

Spatial and Seasonal Variations of Total Petroleum Hydrocarbon in Surface Water and Sediment in Pearl River Delta

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Abstract A field study in the Pearl River Delta of China was conducted in order to describe to the spatial and seasonal variation of occurrence and concentrations of total petroleum hydrocarbon (TPH) in surface water and sediments. Petroleum hydrocarbons and isoprenoid alkanes were quantified by UV spectroscopy and gas chromatography with a mass selective detector. The concentrations of TPH ranged from 4.3 to 68.7 $\mu\text{g L}^{-1}$ in surface water, and from 66.6 to 1445 $\mu\text{g g}^{-1}$ in surface sediments. The ratios of pristine to phytane suggested that the main sources of TPH in the sediment were petroleum importation. The highest concentrations of TPH were present in the spring season. When compared with results from previous studies, it can be concluded that the Pearl River Delta was moderately polluted by TPH. No statistically significant correlations were observed between the concentrations of TPH in surface water and sediments.

Keywords Total petroleum hydrocarbons · Spatial and seasonal effects · Water and sediments · Pearl River Delta

Petroleum hydrocarbons are ubiquitous pollutants in the environment and they have been found to be present in sediment, groundwater, soil and biota (Carine et al. 2014; Teng et al. 2013; Li et al. 2012). Total petroleum hydrocarbon (TPH) enters into the environment through accidents, spills, and leaks from industrial processes, or as by products from commercial or domestic activities (de Mora et al. 2010; Kanaji et al. 2013). The composition of petroleum hydrocarbon is complex. Some of these compounds are carcinogenic and mutagenic. Once entered into a water environment, part of the lower molecular weight fraction of the petroleum hydrocarbon compounds is removed by evaporation. The other part of it gets dispersed in the water phase and poses risks to ecosystems and human health through trophic transfer (Hentati et al. 2013). In addition, the heavier fractions of TPH can accumulate and persist in the sediment, which can cause long-term adverse effects to the aquatic environment (Oros et al. 2007). Thus, the assessment of the degree of contamination by petroleum hydrocarbons in water and sediment is important.

Many recent studies described the spatial and seasonal variations of TPH in waters or surface sediments (Li et al. 2010; Zhou et al. 2014; Venkatachalapathy et al. 2010; Srinivasa et al. 2005). However, little information is available on TPH contamination in both water and sediments. The studies have demonstrated that TPH concentrations in water or sediment varied with seasons, and indicated that low temperature limits the degradation activities of the microorganisms and phytoplankton for TPH. The spatial distributions of TPH are generally site specific. High values are found in highly industrialized

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areas. Recently, Carine et al. (2014) investigated the sources of TPH in the sediments of Todos os Santos Bay, Brazil, and the results suggest that oil is the significant source of contamination. However, the relationship between the concentrations of TPH in water and in sediment has not been well documented. Also few studies have reported the weathering characteristics of TPH in surface sediments.

The Pearl River Delta (PRD) is one of the fastest developing regions in China in recent decades and it is experiencing accelerating environmental deterioration. The PRD has a large number of tributaries and streams, which form a complicated watershed. It has three major tributaries: West River, North River and East River. Each year, approximately $3.3 \times 10^{11} \text{ m}^3$ of freshwater with suspended solids of 7.1×10^7 tons flow into the Pearl River Estuary (PRE) via eight major outlets. Pollutants from the PRD region are discharged into the river networks and finally transported to the South China Sea via the PRE. It is estimated that the annual discharge of industrial and domestic wastewater amounted to $3.0 \times 10^9 \text{ m}^3$. Studies have reported high levels of PAHs, alkylphenols and other persistent organic pollutants in both abiotic and biotic compartments in the PRD (Luo et al. 2008; Chen et al. 2006, 2011). However, studies on the levels of petroleum hydrocarbons in the PRD are relatively rare.

In the present study, the spatial and seasonal distributions of TPH in both water and surface sediments in the Pearl River Delta were investigated. In addition, the degree of contamination and pollution sources of TPH in the PRD from past and present studies was discussed in order to provide a risk assessment and highlight data gaps related to the control of TPH pollution in the PRD.

Materials and Methods

Seven sampling stations in the Pearl River Delta were investigated in this study (Fig. 1). The stations included two stations at Humen (S3, S4) and one station each at Jiaomen (S1), Hongqimen (S2), Modaomen (S5), Yamen (S6) and Jitimen (S7). Forty two sets of surface water and surface sediment samples were collected at January, April, and July in 2013. Among the sample sets, 14 were collected in January (in winter), 14 were collected in April (in spring) and 14 were collected in July (in summer). Each surface water sample (100 cm) was collected by a water sampler and stored at 4°C in sterilized glass jars until further analysis. Each surface sediment sample (0–5 cm) was collected by a gravity corer sampler and stored in polyethylene bags. Sediment samples were transported back to the laboratory and stored at –20°C until further analysis.

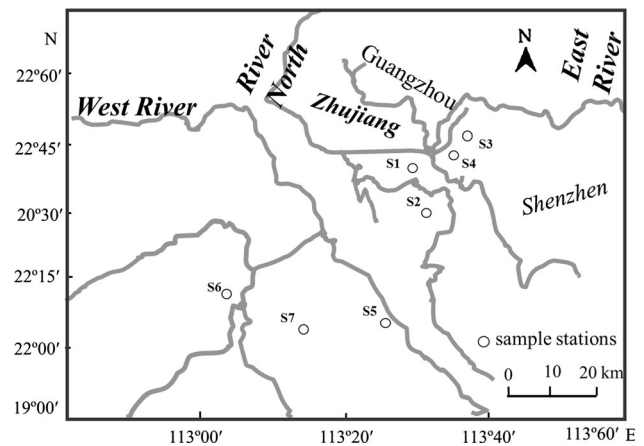


Fig. 1 Study area map

Water samples were filtrated through Millipore 0.45 μm glass fiber filters (Ireland) to remove the particulate fraction and then extracted with *n*-hexane in triplicates, by following China's Marine Monitoring Standards of GB17378.4-2007. The concentrations of TPH in the extracts were quantified by ultra-violet spectroscopy (RF-1501 Spectrofluorophotometer) at a wavelength 225 nm. Sediment samples were air-dried and large debris and fragments of shells were removed before grinding. About 5 g of sediment were extracted with 60 mL of dichloromethane. The extracts were then concentrated to approximately 1.0 mL. The concentrated extracts were cleaned and fractionated on a 10-mm i.d. 1:2 alumina/silica gel glass column packed, from bottom to top, with neutral silica gel (12 cm), neutral alumina (6 cm), and anhydrous sodium sulfate (1 cm). The column was washed with 20 mL of hexane. The extracted solution (1 mL) was added to the top of column and eluted with 20 mL of distilled hexane for petroleum hydrocarbons. Thereafter, the eluted product was evaporated and made to a volume of 1 mL for subsequent gas chromatography with flame ionization detector (GC-FID) analyses.

The *N*-Alkane and isoprenoids were determined with a Trace DSQ. The chromatographic column was a 30 m \times 250 mm i.d.; 0.25 μm film thickness TG-1MS capillary column. The column was programmed from 60 to 90°C at 6°C min^{-1} , and then held at 290°C for 20 min. The flow rate of the carrier gas (helium) was kept constant at 1.0 mL min^{-1} . The MSD was operated in the full scan mode covering the mass range of 35–400. The ion source temperature was 280°C. Method blanks (solvent) and a procedural blank were analyzed in sequence to check for contamination in laboratory. A policy was adopted that if the peak area of the analyte in the laboratory blank is more than 30 % of the peak area in an unknown sample, we censor the data as blank interference. In our present study,

blank values were mostly negligible. Recovery tests were conducted on surface water and sediment samples spiked with target compounds. The average recoveries of TPH were $90.5 \pm 5.3 \%$ and $85.4 \pm 2.2 \%$ in surface water and sediment, respectively, and were $82.8 \pm 4.7 \%$, $82.7 \pm 8.2 \%$, $85.5 \pm 3.8 \%$, $85.0 \pm 9.6 \%$ for n-C17, pristine (Pr), n-C18, phytane (Ph) in sediment. The limits of detection (LOD) were determined to be $6.5 \mu\text{g L}^{-1}$ for TPH, $1.07 \mu\text{g g}^{-1}$ for n-C17, $1.0 \mu\text{g g}^{-1}$ for Pr, $1.20 \mu\text{g g}^{-1}$ for n-C18, and $1.11 \mu\text{g g}^{-1}$ Ph.

Statistical analyses were performed by using IBM SPSS 19.0. Concentrations of target TPHs in each of the two environmental media (surface water and sediment) were analyzed with one-way ANOVA to determine the significance of differences among three seasons. The principal component analysis (PCA) by correlation matrix was used in this work to reveal the similarities and differences of the sampling sites and assess the relationships between TPH, Pr, Ph in surface water and sediment. A concentration value equal to half of the detection limit was used in the calculations when sample concentrations were below the LOD (Stasinakis et al. 2008).

Results and Discussion

Total petroleum hydrocarbon contaminants investigated were found in all surface water samples in the PRD (Table 1). The concentrations of TPH in water fluctuated between 4.3 and $68.7 \mu\text{g L}^{-1}$. The observed variations may be related to variable, but uncertain, oil spills and disposal of land waste and sewage into the study area. The TPH values in almost all of the water samples were under the standard value for First Class of Seawater Quality Standard of China's Marine Monitoring Standard of GB 3097-1997 ($50 \mu\text{g L}^{-1}$). The highest value of $68.7 \mu\text{g L}^{-1}$ was found in station S1 (Jiaomen). Only one sample's value exceeded the standard. The mean TPH concentration in surface water in station S4 ($18.1 \pm 4.4 \mu\text{g L}^{-1}$) was the lowest, whereas

the highest mean value of $38.0 \pm 18.4 \mu\text{g L}^{-1}$ was found in station S1. This might be due to past usage of ships in the station S1 area, as well as continuous receiving of sewage. High values of TPH, ranging from 34.7 to $35.9 \mu\text{g L}^{-1}$, were also found in stations S2 and S3, which are located in the downstream of city of Dongguan. The overall mean concentration of TPH in seven stations was $28 \pm 3.1 \mu\text{g L}^{-1}$ in this study (Table 1). This level is under the standard value of China's Marine Monitoring Standards of GB 3097-1997. However, as some local values obviously exceeded the standard value, there is likely to be potential risks of exposure to the TPH in the water environment.

Total petroleum hydrocarbons were also detected in all of the surface sediment samples (Table 1). The concentrations detected ranged from 66.6 to $1445 \mu\text{g g}^{-1}$ (dry weight), and the mean value was $663 \pm 103 \mu\text{g g}^{-1}$. The distribution of the TPH in surface sediments was not uniform, but varied with different sampling stations. The highest mean value of $997 \pm 217 \mu\text{g g}^{-1}$ was found in station S3 (Humen), which is higher than the standard value of $500 \mu\text{g g}^{-1}$ of First Class of China's Marine Sediment Quality Standard (China's Marine Monitoring Standards of GB18668-2002). The lowest mean value of $288 \pm 49.5 \mu\text{g g}^{-1}$ was detected at station S1. Lower values of TPH, ranging from 438 to $460 \mu\text{g g}^{-1}$, were found in stations of S2 and S7. On the other hand, higher values, ranging from 807 to $947 \mu\text{g g}^{-1}$, were found in stations of S4 and S5, which are the main channel to Jiangmen, Foshan and Zhaoqing and are close to power plants in the area.

Isoprenoid concentrations were detected in the sediment. Pristine (Pr) and phytane (Ph) was detected in every station of the surface sediments. The concentrations of Ph in the stations ranged from 0.086 to $2.233 \mu\text{g g}^{-1}$. As phytane concentrations in sediments unpolluted by oil are generally less than $0.001 \mu\text{g g}^{-1}$ (Seagars and Garlich 2001), the sediments in the study area were mostly likely to be polluted by oil. The ratios Pr/Ph were calculated for the

Table 1 Concentrations of TPH in water and sediments in three seasons

Station no.	Latitude/longitude	Water ($\mu\text{g/L}$)			Sediments ($\mu\text{g/g dw}$)		
		Range	Mean \pm SE	n	Range	Mean \pm SE	n
S1	22°36.822'N/113°39.258'E	4.9–68.7	38.0 ± 18.4	7	197–367	288 ± 49.5	8
S2	22°35.019'N/113°37.159'E	7.0–49.2	34.7 ± 13.8	7	66.6–1030	438 ± 301	7
S3	22°49.118'N/113°35.361'E	26.4–47.7	35.9 ± 6.28	6	666–1410	997 ± 217	7
S4	22°48.871'N/113°34.754'E	10.0–25.2	18.1 ± 4.42	8	508–1445	947 ± 272	7
S5	22°11.003'N/113°24.341'E	4.3–32.9	22.6 ± 9.13	6	265–1250	808 ± 280	9
S6	22°11.796'N/113°05.448'E	7.4–42.8	20.6 ± 11.2	9	252–1096	700 ± 244	6
S7	22°11.163'N/113°18.860'E	13.3–46.3	26.0 ± 10.3	7	130–744	461 ± 179	7

n = number of samples with concentrations higher than the LOD of the method

stations and they were almost all less than 1 (Table 2), which also suggests crude oil input (Commendatore and Esteves 2004). The ratios of nC17/Pr and nC18/Ph were also calculated to estimate the degree of weathering of crude oil. The ratios are in the ranges from 2.0 to 6.2 and from 2.0 to 4.5 in crude oil, respectively. The ratios of nC17/Pr and nC18/Ph in the study area ranged from 0 to 7.06 and from 0.235 to 1.03, respectively (Table 2). The arithmetic means of ratios were 1.74 and 0.63, respectively. Therefore, it can be concluded that petroleum hydrocarbons in sediments in the PRD were moderately weathered. At stations S5 and S6 in spring and S2 and S3 in summer, the ratios of nC17/Pr and nC18/Ph were less than 1 (Table 2), indicating that these local areas were the most weathered.

According to one-way ANOVA analysis conducted on the concentration values, in each of the environmental media (surface water and sediment), significant differences ($p < 0.05$) of TPH concentrations were found between the three seasons at all of the sampling locations. The average concentrations of TPH in the 7 stations varied with season and showed a decreasing order of spring (mean = $42.0 \pm 6.83 \mu\text{g L}^{-1}$) > winter (mean = $22.6 \pm 4.98 \mu\text{g L}^{-1}$) > summer (mean = $19.3 \pm 5.99 \mu\text{g L}^{-1}$) in the surface water, and spring (mean = $919 \pm 188 \mu\text{g g}^{-1}$) > summer (mean = $611 \pm 98.2 \mu\text{g g}^{-1}$) > winter (mean = $328 \pm 153 \mu\text{g g}^{-1}$) in surface sediments. As shown in Fig. 2a, b, the highest concentration of TPH in surface water ($68.7 \mu\text{g L}^{-1}$ S1) and sediments ($1445 \mu\text{g g}^{-1}$ S4) were both found during the spring season. And relatively high concentrations, 47.7, 46.3 and $49.2 \mu\text{g L}^{-1}$ in surface water, 1406 and $1248 \mu\text{g g}^{-1}$ in surface sediments, were found during the spring season. It is possible that the weather factors influenced the activities of the microorganisms and phytoplankton to degrade TPH in surface water and sediment, and the relatively low temperature and high humidity were the main reasons (Faksness and Brandvik 2008). In water, lower TPH concentrations, 4.9 and $4.3 \mu\text{g L}^{-1}$ were both found during the summer season. The reason was that they were being diluted significantly by the high water flow rates. In the summer season,

the rate of evaporation of petroleum hydrocarbons was also higher at high temperatures and low atmospheric pressures. In sediments, lower TPH concentrations, 66.6 and $130.6 \mu\text{g g}^{-1}$ were both present in the winter. It was possibly due to the lack of precipitation and reduced water usage during the dry season (winter) (Li et al. 2013). It is worth noting that, at station S6, the trend was different to others in winter, in which the highest concentrations of TPH were detected in winter. It might be that the temperature impact is greater than the humidity to the degradation bacteria to degrade TPH in these areas, or this station has high strength of shipping activities even in the winter season.

In order to understand the relationship between the concentrations of TPH in water and in surface sediment, correlation analyses were conducted. No statistically significant correlations ($r < 0.5$) between the concentrations in water and sediment were observed. This indicated that the TPH values did not reach equilibrium in Pearl River Delta. The concentrations of TPH in the two matrices varied with the various conditions. It is possible that the petroleum hydrocarbons in surface sediment is released to the water phase again, and cause the secondary pollution to the water environment.

In addition, PCA was conducted on the data set. The score plot (Fig. 3a) revealed the distinct groups of the samples. The site groups of S3, S4, and S6 seemed to be discriminated from S1, S2, S5, S7 sites. The significant discrimination of sites (S3, S4) was related to the high loadings of TPH, Pr, Ph, nC17, and nC18 in sediments (Fig. 3b). This result indicated a close relationship between the contamination levels of TPHs. Furthermore, two main clusters were formed in the loading plot as shown in Fig. 3b. One was formed by the concentrations of TPHs in surface water, and the other was formed by sediments. The high correlation between the TPH, Ph, Pr, nC17 and nC18 concentrations in sediment may indicate a common origin for such compounds.

The concentrations of TPH detected in this study were generally comparable to literature data. For example, TPH concentrations ranging from 23.7 to $508 \mu\text{g L}^{-1}$ in surface

Table 2 Parameter distributions of normal alkanes in the sediments from PRD, China

Station no.	Winter			Spring			Summer		
	nC17/Pr	nC18/Ph	Pr/Ph	nC17/Pr	nC18/Ph	Pr/Ph	nC17/Pr	nC18/Ph	Pr/Ph
S1	1.32	0.64	0.81	2.35	1.03	0.40	1.23	0.63	0.69
S2	1.40	0.58	0.71	1.85	0.77	0.35	0	0.65	0.34
S3	1.44	0.68	1.02	2.00	0.81	0.30	0	0.24	0.88
S4	1.43	0.66	0.66	7.06	0.68	0.10	1.12	0.51	0.64
S5	1.54	0.61	0.61	0	0.50	0.53	1.18	0.63	0.65
S6	1.09	0.52	0.86	0	0.51	0.76	1.17	0.60	0.68
S7	1.08	0.51	0.84	1.17	0.59	0.67	1.23	0.74	0.50

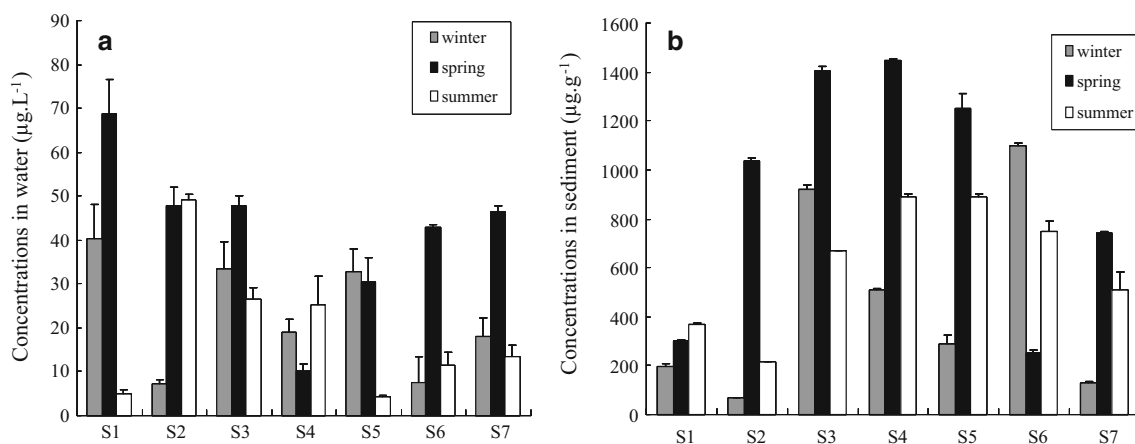


Fig. 2 Concentrations of TPH in water (a) and sediments (b) in 7 stations in three seasons

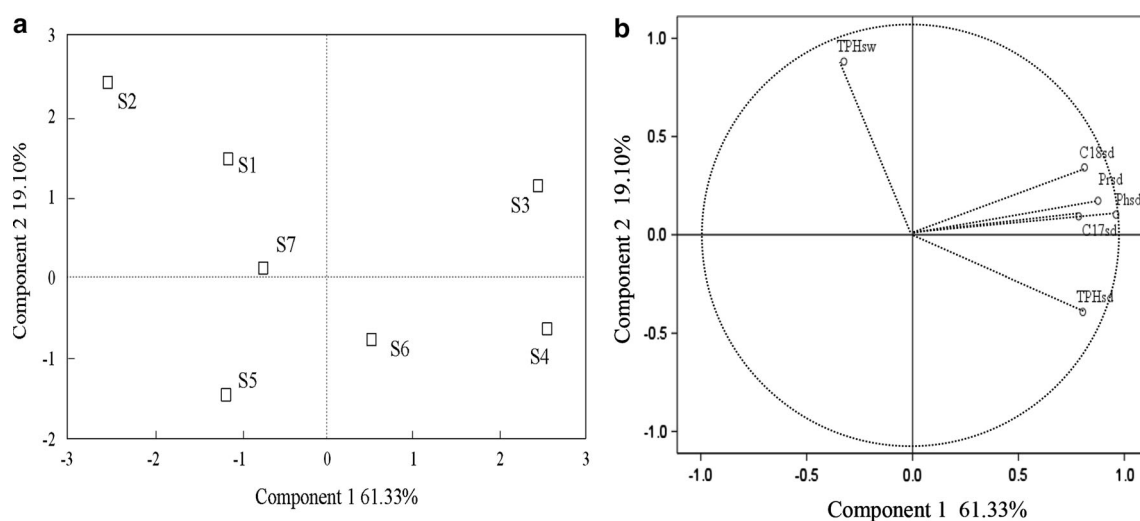


Fig. 3 Score (a) and loading (b) plots of PCA

water and 6.3 to 535 $\mu\text{g g}^{-1}$ in surface sediments were reported from Bohai Bay, China (Li et al. 2010; Zhou et al. 2014). Monitoring data in waters along Tamilnadu coast varied in a wide range of 2.28–14.02 $\mu\text{g L}^{-1}$ (Veerasingam et al. 2011). Concentrations of TPH in surface sediments ranged from 72 to 3000 $\mu\text{g g}^{-1}$ and 0.22 to 40,000 $\mu\text{g g}^{-1}$ (Ye et al. 2007; Carnie et al. 2014), which were much higher than those observed in this study. However, in the Chennai Coast, Bay of Bengal of India, the concentrations ranging from 1.8 to 39.8 $\mu\text{g g}^{-1}$ (Venkatachalapathy et al. 2010) were much lower than those in this study. In unpolluted environment and estuarine sediments, TPH concentrations are almost less than 10 $\mu\text{g g}^{-1}$ (Bouloubassi and Saliot 1993). This suggests that all the studied areas were subjected to different degrees of pollution. When compared with these previous studies, the petroleum hydrocarbon concentrations in this study were in the median ranges.

Conclusions

In this study, the spatial and seasonal variations of TPH in water and surface sediment in 7 sampling stations in Pearl River Delta were investigated. TPH was detected in all water and sediment samples, and the concentration levels of TPH were moderate as compared to those reported in other studies. The ratios of Pr/Ph indicated that most of pollution sources for sediments came from petroleum and its products. The ratios of nC17/Pr and nC18/Ph suggested that oil in the study area was weathering. Seasonal variations of the total petroleum hydrocarbon concentrations were different in water and sediment. The level's decreasing orders were spring > winter > summer in water, and spring > summer > winter in surface sediment, respectively. No statistically significant correlations were observed between the concentrations of TPH in water and in surface sediment.

While the overall mean values of TPH in both water and sediments were lower than the standard values of China, some local values clearly exceeded. Therefore, there is likely to be potential risks of exposure to the TPH in the aquatic environment in the PRD region.

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