



Distributions, Composition Patterns, Sources and Potential Toxicity of Polycyclic Aromatic Hydrocarbons (PAHs) Pollution in Surface Sediments from the Kim Kim River and Segget River, Peninsula Malaysia

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ABSTRACT

This study investigated distributions, composition patterns, sources and potential toxicity of polycyclic aromatic hydrocarbon (PAHs) pollution in surface sediments from the Kim Kim River and Segget River, Peninsular Malaysia. The samples were extracted using Soxhlet extraction, purified using two-step silica gel column chromatography and then analysed by gas chromatography mass spectrometry (GC-MS). The total PAH concentrations ranged from 95.17 to 361.24 ng g⁻¹ dry weight (dw) and 330.09 to 552.76 ng g⁻¹ dw in surface sediments from the Kim Kim and Segget Rivers, respectively. Source type identification using PAH molecular indices and hierarchical cluster analysis (HCA) indicated that PAHs were mostly of pyrogenic origin, while in some stations petrogenic sources had a significant portion. A PAH toxicity assessment using sediment quality guidelines (SQGs), mean effect range-median quotient (M-ERM-Q), benzo[*a*]pyrene (BaP) equivalent concentration and BaP toxicity equivalent quotient (TEQ^{carc}) indicated low probability of toxicity for both the Kim Kim and Segget Rivers. Moreover, the human health risk assessment applying Cancer Risk_{ingestion} and Cancer Risk_{dermal} indicated that probabilistic health risk to humans via ingestion and dermal pathways from sediments of the Kim Kim and Segget Rivers can be categorised as low-to-moderate risk.

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INTRODUCTION

Johor State is one of the most developed states of Malaysia (Alkhadher et al., 2016), located in the southern part of Peninsular Malaysia. It is connected to Singapore by the Straits of Johor. The population size and density of Johor State have increased continuously in the last two decades. In 2012, the population of Johor State rose from 2.76 million in 2000 to 3.46 million (increase over 25%) (DOSM, 2014a, 2014b). Increase in the population size and density, quick development (industrialisation, urbanisation, agriculture and aquaculture) and marine transportation in Johor State and along the coastal areas of the Johor Straits can result in comprehensive physical and chemical changes in the aquatic environment. Industrial development in the region has also resulted in increase in the usage of petroleum and its derivatives as sources of energy. As a consequence, petroleum hydrocarbon contaminants released into this environment became significant. The Kim Kim River and Segget River run through Johor State. Pollutants, including polycyclic aromatic hydrocarbons (PAHs), originating from anthropogenic activities such as petrol combustion, industrialisation and multiple diffuse sources in Johor State and the neighboring area are released into the rivers via dry and wet deposit, urban runoff and direct input. It was reported that the Segget River is grossly polluted by domestic sewage (Lye & Eng, 1988; Tung et al., 2013).

Polycyclic aromatic hydrocarbons (PAHs) are categorised as one of the most important classes of contaminants. They have two or more benzene rings with variable physicochemical properties (Neff, 1979). PAHs are widely distributed in diverse ecosystems, including freshwater and marine, and they have drawn significant attention due to their persistence and toxicity and mutagenic, teratogenic and carcinogenic effects (Cao et al., 2009).

Many studies have reported that urban and riverine runoff (Alkhadher et al., 2015; Lipiatou et al., 1997; Magam et al., 2015; Motelay-Massei et al., 2006) and atmosphere (Liu et al., 2007b; Tsai et al., 2002) are the two most important transportation pathways of contaminants of the aquatic environment. Hoffman et al. (1984) demonstrated that urban runoff comprised 71% HMW PAHs and 36% total PAHs input into Narragansett Bay. This is consistent with Oros et al. (2007), who suggested that 51% of total PAH in the San Francisco Bay was transported by storm water runoff. The sources of PAHs in urban runoff are wear of asphalt surfaces, abrasion of vehicle tires, car exhaust and used crankcase oils (Takada et al., 1991). Incomplete combustion derived PAHs emitted into the atmosphere from different sources enter the water column directly via dry and wet deposition or are indirectly deposited to the terrestrial environment and urban runoff; they include street dust and storm water discharge, which are finally transported to the aquatic ecosystem.

PAHs can originate from natural processes, but the major source of PAH input is generally from anthropogenic activities (Keshavarzifard et al., 2017a; Keshavarzifard and Zakaria, 2015; Keshavarzifard et al., 2015; Masood et al., 2016). As for pollution due to anthropogenic activities, the origins are classified as pyrogenic PAHs (from high temperature incomplete combustion of organic matter) and petrogenic PAHs (from fresh petroleum input). Nevertheless, pyrogenic PAHs i.e. incomplete combustion derived PAHs can be adsorbed or absorbed to airborne organic particles and moved long distances by wind and other atmospheric phenomena.

Due to lipophilic characteristic, PAHs are mostly partitioned with organic matter in the environment and undergo sedimentation (Keshavarzifard et al., 2017b); however, occasionally these contaminants are not partitioned with organic matter, which indicates existence of soot particles in sediments (Gustafsson & Gschwend, 1997). PAH compounds are associated with soot particles and transported in long-range atmospheric pathway (Pereira et al. 1999). Petrogenic PAHs (associated with particles containing organic carbon) and pyrogenic PAHs (partitioned with airborne particles) reach the surface layers of river and marine water columns and pass through the water columns and then settle down in the bottom sediments. It was reported that the sorption of hydrocarbons in sediments is influenced largely by the content of organic carbons, particle size distribution and the octanol-water partition coefficients of the pollutant (Gustafsson et al., 1996). This statement is consistent with those reported by Karickhoff and Brown (1978) and Karickhoff et al. (1979). It was suggested that petroleum hydrocarbons deposited in sediments are more likely to partition into porewater than pyrogenic PAHs associated with soot particles (McGroddy & Farrington, 1995). Usually HMW PAHs i.e. 4-, 5- and 6-ring PAHs, which are heavier than LMW PAHs i.e. 2- and 3-ring PAHs, are placed in the centre of the particles and the LMW PAHs are placed around the particles and become heavier and then undergo sedimentation. Pyrogenic and petrogenic PAHs release into the river and marine ecosystems and then undergo dispersion, evaporation, settlement in the sediments, weathering, chemical changes, photooxidation and microbial degradation (bacteria and fungus) in short- and long-term periods (Aldarondo-Torres et al., 2010; Irwin, 1997; Mai et al., 2002; Neff, 1979; Ünlü et al., 2010).

The objectives of this study were: (a) to assess the concentration and distribution of 16 USEPA PAHs in surface sediment from the Kim Kim and Segget Rivers; (b) to identify the PAH sources using molecular indices; (c) to evaluate the toxicity potency and human health risk of PAHs. The results of this research in conjunction with long-term study of PAHs in the Malacca and Johor Straits enrich our knowledge of the distribution and pathways of PAHs in this region and introduce useful information about PAH pollution in order to improve regulations and consequently the environmental quality of the study area.

METHODOLOGY

Sampling

In May 2013, six surficial sediment samples (0-4 cm) were collected from the Kim Kim and Segget Rivers using an Ekman Dredge sampler. The location of the sampling stations are shown in Figure 1 and Table 1. The top 4 cm of surface layer was carefully removed to identify modern input. After separating the organisms and shell debris from sediments at each sampling station, the samples were immediately placed in previously cleaned aluminum containers and Ziploc bags, and then stored in a cooler box with dry ice. The samples were subsequently freeze-dried and stored at -20°C before analysis.

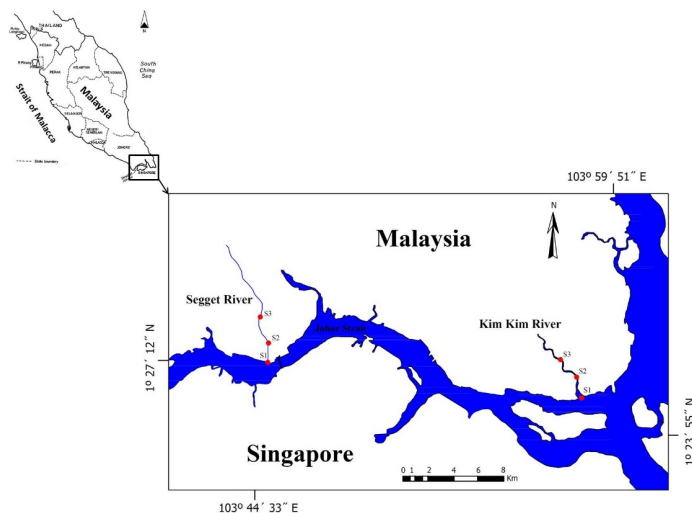


Figure 1. Location of sampling areas

Table 1
Details of water quality data of sampling stations

Sampling Location	Latitude (°N)	Longitude (°E)	DO (mg/l)	Turbidity (NTU)	Temperature (°C)	pH
Kim Kim River S1	01° 25'	103° 58'	5.9	15	29.8	7.55
Kim Kim River S2	01° 20"	103° 58'	4.7	7.32	29.9	7.47
Kim Kim River S3	01° 27'	103° 57'	6.54	8.6	20.3	7.27
Segget River S1	01° 27'	103° 45'	4.5	42.3	29	7.27
Segget River S2	01° 27'	103° 45'	2	54.8	28	7.63
Segget River S3	01° 26'	103° 45'	3.4	48	28.9	7.76

Extraction and Purification

Each sediment sample was extracted, purified and fractionated according to the method explained in Zakaria et al. (2002). In summary, hydrocarbons were extracted by Soxhlet Apparatus using 200 mL dichloromethane (DCM) for 10 h. A volume of 50 µL of 10 µg g⁻¹ surrogate internal standards (SIS), namely naphthalene-d₈, anthracene-d₁₀, chrysene-d₁₂, and perylene-d₁₂, were added to the sample aliquot before extraction. The SIS compounds were used for recovery verification. The extraction was purified and fractionated through first- and second-step silica gel column chromatography. The extract was rotary evaporated and sulfur interference was removed using activated copper. The extract was allowed to pass through a silica gel column containing 5% H₂O-deactivated silica gel to purify non-polar compounds such as hydrocarbons. The hydrocarbon compounds (alkanes, hopanes, linear alkyl benzenes (LABs), PAHs and many more) were fractionated using a fully activated silica gel column (0.47 cm i.d × 18 cm height). A volume of 16 mL rinsing solution (hexane-DCM 3:1 v/v) was added. The

amount of PAH fraction was further reduced by rotary evaporator and a gentle stream of pure N₂ gas. The final fraction was then adjusted to 200 µL with isooctane containing a 10-ppm internal injection standard (IIS), p-terphenyl-d₁₄. Analysis of PAHs was performed by GC-MS Shimadzu QP5050A model and a BPX-5MS fused-silica capillary column (30 m by 0.25 mm i.d., 0.25-µm film thickness). The oven temperature was ramped from 60 to 260°C at 6° min⁻¹, and then held for 15 min. An aliquot 1 µL of purified sample was injected into the GC-MS injector. The oven temperature started at 70°C for 2 min with a ramp of 30°C.min⁻¹ until 150°C. The temperature was further increased to 310°C with an increasing rate of 4°C.min⁻¹ for 15 min. The 16 PAHs detected in sequence were as follows: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Fl), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fluo), pyrene (Py), benzo[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[1,2,3-*cd*]pyrene (InP), dibenzo[*a,h*]anthracene (DBA), and benzo[*ghi*]perylene (BgP).

Total Organic Carbon (TOC) Analysis

TOC content of sediment sample was identified using a LECO CR-412 carbon analyser. Carbonates were eliminated by 2 mL of 1 M HCL at 100°C overnight before TOC identification (Nelson & Sommers, 1996).

Quality Control and Assurance

Quality control of sediment samples was applied to ensure the accuracy and precision of the analytical procedure. Surrogate internal standards were used to examine the recovery percentage of each sample. The recovery rates of 16 individual EPA PAHs ranged from 81% to 110%. Quantification of PAH compounds was processed based on external calibration curves using standard mixtures of PAH. Determination of target compounds was achieved based on matching target compound ionisation and retention times with the standard mixture of PAHs. Internal injection standard (p-terphenyl-d₁₄) was used for quantitation and identifying injection error. Procedural blanks were subjected for every batch of four samples; no contamination was detected in the analytical system and glassware.

RESULTS AND DISCUSSION

Distribution of PAHs in Surface Sediments

Analytical results of the 16 EPA priority PAHs measured at different sampling stations are listed in Table 2. The total PAH concentrations varied from 95.17 to 361.24 ng g⁻¹ dry weight (dw) with a mean value of 218.1 ng g⁻¹ dw in sediments from the Kim Kim River and 330.09 to 552.76 ng g⁻¹ dw with a mean value of 437.5 ng g⁻¹ dw in sediments from the Segget River. Therein, the highest concentrations were determined at Station 1 (estuary) of both the Kim Kim and Segget Rivers and lowest concentrations recorded at Station 3 (upstream) of the Kim Kim and Segget Rivers. PAHs normally partition to lipophilic particles in the water column and as transported downstream, more particles clump together to flocculate. As the particles reach the salinity regime in estuaries, they settle to the bottom sediment, which usually

increases the concentration of PAHs in sediments closer to an estuary. Moreover, tidal current might also influence the PAH distribution pattern. Baumard et al. (1998) categorised the PAHs levels in the aquatic environment from 0-100 (low), 100-1000 (moderate), 1000-5000 (high) and >5000 ng g⁻¹ (very high). According to this categorisation, the PAH levels can be classified as low-to-moderate in the Kim Kim River and moderate in the Segget River.

Table 2
PAHs in surface sediments from the Kim Kim and Segget rivers

	Kim Kim River			Segget River			¹ ERL	² ERM	³ TEL	⁴ mPEL	⁵ TEF
	Station 1	Station 2	Station 3	Station 1	Station 2	Station 3					
Nap	18.38	6.17	6.63	33.68	55.41	34.16	160	2100	34.6	391	0.001
Acy	19.64	7	9.38	6.33	51.1	4.89	44	640	5.87	128	0.001
Ace	39.72	2.74	4.23	5.72	47.77	2.4	16	500	6	88.9	0.001
Fl	18.56	10.45	8.81	7.49	75.55	11.26	19	540	21.2	144	0.001
Phe	16.22	8	19	56.92	10.41	51.98	240	1500	86.7	544	0.001
Ant	36.27	23.26	0.82	4.26	51.47	9.55	85	1100	46.9	245	0.001
Fluo	84.52	18	7.8	55.92	9.96	14.38	600	5100	113	1494	0.01
Py	90	17.31	12.43	52.1	16.19	10.72	665	2600	153	1398	0.001
BaA	14.54	30.8	2.73	47.3	3.22	0.61	261	1600	74.8	693	0.1
Chr	3.69	1.89	3.1	4.28	13.75	0.13	384	2800	108	846	0.01
BbF	3.59	15.63	7.7	44.51	2.15	0.4	-	-	-	-	0.1
BkF	2.1	7.19	1.27	10.99	25.28	46.1	-	-	-	-	0.1
BaP	4.55	24.23	2.31	84.39	28	53.88	430	1600	88.8	763	1
InP	2.25	11.46	1.28	54.91	5.9	15.5	-	-	-	-	0.1
DBA	2.11	6.9	0.8	21.48	4.68	53.64	63	260	6.2	135	1
BgP	5.1	6.75	6.88	62.48	28.86	20.49	-	-	-	-	0.01
⁶ Total PAHs	361.24	197.78	95.17	552.76	429.7	330.09	4000	45000	1684	16770	-
⁷ LMW PAHs	148.79	57.62	48.87	114.4	291.71	114.24	552	3160	-	-	-
⁸ HMW PAHs	212.45	140.16	46.3	438.36	137.99	215.85	1700	9600	-	-	-
⁹ LMW/ HMW	0.7	0.41	1.1	0.26	2.11	0.53	-	-	-	-	-
¹⁰ Py/Fluo	1.06	0.96	1.59	0.93	1.63	0.75	-	-	-	-	-
¹¹ Fluo/ (Fluo+Py)	0.48	0.51	0.39	0.52	0.38	0.57	-	-	-	-	-
¹² BaA/ (BaA+Chr)	0.79	0.94	0.47	0.92	0.19	0.82	-	-	-	-	-
¹³ InP/ (InP+BgP)	0.31	0.63	0.16	0.47	0.17	0.43	-	-	-	-	-
¹⁴ TOC %	1.15	0.9	8.1	2.1	1.89	3.31	-	-	-	-	-

⁶Total PAHs = Sum of the concentrations of 16 USEPA PAHs; ⁷LMW PAHs = Sum of low molecular weight PAH concentrations from naphthalene to fluoranthene; ⁸HMW PAHs = Sum of high molecular weight PAH concentrations from pyrene, benzo[a]anthracene to benzo[ghi]perylene

⁹LMW/HMW = The ratio of low molecular weight PAHs to high molecular weight PAHs; ¹⁰Py/Fluo = The ratio of pyrene to fluoranthene; ¹¹Fluo/Fluo+Py = The ratio of fluoranthene to fluoranthene + pyrene; ¹²BaA/(BaA+Chr) = The ratio of Benzo(a)anthracene to Benzo(a)anthracene+Chrysene; ¹³InP/(InP+BgP) = The ratio of indeno(1,2,3-cd)pyrene to indeno(1,2,3-cd)pyrene+benzo(ghi)perylene; ¹⁴TOC = Total organic carbon

¹ERL = Effects range-low value (Long et al. 1995); ²ERM = Effects range-median value (Long et al. 1995); ³TEL = Threshold effect levels (Macdonald et al. 1996); ⁴mPEL = Probable effect levels (Macdonald et al. 1996); ⁵TEF = Toxic equivalency factor

The PAH concentrations in the Kim Kim River were compared with those of Zakaria et al. (2002), who found that PAH levels ranged from 37 to 41 ng g⁻¹ (Table 3), confirming an increase in the levels of PAHs. However, more investigation is needed to support this statement. Among the PAH levels in the Malaysian aquatic ecosystems (Table 3), the concentrations of total PAH in sediments from the Kim Kim and Segget Rivers are close to those in sediments from the Selangor (Masood et al., 2014), the Merambong and Muar (Vaezzadeh et al., 2015) and the Perlis aquatic ecosystems (Keshavarzifard et al., 2015). However, they are lower than those of the Prai (Keshavarzifard et al., 2014b) and the Klang Rivers (Keshavarzifard et al., 2015). In comparison with other aquatic ecosystems in the world, sediment contaminations by PAHs in the Kim Kim and Segget Rivers are comparable with those of developing countries and much lower than those of developed and fast developing countries (Table 3).

Table 3
Comparison of PAH concentrations in surface sediments of different worldwide locations and this study

Location	Total PAHs (ng g ⁻¹)	^a n	^b Contamination Level	References
Boston Harbour, USA	7300-358000	16	Very high	Wang et al., 2001
Barcelona Harbour, Spain	1740-8420	14	High to very high	Baumard et al., 1998
Cape Cod, Massachusetts, USA	30000	38	Very high	Thorsen et al., 2004
Guanabara Bay, Brazil	91-8035	20	Low to very high	Meniconi et al., 2002
Patos Lagoon Estuary, Brazil	37-11779.9	23	Low to very high	Medeiros et al., 2005
Rybinsk Reservoir, Russia	178-5203	11	High to very high	Siddall et al., 1994
Lake iznik, Turkey	17-835	12	Low to moderate	Ünlü et al., 2010
Bohai Sea, China	0.14-10757	16	Low to very high	Zeng et al., 2013
Hyeongsan River, Korea	5.30-7680	16	Low to very high	Koh et al., 2004
Iranian coast of the Persian Gulf	93-4077	15	Low to high	Mirza et al., 2014
Gorgan Bay, Caspian Sea, Iran	107.9-516	16	Moderate	Araghi et al., 2014
Jakarta Bay, Indonesia	191-1252	18	Moderate to high	Rinawati et al., 2012
Rivers and Estuary of Thailand	6-8399	11	Low to very high	Boonyatumanond et al., 2006
Kim Kim River and Estuary, Malaysia	37-41		Low	Zakaria et al., 2002
Penang Estuary, Malaysia	924		Moderate	
Malacca River (1998), Malaysia	494	15	Moderate	
Klang Estuary, Malaysia	19-431		Low to moderate	
Johor Bahru Coast, Malaysia	239		Moderate	
Chini Lake, Malaysia	248-8098	16	Moderate to high	Bakhtiari et al., 2009
Prai River, Malaysia	1102-7938	16	High to very high	Keshavarzifard et al., 2014b

Table 3 (continue)

Malacca River, Malaysia	716-1210	16	Moderate to high	
Merambong River, Malaysia	38.6-122.8	16	Low to moderate	Vaezzadeh et al., 2015
Muar River, Malaysia	15.5-165.7		Low to moderate	
Selangor River, Malaysia	563-1037	16	Moderate to high	Masood et al., 2014
Perlis River, Malaysia	225.5-293.9		Moderate	Keshavarzifard et al., 2015
Kedah River, Malaysia	195.2-481.2		Moderate	
Merbok River, Malaysia	791.2-1995.4	16	Moderate to high	
Perak River, Malaysia	231.2-426.7		Moderate	
Klang River, Malaysia	3803.2-7442.7		High to very high	
Port Dickson, Malaysia	481.3-976.6		Moderate	Keshavarzifard & Zakaria, 2015
Johor Bahru Coast, Malaysia	650.5-1441.2	16	Moderate to high	Keshavarzifard et al., 2016
Malacca Strait, Malaysia	357.1-6257.1	16	Moderate to very high	Keshavarzifard et al., 2017d
Kim Kim River, Malaysia	95.17-361.24	16	Low to moderate	This study
Segget River, Malaysia	330.09-552.76		Moderate	This study

^aNumber of PAH compounds; ^bScale of level of contamination: low: 0-100; moderate: 100-1000; high: 1000-5000; very high: >5000 (Baumard et al., 1998)

Determination of PAHs Sources Based on PAH Composition

Many investigations have confirmed that based on the characteristics of PAH composition and distribution pattern, the sources of anthropogenic PAHs can be distinguished (Keshavarzifard et al., 2017d; Liu et al., 2009; Vaezzadeh et al., 2014; Zakaria et al., 2002). Of anthropogenic PAHs, the incomplete combustion sources such as vehicle exhaust, domestic heating with coal and biomass burning contain predominantly HMW PAHs including 4-, 5- and 6-ring PAH compounds or pyrogenic PAHs, whereas the uncombusted sources (e.g. crude and fuel oil) contain predominantly LMW PAHs, including 2- and 3-ring PAH compounds or petrogenic PAHs (Bouloubassi et al., 2012; Irwin, 1997; Mai et al., 2002; Zakaria et al., 2002). In Stations 1 and 2 of the Kim Kim River, 4-ring PAHs had the highest portion of PAHs followed by 3-ring PAHs (Figure 2). In Station 3 of the Kim Kim River, 3-ring PAH had the highest portion, followed by 4-ring PAHs (Figure 2). In Stations 1 and 2 of the Segget River, PAHs were dominated by 4- and 3-ring PAHs, respectively. PAHs in Station 3 of the Segget River were dominated by 5-ring PAHs, followed by 6-ring ones (Figure 2). These results displayed that Stations 1 and 3 of the Segget River were significantly impacted by combustion-derived PAHs present in urban atmospheric particles, while Station 2 had received fresh petroleum

hydrocarbon. Based on the results, surface sediments from the Kim Kim and Segget Rivers showed that they contained both pyrogenic and petrogenic PAHs with pyrogenic PAHs dominating.

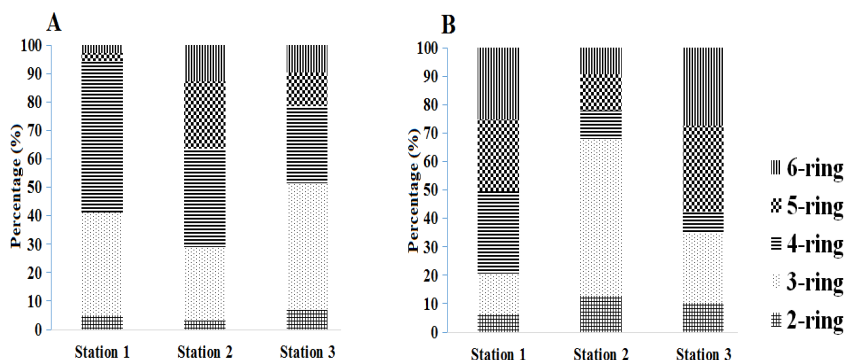


Figure 2. The composition pattern of PAHs by ring size in surface sediments from the Kim Kim River (A) and Segget River (B)

Determination of PAHs Sources Using Diagnostic Ratios

The diagnostic ratios of PAHs such as Py/Fluo, Fluo/(Fluo+Py), BaA/(BaA+Chr) and InP/(InP+BgP) have been introduced as a useful tool to identify and assess the possible sources of PAHs (Keshavarzifard et al., 2014a; Qian et al., 2016; Yunker et al., 2002). For collected sediment samples of PAH isomer pair ratios, Py/Fluo was plotted against LMW/HMW and Fluo/(Fluo+Py) was plotted against BaA/(BaA+Chr) and InP/(InP+BgP) to demonstrate the distribution of PAHs relative to their possible sources (Figure 3 and Figure 4). In general, a ratio of Py/Fluo > 1 indicates petroleum derivative PAHs, while a ratio < 1 suggests a dominance of combustion derivative PAHs (Sicre et al., 1987). Magi et al. (2002) reported that the ratio of LMW PAHs (2- and 3-ring PAHs) to HMW PAHs (4-, 5- and 6-ring PAHs) higher than 1 displays a pollution of petroleum contamination, while a ratio lower than 1 displays pyrogenic-origin PAHs. In this study, Py/Fluo ratios at Station 2 of the Kim Kim River and Stations 1 and 3 of the Segget River were lower than 1, indicating a dominance of pyrogenic PAHs. In the other stations, Py/Fluo ratios were lower than 1, suggesting a dominance of petroleum inputs (Table 2 and Figure 3). This is confirmed by the ratios of LMW/HMW, which were lower than 1 at all stations except Station 1 and 3 of the Kim Kim River and Station 2 of the Segget River (Table 2 and Figure 3).

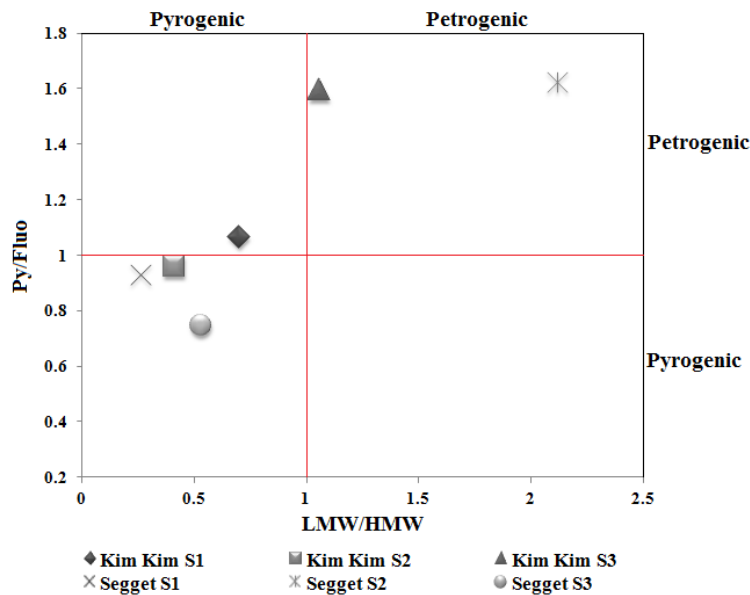


Figure 3. Plots of PAH isomer pair ratios for source identification: LMW/HMW versus Py/Fluo

Sediments with $\text{Fluo}/(\text{Fluo}+\text{Py}) < 0.4$ indicate petrogenic contamination, while $\text{Fluo}/(\text{Fluo}+\text{Py}) > 0.5$ suggests biomass and coal combustion origins and sediments with $0.4 < \text{Fluo}/(\text{Fluo}+\text{Py}) < 0.5$ indicate petroleum combustion sources (Yunker et al., 2002). Those PAHs in sediments with the ratio of $\text{InP}/(\text{InP}+\text{BgP}) < 0.2$ were mainly from petrogenic sources, while those with $\text{InP}/(\text{InP}+\text{BgP}) > 0.2$ were typical of combustion-derivative PAHs (Budzinski et al., 1997; Yunker et al., 2002). Moreover, $\text{BaA}/(\text{BaA}+\text{Chr})$ ratios lower than 0.2, from 0.2 to 0.35 and higher than 0.35 are used as an indicator of petroleum, petroleum/combustion and combustion sources, respectively (Yunker et al., 2002). In this study, the ratios of $\text{Fluo}/(\text{Fluo}+\text{Py})$ at Station 3 of the Kim Kim River and Station 2 of the Segget River were lower than 0.4, indicating dominance of petroleum input, while at Station 1 of the Kim Kim River the ratio was 0.48, suggesting petroleum combustion processes make a more significant contribution than petroleum. Other stations had $\text{Fluo}/(\text{Fluo}+\text{Py})$ ratios more than 0.5, indicating biomass and coal combustion origins (Table 2 and Figure 4). This was confirmed by the results of $\text{InP}/(\text{InP}+\text{BgP})$ ratios (Table 2 and Figure 4). The ratio of $\text{BaA}/(\text{BaA}+\text{Chr})$ at Station 2 of the Segget River was 0.19, indicating petroleum input could be the main source. The $\text{BaA}/(\text{BaA}+\text{Chr})$ ratios in other stations were more than 0.35, implying combustion derivative PAHs played a major role.

Overall, sediments from the Kim Kim and Segget Rivers were affected by both petrogenic and pyrogenic sources, with a greater contribution from pyrogenic sources. This is consistent with Keshavarzifard et al. (2014b) and Keshavarzifard et al. (2015), who suggested dominance of combustion derivative PAHs in sediments from the major rivers of Peninsular Malaysia. Our findings are also consistent with those of industrialised countries (Notar et al., 2001;

Pereira et al., 1999; Stout et al., 2004), where pyrogenic PAHs were reported as mainly being of PAH origin in aquatic ecosystems. For example, recent widescale studies of background anthropogenic hydrocarbons in surficial sediments collected from the Arc River, the Berre Lagoon and an industrialised urban river (Huveaune) in France using PAH ratios to identify the sources of PAHs affirmed the pyrogenic-origin background of PAHs (Kanzari et al., 2014; Kanzari et al., 2012). On the other hand, Zakaria et al. (2002) reported petrogenic sources as a major source in Malaysian aquatic ecosystems including the Kim Kim River, which is in contrast with the results of this study. It can be suggested that the origin of PAHs in the Kim Kim River has shifted from petrogenic to pyrogenic sources; however, more studies are needed to confirm these findings.

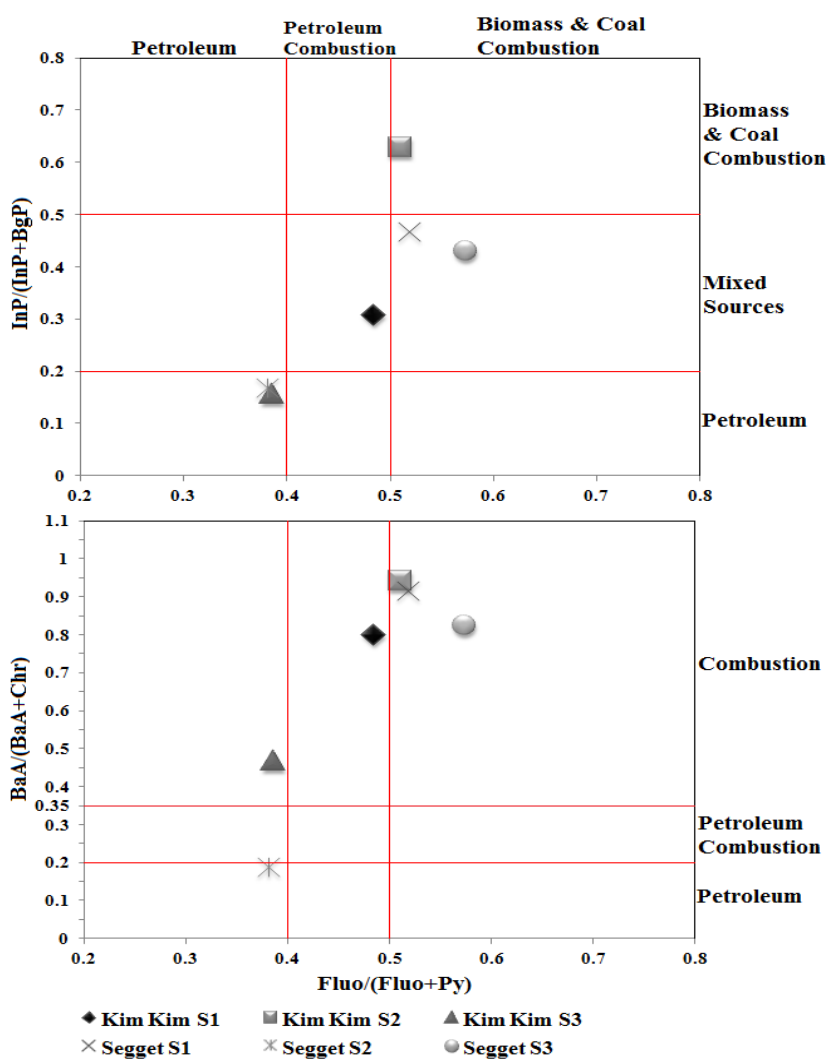


Figure 4. Plots of PAH isomer pair ratios for source identification: Fluo/(Fluo+Py) versus BaA/(BaA+Chr) and InP/(InP+BgP)

Determination of PAH Sources Using Hierarchical Cluster Analysis (HCA)

Hierarchical cluster analysis (Wards clustering method using average linkage between groups and Squared Euclidean distance as measure interval for PAH data normalised by Z score transformation) in package cluster (Maechler et al., 2012) was carried out to identify possible sources of individual PAHs in the surface sediments from the Kim Kim and Segget Rivers. It was assumed that PAHs with chemical configuration or similar ring number would be clustered among the samples. The hierarchical diagram clustered the 16 individual PAHs into major groups based on the similarity of PAH fingerprints (Figure 5 and Figure 6).

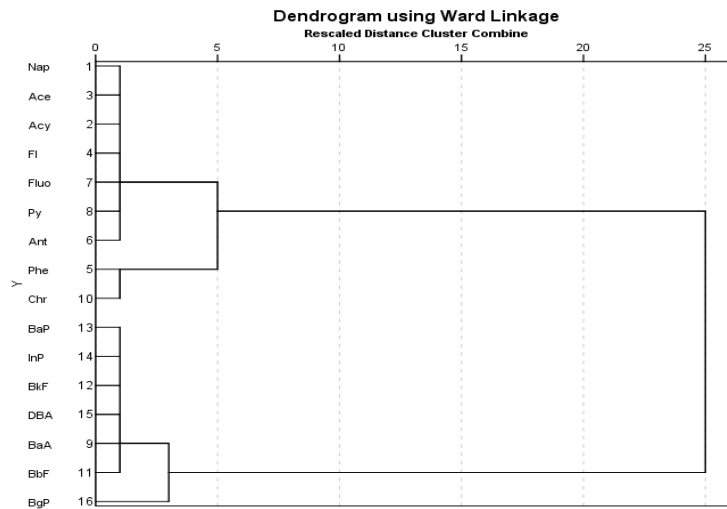


Figure 5. Hierarchical dendrogram for 16 individual PAHs in sediment from the Kim Kim River

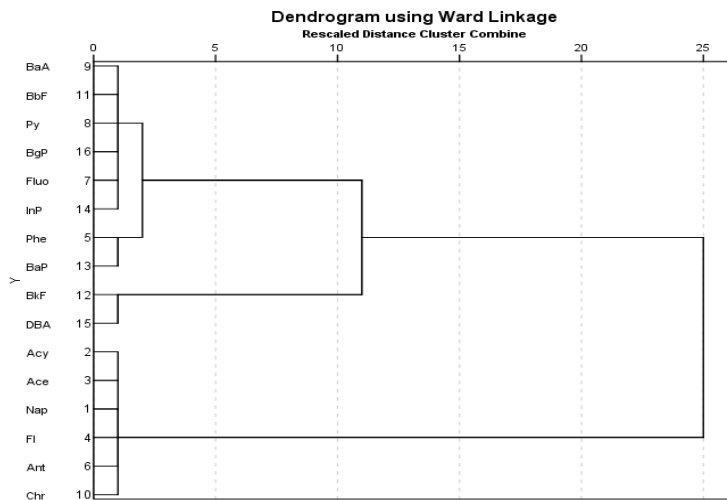


Figure 6. Hierarchical dendrogram for 16 individual PAHs in sediment from the Segget River

For PAHs in the Kim Kim River, cluster analysis led to four major groups. Group 1 included Nap, Ace, Acy, Fl, Fluo, Py and Ant, which belong mainly to lower molecular weight PAHs. Lower molecular PAHs including Nap, Acy, Ace, Fluo and Ant have been reported as markers of spills or crude oil leaks. Also, these compounds can be used as markers for low-temperature pyrogenic processes such as biomass combustion of straw and firewood (Jenkins et al., 1996; Yang et al., 2006; Zhang et al., 2008). As a result, Group 1 was believed to be mixed sources of PAHs derived from petroleum pollution and incomplete combustion of biomass. Group 2 included Phe and Chr, which can be identified as the markers of coal combustion source (Sofowote et al., 2008). The third group included BaP, InP, BkF, DBA, BaA and BbF. This group could be selected to represent emissions from vehicles because it aggregated mainly PAHs of higher molecular weight. Group 4 contained BgP, which identified as a tracer of gasoline emission (Harrison et al., 1996; Li & Kamens, 1993; Miguel & Pereira, 1989). The gasoline- and diesel-fuelled traffic on the road and boats in the study area may have contributed to these groups.

Similarly, cluster analysis represented four major groups for PAHs in the Segget River. The first group included BaA, BbF, Py, BgP, Fluo, InP, which are higher molecular weight PAHs with 4 to 5 rings. Five- and 6-ring PAHs were mostly found in road dust (Liu et al., 2007a; Ren et al., 2006) and vehicular emissions (Zuo et al., 2007). Group 2 was characterised by Phe and BaP. The research literature cited Phe as a tracer for coal combustion (Sofowote et al., 2008), while BaP is produced mostly during pyrolysis processes (Jung et al., 2010; Siddens et al., 2012). Based on field investigations, the Segget River passes through Johor Bahru; therefore, the road traffic in the city and residential and industrial activities near the Segget River may be the main contributors to PAH pollution in the Segget River. The third major group mainly consisted of BkF and DBA. Previous studies suggested that DBA indicates PAH compounds emitted from diesel combustion (Harrison et al., 1996; Larsen & Baker, 2003). Motelay-Massei et al. (2005) reported that BkF is a fossil-fuel combustion marker. Therefore, it is reasonable to attribute this group to vehicular emission. Group 4 was characterised by low molecular weight PAHs with 2 to 3 rings, including Acy, Ace, Nap, Fl and Ant, which are abundant in petrogenic sources and mainly are a result of petroleum spills (Dobbins et al., 2006; Liu et al., 2009). HCA suggested that both petrogenic and pyrogenic sources including traffic emissions, coal combustion, biomass burning and petroleum spills were the major sources of PAH contamination in sediments from the Kim Kim River and Segget River.

In the past two decades, population as well as urbanisation in Malaysia has had continuous growth, which caused an increase in the number of vehicles and road traffic. Possible sources for combustion-derivative PAHs in the sediments from the Kim Kim and Segget Rivers could be attributed to vehicular and industrial emissions, which are transported into the study areas via long-range atmospheric transport. This is consistent with Okuda et al. (2002) and Omar et al. (2002, 2006), who reported road-traffic emissions as the main origin of PAHs in atmospheric aerosols in Kuala Lumpur, Malaysia.

In addition, biomass burning from Sumatra, Indonesia has a portion of combustion-derivative PAHs in the study areas. He et al. (2010) reported higher level of PAHs in Singapore during the month of October 2006 compared to other months, especially two to three times at nighttime. Interestingly, satellite images showed hot spots in Sumatra Island in October, which

implied large biomass burning. The high concentration of retene in October in Singapore's atmosphere was attributed to long-range atmospheric transportation of carbonaceous material originated from biomass combustion in Indonesia (He et al., 2010). This is consistent with Okuda et al. (2002), who demonstrated that the large-scale biomass burning in Indonesia caused haze episodes in Malaysia, with high levels of carbonaceous material containing PAHs. As the Johor State is relatively close to Indonesia, the same influence can be suggested for the Johor State. However, local wood-burning can also be introduced as an important PAH source in the Kim Kim and Segget Rivers. For example, Okuda et al. (2002) reported a high portion (25%-35%) of local wood burning to Malaysian atmospheric PAHs. In other studies the measured PAH concentrations in Kula Lumpur during a hazy day were reported many folds higher than those on clear days (Abas et al., 2004; Omar et al., 2002), indicating the impact of biomass burning to PAH Levels.

Toxic Potency and Ecological Risk Assessment of PAHs

PAHs are widely distributed in diverse ecosystems, including freshwater and marine, and they have drawn significant attention due to their persistence and toxicity and mutagenic, teratogenic and carcinogenic effects. Sediment quality guidelines (SQGs) including effects range-low value (ERL), effects range-median value (ERM), probable effects level (PEL) and threshold effects level (TEL) (Long et al., 1995; Macdonald et al., 1996) are widely used for the purpose of ecological risk assessment of PAHs in aquatic ecosystems. The comparison between the measured concentrations of PAHs from sediments and the ERL, ERM, PEL and TEL values are listed in Table 2. The LMW PAHs, HMW PAHs and total PAH concentrations of sediment samples from the Kim Kim and Segget Rivers were below the ERL, ERM, PEL and TEL levels. It can be suggested that the ecological risk of PAHs was relatively low. However, PAH contamination of sediments from the study areas should be further evaluated for toxic potency assessment and ecosystem impairment due to rapid industrialisation, urbanisation and economy development in the region.

The ecological risk assessment technique of PAHs in marine and estuary surface sediments was also selected in our study (Long et al., 1995). The concentrations of effects range low (ERL) and effects range median (ERM) were applied to evaluate the ecological toxicity of individual PAH by some scholars (Mai et al., 2002). In addition, the quotient technique of average ERM was a complex manner of quantitative prediction for joint toxicity of pollutants in marine and estuary sediments. In the following equations,

$$ERM-Q = \frac{C_i}{ERM_i} \quad (1)$$

$$M-ERM-Q = \frac{\sum ERM-Q}{n} \quad (2)$$

where, C_i represents the measured concentration of given PAHs; ERM-Q is the risk quotient of ERM and M-ERM-Q stands for average risk quotients of ERM concentrations. When

M-ERM-Q is not 0.10, there rarely is observable toxicity to organisms (biological risk chance was less than 10%); when it is within 0.11-0.50, the possibility of ecological risk increases to 30% with low toxicity; when M-ERM-Q is between 0.51-1.50, the probability of ecological risk increases to 50% with high toxicity; when it is more than 1.50, the probability of ecological risk increases to 75% with higher toxicity (Long & MacDonald, 1998). The results indicated that values of M-ERM-Q in all sediment samples from the Kim Kim and Segget Rivers were lower than 0.1, which implied low probability (9%) of ecological effects.

Human Health Risk Assessment

The toxicity of a PAH compound is usually expressed by its BaP equivalent concentration (BaP_{eq}), and the toxicity equivalency factors (TEFs), proposed by Nisbet and LaGoy (1992). BaP is the most toxic PAH among all known potentially carcinogenic PAHs; hence, it is used as the reference chemical and assigned a value of 1 in the TEF system (Nisbet & LaGoy, 1992; Pufulete et al., 2004). The TEF values of other PAHs was calculated based on their carcinogenic level in comparison to that of BaP (Pufulete et al., 2004). The total BaP_{eq} in sediment samples was calculated according to Eq. 3,

$$Total\ BaP_{eq} = \sum(C_i \times TEF_i) \quad (3)$$

where, BaP_{eq} is the equivalent BaP concentration of PAHs, C_i is the concentration of PAH $_i$ and TEF_i is the toxic equivalency factor of individual PAH $_i$ (Table 2). Among 16 PAH compounds, seven PAHs, BaA, Chr, BaP, BbF, BkF, InP and DBA, have high toxic and carcinogenic effects. The potential toxicity of the sediment samples was then assessed using the toxic equivalent quotient (TEQ^{carc}) for seven carcinogenic PAHs calculated according to the following equation (Keshavarzifard et al., 2017c; Tian et al., 2013):

$$TEQ^{carc} = \sum(C_i \times TEF_i) \quad (4)$$

where, C_i is the concentration of an individual carcinogenic PAH ($ng\ g^{-1}$) and TEF_i is the toxic factor of this carcinogenic PAH relative to BaP.

The BaP_{eq} of total PAHs in collected sediment samples from the Kim Kim River and the Segget River ranged from 4.65 to 37.98 $ng\ g^{-1}$ and 37.17 to 123.03 $ng\ g^{-1}$, respectively (Table 4), indicating a relatively low level of BaP_{eq} . Total TEQ^{carc} in the Kim Kim River ranged from 4.44 to 37.66 $ng\ g^{-1}$ - BaP_{eq} , with a mean concentration of 17.01 $ng\ g^{-1}$ - BaP_{eq} . In addition, the sediment samples from the Segget River ($TEQ^{carc} = 36.47$ to 121.68 $ng\ g^{-1}$ - BaP_{eq} , with a mean concentration of 90.64 $ng\ g^{-1}$ - BaP_{eq}) contained slightly higher levels of total TEQ^{carc} than those from the Kim Kim River.

Table 4
Estimated ecological and human health risk caused by PAH contaminated sediment

Parameter	Kim Kim River			Segget River		
	S1	S2	S3	S1	S2	S3
M-ERM-Q	0.018	0.010	0.005	0.024	0.032	0.025
Total BaP _{eq}	10.08	37.98	4.65	123.03	37.17	114.26
TEQ ^{car}	8.94	37.66	4.44	121.68	36.47	113.78
Cancer risk _{ing}	2.26E-04	1.24E-04	5.94E-05	3.45E-04	2.69E-04	2.06E-04
Cancer risk _{der}	1.17E-04	6.41E-05	3.09E-05	1.79E-04	1.39E-04	1.07E-04

In comparison, TEQ^{carc} values in sediments of the Kim Kim and Segget Rivers were lower than those of other areas reported in earlier studies (Table 5). The low BaP_{eq} concentrations suggest potentially low carcinogenicity for both the Kim Kim and Segget Rivers. The contribution of each carcinogenic PAH to the toxic equivalent quotient of sediments in the study area varied according to the following order:

The Kim Kim River: BaP (60.91%) > DBA (19.22%) > BaA (9.42%) > BbF (5.27%) > InP (2.94%) > BkF (2.07%) > Chr (0.17%)

The Segget River: BaP (61.14%) > DBA (29.34%) > BkF (3.03%) > InP (2.81%) > BaA (1.88%) > BbF (1.73%) > Chr (0.07%).

Table 5
The toxic equivalent quotient (TEQ^{carc}) in sediment from different locations around the world

Location	TEQ ^{carc} (ng g ⁻¹ -BaP _{eq})	References
Naples Harbour , Italy	2-4723	Sprovieri et al., 2007
Kaohsiung Harbour, Taiwan	3.9-1970	Chen et al., 2013
Meiliang Bay, China	94 to 856	Qiao et al., 2006
Barents Sea, Russia	18-300	Savinov et al., 2003
Langkawi Island, Malaysia	76.3 to 174.6	Nasher et al., 2013

The results indicated that BaP and DBA had the highest portion of carcinogenic PAHs in sediment from both the Kim Kim and Segget Rivers.

Generally, there are pathways (i.e. ingestion and dermal contact) for humans to be exposed to PAHs via contaminated sediments. The assumption is that ingestion and dermal contact are more important ways. Therefore, the health risk assessments were determined based on the exposure factors and were calculated by assuming the daily intake, exposure time and skin contact (Keshavarzifard et al., 2017c). The health risk assessment through the ingestion and the dermal absorption pathways from sediments was calculated using the following equations:

$$Cancer Risk_{ingest} = \frac{C_s \times IngR \times EF \times ED}{BW \times At} \times CF \times SFO \quad (5)$$

$$Cancer\ Risk_{dermal} = \frac{C_s \times SA \times AF \times ABS \times EF \times ED}{BW \times At} \times CF \times SFO \quad (6)$$

where, $Cancer\ Risk_{ingest}$ is cancer risk through ingestion of sediment; C_s is the concentration of PAH compound; $IngR$ is the ingestion rate: $100\ mg\ day^{-1}$; EF is the exposure frequency: $350\ days\ year^{-1}$; ED is the exposure duration: $70\ years$; BW is the body weight: $70\ kg$; At is the average day: $25,550\ days$; CF is the unit conversion factor: $10^{-6}\ kg\ mg^{-1}$; SFO is the oral slope factor: $7.3\ mg\ (kg\ day)^{-1}$; $Cancer\ Risk_{dermal}$ is cancer risk through the dermal pathway of sediment; SA is the exposed skin surface area: $5700\ cm^2$; AF is the adherence factor from the sediment to skin: $0.07\ mg\ cm^{-2}$; and ABS is the dermal absorption from the sediment: 0.13 . Due to different cancer risk ranges, the characteristics of cancer risk have been qualitatively categorised as $\leq 10^{-6}$ (very low risk), from $10^{-6} < to < 10^{-4}$ (low risk), from $\leq 10^{-4}$ to $< 10^{-3}$ (moderate risk), from $10^{-3} \leq to < 10^{-1}$ (high risk) and $\geq 10^{-1}$ (very high risk) (Man et al., 2013).

The results showed that $Cancer\ Risk_{ingest}$ for sediment samples from Station 3 of the Kim Kim River was between 10^{-6} and 10^{-4} , indicating low risk from this station, while for Stations 1 and 2 of the Kim Kim River and all stations of the Segget River, it was from $\leq 10^{-4}$ to $< 10^{-3}$ (Table 4), implying moderate risk. The results demonstrated that $Cancer\ Risk_{dermal}$ for sediments from Stations 2 and 3 of the Kim Kim River was between 10^{-6} and 10^{-4} , indicating low risk level (Table 4). The $Cancer\ Risk_{dermal}$ for Station 1 of the Kim Kim River and all stations of the Segget River were between 10^{-4} and 10^{-3} , indicating moderate risk. Therefore, probabilistic health risk to humans via ingestion and dermal pathways from sediments of the Kim Kim and Segget Rivers can be categorised as low-to-moderate risk. The highest cancer risk occurred near Station 1 of both the Kim Kim River and the Segget River, which were near the coast. Therefore, people who live and work on the shore would likely have a higher risk of getting cancer.

CONCLUSION

The data from this study elucidated the 16 EPA PAHs contamination features in surface sediments from the Kim Kim River and Segget River, Peninsular Malaysia. The total PAH concentrations ranged from 95.17 to 361.24 , with a mean value of $218.1\ ng\ g^{-1}\ dw$ in sediments from the Kim Kim River and 330.09 to 552.76 with a mean value of $437.5\ ng\ g^{-1}\ dw$ in sediments from the Segget River. The PAH concentrations can be classified as low to moderate in the Kim Kim River and moderate to the level of pollution in the Segget River. The higher concentrations of PAHs in the Segget River can be attributed to the heavy industrial activities and road traffic in Johor City and nearby areas. PAH molecular indices and HCA were applied to determine the possible sources of PAHs. The results indicated both petrogenic and pyrogenic input with significant dominance of pyrogenic PAHs. The concentrations of LMW, HMW and total PAHs were compared with SQGs to assess the potential ecological effects of the sedimentary PAHs in the study areas. The results indicated minimum adverse ecological effect from PAHs. The results of potential toxicity assessment including M-ERM-Q, total BaP_{eq}, and TEQ^{carc} implied low probability of ecological effects in the Kim Kim and Segget Rivers. The human health risk of PAH compounds in the Kim Kim and Segget Rivers through ingestion and dermal

contact of sediments was evaluated; the result was low-to-moderate risk from ingestion and dermal contact.

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