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Biological risk and pollution history of polycyclic aromatic hydrocarbons (PAHs) in Nansha mangrove, South China

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ABSTRACT

Chinese government has taken various measures to alleviate pollution caused by polycyclic aromatic hydrocarbons (PAHs) in the region of Pearl River Delta since the economic reform in 1978, but the effectiveness of these measures remains largely unknown. This study aimed to elucidate the biological risk and pollution history of PAHs by measuring the concentrations of 28 PAHs in the surface and core sediments, respectively, in Nansha mangrove. Results found that the biological risk of PAHs was low without obvious spatial variation. The PAH concentration along the depth gradient indicated that PAH pollution was stabilized since the early 1990s while the source of PAHs has gradually changed from combustion of coal to petroleum products. This implied that the mitigation measures taken by the Chinese government were effective. Compared to marine bottom sediment, we propose that using mangrove sediment can provide a more accurate and precise estimate of pollution history of PAHs.

1. Introduction

The population and economy in China have grown expeditiously over the last few decades. The Pearl River Delta (PRD), in particular, has become the most populated and economically dynamic region in China since the late 1970s (Wang et al., 2011). Meanwhile, the rapid economic development has made China become the biggest coal and petroleum consumer worldwide (NBSC, 2005), generating a substantial amount of pollutants to the environment. Among various pollutants, polycyclic aromatic hydrocarbons (PAHs) are of special concerns due to their toxicity, mutagenicity and carcinogenicity to living organisms, as well as their persistence in the environment (Tam et al., 2001; Zheng et al., 2002; Xu et al., 2006; Sehili and Lammel, 2007; Zhang and Tao, 2009). In light of the elevated number of factories, vehicles and power plants in the PRD since the late 1970s (Zhang et al., 2013a), PAH pollution has deteriorated gradually via combustion of fossil fuel and discharge of petroleum products. To eradicate PAH pollution, efforts were made by the Chinese government, such as promoting energy-saving coal and replacing coal with petroleum or natural gas, from the early 1990s onwards (Wang, 2002; NBSC, 2005; Xu et al., 2006). The effectiveness of these measures by monitoring PAH pollution over time has, however, not been extensively studied.

Sediment is often the final repository of environmental pollutants, including PAHs, due to its strong adsorption capacity. As such, the pollution history of PAHs can be retrieved by analyzing the PAH concentrations in sediment along the depth gradient as long as the sedimentation rate is known (Hites et al., 1977). This monitoring technique for PAH pollution has been applied in numerous studies but mostly using marine bottom sediment (Lima et al., 2003; Liu et al., 2005; Azoury et al., 2013; Zhang et al., 2013b). Nevertheless, using mangrove sediment to reflect pollution history of PAHs may be more promising since the high organic matter and anoxic condition in mangrove sediment can facilitate the adsorption of PAHs (Tam et al., 2001; Ke et al., 2005). On the other hand, mangrove sediment can potentially lead to over-accumulation of PAHs owing to its strong adsorption capacity (Nagelkerken et al., 2008), the biological risk of PAHs should not be overlooked.

The present study aimed to investigate (1) the biological risk of PAHs in Nansha mangrove, the largest one in the PRD, by measuring their concentrations in the surface sediment, and (2)

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the pollution history of PAHs and their potential sources since the late 1970s by measuring their concentrations along the depth gradient. The findings could provide insight into (1) how socio-economic development affects PAH pollution, (2) the effectiveness of the mitigation measures taken by the Chinese government to control PAH pollution, and (3) the accuracy and precision of using mangrove sediment to reflect the pollution history of PAHs. The authorities concerned can, therefore, implement effective policies to control the potential sources of PAHs.

2. Materials and methods

2.1. Study site and sampling design

Nansha mangrove (113°33′00″E, 22°39′14″N) with an estimated area of 55 hm^2 in Sanmin Island, South China, was chosen as the study site (Fig. 1). Based on our field observation in December 2011, this mangrove was highly dominated by the introduced, mature, true mangrove plant *Sonneratia apetala*. The individuals, which appeared healthy with greenish leaves, were uniformly distributed with density of approximately 230 ind hm^{-2}. Their average height and average diameter at breast height (DBH) were 8 m and 11.8 cm, respectively. Apart from mangrove plants, sedge *Cyperus malaccensis* was ubiquitously distributed in the open mud-flat area while water hyacinth *Eichhornia crassipes* was a rare plant species in the study site.

Field collection was launched in December 2011. The study site was uniformly divided into 20 sampling points along the Island with a distance of about 100 m between two consecutive sampling points (Fig. 1). At each sampling point, three random replicates of surface sediment samples were collected by a rectangular sampler (10 cm long × 10 cm wide × 15 cm deep). To evaluate the pollution history of PAHs, three core sediment samples were randomly collected by a PVC core (10 cm in diameter × 66 cm deep) at sampling points N5, N10 and N15 (Fig. 1). According to the previous results of ^{210}Pb dating, the sedimentation rate in the study area is approximately 2 cm yr^{-1} (Chen and Luo, 1991; Zhang et al., 2002; Liu et al., 2010). Hence, the sediment samples in each core were cut into 33 layers from the top at 2 cm depth interval by a PVC knife to represent the pollution history from 1979 to 2011. All the samples were put into an ice box, transported to laboratory and finally stored at −20 °C before analyses.

2.2. Chemical analyses of sediment

The sediment samples were freeze-dried, ground into powder and passed through a 2 mm sieve. Total organic carbon (TOC) was determined by a CHN elemental analyzer (Perkin-Elmer 2400, Perkin Elmer Corp., USA). The concentrations of 28 PAHs, including 16 USEPA priority PAHs, were examined in the present study (Table 1). The method described by Mai et al. (2002) was applied for PAH extraction. Before extraction, a mixture of deuterated PAH compounds (naphthalene-d_8, acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12} and perylene-d_{12}) was added to all the samples as recovery surrogate standard whereas hexamethylbenzene was added to the extract as internal standard.

The concentrations of PAHs were analyzed using Agilent 7890A/5975C Gas Chromatography–Mass Spectrometry (GC–MS), equipped with Agilent DB-5MS column (30 m × 0.25 mm × 0.25 μm). Regarding the temperature programming, the column temperature firstly increased from room temperature to 80 °C, followed by a 1 min hold. Then, the temperature further increased to 235 °C at a rate of 10 °C min^{-1}, prior to the last stage where the temperature continued to climb to 300 °C at a rate of 4 °C min^{-1}. Pure helium was used as the carrier gas at a flow rate of 1.5 ml min^{-1}. 1 μl sample was injected into the injector with an initial temperature of 290 °C using splitless injection mode while the interface temperature was maintained at 230 °C. The mass

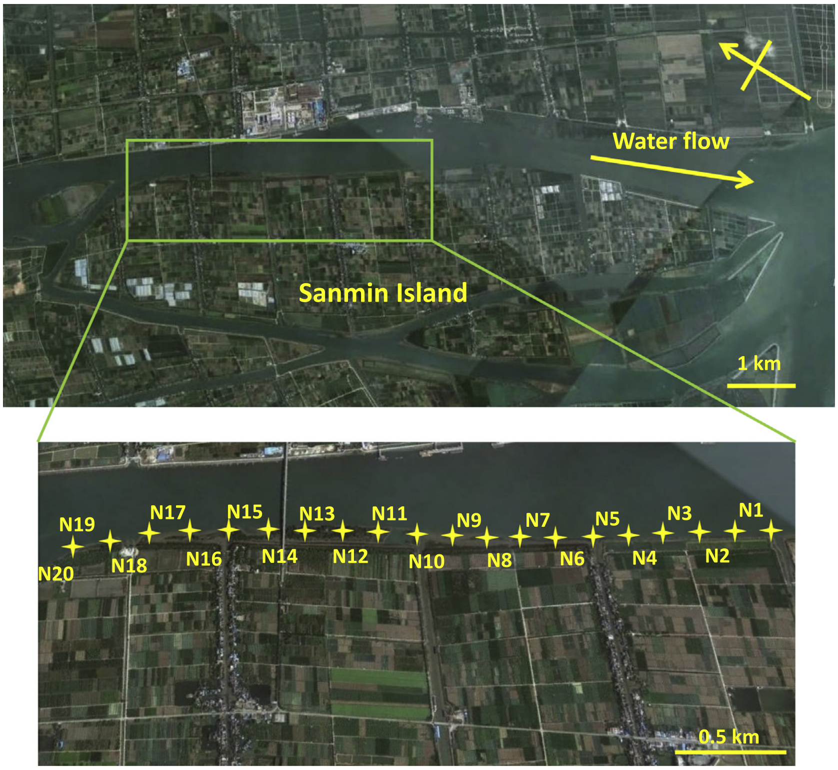

Fig. 1. The sampling points, shown by ✦, in Nansha mangrove in the present study (retrieved from Google Earth). At each sampling point, three random replicates of surface sediment samples were collected. Three random replicates of core sediment samples were collected at sampling points N5, N10 and N15.

Table 1
The 28 PAHs analyzed in the present study. The USEPA priority PAHs are indicated by an asterisk (*).

Number of rings	PAH	Molecular weight
2 rings	Naphthalene*	128.17
	l-Methylnaphthalene	142.20
	2-Methylnaphthalene	142.20
	Biphenyl	154.21
	2,6-Dimethylnaphthalene	156.22
	2,3,5-Trimethylnaphthalene	170.25
3 rings	Acenaphthylene*	152.20
	Acenaphthene*	154.21
	Fluorene*	166.22
	Anthracene*	178.23
	Phenanthrene*	178.23
	1-Methylphenanthrene	192.26
	2-Methylphenanthrene	192.26
	2,6-Dimethylphenanthrene	206.11
4 rings	Pyrene*	202.25
	Fluoranthene*	202.25
	1H-Benzo[b]fluorene	216.28
	Chrysene*	228.29
	Benzo(a)anthracene*	228.29
5 rings	Perylene	252.31
	Benzo[a]pyrene*	252.31
	Benzo[e]pyrene	252.31
	Benzo[b]fluoranthene*	252.31
	Benzo[k]fluoranthene*	252.31
	Dibenzo[a,h]anthracene*	278.35
	9,10-Diphenylanthracene	330.42
6 rings	Benzo[g,h,i]perylene*	276.33
	Indeno[l,2,3-c,d]pyrene*	276.33

spectra were obtained using electron ionization mode at 70 eV and 230 °C. The SIM scanning ranged between 50 and 550 amu at 2.4 scan s^{-1}. A standard reference material (National Institute of Standards and Technology, SRM 1941) was used to measure the accuracy of the method. Surrogate recoveries were 50.7 ± 9.1% for naphthalene-d_8, 66.1 ± 8.1% for acenaphthene-d_{10}, 85.6 ± 8.9% for phenanthrene-d_{10}, 87.3 ± 10.6% for chrysene-d_{12} and 89.4 ± 15.2% for perylene-d_{12}.

2.3. Statistical analyses

The ratio of fluoranthene/fluoranthene and pyrene (Fl/(Fl + Py)) was calculated to determine the source of PAHs. It can be categorized into three groups (Yunker et al., 2002): Fl/(Fl + Py) < 0.4: oil pollution; 0.4 < Fl/(Fl + Py) < 0.5: combustion of oil and refined products; Fl/(Fl + Py) > 0.5: combustion of wood and coal. Linear regression analysis was used to estimate whether TOC, total concentration of PAHs and Fl/(Fl + Py) were significantly correlated with sediment depth. Pearson correlation analysis was used to determine the correlation between TOC and total concentrations of PAHs in both surface and core sediments. Statistical analyses were performed using SigmaPlot 12.0 for Windows.

3. Results

3.1. Spatial variation of TOC and PAHs in the surface sediment

Excluding N3, TOC in the surface sediment fluctuated between 2.5% and 4.2% from N1 to N13, but it was remarkably reduced from N14 to N20, ranging from 2.2% to 0.7% (Fig. 2). The spatial variation of PAHs is illustrated in Fig. 3. The total concentration of 28 PAHs, ranging approximately from 330 ng g^{-1} to 1270 ng g^{-1}, highly fluctuated from N1 to N20 with a slightly decreasing trend (Fig. 3a). Similar pattern was observed for the total concentration of 16 USEPA priority PAHs, ranging from 150 ng g^{-1} to 610 ng g^{-1} (Fig. 3b). TOC was positively correlated with the total concentration of PAHs in the surface sediment (28 PAHs: Pearson correlation coefficient = 0.39, $p < 0.01$, $n = 60$; 16 PAHs: Pearson correlation coefficient = 0.34, $p < 0.01$, $n = 60$). As for the potential source, it is manifest that Fl/(Fl + Py), with a value about 0.5, did not vary

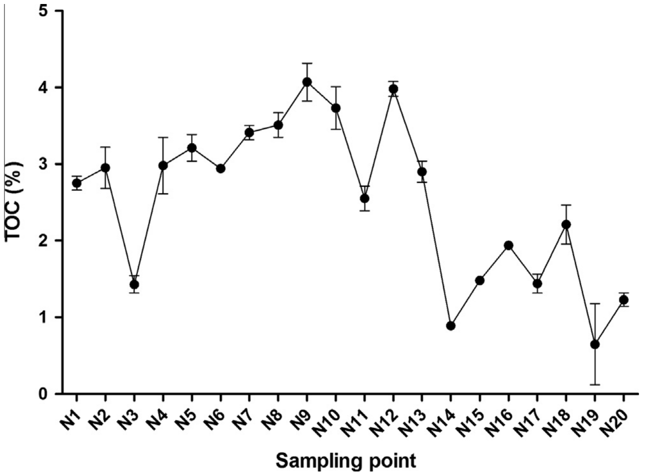

Fig. 2. The TOC (%) in the surface sediment at different sampling points in Nansha mangrove (mean ± S.D., $n = 3$).

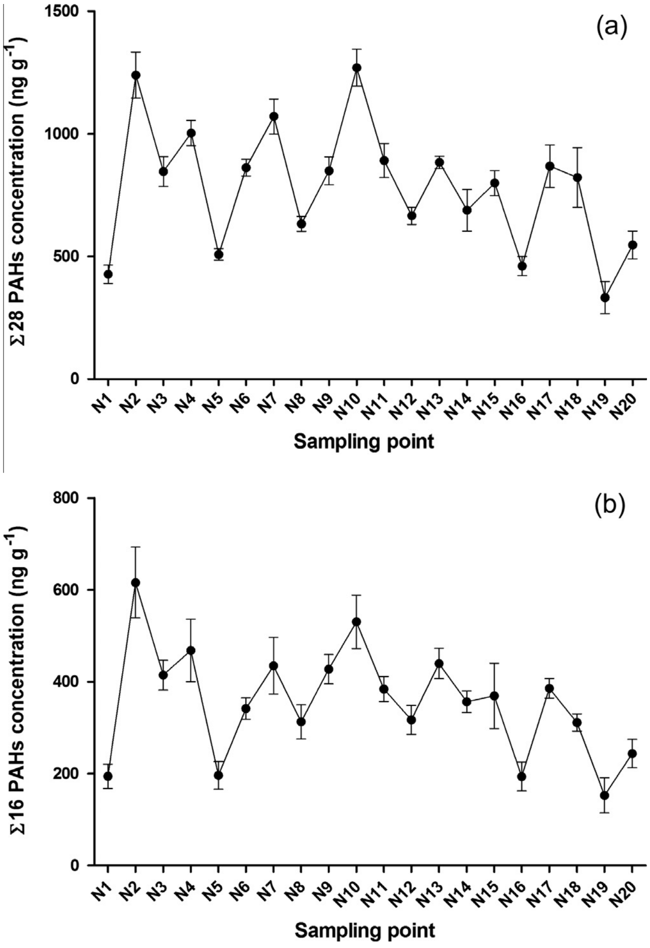

Fig. 3. The total concentration (ng g^{-1}) of (a) 28 PAHs and (b) 16 USEPA priority PAHs in the surface sediment at different sampling points in Nansha mangrove (mean ± S.D., $n = 3$).

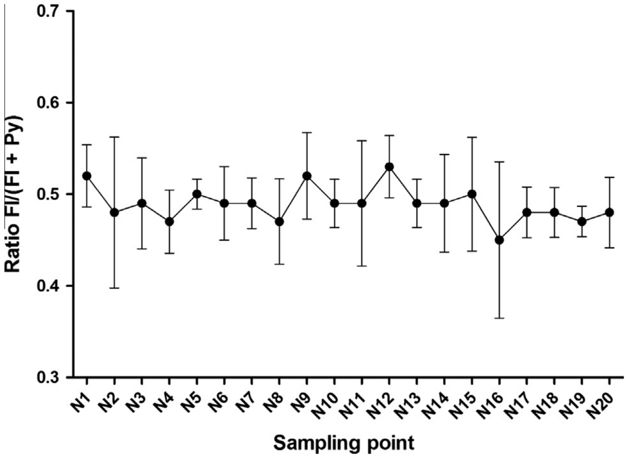

Fig. 4. The ratio of Fl/(Fl + Py) in the surface sediment at different sampling points in Nansha mangrove (mean ± S.D., $n = 3$).

significantly from N1 to N20 (Fig. 4), suggesting that the PAHs in the surface sediment were mostly originated from the combustion of wood, coal and petroleum products. Fig. 5 shows the proportion of PAHs with different numbers of benzene rings. PAHs with 5–6 rings were dominant, contributing more than 50% total PAHs. PAHs with 2–3 rings had a similar proportion as those with 4 rings (about 20–25 %). The high molecular weight (HMW) PAHs (4–6 rings) accounted for about 80% total PAHs in the surface sediment. Overall, the spatial variation of PAHs in the surface sediment was not obvious, in terms of total concentration, source and composition.

3.2. TOC and PAHs along the depth gradient

TOC decreased gradually with depth from 4% at −2 cm to 1% at −66 cm (Fig. 6). The concentration of each PAH along the depth gradient is shown in Appendix. Although no significant overall trends in both the total concentrations of 28 PAHs and 16 USEPA priority PAHs were found (Figs. 7a and b), two patterns were observed: (1) The concentrations highly fluctuated between −2 cm and −38 cm, and (2) The concentrations increased gradually from −66 cm to −40 cm. Specifically, three sudden drops were observed at −28 cm, −16 cm and −8 cm, which are approximately corresponding to the years 1998, 2004 and 2008, respectively. No significant correlation between TOC and total concentration of PAHs in the core sediment was found (28 PAHs: Pearson correlation coefficient = −0.16, $p > 0.05$, $n = 99$; 16 PAHs: Pearson correlation coefficient = 0.012, $p > 0.05$, $n = 99$). Regarding the source, Fl/(Fl + Py) decreased gradually from 0.55 at −66 cm to 0.47 at −2 cm (Fig. 8), suggesting that the source has changed from combustion of wood and coal to combustion of petroleum products. PAHs with 5–6 rings were dominant, accounting for more than 50% total PAHs, except those at −22 cm (Fig. 9). The proportion of PAHs with 4 rings remained quite stable along the depth gradient (about 10%). Overall, HMW PAHs were more dominant than the low molecular weight (LMW) PAHs (2–3 rings) in the core sediment.

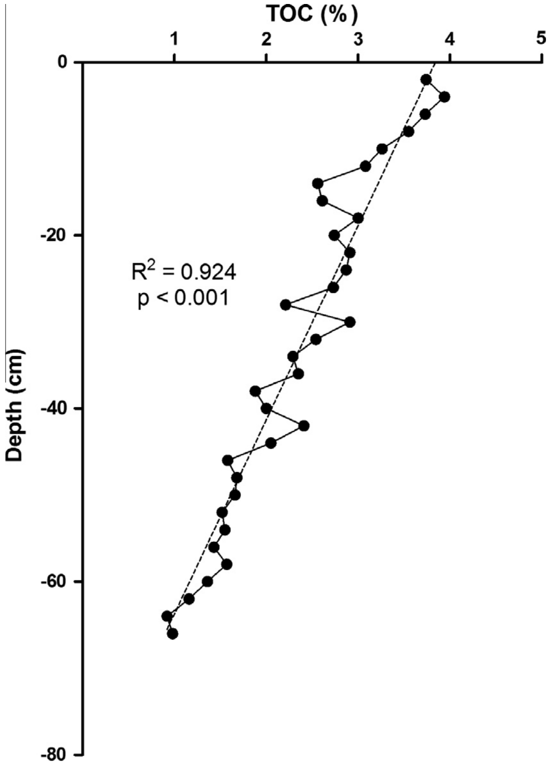

Fig. 6. The TOC (%) in the sediment along depth gradient (cm) (mean, $n = 3$). The dash line represents the best-fit line according to linear regression analysis.

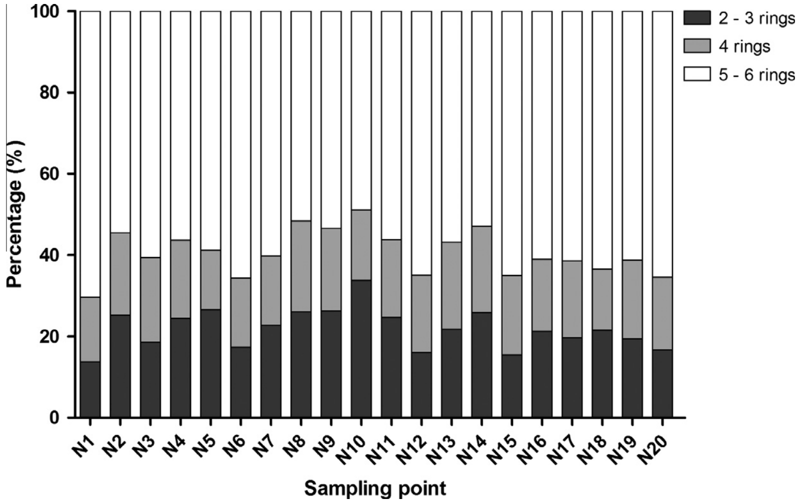

Fig. 5. The proportion (%) of PAHs with 2–3 rings, 4 rings and 5–6 rings in the surface sediment at different sampling points in Nansha mangrove (mean, $n = 3$).

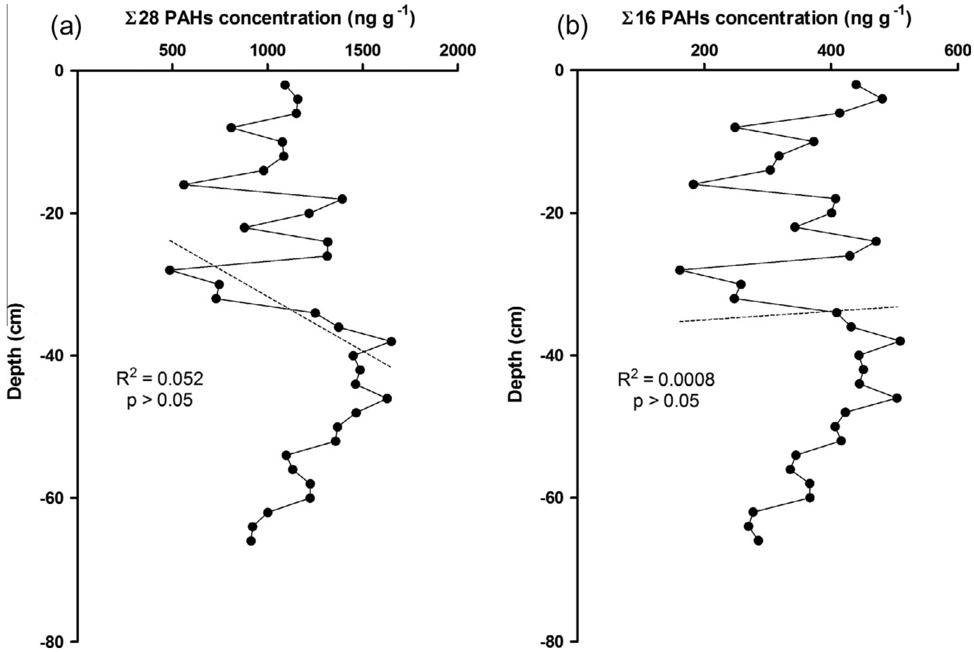

Fig. 7. The total concentration (ng g^{-1}) of (a) 28 PAHs and (b) 16 USEPA priority PAHs in the sediment along depth gradient (cm) (mean, $n = 3$).

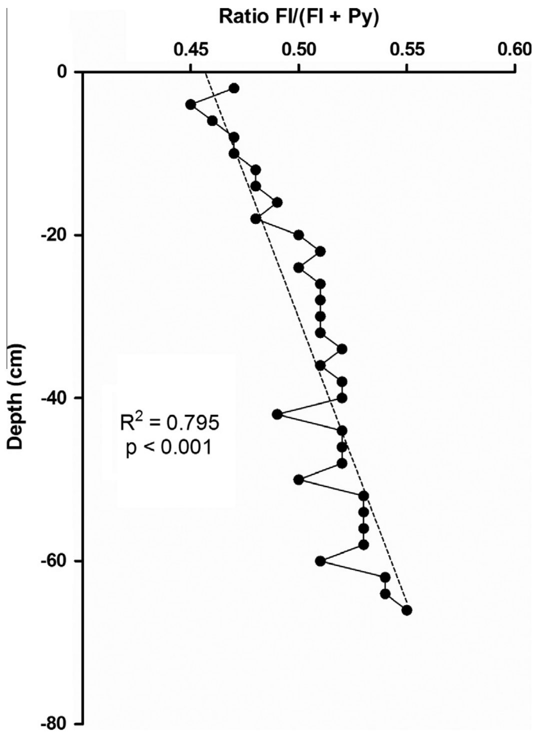

Fig. 8. The ratio of Fl/(Fl + Py) in the sediment along depth gradient (cm) (mean, $n = 3$).

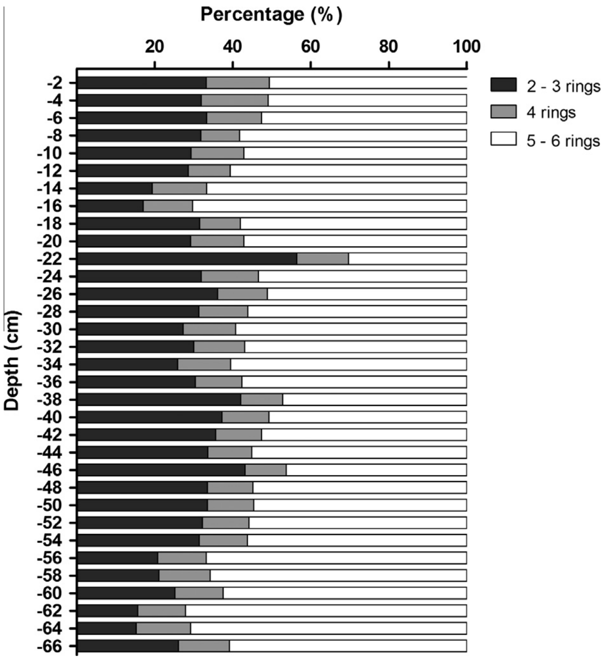

Fig. 9. The proportion (%) of PAHs with 2–3 rings, 4 rings and 5–6 rings in the sediment along depth gradient (cm) (mean, $n = 3$).

4. Discussion

4.1. Spatial variation of PAHs in the sediment

The spatial variation of PAHs in the surface sediment was generally not remarkable, except that slightly higher concentrations were found between N1 and N13 than between N14 to N20. The wider river between N1 and N13 than between N14 and N20 led to slower water flow. As a result, more PAH-laden sediment or organic matter in the water could deposit in the mangrove sediment (Yang, 2000; Santschi et al., 2001). This also explains why a weak positive correlation was found between TOC and total concentration of PAHs. Apart from high TOC, the silty sediment in Nansha mangrove can also facilitate the sorption of PAHs in view of the high surface area to volume ratio (Wang et al., 2001). Nevertheless, the fluctuation of PAHs with high precision indicated that the sources of PAHs in the mangrove were rather localized.

4.2. Biological risk of PAHs in Nansha mangrove

To evaluate the biological risk of PAHs in the sediment, Long et al. (1995) has put forward reliable guideline values based on the percentage of biotas adversely affected in the exposure. The effects range-low (ERL) refer to the lower 10th percentile of the effects data for each chemical whereas the effects range-median (ERM) is the 50th percentile of the effects data. In other words, adverse effects are rarely observed if the concentration is lower than ERL. The 16 USEPA priority PAHs are of special concern due to their persistence and toxicity, and therefore they have been conventionally used to evaluate the anthropogenic impact on the environment (Pereira et al., 1996; Tolosa et al., 1996). Although Long et al. (1995) only reported 13 PAHs in which 12 of them are USEPA prioritized, the total concentration of 28 PAHs in the present study (max. 1650 ng g^{-1}) was still far lower than the ERL of the total 13 PAHs (4022 ng g^{-1}). Furthermore, the concentration of each PAH was far below the ERL, except fluorene which was slightly higher the ERL for some samples. Overall, the biological risk of PAHs in Nansha mangrove was very low.

4.3. Pollution history of PAHs in Nansha mangrove

The pollution history of PAHs was precisely depicted in the core sediment although no significant correlation between TOC and total concentration of PAHs was found. The insignificant correlation implied that PAHs in the sediment were subject to various biochemical or anthropogenic processes rather than solely sequestration with TOC (Tao et al., 2012). Likewise, TOC in the sediment also depends on several processes. For example, TOC decreased with depth probably because the organic matter in the sediment was broken down by anaerobic bacteria (Kristensen et al., 1995); the incessant input of leaf litter and suspended particles from the tide could enrich TOC in the surface sediment.

Regarding the PAH profile, two patterns were observed which reflects the emission and control of PAHs, as well as the economic status in the PRD from 1979 to 2011. The total concentration of PAHs gradually increased from −66 cm to −38 cm, representing the PAH pollution from 1979 to 1991. During that period, the demand of energy surged for economic development, leading to the concomitant increase in combustion of coal and hence total concentration of PAHs. On the other hand, many factories in Hong Kong moved to China, especially the PRD, during the 1980s to 1990s (Tanner et al., 2000). The elevated number of vehicles and factories in the PRD could consequently increase the total concentration of PAHs (Xu et al., 2006). Since then, the Chinese government has tightened the emission standard of PAHs, and therefore the increase ceased at −38 cm and the total concentration of 28 PAHs fluctuated at around 1000 ng g^{-1} from −38 cm to −2 cm. There were three dramatic drops at −28 cm, −16 cm and −8 cm, which were approximately corresponding to the years 1998, 2004 and 2008, respectively. China was crisis-stricken in these years (e.g. Asian Financial Crisis in 1997; outbreak of severe acute respiratory syndrome in 2003; Global Financial Crisis in 2008) (Wang et al., 2011; Wei, 2013), culminating in reduced productivity and hence emission of PAHs in the PRD. Based on these results, we suggest that using mangrove sediment can provide a precise and accurate pollution history of PAHs, probably ascribed to higher sedimentation rate and closer proximity to the source. Compared to subtidal environments (e.g. Liu et al., 2005, 0.61 cm yr^{-1}), the higher sedimentation rate in mangrove habitats (2 cm yr^{-1}) can greatly minimize the sampling error (i.e. cutting the core sediment into layers) caused by annual variation of sedimentation rate.

4.4. Source and composition of PAHs in Nansha mangrove

PAH composition is frequently used to identify the potential sources (Yunker et al., 2002). The findings herein unequivocally demonstrated that the source of PAHs changed over time, indicated by change of Fl/(Fl + Py) along the depth gradient, implying. The decrease in Fl/(Fl + Py) from 0.55 at −66 cm to 0.47 at −2 cm suggests that the source has gradually changed from combustion of coal to petroleum products from 1979 to 2011. Such change generally matches the energy source policy in Guangdong province in which biomass and coal were substituted by natural gas or petroleum as the major source of energy for heating facilities since 1990s (Xu et al., 2006). It is estimated that such heating facilities, which are expected to cover up to 40% in China by 2010 (National Development and Reform Committee, 2004), could not only reduce approximately 49 million tons of coal per year, but also reduce 1850 tons of annual PAH emission (Xu et al., 2006). This could further explain why the total concentration of PAHs was relatively stable in the early 1990s and even lower in the 2000s.

The present study also revealed that the sediment in Nansha mangrove was dominated by HMW PAHs, regardless of temporal variation. Previous studies generally concluded that HMW PAHs are generated by high temperature combustion, including vehicle emissions and power generation (O'Connor and Lauenstein, 1996). For instance, the aerosols produced by high temperature combustion in vehicles contain high proportion of HMW PAHs (Liu et al., 2005; Okuda et al., 2010). The dominance of HMW PAHs in Nansha mangrove, therefore, corroborated the proposition that combustion of fuel in power plants and vehicles was the major source of PAHs in Guangdong province (Bi et al., 2003; Xu et al., 2006). Furthermore, the low proportion of LMW PAHs could be due to the degradation by anaerobic bacteria as LMW PAHs are more readily degraded than HMW PAHs (Qiu et al., 2009). Although HMW PAHs are less resistant to photo-oxidation (Garrett et al., 1998), they were protected by the shading provided by the mangrove plants. All of the aforesaid reasons explained why HMW PAHs prevailed in Nansha mangrove.

Among all the PAH members, a unique depth profile of perylene concentration was observed (Appendix), where the concentration of perylene at the bottom was higher than that near the surface. Such depth profile has been widely recorded (reviewed in Venkatesan, 1988). Silliman et al. (2001) suggested that the anoxic condition and higher content of recalcitrant organic matter in the deeper sediment promote the dominance of perylene-forming microorganisms which are out-competed by other microorganisms in the surface sediment. The biogenic origin of perylene in the present study was further substantiated by the high ratio of perylene to total penta-PAHs (0.66–0.91) (Baumard et al., 1998). Nevertheless, the concentration of perylene highly fluctuated from −30 cm to −2 cm, dictating the overall pattern of the total PAH concentration (e.g. three sudden drops simultaneously observed at −28 cm, −16 cm and −8 cm). Therefore, anthropogenic origins of perylene, which could be from incineration, oil refining and fossil fuel combustion (Venkatesan, 1988; Silliman et al., 2001), have also played a role in the pollution history of PAHs in Nansha mangrove.

To conclude, the present study succinctly demonstrated that mangrove core sediment can precisely reflect the change of PAHs in the PRD over the last three decades, probably due to high sedimentation rate and closer proximity to the PAH source. In the early 1980s, the increase in total concentration of PAHs was linked to the rapid socio-economic development in the PRD. The mitigation measures for PAHs taken by the Chinese government in the early 1990s were effective and therefore the biological risk of PAHs was low. The PAH depth profile indicated the gradual change of

PAH source from combustion of coal to petroleum products in the PRD.

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Appendix A. Supplementary materials

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.marpolbul.2014.06.014.

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