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ORIGINAL ARTICLE

Composition and source identification of polycyclic aromatic hydrocarbons in mangrove sediments of Peninsular Malaysia: indication of anthropogenic input

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Abstract This is a comprehensive study of the composition, origin and sources of specific polycyclic aromatic hydrocarbons (PAHs) in sediments of mangrove estuary in the western part of Peninsular Malaysia. Mangrove sediments were analyzed for 17 PAHs by gas chromatography–mass spectrometry. Total PAH concentrations in the sediments ranged from 20 to 112 ng/g on a dry-weight basis. High molecular weight PAHs were abundant in the sediments. Parent PAH ratios revealed that pyrogenic input has important contribution to the sedimentary PAHs. Ratios of alkylated PAHs indicate that the sedimentary PAHs were influenced by petrogenic PAHs, which implies that petrogenic input has contribution to the sedimentary PAHs but that it is not a major factor in distribution of PAHs within the estuary. Combustion-derived PAHs show a positive and very strong correlation with total PAHs ($R^2 = 0.926$, $p < 0.05$). Total methylphenanthrenes show very weak correlation with total PAHs ($R^2 = 0.0928$, $p < 0.05$). The PAH concentrations were found to increase with distance from the upstream of the estuary to the coastal area of the Straits of Malacca. For the assessment of sediment contamination using biological thresholds, none of the individual studied PAH compounds exceeded the values of the effect range low–effect range median guideline and the threshold effects level–probable effects level guideline. This study demonstrates that the sediments of the mangrove ecosystem facing the Straits of Malacca and Sumatra are influenced by anthropogenic PAH inputs as a result of human activities such as biomass burning, vehicle emissions and boating activities.

Keywords Mangrove sediments · PAHs composition · PAHs sources · Anthropogenic activities · Peninsular Malaysia

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Introduction

Mangrove forests in Southeast Asia are the best-developed and most biodiverse ecosystems in the world which can be found along coastal areas and river estuaries (Giesen and Wulffraat 1998). In the region where land meets the sea, the mangrove ecosystem plays significant roles particularly in coastal erosion prevention as well as control of floods and sediments (Agrawala et al. 2004). The features of mangrove ecosystems such as richness in organic carbon content, the anoxic condition in the sediments (Bernard et al. 1996), abundance in detritus materials and being protected from strong wind and sea wave make them preferential sites for deposition and accumulation of pollutants. Along with 42 mangrove species, Malaysia ranks as the country with the second largest mangrove areas (11.7 %) in Southeast Asia (Giesen et al. 2007). The mangrove ecosystem in Peninsular Malaysia, particularly in the coastline of the Strait of Malacca, is exposed to the threat of pollutants mainly derived from anthropogenic activities. It is likely that this threat will grow in the future (Alongi 2002). It is recorded elsewhere that anthropogenic activities have polluted mangrove sediments, especially with PAHs derived from both pyrogenic and petrogenic sources (Tam et al. 2001; Ke et al. 2005; Zhi-qiang et al. 2005; Cavalcante et al. 2009).

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Polycyclic aromatic hydrocarbons (PAHs) are widespread organic pollutants. The United States Environmental Protection Agency (USEPA) has listed 16 PAH compounds as priority pollutants due to their carcinogenic and mutagenic characteristics. Anthropogenic inputs of PAHs in the marine and estuarine environments are mostly derived from urban runoff, wastewater and industrial discharge, oil spills and ship traffic, and atmospheric fallout of vehicle exhaust and industrial emissions (Tam et al. 2001; Zakaria et al. 2002; Boonyatumanond et al. 2006). Once PAHs enter the environment, they accumulate and degrade slowly under anoxic conditions (Neff 1979; Bernard et al. 1996), thus persisting in the mangrove sediments for long periods of time.

While most studies of PAHs in the environment focus on marine coastal sediments, studies of distribution of PAHs in mangrove sediments are few. This is true in Malaysia where the majority of PAHs studies were conducted in sediments collected from rivers, estuaries and marine environments (Zakaria et al. 2002; Elias et al. 2007; Sakari et al. 2008; Bakhtiari et al. 2010; Sakari et al. 2010; Bakhtiari et al. 2011; Mirsadeghi et al. 2011; Tahir et al. 2011). The objective of the present study is to determine the composition and major sources of PAHs in mangrove sediments in a riverine estuary located along the coastline of the Straits of Malacca. Several PAH ratios have been applied to identify the specific PAH sources. In order to enhance the understanding of PAHs distribution, this paper also discusses the relationship between sedimentary PAH levels and OC content in the sediments.

Materials and methods

Study area

The mangrove swamp forest in Rembau–Linggi estuary is located at the coastline of the Strait of Malacca in Peninsular Malaysia. It lies at the boundaries of two adjoining states: Negeri Sembilan and Malacca. This area was chosen for its strategic position facing the Strait of Malacca and the Indonesian island of Sumatra. The Strait of Malacca is a narrow channel between Peninsular Malaysia and Sumatra of Indonesia and is recognized as one of the busiest waterways in the world (Abdullah et al. 1999; Thia-Eng et al. 2000). The island of Sumatra experiences biomass burning regularly, especially during the dry season from July to September (Wang et al. 2004). Additionally, Kuala Linggi Port is located in the estuarine mouth where goods exchange is active. This mangrove estuary consists of Rembau River and Linggi River that provide raw water for Port Dickson District and the surrounding area which are part of Linggi River Basin, Negeri Sembilan. Linggi River Basin is surrounded by rubber and palm oil plantations, residential areas and mangrove swamp forests downstream of the basin. Hence, it receives significant inputs of various pollutants (Ibrahim and Mustafa 2010; Nazli and Hashim 2010). According to the River Water Quality Report 2010 issued by the Department of Environment of Malaysia, Linggi River Basin is categorized as a slightly polluted region.

Sample collection

Twenty-one surficial mangrove sediments (0–5 cm) were collected in January 2010 using Eijkelkamp hand auger from nine sampling stations in Rembau–Linggi estuary, Negeri Sembilan, Malaysia (Fig. 1). Stations 1–3 were located in Rembau River, whereas stations 4–9 were located in Linggi River. The samples were taken only from the *Sonneratia* species area with pneumatophores in order to avoid environmental variations. The samples were immediately transferred to stainless steel containers and transported to the laboratory in ice boxes. The samples were then stored at −20 °C until further analysis. All the materials in contact with the samples were previously rinsed with distilled water, methanol, acetone and hexane.

Analytical procedure for PAHs analysis

Plant roots and detritus contained in the sediment samples were discarded to remove non-sedimentary PAHs from the sediment samples. The entire procedure for the extraction, purification and fractionation of PAHs is similar to the procedures described by Zakaria et al. (2002) and Boonyatumanond et al. (2006) with minor modification. Briefly, about 20 g [dry weight (dw)] of each sediment sample were homogenized with anhydrous sodium sulphate (Na_2SO_4) to remove excess water from the samples. The samples were extracted with a soxhlet extractor using 300 mL of dichloromethane (DCM) for 8 h. Seventy microliter of the PAH surrogate internal injection standard mixture (10 μg/g mixture of phenanthrene-d_{10} and chrysene-d_{12}) were added prior to the extraction. The sample extracts were then purified and fractioned into an aromatic fraction through silica gel column chromatography. Briefly, the samples were concentrated and transferred to the top of 5 % H_2O deactivated silica gel column. The sample extracts were then further fractioned using fully activated silica gel in order to obtain the PAHs fraction. Prior to sample injection into the gas chromatography–mass spectrometry (GC–MS), *p*-terphenyl-d_{14} was spiked into each sample as a gas chromatography (GC) internal standard.

Quantification of the PAHs was carried out using Hewlett-Packard Model 5972 quadrupole mass spectrometer

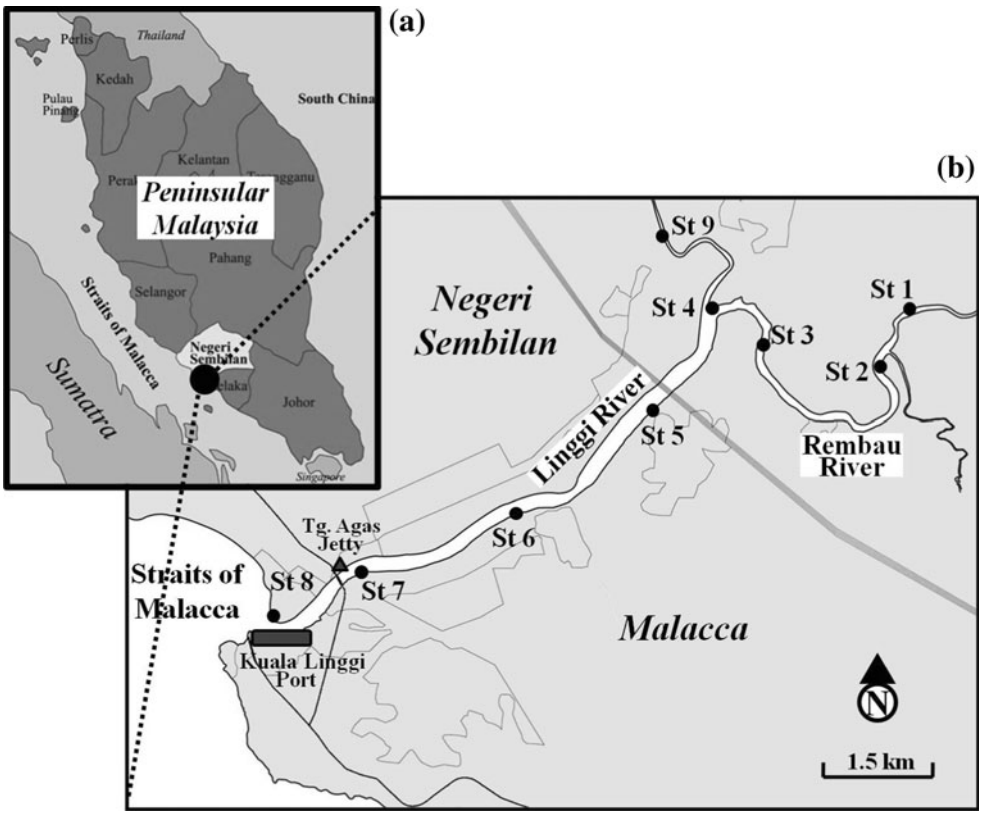

Fig. 1 Mangrove forest in Rembau–Linggi estuary, Negeri Sembilan, Malaysia: **a** the coastline of the Strait of Malacca facing Sumatra, Indonesia; **b** *circles* represent sampling stations; *rectangles* represent ports; and *triangles* represent jetties

(MS) detector coupled with Hewlett-Packard Model 5890 GC. A 30 m × 0.25 mm i.d. DB5MS fused silica capillary column and helium, as the carrier gas, were used in the analysis. The MS operating conditions were set at an ionization potential of 70 eV and an interface temperature of 280 °C. The temperature of the injection port was maintained at 310 °C and the sample (2 μL in volume) was injected in the splitless mode. The column temperature was held at 60 °C for 2 min and raised at a rate of 6 °C/min to 300 °C and held for 13 min (Yim et al. 2005). Seventeen PAH congeners were analyzed: phenanthrene, anthracene, 3-methylphenanthrene, 2-methylphenanthrene, 9-methylphenanthrene, 1-methylphenanthrene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo [k]fluoranthene, benzo[e]pyrene, benzo[a]pyrene, indeno [1,2,3-cd]pyrene, benzo[g,h,i]perylene, and dibenzo[a,h] anthracene.

Quality control

One procedural blank was made for every batch of samples and processed together with the samples from the extraction until the instrumental analysis. *P*-Terphenyl-d_{14} was used as GC internal injection standard (IISTD) and was spiked with the samples prior to their injection into the GC–MS. Concentrations of the target PAH congeners were quantified using the average response factors (RFs) generated from a five-point calibration curve which was constructed for the concentration range of 0.015–1.5 μg/g. For each sample, the concentrations obtained were recovery-corrected using the spiked surrogate standards. Concentrations of PAHs ranging from phenanthrene to pyrene and benzo[a]anthracene to benzo[g,h,i]perylene were corrected based on the recoveries of phenanthrene-d_{10} and chrysene-d_{12}, respectively. Standards for 2-, 3- and 9-methylphenanthrene were not available. Therefore, the response factors of these compounds were estimated based on the response of 1-methylphenanthrene.

Total organic carbon analysis

The analytical procedure for assessment of the total organic carbon (TOC) content is similar to the procedure described by Bakhtiari et al. (2010) and Raza et al. (2011). Plant roots and detritus were discarded from the sediments to distract their contribution of TOC to the sediment samples. The sediment samples were dried overnight at 60 °C in an oven and then ground and homogenized using mortar and pestle. In order to eliminate any inorganic carbon (carbonates) contained in the samples, an acidification procedure was employed. About 1–2 g of each sediment sample was weighed and 1–2 mL of 1 M hydrochloric acid (HCl) was

added until the sample was totally moist with HCl. After that the samples were dried at 100 °C for 10 h to remove HCl. About 1 g of each sample was reweighed and then analyzed using LECO CR-412 Carbon Analyzer at 135 °C for 60 s to determine the TOC percentage. The TOC is reported in mg/g unit.

Results and discussion

Composition and distribution of PAHs

The total concentrations of the 17 PAHs (Σ PAHs) in the mangrove sediments varied significantly ($p < 0.05$) among the stations and ranged from 20 to 112 ng/g dw (Table 1). The highest concentration of total PAHs was recorded in station 7 which is located near the Kuala Linggi Bridge and Tanjung Agas jetty (Fig. 1). The surrounding areas of station 7 are protected from wind and the sea waves, particularly from Strait of Malacca. Besides this, the abundance of *Sonneratia* and *Rhizophora* roots is likely to reduce water flows and enhance sediments settling (Zheng et al. 2002). This would promote deposition of the PAHs in the suspended particles that are transported through the river in this. The second highest concentration of total PAHs was recorded in station 8 which is located in the coastal area of the Strait of Malacca facing the Kuala Linggi port. The lowest total PAHs concentration was found in station 1 which is located at Rembau River. Station 1 and station 9, which is located in the upstream of the mangrove estuary, were compared in terms of PAHs composition. It was found that station 9 in Linggi River had a mean Σ PAHs concentration of 34.5 ng/g which is three times significantly ($p < 0.05$) higher than the respective concentration found in station 1 (11.69 ng/g). Both these stations were dominated by four-, and five-ring PAHs (Table 1; Fig. 2). A strong correlation ($r = 0.703$, $p < 0.05$) was found between Σ PAHs concentrations of station 1 and station 9, which indicates that contamination with PAHs in both stations might be derived from the same, or similar, sources.

It is not uncommon for the total PAH concentrations to increase towards the downstream of an estuary. The increasing PAH concentrations with distance from upstream

Table 1 Concentrations of PAHs and total organic carbon (TOC) content

PAH	Station 1	Station 2	Station 3	Station 4	Station 5	Station 6	Station 7	Station 8	Station 9
Phe	1.45	2.20	1.69	2.76	2.83	2.87	3.59	4.94	4.17
Ant	0.24	0.77	0.51	0.58	0.47	0.51	0.56	0.82	1.95
3-MP	0.59	3.69	2.34	2.56	3.45	3.43	2.27	2.98	2.35
2-MP	0.93	2.16	1.90	2.79	2.55	2.36	2.08	1.98	3.89
9-MP	0.58	4.21	1.67	2.27	3.76	1.89	1.43	2.12	1.75
1-MP	0.89	3.71	1.55	2.26	2.19	1.55	1.38	1.36	1.76
Flt	2.44	4.66	6.47	8.61	5.07	3.57	5.56	6.76	4.80
Pyr	2.20	4.73	6.13	8.32	5.30	4.29	4.86	6.42	4.19
B[a]A	1.35	2.39	2.71	3.01	7.83	12.4	10.7	5.48	6.05
Chr	1.91	2.58	2.75	4.25	4.71	3.93	5.76	7.04	6.59
B[b]F	2.65	3.83	5.04	6.49	8.30	6.86	13.0	11.0	6.31
B[k]F	0.74	0.92	1.46	1.78	2.11	1.76	3.80	3.80	4.43
B[e]P	1.22	1.46	2.30	3.51	3.86	3.08	5.91	5.16	3.69
B[a]P	0.92	1.21	2.35	3.09	3.14	2.19	5.93	6.22	5.69
I[c,d]P	1.78	2.43	3.59	5.48	6.01	4.53	14.3	7.96	4.15
DB[a,h]A	0.77	0.99	1.37	1.87	2.41	1.83	2.74	1.78	2.80
B[g,h,i]P	2.69	3.33	4.45	6.49	7.85	7.54	12.9	7.16	4.47
Total PAHs	23.4	45.3	48.3	66.1	71.8	64.6	96.8	83.0	69.1
Total LMW	4.68	16.7	9.66	13.2	15.3	12.6	11.3	14.2	15.9
Total HMW	18.7	28.5	38.6	52.9	56.6	52.0	85.5	68.8	53.2
CombPAH	17.9	27.5	37.3	51.0	54.2	50.1	82.8	67.0	50.4
TOC	18.4	21.2	18.9	13.3	19.5	21.8	27.3	16.7	12.2

Total PAHs: Sum of 17 PAHs ranging from phenanthrene to benzo[g,h,i]perylene (ng/g dw); Total LMW: sum of PAHs ranging from phenanthrene to 1-methylphenanthrene; Total HMW: sum of PAHs ranging from fluoranthene to benzo[g,h,i]perylene; CombPAH: Sum of fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzofluoranthenes, benzo[e]pyrene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene and benzo[g,h,i]perylene; TOC: total organic carbon (mg/g)

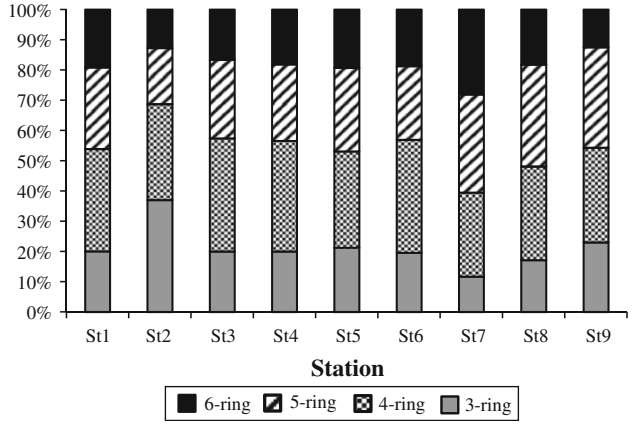

Fig. 2 Percentages of three- to six-ring PAHs in mangrove sediments of the nine sampling stations in Rembau–Linggi estuary

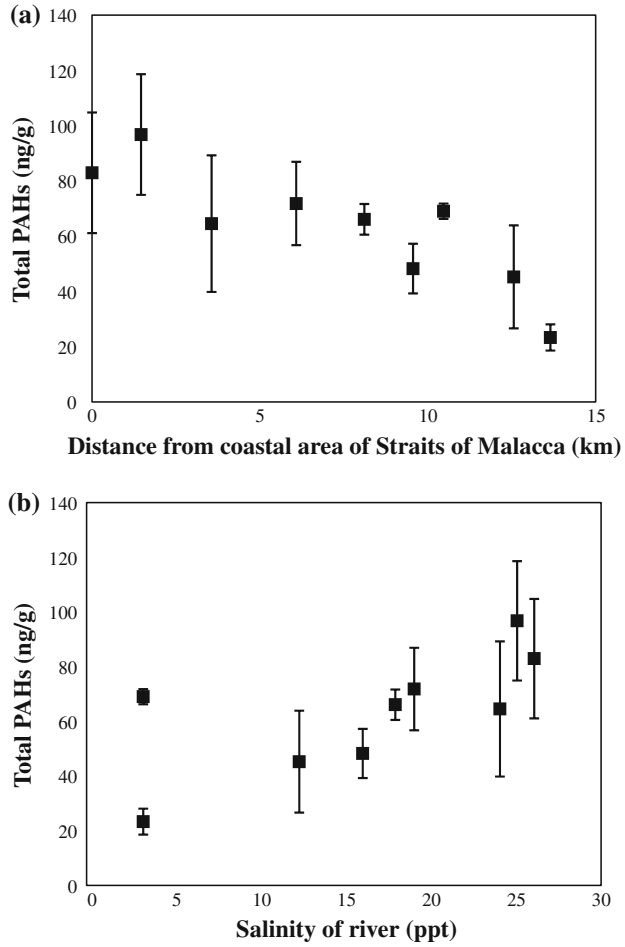

Fig. 3 Scatter plot showing: **a** total PAH concentrations and the distances of the sampling stations from the coastal area of the Strait of Malacca; **b** total PAH concentrations and salinity levels in the studied rivers

of the estuary to the coastal area of Strait of Malacca are shown in Fig. 3a. Since the highest PAHs concentration was recorded in the mouth of the estuary, it can be concluded that river discharge and urban runoff upstream of Rembau River and Linggi River might not be the principal sources of sedimentary PAHs. Instead, this finding suggests that urban runoff from the residential areas adjacent to the coastline of Strait of Malacca (for example, Kampung Kuala Linggi, Kampung Nelayan and Kampung Tanjung Agas) is the probable source of PAHs in the sample sediments. Conversely, several studies (Witt and Trost 1999; Tam et al. 2001; Boonyatumanond et al. 2006) found that the PAHs concentrations decreased in the direction of the sea and these researchers concluded therefore that river discharge and urban runoff were the principal sources of sedimentary PAHs.

It has been suggested that the pattern of increasing total PAHs concentrations downstream of a river were influenced by microbial biodegradation in the aerobic layer of the mangrove surface sediments (DeLaune et al. 1980). Several studies found that the rate of microbial biodegradation of PAHs and crude oil in sediments decrease with increasing salinity (Tam et al. 2001; Chen et al. 2009; Haritash and Kaushik 2009; Minai-Tehrani et al. 2009) since the high salinity can reduce the microbial metabolic rates (Ward and Brock 1978; Haritash and Kaushik 2009). The current study obtained similar results where the total PAH concentrations increased downstream of the estuary with the increasing salinity of the sea water (Fig. 3b).

The percent composition of PAHs by ring size (3–6) in nine sampling stations is shown in Fig. 2. High molecular weight (HMW) PAHs, specifically four-ring and five-ring PAHs, were relatively more abundant in all stations (33 and 28 %, respectively) excluding station 2. In contrast, sediments collected from station 2 were dominated by three-ring PAHs (37 %), ranging from phenanthrene to methylphenanthrenes. Referring to the PAH composition in Rembau River and Linggi River, it was found that the PAHs in both rivers were dominated by four-ring PAHs (34 and 32 %, respectively) (Fig. 2). The three-ring PAHs were the second most abundant PAHs in Rembau River due to abundance of phenanthrene, anthracene and methylphenanthrenes in station 2.

The most dominant individual PAH compounds were benzo[b]fluoranthene (five-ring PAH) and benzo [g,h,i]perylene (six-ring PAH), representing 10 and 9 % of the total PAH concentration, respectively. The findings of the present study are consistent with those of some other studies (e.g., Al-Saad and Al-Timari (1989); Bakhtiari et al. (2010)) where sedimentary PAHs showed abundance of HMW PAHs. This is due to their high particle affinities and low microbial degradation rates (Cerniglia 1992; Mahro et al. 1994) which lead to their accumulation in sediments (Al-Saad and Al-Timari 1989).

Correlation between PAHs and organic carbon

Mangrove sediments are preferential sites for uptake and preservation of hydrocarbons due to their high organic carbon content (Bernard et al. 1996). Organic carbon is believed to be an important controlling factor of the sorption of PAHs on sediments (Karickhoff et al. 1979). Several previous studies have found a significant positive correlation between PAHs and organic carbon (OC) in sediments collected from mangrove swamps (e.g., Ke et al. (2005); Liang et al. (2007)). In the current study, the OC content of the mangrove sediments of the estuary ranged from 10 to 27 mg/g (Table 1). The highest sediments' OC content was observed in station 7 and the lowest one was detected in station 9, which are both located in Linggi River. The area around station 9 is dominated by mudflats where the mangrove plants are lacking due to the deforestation of mangroves. Therefore, the mudflats were not affected or influenced by the mangrove roots and leaves which would be the main source of organic matter in the sediments. In general, the spatial distribution of OC in Rembau–Linggi estuary is influenced by the different land use types that exist within this estuary. Different land use patterns and changes in the mangrove estuary have resulted in the varied content of OC in the sediments.

However, no correlation was found between PAHs in the sediments and sediments' OC content in the present study (Fig. 4). This is in agreement with several previous works (Tam et al. 2001; Ke et al. 2005; Farias et al. 2008; Cavalcante et al. 2009) where the OC content did not correlate with the PAH concentrations in the mangrove sediments. Simpson et al. (1996) concluded that correlation of PAHs in the sediments with sediments' OC is only significant for highly contaminated sites where the total PAH concentrations were >2,000 ng/g. In the present study, the sediment in all stations had total PAH concentrations less than this value (Table 1). Hence, it is concluded that the distributions and concentrations of PAHs in sediments of this mangrove estuary are not controlled by the sediments' OC content.

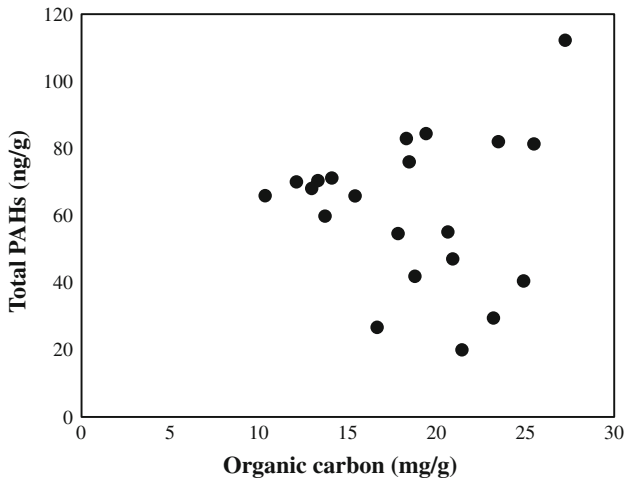

Fig. 4 Scatter plot showing the relationship between total PAH concentration and organic carbon content in mangrove sediments

Identification of the sources of PAHs

Molecular ratios of PAHs have been widely used in hydrocarbons studies for source identification of PAH contamination in the environment. Using the ratio of parent or alkylated PAHs, the origins of PAHs can be identified as either petrogenic PAHs (petroleum and petroleum products) or pyrogenic PAHs (incomplete combustion of coal and petroleum, or biomass burning). Petrogenic PAHs are abundant in low molecular weight (LMW) PAHs and alkylated PAHs, while the pyrogenic PAHs consist of high concentrations of HMW PAHs and low concentrations of alkylated PAHs (Zakaria et al. 2002; Saha et al. 2009; Dsikowitzky et al. 2011).

In order to identify the sources and origin of PAHs contamination in this study, several PAHs ratios were applied. Parent PAHs ratios, namely, ratio of anthracene/(anthracene + phenanthrene) (Ant/(Ant + Phe)), ratio of fluoranthene/(fluoranthene + pyrene) (Flt/(Flt + Pyr)), ratio of benzo[a]anthracene/(benzo[a]anthracene + chrysene) (B[a]A/(B[a]A + Chry)) and ratio of indeno[1,2,3-cd]pyrene/(indeno[1,2,3-cd]pyrene + benzo[g,h,i]perylene) (I[c,d]P/(I[c,d]P + B[g,h,i]P)) have been used widely in many PAHs studies for source identification. The ratios of Ant/(Ant + Phe) and B[a]A/(B[a]A + Chry) enable distinction between the petroleum and petroleum combustion sources of PAHs, whereas the ratios of Flt/(Flt + Pyr) and I[c,d]P/(I[c,d]P + B[g,h,i]P) can be used to distinguish between PAHs derived from petroleum, petroleum combustion as well as combustion of charcoal, grass and wood (Yunker et al. 2002; Zemo 2009). As proposed by Youngblood and Blumer (1975), the ratio of methylphenanthrenes to phenanthrenes (MP/P) has been employed to distinguish between petrogenic and pyrogenic sources of PAHs in many previous studies (e.g., Zakaria et al. (2002), Boonyatumanond et al. (2006), Sakari et al. (2008), Saha et al. (2009); Bakhtiari et al. (2010)). The methylphenanthrenes consist of 3-methylphenanthrene, 2-methylphenanthrene, 9-methylphenanthrene and 1-methylphenanthrene which are divided by phenanthrene concentration.

In the present study, the values of Ant/(Ant + Phe) and Flt/(Flt + Pyr) ranged from 0.10 to 0.35 and from 0.44 to 0.55, respectively. Whereas, the values of B[a]A/(B[a]A + Chry) and I[c,d]P/(I[c,d]P + B[g,h,i]P) ranged from 0.41 to 0.81 and from 0.33 to 0.53, respectively (Fig. 5). Interestingly, it was found that all the values of

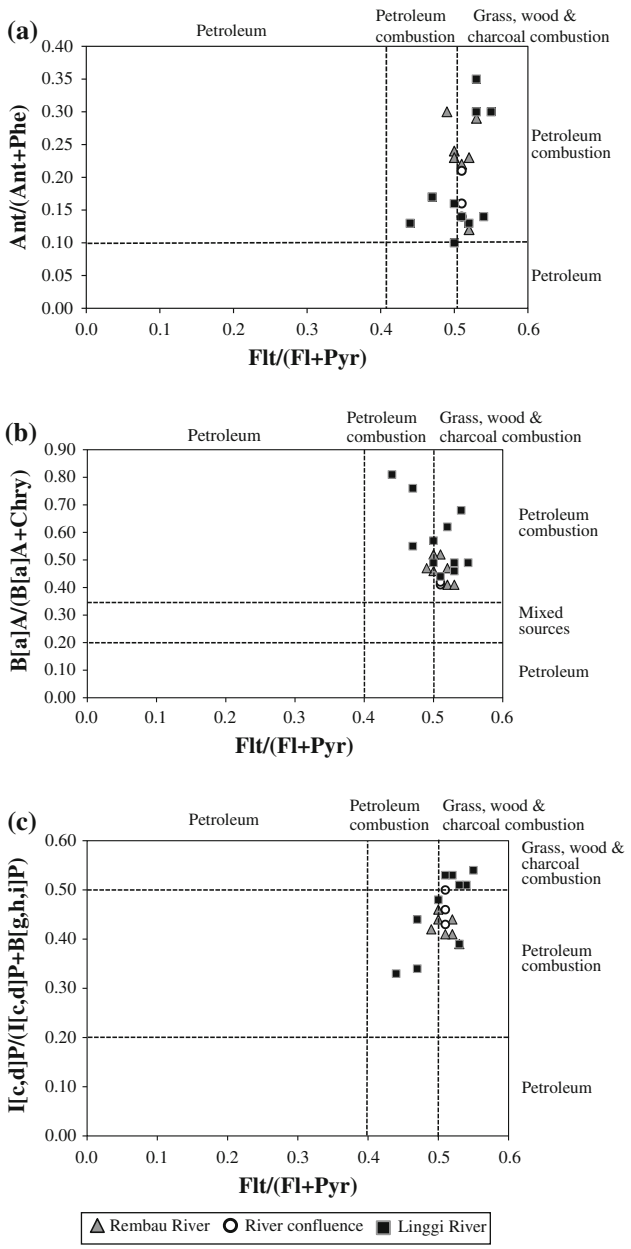

Fig. 5 Scatter plot of PAH ratios and their sources: **a** ant/(Ant + Phe) versus Flt/(Flt + Pyr); **b** B[a]A/(B[a]A + Chry) versus Flt/(Flt + Pyr); (c) I[c,d]P/(I[c,d]P + B[g,h,i]P) versus Flt/(Flt + Pyr)

Flt/(Flt + Pyr), B[a]A/(B[a]A + Chry) and I[c,d]P/(I[c,d]P + B[g,h,i]P) fell in the pyrogenic zone, thus indicating that the mangrove sedimentary PAHs in the nine study stations originate from combustion activities (Fig. 5). This demonstrates that PAHs in the mangrove sediments, both in Rembau and Linggi rivers, derived from pyrogenic processes.

Specifically, the ratios of Flt/(Flt + Pyr) and I[c,d]P/(I[c,d]P + B[g,h,i]P) indicate that PAHs in station 2, station 5 and station 6 originated from petroleum combustion, whereas the PAHs in station 7, station 8 and station 9 originated from combustion of charcoal, grass and wood. Sedimentary PAHs in station 2 and station 9 derived from different combustion processes as these stations are located in the upstream areas of two different rivers. Conversely, sedimentary PAHs in station 9 and station 8 which were located upstream and downstream of the same river, respectively, originated from combustion of charcoal, grass and wood. Although station 8 is located in the coastal area of Strait of Malacca facing the Kuala Linggi port, there is no indication that this station is under threat of oil pollution from sea-based sources. Based on these findings, sedimentary PAHs mainly originated from combustion processes and they have been brought by long-range atmospheric transports and deposited into this mangrove estuary.

Moreover, the location of Rembau–Linggi estuary which is facing the Strait of Malacca and Sumatra (Indonesia) experienced biomass burning originating from southern Sumatra (Radzi bin Abas et al. 2004). It is expected that PAHs derived from biomass burning during haze episode and associated with atmospheric particles were transported by air masses during the monsoon season (He et al. 2010). The aerosols in Peninsular Malaysia were found to be dominated by pyrogenic PAHs originating from automotive exhaust emissions (Okuda et al. 2002) and biomass burning (Omar et al. 2006; Bahry et al. 2009). Contamination with PAHs in urban and remote areas where PAHs were transported through long-range atmospheric transport has been reported by many researchers (Okuda et al. 2002; Yang et al. 2007; He et al. 2010).

Since the alkylated homologues of PAHs generally originate from petrogenic sources (Zakaria et al. 2002; Dsikowitzky et al. 2011), the PAH ratios of MP/P and ratio of the sum of 3-methylphenanthrene and 2-methylphenanthrene divided by 9-methylphenanthrene plus 1-methylphenanthrene (3-MP + 2-MP)/(4/9-MP + 1-MP) were calculated in order to identify the contribution of petrogenic PAH input to the mangrove sediments. The MP/P ratios for all sediment samples ranged from 1.67 to 11.69 which indicate that sedimentary PAHs in all stations are mainly petrogenic PAHs (Fig. 6). Furthermore, it was found that the ratio of (3-MP + 2-MP)/(4/9-MP + 1-MP) had values ranging from 0.45 to 2.10 in the sediments of most of the petrogenically affected stations, hence demonstrating that these sediments are affected by similar petrogenic sources. This result implied that petrogenic input had a contribution to the sedimentary PAHs, but that it was not a major controlling factor in distribution of PAHs within the estuary. This finding is consistent with that of Zakaria et al. (2002) who found that the riverine, estuarine and coastal sediments in the west coast of Peninsular Malaysia were generally polluted by petrogenic PAHs where used crankcase oil was the major source of sedimentary PAHs in urban areas. This study suggests that petrogenic sources of PAHs in this

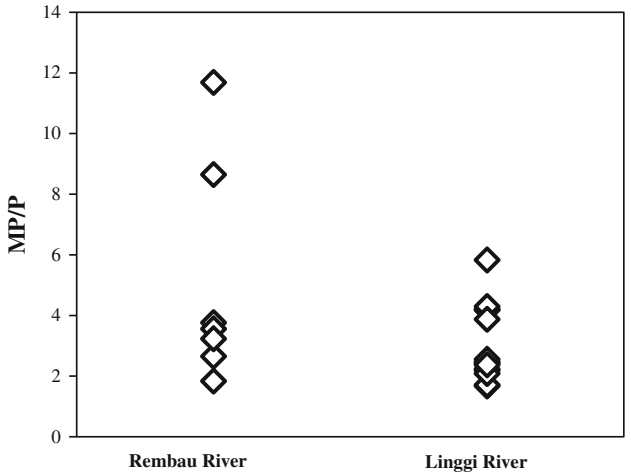

Fig. 6 The ratio of MP/P. Methylphenanthrenes (MP) consist of 3-methylphenanthrene, 2-methylphenanthrene, 9-methylphenanthrene, and 1-methylphenanthrene

estuary originate from ship and boat activities (Maher and Aislabie 1992) as well as from used crankcase oil brought to the estuary by urban runoff from residential areas near the coastline of the Strait of Malacca.

Regarding that some sites in the current study showed both petrogenic and pyrogenic signatures, the regions having higher pyrogenic inputs are likely to have a petrogenic signature in the phenanthrene series only (Saha et al. 2009). Thus, Saha et al. (2009) suggested that the differential homolog distribution among petrogenic and pyrogenic sources is the main reason for the source difference among ratios. Since the pyrogenic sources of PAHs have contributed tangibly to the sediments of this estuary, the relationship between combustion-derived PAHs (CombPAH) and total PAHs was examined. CombPAH is represented by the sum of fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzofluoranthenes, benzo[e]pyrene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene and benzo[g,h,i]perylene (Prahl and Carpenter 1983). CombPAH showed a positive and very strong correlation with total PAHs ($R^2 = 0.926$, $p < 0.05$) (Fig. 7a), suggesting that pyrogenic sources of PAHs had important contribution to the sedimentary PAHs in this mangrove estuary. Additionally, the relationship between total methylphenanthrenes and total PAHs was studied to elucidate the influence of petrogenic input to sedimentary PAHs. Total methylphenanthrenes showed very weak correlation with total PAHs ($R^2 = 0.0928$, $p < 0.05$) (Fig. 7b), implying that petrogenic input had some contribution to the sedimentary PAHs but that it was not a major controlling factor in distribution of PAHs within estuary. The findings of this study demonstrates that the mangrove estuary was exposed to PAH contamination from both petrogenic and pyrogenic sources.

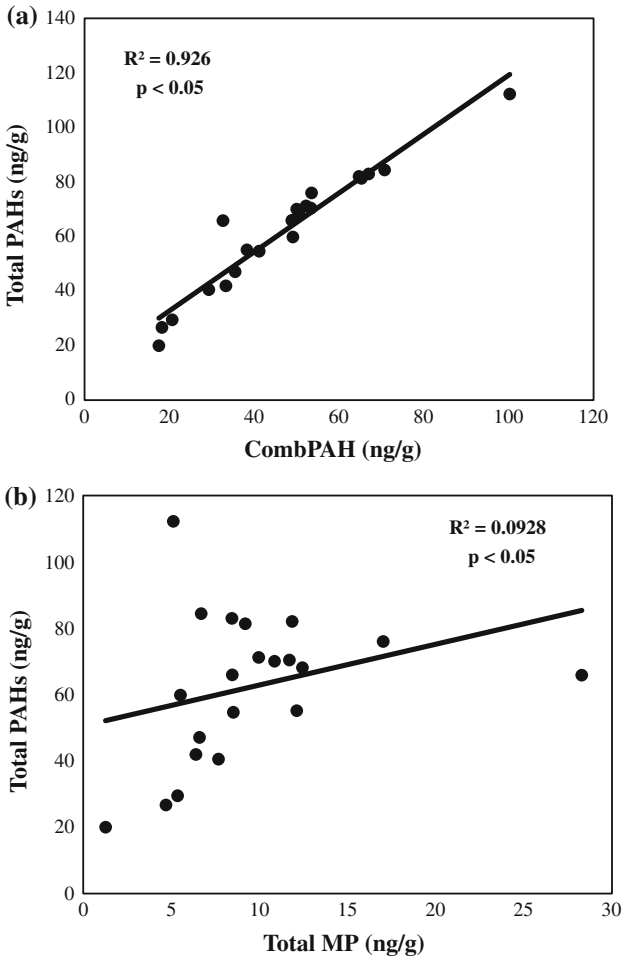

Fig. 7 Scatter plot showing the relationships between: **a** total PAH and CombPAH concentrations; **b** total PAH and total methylphenanthrenes (MP) concentrations

Comparison with other mangrove forest worldwide

Not many studies have been carried out globally to assess the levels and distribution of PAHs in mangrove ecosystems. Most of PAH studies were conducted on marine water, bottom sediments and biota. Studies of PAHs in mangrove forests were mainly conducted in China (Table 2). In Malaysia, there is lack of studies on PAHs in mangrove ecosystems. Furthermore, there are no data on the levels and distribution of PAHs in mangrove sediments of the western part of Peninsular Malaysia. The mangrove sediments investigated in this study demonstrated slight pollution in comparison with other mangrove sites (Table 2). The concentrations of the 17 PAHs in the present study ranged from 20 to 112 ng/g dw. These levels are relatively low as compared to mangroves in Mai Po, Tolo, Sai Keng and Ho Chung in China where the values ranged from 356 to 11,098 ng/g (Tam et al. 2001). Moreover, Tam et al. (2001) found that the concentrations of

PAHs in surface sediments of mangroves were generally higher than marine bottom sediments of harbors. Other mangrove sites like Fortaleza in Brazil recorded high values (>2,000 ng/g dw) of total PAH concentrations as well (Cavalcante et al. 2009).

In a study by Zakaria et al. (2002), estuarine sediments in Pinang Estuary which is located in the West Coast of Peninsular Malaysia, contained about 924 ng/g of PAHs. Zakaria et al. (2002) also found that the levels of PAHs in estuarine and coastal sediments collected from Klang Estuary and offshore of the Strait of Malacca ranged from 19 to 431 ng/g and from 4 to 73 ng/g, respectively. Additionally, total PAHs concentrations were reported to range from 260 to 590 ng/g in coastal sediments from the East Coast of Peninsular Malaysia (Elias et al. 2007) which is facing the South China Sea. These levels are relatively higher than the levels detected in the present study. These findings therefore indicate that PAH contamination in Peninsular Malaysia is diverse and influenced by the surrounding environment.

Assessment of sediment contamination using biological thresholds

The effect-based guidelines can be used to compare and evaluate the levels of pollutants in areas that have the potential for biological effects, especially for aquatic organisms that are closely associated with sediments. Two preferred sediment quality guidelines (SQGs) were applied in this study in order to evaluate the toxicity potential of PAHs in mangrove surface sediments of Rembau–Linggi estuary to aquatic organisms. The two SQGs are the effect range low (ERL)–effect range median (ERM) guideline proposed by Long et al. (1995) and the threshold effects level (TEL)–probable effects level (PEL) guideline proposed by Macdonald et al. (1996).

According to Long et al. (1995), concentrations below the ERL value represent minimal-effects range; a range intended to estimate conditions in which effects would be rarely observed. Concentrations equal to or above the ERL, but below the ERM, represent a possible-effects range within which effects irregularly happen. Lastly, concentrations equivalent to or above the ERM represent a probable-effects range within which effects would frequently occur (Long et al. 1995). On the other hand, Macdonald et al. (1996) interpreted the TEL as concentrations below which adverse biological effects rarely occur and so the TEL are considered to provide a high level of protection for aquatic organisms. Moreover, they interpreted the PEL as the concentrations above which adverse biological effects frequently occur and thus the PEL are considered to provide a lower level of protection for aquatic organisms.

Referring to Table 3, the mean total PAHs concentration in the mangrove sediments at the nine sampling stations is 50.3 ng/g, which is much lower than the ERL and TEL values which correspond to 4,022 and 655 ng/g, respectively.

Table 2 Comparison of PAHs concentration (ng/g dw) between the study area and other mangrove forests worldwide

Study area	Sample	No. of PAHs	Total PAHs (ng/g dw)	References
Worldwide				
Mai Po, Tolo, Sai Keng and Ho Chung, China	Mangrove sediments	15	356–11,098	Tam et al. (2001)
Hong Kong SAR, China	Mangrove sediments	16	56–3,758	Ke et al. (2005)
Jiulong River Estuary, China	Mangrove sediments	16	193–271	Zhi-qiang et al. (2005)
Deep Bay, China	Mangrove sediments	16	238–726	Zhang et al. (2004)
Mai Po, Hong Kong	Mangrove sediments	15	212–1,042	Zheng et al. (2000)
Ceara and Coco River, Fortaleza, Brazil	Mangrove sediments	17	96–2,235	Cavalcante et al. (2009)
Caribbean Island of Guadeloupe	Mangrove sediments	13	103–1,675	Bernard et al. (1996)
Sundarban Mangrove Wetland, India	Mangrove sediments	16	241–1,376	Domínguez et al. (2011)
Sundarban Mangrove Wetland, India	Mangrove sediments	19	9–4,223	Sarkar et al. (2012)
Malaysia				
Rembau–Linggi Estuary, Malaysia	Mangrove sediments	17	20–112	This study
Pulau Cik Wan Dagang, Terengganu	Mangrove sediments	16	120–1,420	Tahir et al. (2011)
West Coast of Peninsular Malaysia	Coastal sediments	15	4–924	Zakaria et al. (2002)
East Coast of Peninsular Malaysia	Coastal sediments	17	260–590	Elias et al. (2007)
West Coast of Peninsular Malaysia	Mudflat sediments	7	0.8–163	Mirsadeghi et al. (2011)
Kelantan and Terengganu River, Malaysia	Riverine & estuarine sediments	17	59–1,689	Sakari et al. (2008)
Klang River, Malaysia	Riverine sediments	19	1,304–2,187	Bakhtiari et al. (2010)

The same finding applies to the maximum PAH concentrations as well. The maximum of total PAH concentrations in the mangrove sediments at the nine stations is 144 ng/g, which is also much lower than ERL and TEL values, that is, 4,022 and 655 ng/g, respectively. None of the individual PAH compounds exceeded the values of ERL–ERM and TEL–PEL guidelines (Table 3). The highest concentration among individual PAH compounds was that of benzo[a]anthracene (20.9 ng/g dw). However, this value is much lower than the ERL and TEL of benzo[a]anthracene, namely, 261 and 75, respectively. On the other hand, the maximum concentration of dibenzo[a,h]anthracene, 4.74 ng/g, is close to the TEL guideline value of 6 ng/g. The value of 4.74 ng/g of dibenzo[a,h]anthracene was recorded in Station 9 which is located in Linggi River.

Therefore, the PAH levels in mangrove sediments in this estuary correspond to minimal-effects and, consequently, the PAHs are unlikely to cause any adverse biological effects to aquatic organisms (Long et al. 1995; Macdonald et al. 1996). It is important to evaluate the sedimentary PAH concentrations with reference to related guidelines since aquatic organisms such as mangrove oysters are able to bioaccumulate PAHs from surface sediments (Dsikowitzky et al. 2011; Mirsadeghi et al. 2011). As a conclusion, the PAH levels in the mangrove surface sediments in Rembau–Linggi estuary are unlikely to cause any adverse biological effects to aquatic organisms, including those that are closely associated with sediments.

Conclusion

In conclusion, the PAHs found in the mangrove sediments in Rembau–Linggi estuary are dominated by HMW PAHs as well as influenced both by pyrogenic and petrogenic sources. The pyrogenic sources of PAHs include particulate PAHs resulting from combustion activities which are brought by long-range atmospheric transport, deposited into this mangrove estuary and accumulated mostly in the downstream and estuarine mouth regions. Additionally, this study also revealed that the sediments of the mangrove ecosystem facing the Strait of Malacca and Sumatra are influenced by anthropogenic PAHs as a result of human activities (e.g., such as biomass burning, vehicle emissions and boating activities) in surrounding areas. Another important finding of the current study is the PAH concentrations were found to increase with distance from upstream of the estuary to the coastal area of the Strait of Malacca. This suggests that petrogenic sources of PAHs in this estuary originate from ship and boat activities as well as from used crankcase oil that is brought by urban runoff. Although a number of PAH compounds have been detected in the sediments, the data indicate that Malaysian mangrove forests are less polluted than mangroves in other countries. Furthermore, none of the individual PAH compounds exceeded the values of ERL–ERM and TEL–PEL guidelines, thus the sedimentary PAHs of this mangrove estuary are unlikely to cause any adverse biological effects

Table 3 Proposed guidelines for levels of PAHs in sediments

PAHs	Sediment quality guidelines		Concentration of PAH in sediments	
	ERL–ERM (ng/g dw)	TEL–PEL (ng/g dw)	Mean (ng/g dw)	Maximum (ng/g dw)
Naphthalene	160–2,100	35–391	–	–
Acenaphthylene	44–640	6–128	–	–
Acenaphthene	16–500	7–89	–	–
Fluorene	19–540	21–144	–	–
Phenanthrene	240–1,500	87–544	2.51	4.94
Anthracene	85–1,100	47–245	0.64	2.28
Fluoranthene	600–5,100	113–1,494	5.45	11.32
Pyrene	665–2,600	153–1,398	5.30	10.76
Benzo[a]Anthracene	261–1,600	75–693	5.61	20.94
Chrysene	384–2,800	108–846	4.61	16.02
Benzo[b]fluoranthene	–	–	6.83	17.94
Benzo[e]pyrene	–	–	3.24	7.80
Benzo[a]pyrene	430–1,600	89–763	3.19	7.56
Indeno[1,2,3-cd]pyrene	–	–	5.04	20.84
Dibenzo[a,h]anthracene	63–260	6–135	1.92	4.74
Benzo[g,h,i]perylene	–	–	6.0	18.54
Total PAHs	4,022–44,792	655–6,676	50.34	143.68

ERL effect range low, *ERM* effect range median, *TEL* threshold effects levels, *PEL* probable effects level

to aquatic organisms. The current paper represents a baseline study for further studies which should include the whole of Peninsular Malaysia and the East Malaysia region in order to enhance our understanding of the distribution of PAHs in mangrove ecosystems.

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