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Polycyclic aromatic hydrocarbons (PAHs): contamination level and risk assessment in urban areas, Kuala Lumpur, Malaysia

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ABSTRACT

We are reporting the distribution of 15 polycyclic aromatic hydrocarbons (PAHs) in roadside soils and road dusts of five industrial areas around Kuala Lumpur city, namely Chan Saw Lin, Kuchai, Segambut, Kepong-1, and Kepong-2 using GC-MS and assessing their toxicological risk. The total PAH concentrations of PAHs in roadside soils of the industrial areas ranged from 2804 ± 2645 μg/kg (Kepong-2) to 18384 ± 12324 μg/kg (Chan Saw Lin), while for the road dusts, it ranged from 3056 ± 1929 μg/kg (Kepong-2) to 13581 ± 5295 μg/kg (Segambut). High molecular weight PAHs dominated the distribution of PAHs in all sampling locations for roadside soils and in road dust except for road dust of the Segambut industrial areas. Result of isomeric pair ratios and PCA used for the identification of the PAHs sources, suggested that the contribution of petrogenic sources is greater than the pyrogenic sources in almost all studied locations. Total B[*a*]P_{eq} for roadside soils and road dusts of studied areas, recorded highest in Chan Saw Lin and lowest in Kepong-2. Cancer risks caused by a long-period exposure to the PAHs in roadside soils of studied areas are still considered very low.

Keywords: PAHs; Roadside soils; Road dusts; Risk assessment; Cancer risk

1. Introduction

Roughly, cancer has become the second highest cause of death in Malaysia for 2008 with 6.9% [1]. It is also the second most common cause of death in the United States of America with approximately 1,500 people dying of cancer every day [2]. Meanwhile, in the United Kingdom, the ratio of death caused by cancer is one for every four persons [3]. Cancer researchers estimated that as many as two in three cases of cancer (67%) are linked to some type of environmental factor, including pollution in air, water, and soil [4].

Polycyclic aromatic hydrocarbons (PAHs) are one of the environmental pollutants that have potential to cause cancer in animals and humans.

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PAHs have two or more aromatic rings causing it to have a stable molecular structure [5]. It shows that its photochemical decomposition rates and its biodegradation process contribute to the persistence of PAHs in the environment. PAHs are derived from incomplete combustion of both natural processes such as volcanic eruptions, forest burning, etc. and anthropogenic processes such as vehicle exhaust gases, coal combustion, incinerator waste, petroleum spillage, etc. [6]. The amount of PAHs released in the air increases with decreasing the efficiency of the combustion processes. According to Khan et al. [7], the reduced efficiency of burning processes increases the amount of PAHs released in the air. The most favorable temperatures for their production are between 660 and 740℃ [8]. Urban areas have become the supply region of PAHs in the environment due to its huge utilization of energy sources [9]. There are more than 160 PAHs identified in the environment, however, only 16 were listed as priority pollutants by the United States Environmental Protection Agency (US EPA) [7,10].

Vehicle exhausts releases gaseous and particulate matter and has become the main contributor of PAHs and other toxic substances such as benzene and metals [11]. The increased numbers of vehicles recorded every year is worrying as this will increase the release of toxic substances into the atmosphere, which will then be deposited into soils and water reservoirs with rainfall. In Malaysia, the number of vehicle registered has increased from 13.8 to 18.0 million between 2004 and 2008 [1], and the amount of rainfall recorded in Kuala Lumpur ranged from 136 to 424 mm and 110 to 537 mm in 2009 and 2010, respectively [12].

According to Pirjola et al. [13], resuspension of surface particle loading is an important source of road dust emission. However, the main mechanisms leading to road dust emissions are still unidentified. Other than heavy metals, PAHs are also known as the major contaminants in road dust [14]. The contaminations from PAHs in road dust are caused by incomplete pyrolysis of organic materials from varying urban sources [15,16]. Studies have shown that the traffic density and the rate of deposition influence the concentration of PAHs in street dust [17–19]. Previous studies have reported that the origin of PAHs in road dust were mainly from automobile exhaust [14,20], road pavement material or asphalt [14,21], tire rubber, and lubricant oil [21]. Similar to aerosols, road dust will be deposited onto the surface water by rain [22,23] bringing along the PAHs into the water reservoirs. The concentrations of PAHs in road dust vary with the distance of its sources [24]. According to Zhao et al. [25], PAHs in street dust particles can be washed into nearby water reservoirs and become one of PAHs important sources in sediments.

On the other hand, for the soil, atmospheric depositions are known to be the most common source of the pollution [24,26]. Contamination of PAHs are believed to be greater in urban areas than the rural, forest, and agricultural areas as the urban areas have normally a higher density of automobiles and most of the PAHs received are expected to accumulate in surface soil [27,28]. Previous studies have shown that approximately 95% of the PAHs released can be found in soils and only about 0.2% of it can be found in the air [15]. The soil system seems to be a suitable medium to study environmental pollution. It is an important long-term repository for the PAHs [29,30]. Moreover, studies have found that humans are greatly exposed to the PAHs through contact with soils rather than air or water [27,31].

According to He et al. [32], the properties of PAHs and the environmental medium play an important role in determining the destiny of PAHs in the environment. Meanwhile, location controls the distribution rather than size fractions and sampling times as suggested by Murakami et al. [33]. Other than urban areas, industrial areas are the locations which frequently have high concentration of PAHs [34]. PAHs in industrial areas are derived from the surrounding industries such as the coal tar production plants, coking plants, coal tarring facilities, municipal trash incinerators, etc. PAHs in industrial areas can also be found in other facilities, where petroleum and petroleum products are used or where coal, oil, wood, or cellulose was burned [26]. The contributions of vehicular traffic are also important for the PAHs in industrial areas. Thus, it is expected that industrial areas located in urban areas have greater contamination of PAHs rather than in any other part of the city as it receives PAHs from both urban and industrial activities.

PAHs can enter the human body through lungs while the air is inhaled, through drinking water and swallowing food, soil or dust particles that contain PAHs. Under normal conditions of environmental exposure, PAHs also could enter the human body when skin comes into contact with soil or dust that contains high levels of PAHs [35]. Studies have shown that low molecular weight (lighter) PAHs can be absorbed through the skin, but the absorption of high molecular weight (heavy) PAHs is quite limited [26,36]. Accumulations of PAHs in soils can also disrupt food chains and contaminate vegetables [37,38]. It also could become the source of PAHs contamination in the atmosphere and groundwater through leaching, evaporation, and migration processes [39,40]. Therefore, it is important to know how contamination of

PAHs in soils as well as the road dust takes place as it can help us to eliminate food production risks and to prevent the harmful effect of these contaminants.

To date in Malaysia, most studies have focused mostly on the levels of PAHs in river water, ground water, leachate [41], river sediments [42], natural lake [43], biomass smoke [44,45], soils [46–48], tarballs [49,50], tobacco smoke [51], coastal sediments [52], and atmospheric particles [46,53,54]. Since there are no previous data focusing on the concentrations of PAHs in the industrial areas of Kuala Lumpur, this study was aimed to determine the level of PAHs in road dusts and roadside soils to obtain information on the potential sources as well as to evaluate the risk of PAHs in the industrial areas of Kuala Lumpur, Malaysia. Moreover, the results of this study will provide essential reference information to setup the background value for PAHs in order to be compared with data obtained in future surveys.

2. Materials and methods

2.1. Site description

The Federal Territory of Kuala Lumpur is the capital city of Malaysia which covers only an area of 0.07% of the country; hence, its size is only 243 km^2 [1]. However, the population of the country has increased dramatically from 25.6 million in 2004 to 28.3 million in 2009 and 4.63% of the population lives in Kuala Lumpur [1]. The five areas marked as industrial areas in Kuala Lumpur by Kuala Lumpur City Hall in their land use map were chosen for this study (Fig. 1). It includes areas in Chan Sow Lin, Kuchai Lama, Segambut and two industrial areas in Kepong. These areas are among the largest industrial areas in Kuala Lumpur and are separated from one another. Table 1 shows the exact coordinates of the sampling locations and total extracted lipid (TEL) gained from the samples.

2.2. Chemicals

Standard mixture containing 16 PAHs (16 compounds specified in EPA Method 610, each at 100–2000 mg/L), consisting of naphthalene (Naph), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phen), anthracene (Ant), fluoranthene (Flt), Pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Crys), benzo[b]fluoranthene (BbF), benzo[k] fluoranthene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h] anthracene (DbA), benzo[ghi]perylene (BgP), and indeno[1,2,3-cd]pyrene (InP), and deuterated PAHs internal standards (IS) mixture (dichlorobenzene-d$_4$, naphthalene-d$_8$, acenaphthene-d$_{10}$, phenanthrene-d$_{10}$, chrysene-d$_{12}$, and perylene-d$_{12}$) with each compound at 2,000 mg L^{-1} were procured from Supelco (Bellefonte, PA, USA). All solvents (dichloromethane, ethyl acetate, hexane, and methanol) used for sample processing and analyses were of HPLC grade from Merck (Oslo, Norway). Water used for the analysis was high purity deionized water taken from the Mili-Q system.

2.3. Sampling and sample pretreatment

Generally, experimental steps in this study have been adopted from Omar et al. [46] with a few adjustments to suit present experimental situation. In this experiment, surface roadside soils (0–5 cm depth) were taken with a stainless steel soil auger, pooled, and homogenized to provide a composite sample. The uppermost plant cover, twigs, and stones were removed before it was kept in 500 mL glass bottles covered with aluminum foil and capped. Conversely, road dusts were collected using sweeping tools and stored in Methanol and DCM prewashed glass bottles which was also covered with aluminum foil before being capped. All samples were preserved at 4˚C till further processing. In the laboratory, the roadside soils and the road dusts were stored at −20˚C before freeze-drying overnight. Both types of samples were then sieved through a 300 μm mesh and left in a desiccator before extraction.

2.4. Sample extraction for PAHs

Four grams of roadside soils and road dust samples were used after coning and quartering process. The samples were spiked with deuterated IS and extracted three times by ultrasonicator for a 15-min period each with 35 mL of dichloromethane in a cool water bath (10–15˚C). The sonication method is a method developed and recommended by various authors [55–57]. Filtered extracts were then shaken overnight with activated copper wire to eliminate sulfur. The extracts were then concentrated to about 2.0 mL using rotary centrifuge. The volumes of the extracts were then altered to accurately 2.0 mL by adding dichloromethane. Five hundred microliter of the extracts were used in a fractionation step, while another 1 mL of the extracts were shifted to a preweighted vial and left to dry before determining the TEL of the samples.

The extracts were fractionated using silica–alumina column chromatography. A glass column (20 cm × 1.0 m i.d.) was used with 5 g of partially deactivated silica gel (Merck, Pharmprep 60 CC, 40–63 μm) and 10 g of neutral alumina (on top) (Merck, 70–230 mesh).

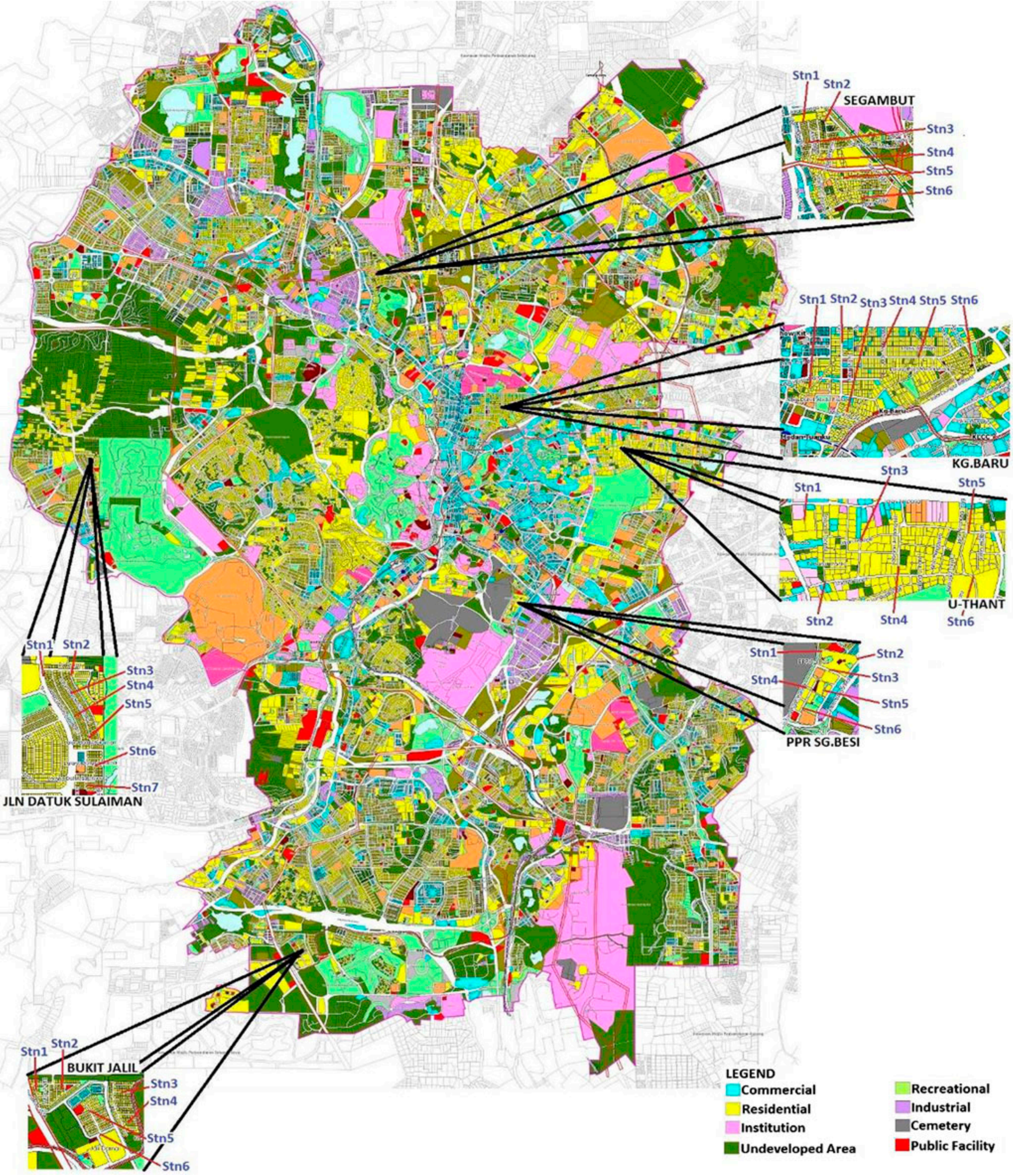

Fig. 1. Study areas and sampling locations in Chan Saw Lin (CSL), Kuchai, Segambut, Kepong-1, and Kepong-2 industrial areas of Kuala Lumpur City, Malaysia (Land use map was provided by Kuala Lumpur City Hall, Ref: DBKL/JPI/BPPM/PS/Ogos09/WPKI).

Table 1
Coordinates and concentration of extracted lipid from road dusts and roadside soils of industrial areas of Kuala Lumpur City

				TEL (mg/g)	
Station		Coordinate		Road dust	Roadside soil
Chan Saw Lin	Station 1	N03°07.533´	E101°42.870´	2.43	2.04
	Station 2	N03°07.290´	E101°.42.709´	6.57	14.5
	Station 3	N03°07.126´	E101°42.582´	13.7	0.72
	Station 4	N03°06.886´	E101°42.735´	7.02	22.5
	Station 5	N03°07.004´	E101°42.827´	2.87	6.52
	Station 6	N03°07.310´	E101°42.575´	9.27	21.0
Kuchai	Station 1	N03°05.583´	E101°41.650´	9.67	0.50
	Station 2	N03°05.617´	E101°41.577´	8.25	0.55
	Station 3	N03°05.592´	E101°41.595´	41.1	0.55
	Station 4	N03°05.557´	E101°41.517´	9.84	1.05
	Station 5	N03°05.407´	E101°41.567´	6.97	1.99
	Station 6	N03°05.358´	E101°41.589´	6.80	1.71
	Station 7	N03°05.316´	E101°41.596´	15.6	0.99
Segambut	Station 1	N03°11.420´	E101°40.628´	9.60	5.47
	Station 2	N03°11.280´	E101°40.741´	2,889	11.0
	Station 3	N03°11.168´	E101°40.510´	10.1	7.46
	Station 4	N03°11.346´	E101°40.375´	9.69	2.27
	Station 5	N03°11.060´	E101°40.588´	9.25	6.80
	Station 6	N03°11.006´	E101°40.408´	10.1	3.10
Kepong 1	Station 1	N03°12.424´	E101°39.138´	8.14	4.54
	Station 2	N03°12.330´	E101°39.262´	4.31	2.71
	Station 3	N03°12.188´	E101°39.187´	6.74	0.72
	Station 4	N03°12.258´	E101°39.016´	2.32	6.63
	Station 5	N03°12.378´	E101°39.020´	7.29	0.44
	Station 6	N03°12.301´	E101°39.141´	6.35	3.71
Kepong 2	Station 1	N03°11.688´	E101°38.499´	1.94	3.82
	Station 2	N03°11.729´	E101°38.597´	4.37	0.66
	Station 3	N03°11.768´	E101°38.660´	2.71	0.44
	Station 4	Nil	Nil	6.64	1.44
	Station 5	Nil	Nil	5.53	3.15
	Station 6	Nil	Nil	2.21	8.24

Before use, both silica gel and alumina were activated at 200°C for 4 h and deactivated with 5% of deionized water. The narrow end of the column was plugged with precleaned glass wool, while the top of the column was capped with 1.0 g of anhydrous sodium sulfate. After placing the extracts on the top of the column, the column was first eluted with 14 mL of hexane and the elution was discarded. The second fraction, rich with PAHs were then obtained by eluting the extracts with 30 mL of 10% dichloromethane in hexane followed by 20 mL of 50% dichloromethane in hexane. This fraction was concentrated to 200 μL using a rotary evaporator at 40°C under reduced pressure, and transferred into a 2 mL Teflon-lined vial for gas chromatography–mass spectra (GC-MS) analysis.

2.5. Instrumental analysis

The determination of PAHs was performed on a GC-MS QP2010 Plus instrument (Shimadzu, Japan) equipped with RTX-5MS (Crossbond 5% diphenyl/ 95% dimethyl polysiloxane) column (30 m × 0.25 mm i.d., 0.25 μm film thickness). The carrier gas was helium with flow rate at 1.20 mL/min. The oven temperature program was as follows: initial temperature at 70°C was held for 2 min, increased at a rate of 30°C/min to 150°C, and then increased at a rate of 4°C/min to 310°C before being held isothermal for 10 min. The injection port was set at 300°C. One microliter of sample extract was injected in the splitless mode. The ionization was carried out in the electron impact mode at 70 eV and the data were acquired under selected ion monitoring mode. Identification of PAHs was based on the selected ions and the comparison of retention time between samples and the standard solution containing individual PAHs. Quantitative determination of individual PAHs was performed using the most abundant ion. Table 2 shows the group list of PAHs, the quantification ion, and the confirmation ions for each PAH identified and quantified.

2.6. Quality assurance

All the analytical data were subjected to strict quality control. The procedural blanks, spiked blanks, sample duplicates were routinely analyzed with samples and all data were blank corrected. Limit of detection (LOD) was calculated as three times the noise level of the chromatogram in blank sample. LODs of PAHs were in the range of 0.06–0.37 g kg^{-1} dry weights. The surrogate recoveries were determined by spiking the samples with five surrogate standards (naphthalene-d8, acenaphthene-d10, phenanthrene-d10, chrysene-d12, and perylene-d12) and the surrogate recoveries for the roadside soils were 63 ± 20% for acenaphthene-d10, 76 ± 21% for phenanthrene-d10, 60 ± 10% for chrysene-d12, and 76 ± 17% for perylene-d12; while the surrogate recoveries for the road dusts were 80 ± 25% for acenaphthene-d10, 88 ± 20% for phenanthrene-d10, 89 ± 254% for chrysene-d12, and 79 ± 27% for perylene-d12. Surrogate recovery for naphthalene was eliminated as the results in both roadside soils and road dusts were too low. The method recoveries were between 40 to 116% for roadside soil samples, and between 57 to 100% for the road dust samples; and the method detection limits ranging from 1—19 μg/kg dry weight for the roadside soils and 3—15 μg/kg dry weight for the road dusts.

Table 2
List of groups of PAHs formed for analysis, the quantification ion, and the confirmation ions for SIM GC-MS mode in PAHs analyses

Group	Chemical	Time interval (min)	Quantification ion	Confirmation ions
1	Naphthalene	4.00–7.00	128	102, 125, 127, 129
	Naphthalene-d8		136	136
2	Acenaphthalene	7.00–9.50	152	125, 151, 153
	Acenaphtene		154	125, 152, 153
	Acenaphtene-d10		164	164
3	Fluorene	9.50–12.0	166	139, 163, 165
4	Phenanthrene	12.0–16.0	178	152, 176, 179
	Anthracene		178	152, 176, 179
	Phenanthrene-d10		188	188
5	Fluoranthene	16.0–25.0	202	200, 201, 203
	Pyrene		202	200, 201, 203
6	Chrysene	25.0–30.0	228	200, 227, 229
	Benzo(a)anthracene		228	202, 226, 229
	Chrysene-d12		240	240
7	Benzo(b)fluoranthene	30.0–37.0	252	125, 126, 250, 253
	Benzo(k)fluoranthene		252	125,126,250,253
	Benzo(a)pyrene		252	125,126,250,253
	Perylene-d12		264	264
8	Indene(1,2,3-c,d)pyrene	37.0–45.0	276	138, 274, 277
	Dibenzo(a,h)anthracene		278	139, 276, 279
	Benzo(g,h,i)perylene		276	138, 274, 277

Industrial soil Certified Reference Material No. 524 (Sample Identification No. 309) from the European Commission, Community Bureau of Reference was also analyzed and the results fall in good agreement with certified values.

3. Result and discussion

3.1. PAHs distributions

3.1.1. PAHs distributions in roadside soils

Table 3(a) shows the summary of PAHs in roadside soils of the industrial areas studied and the corresponding average values. Chan Saw Lin, Segambut, Kepong-1, and Kepong-2 industrial areas were represented by six sampling sites each, while Kuchai industrial area had seven sampling sites. The highest total PAHs concentration detected was in Chan Saw Lin industrial area (18,384 ± 12,324 μg/kg), while the lowest value was detected in Kepong-2 industrial area (2,804 ± 2,645 μg/kg). The standard concentrations for PAHs are very limited worldwide and in Malaysia, there are still no standards limits of PAHs available. It is known that other than industrial emissions, PAHs contributed the most by vehicle emissions [11]. It was observed that among the studied industrial areas, Chan Saw Lin, Segambut, and Kuchai industrial areas do have higher traffic densities compared with Kepong-1 and Kepong-2 industrial areas. PAHs in Chan Saw Lin industrial area might be due to high vehicle densities using Tun Razak Street which is located adjacent to the area. Tun Razak Street is a major road in Kuala Lumpur and it skirts the western part of the inner city.

In terms of individual PAHs, InP has been found to be the most abundant in Chan Saw Lin, Segambut, and Kepong-1 at 4,616, 1,527, and 751 μg/kg, respectively. Meanwhile, phenanthrene has been found to be the most abundant individual PAH in roadside soils of the Kuchai industrial area with 1,336 μg/kg, while Kepong-2 has pyrene as the most abundant individual PAH in its industrial area (448 μg/kg). Even though roadside soils of Kuchai and Kepong-2 industrial areas were not dominated by the same individual PAHs as others, InP were their second most abundant individual PAH. According to Dallarosa et al. [58] and Guo et al. [59], the abundance of InP indicate that there is a strong contribution of traffic emissions and associated emissions from incomplete motor combustion as in other high molecular weight PAHs. On the whole, more than half of the individual PAH compounds in soil samples were higher than the 100 μg/kg limit set by polish regulations [60].

3.1.2. PAHs distributions in road dusts

The summary of PAHs in road dusts of studied areas is shown in Table 3(b). The concentrations ranged from 13,581 ± 5,295 μg/kg in Segambut to 3,056 ± 1,929 μg/kg in Kepong-2. The highest PAHs concentration in road dusts was recorded in Segambut which was different from the highest PAH recorded in roadside soils in Chan Saw Lin. This might be due to a few reasons such as, (1) PAHs sources such as weathered materials of road surfaces, automobile exhaust, lubricating oils, gasoline, diesel fuel, tire particles, construction materials, and atmospherically deposited materials in Segambut play a greater role as the source of PAHs in road dusts rather than the soils, (2) the Segambut industrial area received greater foreign road dusts than other industrial areas as the road dusts were subjected to transportation from one place to another [61,62]. However, this might not remain for long as road dust. This is because road dusts are subjected to evaporation which might bring the PAHs into the atmosphere [63,64], and/or be transported to other places by the wind, and/or involved in runoff process which might remove the road dusts together with the PAHs into the water reservoir during rainfall [65–67].

As for the individual PAH observed in road dust of the studied industrial areas, InP was also the individual PAHs that contributed the most in road dusts of Chan Saw Lin and Kuchai industrial areas with 2,296 and 1,238 μg/kg, respectively. However, the situation was different in Segambut, Kepong-1, and Kepong-2. In Segambut, the highest individual PAHs was contributed by acenaphthalene (5,331 μg/kg); while in Kepong-1 and Kepong-2, the highest concentration of individual PAHs was contributed by phenanthrene and pyrene with 592 and 612 μg/kg, respectively.

3.1.3. Comparison between total distribution of PAHs in roadside soils and road dusts of studied areas and other studies worldwide and locally

Table 4(a) shows the concentrations of PAHs detected in industrial soil and industrial road dust worldwide, while Table 4(b) shows the concentrations of PAHs in soil and road dust studied locally. It is shown from the table that PAHs in soils of studied areas are high compared with other studies worldwide, while the concentrations of PAHs detected in road dust was intermediate. Comparison with other data within Malaysia shows that PAHs in roadside soil of studied areas were quite high, but still lower than the highest value recorded in urban Kota Bharu,

Table 3a
Concentrations of PAHs in roadside soils of industrial areas around Kuala Lumpur City

Compound	Chan Saw Lin (µg/kg dw, $n = 6$)			Kuchai (µg/kg dw, $n = 7$)			Segambut (µg/kg dw, $n = 6$)			Kepong-1 (µg/kg dw, $n = 6$)			Kepong-2 (µg/kg dw, $n = 6$)			Total ± SD
	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	
Acy	1,867 ± 1,258	329	3,317	62.9 ± 51.8	0.00	128	678 ± 226	315	947	151 ± 40.3	104	202	141 ± 34.7	110.6	202	2,909.02 ± 760.10
Ace	25.3 ± 29.0	0.00	64.5	7.25 ± 7.01	0.00	15.5	63.4 ± 22.4	39.9	101	17.8 ± 15.2	0.00	36.7	30.3 ± 8.81	23.0	45.1	144.04 ± 21.20
Flu	80.2 ± 56.6	31.9	166	48.8 ± 55.4	0.00	131	111 ± 68.4	46.1	197	59.4 ± 41.6	10.6	114	68.1 ± 35.8	36.0	123	368.25 ± 24.23
Phen	707 ± 400	114	1,287	1,336 ± 1946	70.4	5,519	592 ± 187	330	806	383 ± 208	159	659	393 ± 228	191	726	3,413.95 ± 390.11
Ant	281 ± 267	0.00	600	124 ± 87.8	6.61	288	224 ± 164	65.4	520	114 ± 85.6	32.1	241	80.8 ± 57.2	35.5	183	825.53 ± 84.25
Flt	1,250 ± 1,322	102	3,829	554 ± 647	62.6	1951	263 ± 149	77.0	446	295 ± 246	64.9	606	299 ± 385	43.3	1,056	2,663.67 ± 418.03
Pyr	2,401 ± 2,460	374	6,926	343 ± 278	36.3	875	464 ± 321	118	889	374 ± 301	59.0	805	448 ± 509	47.1	1,424	4,033.12 ± 893.05
BaA	613 ± 826	113	2,264	123 ± 126	10.8	343	143 ± 118	35.3	304	118 ± 103	7.75	268	97.4 ± 149	20.2	402	1,096.70 ± 221.14
Crys	2,248 ± 1,529	390	4,546	352 ± 300	63.7	827	523 ± 418	140	1,125	364 ± 272	23.6	645	333 ± 362	39.6	957	3,822.57 ± 832.86
BbF	1,733 ± 1856	145	5,264	397 ± 405	36.9	1,064	431 ± 367	121	1,050	303 ± 286	27.2	731	265 ± 422	38.9	1,124	3,132.26 ± 622.71
BkF	1,007 ± 643	131	1,686	166 ± 136	17.1	312	277 ± 369	44.4	1,019	151 ± 131	11.7	312	80.9 ± 112	9.22	306	1,684.62 ± 381.55
BaP	690 ± 788	54.0	2,232	114 ± 103	9.38	280	174 ± 183	28.8	516	100 ± 100	0.00	257	80.4 ± 130	1.80	345	1,160.85 ± 258.85
DbA	627 ± 595	79.2	1,704	87.2 ± 79.8	0.00	180	179 ± 220	0.00	570	85.9 ± 76.5	0.00	167	40.8 ± 61.6	0.00	158	1,020.46 ± 241.99
BgP	230 ± 281	17.8	752	31.1 ± 27.5	0.00	65.2	35.1 ± 57.7	0.00	135	18.5 ± 20.3	0.00	39.1	0 ± 0.00	0.00	0.00	315.31 ± 94.62
InP	4,616 ± 3,460	1,098	9,116	888 ± 959	40.6	2,314	1,527 ± 1,327	146	3,801	751 ± 664	73.3	1,760	444 ± 391	26.2	935	8,228.18 ± 1,707.13
Total ± SD	18,384 ± 12,324			4,639 ± 3,012			5,690 ± 3,938			3,292 ± 2,366			2,804 ± 2,645			34,811.53 ± 6,485.04

Table 3b
Concentrations of PAHs in road dusts of industrial areas around Kuala Lumpur City

	Chan Saw Lin (µg/kg dw, *n* = 6)			Kuchai (µg/kg dw, *n* = 7)			Segambut (µg/kg dw, *n* = 6)			Kepong-1 (µg/kg dw, *n* = 6)			Kepong-2 (µg/kg dw, *n* = 6)			
Compound	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Total ± SD
Acy	1,092 ± 1,059	358	3,155	122 ± 102	47.3	273	5,331 ± 3,236	2,818	10,681	303 ± 190	154	668	135 ± 52.9	93.2	238	6,985.62 ± 2,235.50
Ace	62.2 ± 33.0	19.4	111	65.6 ± 45.0	33.2	151	765 ± 451	394	1,532	30.2 ± 24.2	0.00	70.8	32.3 ± 7.58	21.7	42.5	955.61 ± 321.42
Flu	62.0 ± 36.7	26.1	118	73.4 ± 58.7	28.2	170	527 ± 286	280	1,014	57.6 ± 36.3	9.32	107	74.5 ± 27.9	47.7	124	794.73 ± 205.99
Phen	652 ± 569	221	1,732	773 ± 516	299.36	1,461	1,251 ± 302	758	1,625	592 ± 392	325	1,364	478 ± 321	256	1,114	3,749.35 ± 300.01
Ant	218 ± 241	58.2	691	159 ± 128	52.66	391	539 ± 212	295	848	158 ± 133	67.4	424	81.4 ± 56.6	37.8	190	1,156.88 ± 179.17
Flt	883 ± 1,031	193	2,913	521 ± 354	170.72	1,234	644 ± 233	246	875	481 ± 404	228	1,292	330 ± 244	108	779	2,861.07 ± 206.71
Pyr	1,124 ± 1,437	206	3,994	719 ± 591	155.92	1,954	916 ± 327	335	1,306	536 ± 449	191	1,405	612 ± 378	240	1,314	3,909.47 ± 239.214
BaA	244 ± 294	31.9	818	137 ± 130	28.82	418	175 ± 94.0	53.5	324	114 ± 125	29.3	356	104 ± 87.3	22.0	227	776.82 ± 56.86
Crys	1,490 ± 1,457	345	4,265	906 ± 774	273.31	2,530	1,046 ± 485	382	1,809	552 ± 525	24.1	1,541	452 ± 307	20.6	893	4,447.73 ± 415.56
BbF	700 ± 751	137	2,184	372 ± 358	90.22	1,126	467 ± 200	214	729	297 ± 305	118	916	192 ± 160	67.3	428	2,030.14 ± 192.76
BkF	504 ± 572	70.0	1,638	279 ± 238	88.10	751	254 ± 103	157	427	133 ± 117	41.4	349	88.2 ± 94.7	8.06	238	1,260.81 ± 162.36
BaP	592 ± 939	98.4	2,478	124 ± 126	26.84	400	151 ± 81.7	61.4	270	71.5 ± 75.9	19.1	223	85.6 ± 91.1	15.5	265	1,025.97 ± 218.65
DbA	190 ± 209	38.7	606	80.5 ± 80.3	17.97	234	113 ± 79.0	41.6	231	39.3 ± 36.7	14.3	109	37.8 ± 54.8	0.00	147	461.52 ± 63.31
BgP	59.5 ± 97.2	0.00	253	40.9 ± 45.4	0.00	116	35.5 ± 22.6	17.0	77.6	13.1 ± 21.3	0.00	49.5	5.45 ± 10.7	0.00	26.7	154.36 ± 21.81
InP	2,296 ± 2,969	282	8,242	1,238 ± 1,154	265.62	3,412	1,360 ± 904	524	2,787	396 ± 303	87.7	883	344 ± 262	93.4	840	5,636.17 ± 803.33
Total ± SD	10,174 ± 11,596			5,615 ± 4,396			13,581 ± 5,295			3,777 ± 2,968			3,056 ± 1929			36,206.25 ± 4,498.80

Table 4a
Comparison between PAHs in road dusts and roadside soils of studied areas and industrial areas studied worldwide

Industrial soil		Industrial road dust	
Location	Concentration (μg/kg dw)	Location	Concentration (μg/kg dw)
Angren, Uzbekistan [92]	118–5,913	Taichung industrial park, Taiwan [96]	26,700
El Paso, Texas, USA [93]	0.1–2,226	Industrial site, Lahore, Pakistan [97]	200
Seine River basin, France [94]	450–5,650	Ulsan (industrial city, Korea) [75]	11,840–245,120
Taragona County, Spain [95]	166–1,002	Shanghai industrial area [98]	14,247 (summer) 31,163 (winter)
This study	18,384 ± 12,324–2,804 ± 2,645	This study	13,581 ± 5,295–3,056 ± 1929

Table 4b
Comparison between PAHs in road dusts and roadside soils of studied areas and studied around Malaysia

Soil		Road dust	
Location	Concentration (μg/kg dw)	Location	Concentration (μg/kg dw)
Urban Kuala Lumpur [46]	224 ± 108	Kuala Lumpur [50]	1,080–4,550
Urban Kemamam [99]	6.30–176		
Urban Kota Bharu [48]	22–24,060		

Kelantan. Meanwhile, PAHs detected in road dust of studied areas was recorded higher than available data in urban Kuala Lumpur.

3.2. PAHs profile

3.2.1. PAHs pattern in roadside soils

Fig. 2(a) shows the composition of PAHs in roadside soils based on their number of rings in studied areas. Roadside soils in Chan Saw Lin industrial area was dominated by four rings PAHs with 35.4%, followed by six rings PAHs (26.4%), five rings PAHs (22.1%), and three rings PAHs (16.1%). On the other hand, in Kuchai roadside soils, three rings PAHs dominated with 34.0% followed by four rings PAHs (29.6%), six rings PAHs (19.8%), while the lowest was five rings PAHs with 16.5%. In Segambut, the major contributor of PAHs came from three rings PAHs with 29.4%, followed by six rings PAHs with 27.5%, four rings PAHs with 24.5%, and five rings PAHs with 18.7%. Kepong-1 industrial area was dominated by four rings PAHs (35.0%), six rings PAHs (23.4%), three rings PAHs (22.1%), and five rings PAHs (19.5%). On the other hand, four rings PAHs was found to be the most abundant PAHs in Kepong-2 with 42.0%. The concentrations of the PAHs were also contributed by three rings PAHs with 25.5%, four rings PAHs with 16.7%, and six rings PAHs with 15.8% from the concentrations. In general, the distribution of PAHs based on their number of rings are diverse from one sampling locations to another.

Moreover, the ratio between high molecular weight (HMW) PAHs to the low molecular weight (LMW) PAHs in roadside soils of all sampling locations are shown in Fig. 2(b). HMW PAHs were dominating the concentrations of PAHs in all sampling locations with 83.9% in Chan Saw Lin, 66.0% in Kuchai, 70.7% in Segambut, 77.9% in Kepong-1, and 74.5% in Kepong-2. These results suggest that all the studied industrial areas have been contaminated by the deposition of PAHs from vehicular emission [68]. Furthermore, these results also suggest that the PAHs sources in studied roadside soils came from the surrounding areas as the rise in molecular weight will reduce the ability of the PAHs to travel [37,69].

3.2.2. PAHs pattern in road dusts

Fig. 3(a) shows PAHs percentage based on the number of rings in road dusts of studied industrial areas. The summary of the results were as follows; Chan Saw Lin: four rings (36.8%) > six rings (23.2% > three rings (20.5%) > five rings (19.5%), Kuchai: four rings (40.7%) > six rings (22.8%) > three rings (21.3%) > five rings (15.3%), Segambut: three rings (62.0%) > four rings (20.5%) > six rings (10.3%) > five rings (7.27%), Kepong-1: four rings (44.6%) > three rings (30.2%) >

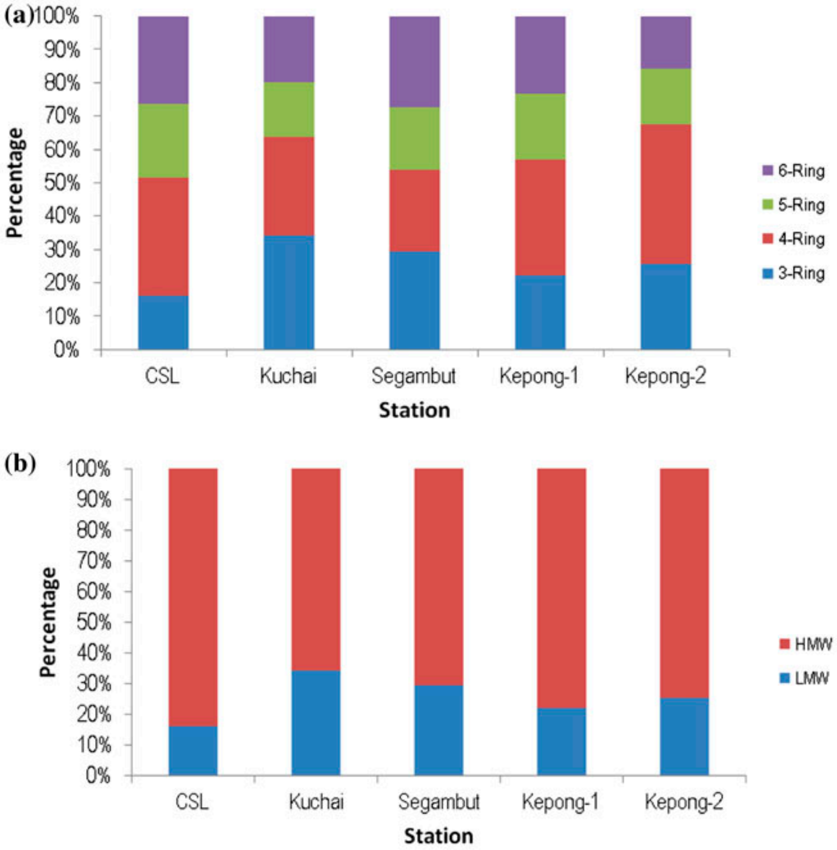

Fig. 2. (a) Composition of PAHs in roadside soils based on their number of rings in studied areas; (b) ratio between HMW PAHs and the LMW PAHs in roadside soils of studied areas.

five rings (14.4%) > six rings (10.9%), Kepong-2: four rings (49.1%) > three rings (26.2%) > five rings (13.2%) > six rings (11.4%). Unlike the distributions of PAHs in roadside soils, PAHs concentrations in road dusts of studied areas shows the major contribution by four rings PAHs except for Segambut which was mainly contributed by three rings PAHs.

In addition, Fig. 3(b) shows the graph of HMW PAHs in road dusts of studied areas over the LMW PAHs. HMW PAHs dominating Chan Saw Lin by 79.5%, Kuchai by 78.7%, Kepong-1 by 69.8%, and Kepong-2 by 73.8%. Only Segambut were dominated by LMW PAHs with 62.0%. The situation in Segambut industrial area suggests that long-range atmospheric transport was the biggest source of PAHs in that area [70,71].

3.3. Relationship between PAHs in roadside soils and road dusts

The relationship of PAHs in roadside soils and road dusts shows that the relationships were variable between locations (Fig. 4). PAHs in roadside soils and road dusts of Chan Saw Lin ($r = 0.60$) and Kepong-1 ($r = 0.55$) areas were at marked, moderate relationship while the relationship in Segambut were low ($r = 0.27$) and almost negligible in Kuchai ($r = 0.18$) and Kepong-2 ($r = 0.10$). Overall, the results shows that the relationships between PAHs in roadside soils and road dusts can be neglected in PAHs analysis as there were no strong bond between them.

3.4. Sources of PAHs

PAHs can be derived from natural and anthropogenic sources. Examples of natural sources of PAHs are forest fires and natural petroleum seepage. However, most of the sources of PAHs in our environment came from anthropogenic sources which can be divided into two: pyrogenic (combustion of biomass and fossil fuels) and petrogenic (input of petroleum products and crude oil).

Abundance of PAHs with certain number of rings can give some general description on their sources.

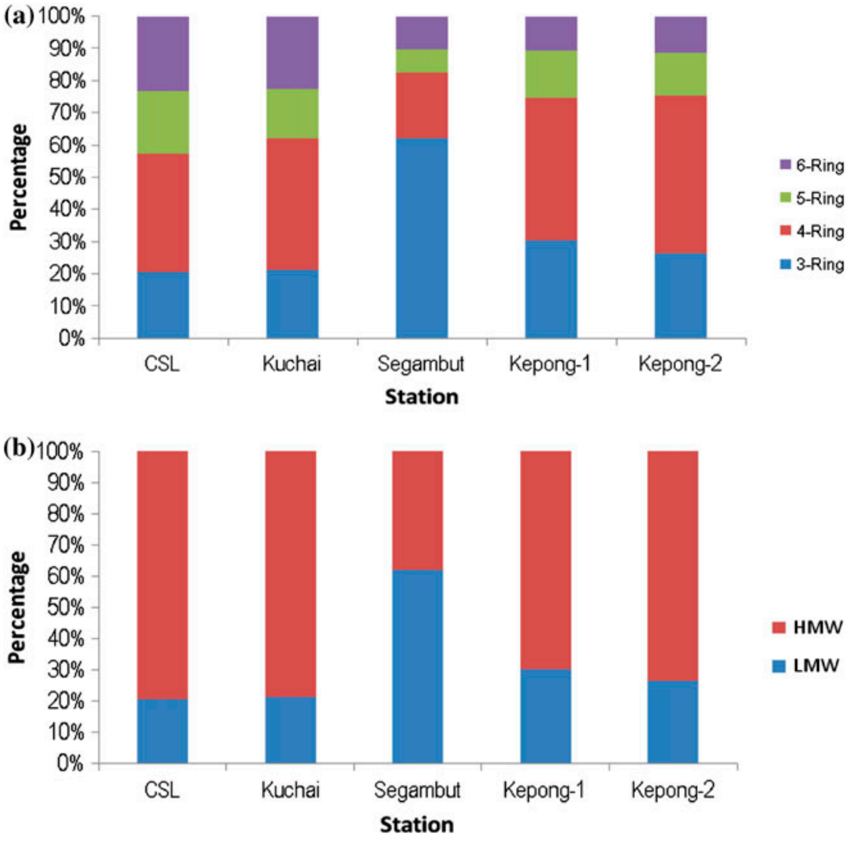

Fig. 3. (a) Composition of PAHs in road dusts based on their number of rings in studied areas; (b) ratio between HMW PAHs and the LMW PAHs in road dusts of studied areas.

PAHs rich with three and four rings are known to originate from petroleum residues [72], while PAHs rich in higher rings are derived from combustion process [73,74]. From the results, since the amount of three and four rings PAHs in roadside soils of studied areas were greater or equal to 50% of total amount of concentrations, it is assumed that the roadside soils received about equal amount of pyrogenic sources of PAHs and petrogenic sources of PAHs (Chan Saw Lin and Segambut) or received greater amount of petrogenic sources PAHs than the pyrogenic PAHs sources (Kuchai, Kepong-1 and Kepong-2). Meanwhile, all samples of road dusts were shown to have high amount of three and four rings PAHs (>50%), which suggest high contribution of petrogenic PAHs. High amounts of petrogenic PAHs in all sampling stations might have come from petroleum residues from workshops around all sampling locations.

Ratios of individual PAHs can also be used to elucidate sources of PAHs. Fig. 5(a) shows the ratios between Fl/Pyr over Phen/Ant to estimate the possible sources of PAHs. Value of Fl/Pyr lower than one suggests that the PAHs came from petrogenic sources, while value higher than one suggests that the PAHs came from the pyrogenic activities [75]. Conversely, the value of Phen/Ant less than 10 suggests that the PAHs came from combustion processes, while values higher than 10 implies PAHs that came from diagenetic process or from a petroleum input [76]. PAHs sources can be understood better by studying the graph of InP/(InP + BgP) against Fl/(Fl + Pyr) as in Fig. 5(b), which can be used to describe the transition point of PAHs in studied areas. Value of Fl/(Fl + Pyr) lower than 0.4 indicate that the PAHs originated from petroleum pollutants. Moreover, value at 4.0 indicates that the PAHs are in the transition of petroleum/combustion process, while value greater than 0.5 indicate that the PAHs originated from wood or coal combustion. Furthermore, the value from 0.4 to 5.0 indicates that the PAHs came from the process of liquid fossil fuel combustion, such as combustion from vehicle and crude oil. On the other hand, the value of InP/(InP + BgP) ratio lower than 0.2 indicate that the PAHs

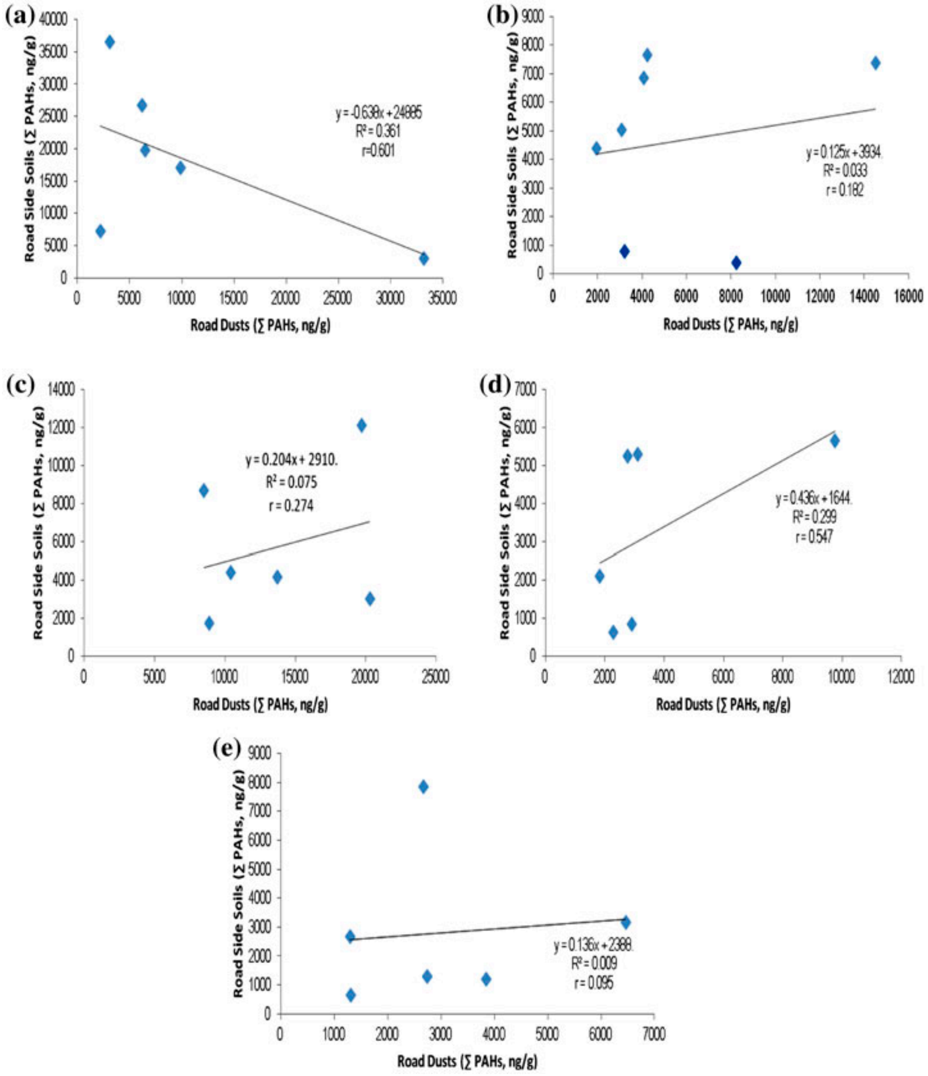

Fig. 4. Graphs of relationship between roadside soils and road dusts of studied industrial areas: (a) Chan Saw Lin, (b) Kuchai, (c) Segambut, (d) Kepong-1, and (e) Kepong-2.

originated from a contribution of petroleum input/discharge. The value greater than 0.5 shows that the PAHs was from wood and coal combustion, while value in between 0.2 and 0.5 suggest that the PAHs were contributed from petrochemical fuel combustion [77,78].

Based on Fig. 5(a) and (b), the values of Fl/Pyr for road dust samples and most of the roadside soil samples were lower than 1. The ratios of Phen/Ant were higher than 10 for all road dust samples and almost all of roadside soil samples. The Fl/(Fl + Pyr) ratios for road dust samples ranging from 0.35 to 0.47, while in roadside soil samples it is ranging from 0.34 to 0.62. The ratios of InP/(InP + BgP) varied from 0.97 to 0.98 for road dust samples and from 0.95 to 1.00 for roadside soil samples. The above data shows that the PAHs in roadside soils and road dusts of industrial areas around Kuala Lumpur city are derived from all petroleum, petroleum combustion, and biomass and coal combustion.

3.5. Principal component analysis (PCA)

To further understand the sources of the PAHs, PCA was then performed. PCA is a multivariate diagnostic tool used to shrink a set of original variables and to dig out a small number of dormant factors to analyze the relationship among observed variables.

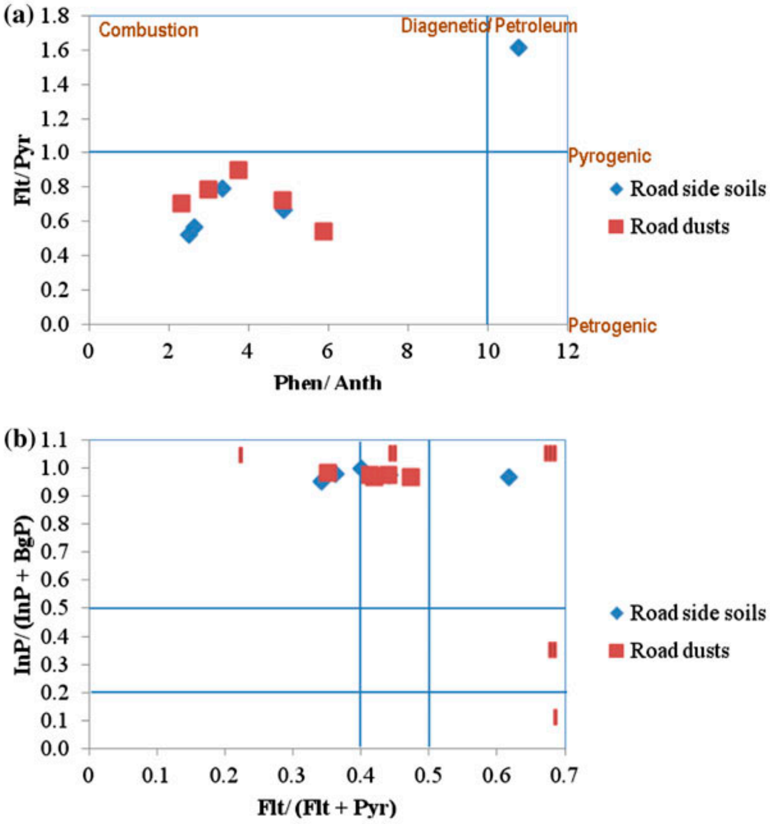

Fig. 5. (a) Plot of the isomeric ratios Phen/An vs. Fl/Pyr; (b) plot of the isomeric ratios Fl/(Fl + Pyr) vs. InP/(InP + BgP).

Fig. 6(a) and (b) indicate that variances for the first two principal components (PC1 and PC2) of the road dusts and roadside soils of studied areas accounted for 81% of the variances.

PC1 indicates 60% of the total variance. Higher loadings of fluoranthene (Flt), Pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Crys), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DbA), BgP, and InP were identified in this factor (Fig. 6(a)), which are common markers of pyrolysis or incomplete combustion. BaP and BgP are suitable to be used as a tracer of vehicle exhaust emission as they have been found to be in abundance in a traffic tunnel in the previous studies [66,79,80]. On the other hand, Flt, Pyr, BaA, Chrys, and BaP can be used as the markers of coal combustion [80,81]. The existence of BbF and BkF point out that there was a contribution of fossil fuel in the PAHs concentrations as these two components have been found to be the products of the process [83,84]. Meanwhile, the combustion of both diesel and natural gas result in the releases of BaA and Chry [83,85]. The existence of InP which is usually found in petroleum indicate the contribution of diesel vehicles [84], while DbA together with InP and BaP indicate traffic emission as the sources [86]. As for the benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), and benzo[a]pyrene (BaP), they are the main compounds in particulate samples of roadside atmosphere [66]. Overall, PC1 shows that pyrogenic source was the main sources of PAHs in road dusts and roadside soils as higher concentrations of PAHs have been observed in Chan Saw Lin (Fig. 6(b)) which has high traffic densities and a lot of petrogenic-related activities taking place. As for other sampling sites, it shows that their scores were almost similar which suggest that most of the sampling sites in the studied industrial areas were polluted by PAHs that came from the same sources.

PC2 with the total variance of 21% shows that PAHs in studied areas are positively dominated by LMW PAHs, such as Acy, Ace, Flu, Phen, and Ant. According to Jenkins et al. [87], pyrogenic activities in low temperature will release a significant amount of LMW PAHs in soil. Among LMW PAHs in the environment, it has been shown that Phen, Ant, and Flu

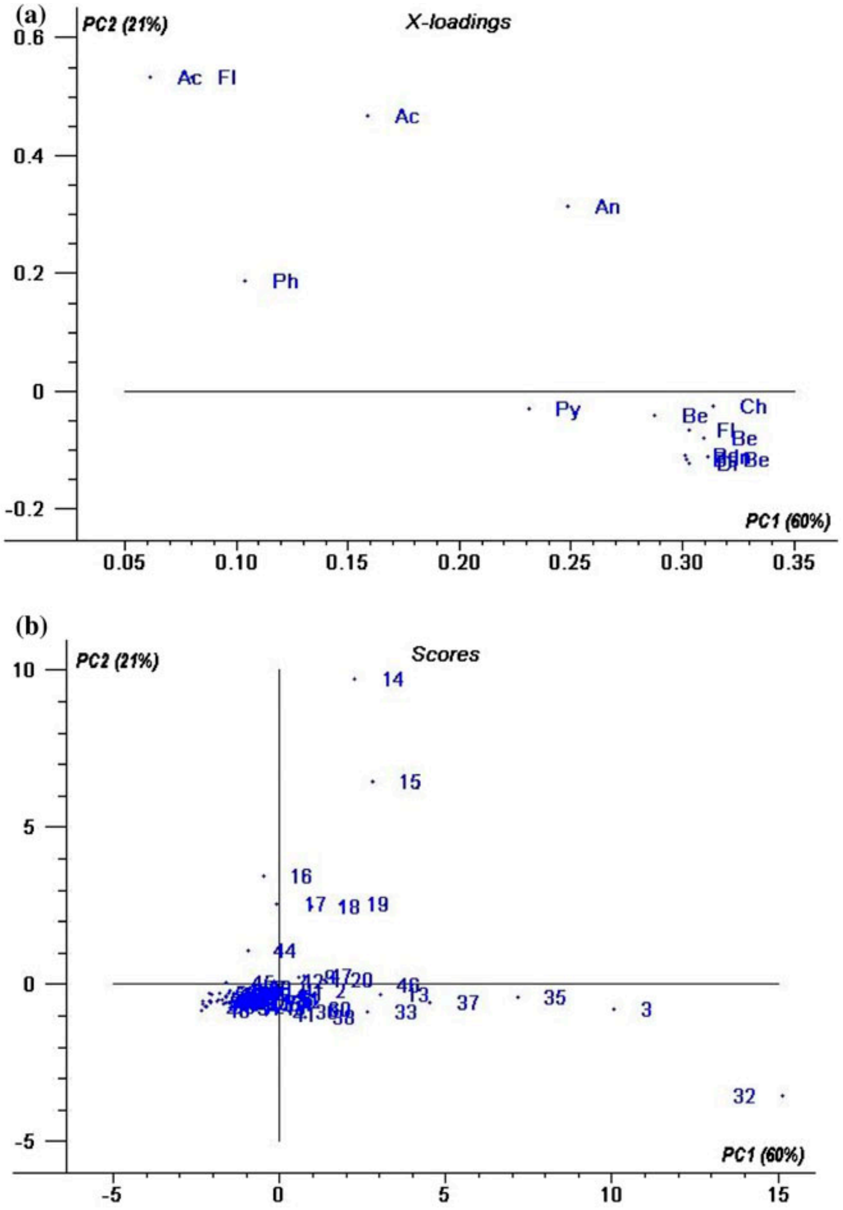

Fig. 6. Plots with PC1 and PC2 from PCA. (a) Factor loadings of 15 PAHs on PC1 and PC2, and (b) factor scores of 62 sampling sites on the PC1 and PC2. 1–6: Road Dust (RD), Chan Saw Lin; 7–13: RD, Kuchai; 14–19: RD, Segambut; 20–25: RD, Kepong-1; 26–31: Kepong-2; 32–37: RoadSide Soil (RSS), Chan Saw Lin; 38–44: RSS, Kuchai; 45–50: RSS, Segambut; 51–56: RSS, Kepong-1; 57–62: RSS, Kepong-2.

can be produced by coke oven [82]. Thus, PC2 represent the contribution of LMW PAHs in the industrial areas and high scores of these PC2 were found to be dominated in road dusts of Segambut.

3.6. Risk assessment

Humans are expected to be exposed to contaminated soil or dust mainly through accidental ingestion and inhalation. PAHs especially BaP has been proved to cause tumors to animals tested in the laboratory by breathing in the compounds, skin contact with the compounds, and eating the compounds. On the other hand, studies by ATSDR [26] had shown that long exposure of humans to the PAHs in any mixtures through inhalation and skin contact can also cause cancer. According to EPA, taking 0.30 mg of anthracene, 0.06 mg of acenaphthene, 0.04 mg of fluoranthene, 0.04 mg of fluorene, and 0.03 mg of pyrene per kg of our body weight daily won't cause any bad health effects [26,88] which might be due to human rapid metabolism.

The standard concentrations for PAHs are very limited worldwide and in Malaysia, there are still no standards limits of PAHs available. In this study, the total concentrations of PAHs recorded fall within the range of a few available standards, such as Mexican standards (0-6,000 μg/kg) and Polish standards (200–10,000 μg/kg). However, the concentrations recorded have exceeded the limits stated by the Dutch [27].

PAHs have also been detected in agricultural products [26] and it was believed that one of the main routes of the PAHs accumulation in the agricultural products was through nutrient uptake from the soil. The concentrations of PAHs in the studied areas are much higher than the typical concentrations found to be suitable in soils for agriculture which is 200 μg/kg [89]. Thus, the growing of vegetables or any other food products are believed to be not suitable in these areas as ingestion of PAHs-contaminated products may cause further PAHs accumulation in the human body.

3.6.1. Roadside soils and road dusts toxicity

Toxic equivalency factors (TEF) are often used in measuring toxicity of the environmental samples. TEF test were also used in this study. TEF used in this study were calculated to measure the carcinogenicity of PAHs relative to benzo[a]pyrene. According to Peters et al. [90], only benzo[a]pyrene toxicological data are sufficient to be used in measuring the carcinogenic potential of PAHs studied. According to Duke and Albert [91], TEFs value for acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, Pyrene, benzo[a]anthracene, chrysene, benzo [b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenzo[a,h]anthracene, BgP, and InP are 0.001, 0.001, 0.001, 0.001, 0.01, 0.001, 0.001, 0.1, 0.01, 0.1, 0.1, 1, 1, 0.01, and 0.1, respectively. Benzo[*a*]pyrene-equivalent concentration (B[*a*]Peq) was calculated as Eq. (1):

$$\text{Total B}[a]\text{P}_{\text{eq}} = \sum i \, C_{\text{i}} \times \text{TEF}_i \tag{1}$$

C_{i} = individual PAHs concentration; TEF_{i} = corresponding toxic equivalency factors [27].

From the results, total B[*a*]P_{eq} of roadside soils of studied areas are as follows: Chan Saw Lin (1,397 μg/kg) > Segambut (337 μg/kg) > Kuchai (239 μg/kg) > Kepong-1 (198 μg/kg) > Kepong-2 (138 μg/kg). Meanwhile, for the road dusts of studied areas, the results are as follows: Chan Saw Lin (859 μg/kg) > Segambut (327 μg/kg) > Kuchai (278 μg/kg) > Kepong-1(156 μg/kg) > Kepong-2 (143 μg/kg). The trends shown in roadside soils and road dusts were same, hence the highest carcinogenic potential is found in Chan Saw Lin and the lowest at Kepong-2.

3.6.2. Cancer risk assessment for the contaminated soil

Cancer risks for the workers in studied industrial areas were determined by using exposure models and risk factors developed by ATSDR and the US EPA. Only increased risk for PAHs in soil (through ingestion) can be determined as data related to cancer by PAHs in dust (through inhalation) are unavailable.

The formula of the oral ingestion cancer risk assessment is as Eq. (2):

$$\begin{aligned} &\text{Exposure dose (mg/kg/d)} \\ &\quad \times \text{ ingestion cancer slope factor (mg/kg/d)}^{-1}) \\ &\quad \times \text{ estimated years exposed/BW)} \\ &\quad = \text{ cancer risk (unitless)} \end{aligned} \tag{2}$$

Assumptions:

- Contaminant concentration is the sum of the PAH–TEF products and does not change from day to day. This is the maximum PAH concentration.
- Ingestion rate is 100 milligrams of soil per day.
- EF = exposure factor, exposure is assumed to be 2 d/wk every week of the year.
- 10–6 kg/mg is a necessary conversion factor for soil.
- The adult body weight is 70 kg.
- Estimated year exposed = 10 years.

where exposure dose (D)—(C × IR × EF × 10–6 kg/mg)/BW; C—contaminant concentration (mg/kg); IR—intake rate of contaminated soil (mg/d); EF—exposure factor = (2 d/wk × (52 wk/y) × (365 d/y) = 0.28; BW—body weight (kg); and EPA ingestion cancer slope factor—7.30 mg/kg/d^{-1} [88].

The results of the assessment shows that cancer risk for the workers in Chan Saw Lin is about 4×10^{-7} or 4 in 100,000,000 people; while in Kuchai and Segambut, the risk is at 1×10^{-7} or 1 in 100,000,000 people. Meanwhile, the cancer risk in Kepong-1 and Kepong-2 are 7×10^{-8} and 6×10^{-8} or 7 and 6 in 1,000,000,000 people, respectively. From the results, it is shown that increased cancer risk in the studied industrial areas can still be considered "extremely low."

4. Conclusion

In conclusion, the concentrations of PAHs in these studied industrial areas are high in comparison to a few guidelines established worldwide and also as noted from the literature. Any food-related plantation activities in these areas are not recommended. Contribution of HMW PAHs is higher than LMW PAHs in both roadside soils and the road dusts except for road dust in Segambut, which support the fact that these industrial areas were highly polluted by petrogenic sources. Even though the toxicity risk test has proved that PAHs concentration of these areas is still low even under a constant long exposure, the result of these studies will be an important input for present knowledge on PAHs and calls for the local environmental authority to take note and enforce stronger legal regulation for a better environmental control in future.

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