticide-residue quality (Tedia).

2.3 Sample cleanup

Silica gel liquid chromatography was used for sample cleanup. Silica gel (particle size 100–200 meshes)

Fig. 1 Location map of sampling stations: *A*: ZZ paper mill; *B*: TH River; *C*: DY petroleum oil field
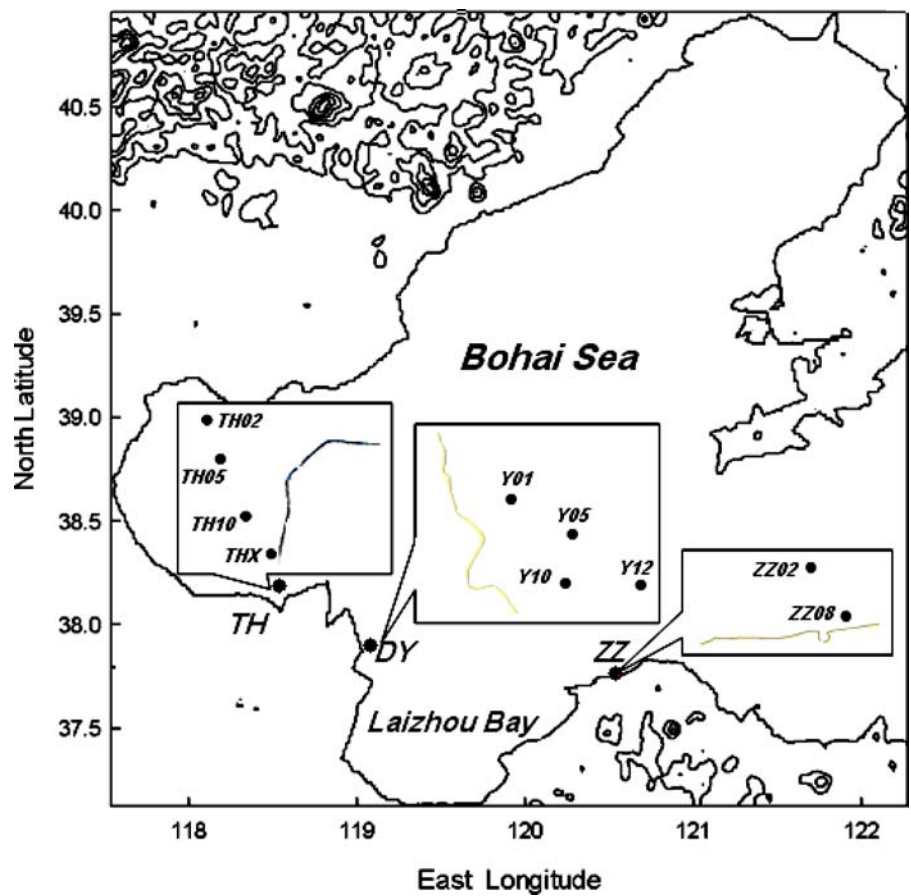
was activated at 300°C for 4 h in a porcelain dish. A glass chromatography column (30×1.2 cm i.d) consisting of 6 g silica gel (deactivated with 5% distilled water) and 1 g of sodium sulfate on column top was equilibrated with 20 ml hexane. The sample extract aliquot was then added to the column and the fraction of aliphatic hydrocarbons (F1) was eluated with 18 ml

distilled hexane. The column was further eluted with 16 ml of a mixture of DCM and hexane (1:1) to separate out the PAHs fraction (F2). These two fractions were concentrated under a gentle stream of nitrogen to the appropriate volumes. F2 was spiked with internal standards (*p*-terphenyl-d_{14}) and then both fractions were adjusted accurately to appropriate

Table 1 Locations of sediments collected from the marine waters near the major drainage outfalls, namely DY petroleum oil field, ZZ paper mill and TH River along the coastal of Laizhou Bay

Drainage outfalls	Sample	Long./Lat.	
DY	Y01	119°6.24′	37°54.30′
	Y05	119°2.44′	37°53.36′
	Y10	119°6.60′	37°51.65′
	Y12	119°8.76′	37°51.62′
ZZ	ZZ08	120°30.54′	37°45.23′
	ZZ02	120°30.54′	37°46.38′
TH	THX	118°33.98′	38°2.03′
	TH10	118°38.22′	38°3.02′
	TH05	118°37.44′	38°3.95′
	TH02	118°37.44′	38°4.63′

volumes (1 ml) for GC-MS (gas chromatography mass spectroscopy) analysis.

2.4 GC/MS analysis

An Aglient 6890 series gas chromatograph with a HP-5 MS capillary column (30 m, 0.25 mm diameter and 0.25 um thickness film, 95% dimethyl-5%dephenyl-polysiloxane) was used for the analysis of petroleum hydrocarbons. For F1 (*n*-alkanes(C14–C35) and biomarkers) determination the GC oven temperature program was: 50°C for 2 min, 6°C/min to 300°C, and finally held constant for 10 min. The MSD detector for the GC was set to selective ion monitoring mode (SIM) and the *n*-alkanes were extracted from the chromatograms using the *m/z* 85 ion. *N*-alkanes were quantified using *n*-C14–C35 aliphatic hydrocarbon standards. Standard solution of each hydrocarbon was prepared in hexane and injected to obtain a standard curve of peak area against concentrations. UCM concentrations were determined based on the calibration curve of C16. Extraction efficiency for F1 of blank sediment has an average value of 83.2%. Biomarker profile of triterpanes was extracted from the chromatogram using *m/z* 191 ion.

For the composition and concentrations of individual PAHs (F2), the GC oven temperature program was: 60°C for 2 min, then 4°C/min to 260°C, held constant for 3 min and 8°C/min to 300°C, held for 5 min. A mix standard of 11 typical PAHs containing fluorene (F), phenanthrene (P), anthracene (A), fluoranthene (FL), pyrene (Py), benz(a)anothrance (BaA), chrysene (C),

benzo(b)fluoranthene (BBF), benzo(k)fluoranthene (BKF), benzo(a)pyrene (BAP) and benzo(ghi)perylene (B(ghi)P) was used for calibration and identification of each PAH peak in the samples. Individual *m/z* ions specific to the respective PAHs were used in selected ion monitoring mode for data collection. Compound identification was carried out by comparison of retention time with corresponding standards. The corresponding response factor for individual PAHs were estimated based on a calibration line involving five separated injections of the mixed external standards. The internal standard (*p*-terphenyl-d_{14}) was used to calculate the peak area of each individual PAH in the sample. The quantifications of seven alkylated PAHs, i.e., C1-fluorenes(C1-F),C2-fluorenes(C2-F), C1-phenanthrenes(C1-P),C2-phenanthrenes(C2-P), C3-phenanthrenes(C3-P),C1-fluoranthenes(C1-Fl), C1-Chrysenes(C1-C) were approximated by the respective response factors of their parent PAHs. Recovery for the whole procedure for F2 fractions were between the range of 74–104% based on the calculation of the spiked surrogate internal standard. The concentrations of individual hydrocarbons such as PAHs in the sediments are in general much higher than their respective detection limits using our analytical methods, which range from 0.1 to 1 ng/g.

3 Results and Discussion

The types and concentration levels of *n*-alkanes and PAHs in the three drainage outfall sediment samples

Table 2 Concentrations (μg/g dry wt.) and characteristics of petroleum aliphatic hydrocarbons in marine waters near the major drainage outfalls, namely DY petroleum oil field, ZZ paper mill and TH River along the coastal of Laizhou Bay

Drainage outfalls	Station	UCM	*n*-Alk	Total F1	UCM/*n*-Alk	*n*-Alk/C16	MH	LMW/HMW	CPI
ZZ	ZZ08	/	1.87	1.87	/	80	c29	0.09	3.68
	ZZ02	3.87	4.15	8.02	0.93	26.2	C17	0.28	3.08
TH	THX	2.91	5.82	8.73	0.50	33.4	C17,C23,C29	0.16	1.82
	TH10	2.10	3.14	5.24	0.67	18.7	C29	0.39	2.33
	TH05	0.31	1.42	1.73	0.22	49.3	C29	0.16	3.24
	TH02	/	1.11	1.11	/	23	C17	0.29	3.76
DY	Y01	0.55	1.80	2.35	0.31	7.8	c17	0.53	3.26
	Y05	0.81	1.24	2.05	0.65	17.5	c17	0.56	2.72
	Y10	/	0.40	0.40	/	23.4	C17	0.36	2.31
	Y12	/	0.60	0.60	/	35.6	C17	0.22	1.52

UCM, unresolved complex mixture; *total F1*, UCM+*n*-Alk; *n*-Alk, $\sum$*n*-C14–*n*-C35; *MH*, major hydrocarbon; *LMW/HMW*, $\sum \leq n - 20 / \sum \geq n - C21$; *CPI*, $2(C27 + C29)/(C26 + 2C28 + C30)$

are compared and the characteristics of the contaminants are examined using a series of hydrocarbon indexes.

3.1 Aliphatic hydrocarbons

Table 2 shows that the carbon number of the *n*-alkanes for sediments ranged between C14 and C35. The highest total F1 concentration (8.73 µg/g dry wt.) was found in Station THX in the estuary of TH River, indicating the deposition of high proportion of organic compound by the river. The total weight of aliphatic material F1 in the sediment of sea area of TH River decreases with increasing distance from shore. The sediment samples in marine waters near the DY petroleum oil field contain relatively less total F1. And sediments near the ZZ paper mill contain less total F1 in the costal stations (Station ZZ08) than in the outer stations (Station ZZ02).

The *n*-alkane found in sediment could be originated from man-made pollution or natural, biogenic sources such as plants, bacteria and algae. Typically, C15, C17 or C19 are indicators for marine algae, whereas C27, C29, C31 are indicators for vascular land plants, and both with odd carbon predominance (Nishigima et al., 2001). The indicator for petroleum products is often around C18 (Colombo et al., 1989), and non-biodegraded crude oils contain a range of *n*-alkanes, which decrease in concentration with increasing carbon number. The *n*-alkanes in petroleum contaminants do not have odd carbon predominance and can therefore be distinguished from the biogenic ones. In this study, data of MH suggests the presence of petroleum oil in most of the sediments stations (ZZ02, TH2, Y01, Y05, Y10 and Y12, the chromatogram for Y05 was shown in Fig. 2c), whereas stations TH10 and TH5 and the coastal station ZZ08 (Fig. 2a) are dominated by vascular plants. Station THX have more than one major hydrocarbons, suggesting the presence of mixed sources (Fig. 2b).

The unresolved complex material (UCM) appearing as unresolved broad humps in the chromatogram (Fig. 2b and c) is due to the more recalcitrant components in the degraded oil, mainly branched and cyclic compound and some partially oxidized hydrocarbons. UCM is a strong characteristic of petrogenic contamination. The occurrence of high concentrations of UCMs in the sediments would mean
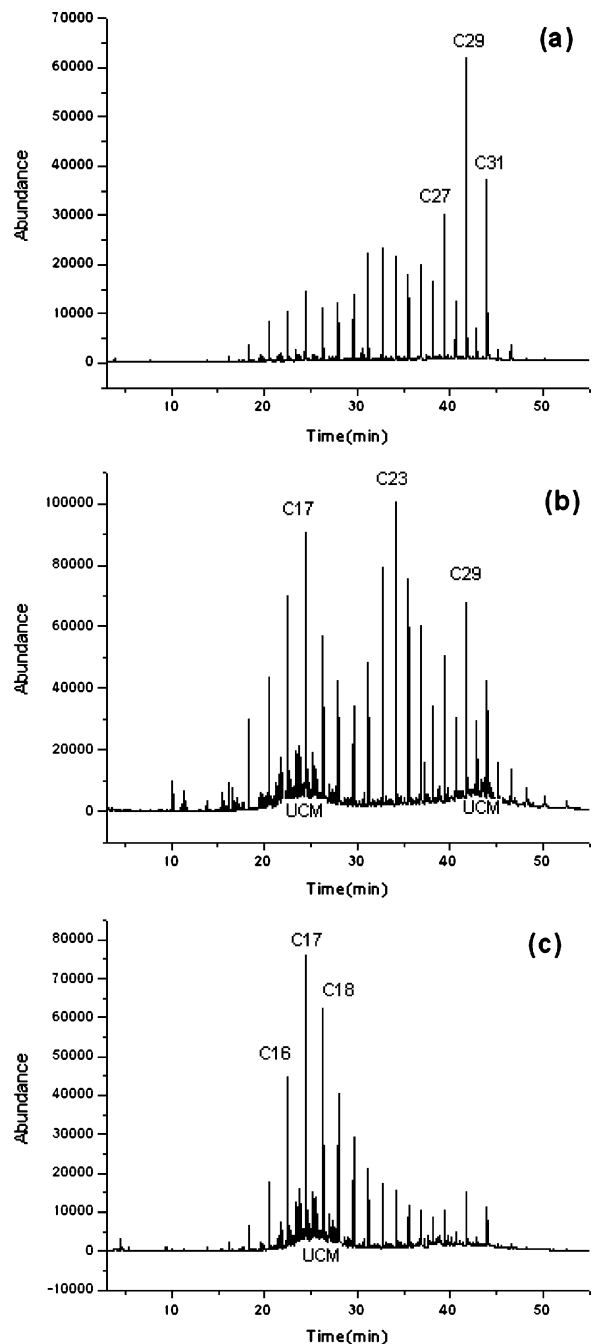

Fig. 2 **a** The aliphatic hydrocarbon profile of a typical sediment sample near paper mill analysed by GC-MSD. The *m/z* 85 ion chromatogram was extracted. The dominance of long chain *n*-alkanes (C27, C29, and C31) suggests vascular plant input. **b** The aliphatic fraction of Station THX was analysed by GC-MSD. The appearance of a mix of major *n*-alkanes indicates mixed sources. **c** The aliphatic hydrocarbon profile of typical sediment near DY petroleum oil field analysed by GC-MSD, The *m/z* 85 ion chromatogram was extracted. The dominance of C17 and C18, UCM and series of long chain *n*-alkanes showing no odd carbon predominace suggest petrogenic input

that the area was polluted by weathered petroleum hydrocarbons, as oppose to areas affected by fresh oil contamination which tend to have relatively higher concentrations of n-alkanes. Thus, the ratio of UCM to n-alkanes can provide useful information on the pollution history of an area due to petroleum contaminations. The UCM and MH indexes in Table 2 indicate that petroleum related contamination in Laizhou Bay sediment is common for all sites studied, but Stations ZZ02, THX, TH10, TH5, Y01 and Y05 are primarily contamination by more extensively weathered petroleum oil, while Stations TH2, Y10 and Y12 are dominated by less weathered oils.

Carbon preference index (CPI) is an additional means of determining the source of n-alkanes in sediment. CPI is defined as the ratio of $2(C27 + C29)/(C26 + 2C28 + C30)$. Petrogenic hydrocarbons show values around 1, while those of vascular plants and uncontaminated sediments range from 3 to 6 (Giger, Reinhard, & Schaffner, 1974). High values from Station ZZ08 confirm its vascular plant origin drained by the paper mill sewage.

The ratio of low to high molecular weight hydrocarbon (LMW/HMW) is the ratio of the sum of n-alkane≤n-C20 over the sum of n-alkane≥n-C21. This ratio is usually close to 1.0 in algae, plankton and crude oil, while sedimentary bacteria, marine animals, higher plants and sediments show lower values (Colombo et al., 1989). Most stations have a LMW/HMW ratio much less than 1, and the value for Station ZZ08 is extremely low, confirming its dominance by the nearby paper plant contamination.

In summary, n-alkane indexes reflect varying degree of petroleum contaminations in the coastal waters near DY petroleum oil field and the remote station of paper mill (Station ZZ02). Station ZZ08 near the paper mill is dominated by vascular plant. Finally, the contamination in the estuary of TH River may be ascribed to different sources such as territorial non-point pollutions, domestic sewages, and storm-water runoffs.

3.2 Triterpanes

The aliphatic fraction of the sediment identified as having petrogenic contamination from their aliphatic profiles was confirmed by their geochemical biomarkers (the m/z 191 ion chromatogram was extracted). Geochemical biomarkers such as Triter-

panes and steranes are derived from once living organisms. These compounds are more resistant to biodegradation than n-alkanes, isoprenoids and PAHs. They are present in relatively high concentrations in crude oils (Daling & Faksness, 2002; Wang et al., 1999) and can serve as marker species for their identification.

Petroleum oils give a characteristic biomarker profile, which can be used to identify oil contaminations. The doublet peaks in the m/z 191 mass chromatogram (triterpane profile) are due to the C-22S and C-22R diastereoisomers of each of the C31–C35 homohopanes. They are characteristic of all crude oils and the doublets decrease in size with increasing carbon number (Webster et al., 2003). From the sediment profiles of Station THX (Fig. 3a), Y05
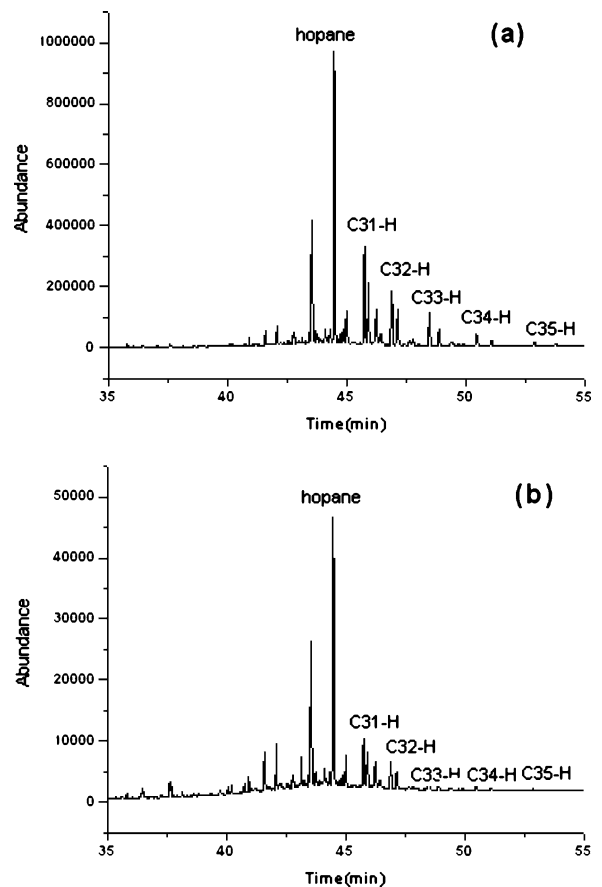

Fig. 3 GC-MS analysis of the aliphatic fraction of sediment samples showing the presence of geo-chemical biomarker. The series of homohopance doublet peaks (C31–C35) in the triterpane profile (m/z191) indicate crude oil contamination. **a** Station THX **b** Station Y05

(Fig. 3b), we can confirm that these stations are contaminated by petroleum.

3.3 PAH profile

The concentrations of PAHs and the related indexes are summarized in Table 3. 18 individual PAHs and selected alkylated species were determined in the sediment samples. The main PAHs detected include pyrene, fluoranthene, phenathene, benzo(a)anthracene and their alkylated homologues. Four- to six-ring parent aromatic hydrocarbons are also found in most of the sediments.

The highest value of total PAHs (based on 11 typical parent PAHs and seven alkylated PAHs) was found in the sediments of the TH River estuary (Station THX; 296.93 ng/g dry wt), followed by Stations ZZ02 and Y05 both with muddy clay material. And the lowest values were in sandy sediment collected in Station Y12. Sediment retention capacity, the ability of sediment solids to reserve organic material, may be related to physico-chemical properties such as the grain size and organic content of the sediment particles, which is a very important variable related to PAH pollution. Higher total PAH concentrations tend to be found in fine muddy sediments with high organic carbon content (Boehm & Farrington, 1984).

Atmospheric deposition, coastal sewage dumping, continental runoff, river outflows and accidental oil spills are the expected contributors of PAH to the marine environment. In general, PAH can be broadly divided into two mainly sources: pyrolytic and petrogenic. The use of several molecular markers and related indexes derived for PAHs for assessing the relative contributions to the environment of hydrocarbon sources has been common and PAH ratios such as the ratios of P/A, MP/P, Fl/Py, % parent PAH relative to total PAHs, etc. are especially useful in revealing the sources of PAHs as pryolytic or petrogenic. PAH ratios as indicators of their source and composition are exemplified in the study of Fraser River basin by Yunker and Macdonald et al. (Yunker et al., 2002).

PAHs from a petroleum-related sources tend to contain a lot of alkylated and lighter molecular weight PAHs such as naphthalene, acenaphthylene etc. PAHs originated from incomplete combustion of coal, oil or organic materials tend to contain little or no alkylated and light molecular weight PAHs, but a high percentage of PAHs with four to six rings such as chrysene, benzo(a)pyrene etc. (Ou et al., 2004). On this basis, sediments that contained predominantly alkylated PAHs and light molecular weight PAHs were likely to be affected by petroleum-related pollution, while sediments characterized by high molecular weight PAHs were likely to be under the influence of pyrolytic sources (Latimer & Zheng, 2003).

The general PAH distribution pattern reflects a high contribution of pyrolytic sources in several sediments in this study because of the dominance of parent PAHs over their alkylated derivatives. The proportion of parent PAHs was high (>60%) in four

Table 3 Concentrations (ng/g dry wt.) and characteristics of PAHs in marine waters near the major drainage outfalls, namely DY petroleum oil field, ZZ paper mill and TH River along the coastal of Laizhou Bay

Drainage outfalls	Station	unsubPAH	subPAH	Total F2	%unsub PAH	FL/Py	P/MP	FL/FL+P
ZZ	ZZ08	4.84	1.59	6.43	0.75	1.40	0.70	0.58
	ZZ02	20.29	5.67	25.96	0.78	1.85	0.83	0.64
	THX	7.14	289.79	296.93	0.02	0.36	0.02	0.26
TH	TH10	3.23	2.58	5.81	0.56	0.85	0.41	0.46
	TH5	3.66	3.91	7.57	0.48	1.00	0.34	0.5
	TH2	10.31	3.67	13.98	0.74	1.50	0.66	0.6
	Y01	1.94	1.11	3.05	0.63	0.45	1.66	0.31
DY	Y05	12.83	16.79	29.62	0.43	1.10	0.19	0.5
	Y10	2.91	2.64	5.55	0.52	0.72	0.41	0.42
	Y12	1.53	2.04	3.57	0.43	0.72	0.51	0.42

F, fluorene; *P*, Phenathene; *A*, anthracene; *Fl*, fluoranthene; *Py*, pyrene; *BA*, benzo(a)anthracene; *C*, chrysene; *BBF*, benzo(b) fluoranthene; BKF, benzo(k)fluoranthene; *BaP*, benzo(a)pyrene; *BghiP*, benzo(ghi)perylene; *M*, methyl; *unsub*, F+P+A+F1+Py+BA+C+BBF+BKF+Bap+BghiP; *sub*, (C1−F+C2−F+C1−P+C2−P+C3−P+C1−F1+C1−C)

Fig. 4 Plot of the fluoranthene/pyrene ratio (*Fl/Py*) against methyl phenathene/phenathene ratio (*MP/P*) of the sediments
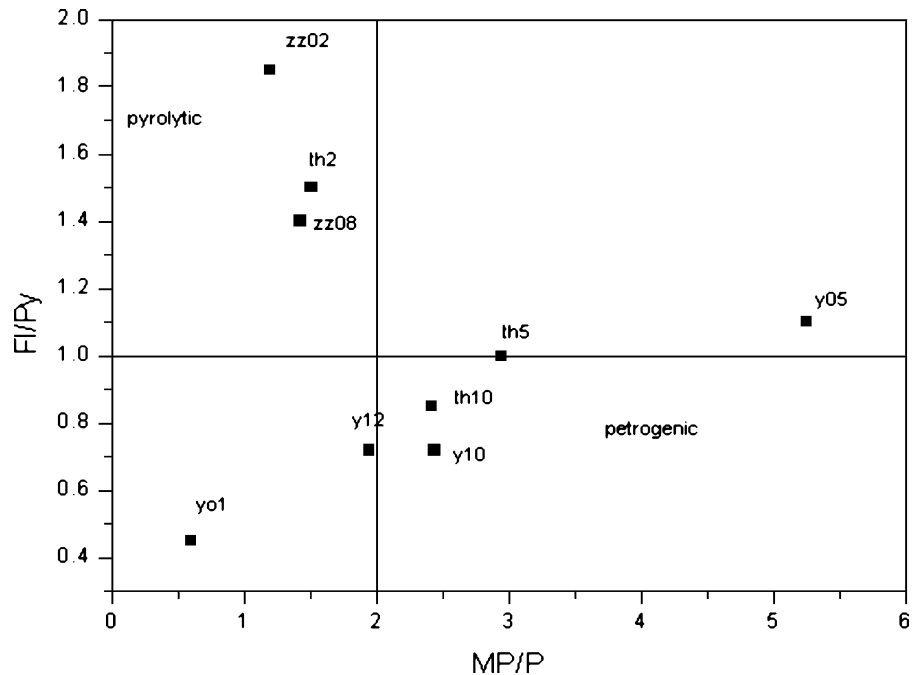
of the sediments including Station ZZ08, ZZ02, TH2 and Y01.The value is especially high in Stations ZZ02 and ZZ08 near paper mill and Station TH2 away from the shore of TH River, and this may be caused by Atmospheric deposition carrying PAH produced by fuel combustion. On the other hand, Station THX sediment contains very low proportion of parent PAHs (only 2%), suggesting strongly its petrogenic input from sewage sources closeby.

The Fl/Fl+Py (fluoranthene to fluoranthene plus pyrene) ratio of 0.5 is usually used as the transition point between petroleum and combustion sources. The ratio is below 0.5 for most petroleum samples and above 0.5 in kerosene, grass, and most coal and wood combustion samples (Budzinski et al., 1997; Yunker et al., 2002). An examination of this ratio in our data strongly suggests that Station THX is dominated by petrogenic sources.

A Fl/Py (fluoranthene/pyrene) ratio of greater than 1 or a MP/P ratio <2 are indicative of a petrogenic source of PAHs (Webster et al., 2003). The Fl/Py ratio was plotted against MP/P ratio as shown in Fig. 4. The distribution pattern shows that the sediments near the paper mill (Station ZZ08 and ZZ02) and the outer station of the TH River such as StationTH2 were situated in the pyrolytic zone. The estuary of TH River including the inner station THx, TH10, TH5 are in the petrogenic zone. Sediment out

of the pyrolytic or petrogenic zone was indicates a mixed source of PAHs. However, using the PAH concentration ratios alone is not sufficient for classifying hydrocarbon sources, and other compositional parameters need to be examined for substantiation and verification.

4 Conclusion

In conclusion, the characteristics of the concentration level, distribution and ratio indexes of hydrocarbons such as *n*-alkanes, PAHs and biomarkers accumulated in marine sediment near the three major drainage outfall in Laizhou Bay can be used concertedly as indicators to source petroleum related contamination.

Our data indicated that the sediment near DY petroleum oil filed and some out-shore sites of ZZ paper mill have a certain degree of petroleum oil pollution. Coastal station near the paper mill (Station ZZ08) is dominated by vascular plant and has little petroleum oil contamination. The hydrocarbon contamination of the marine sediment near TH River may be ascribed to mixed sources.

PAH ratios show that the PAH source is pyrolytic near ZZ paper mill (Station ZZ02) and outer shore stations of TH River (Station TH2); But petrogenic in

the coastal station (Station THX, TH10, TH05) of TH River. The marine sediment in most DY petroleum oil filed may have a mixed source of PAHs both pyrolytic and petrogenic origins.

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