

CONCENTRATION AND DISTRIBUTION OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) IN THE TOWN OF KOTA BHARU, KELANTAN DARUL NAIM

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Abstract

This study was carried out to determine the concentration and distribution of polycyclic aromatic hydrocarbons (PAHs) in soils of Kota Bharu, Kelantan Darul Naim. A total of 20 sampling sites were chosen covering the town center and surrounding suburban area of this district. PAHs in soil matrix were extracted using ultrasonic agitation technique with dichloromethane as extraction solvent. Fractionation of PAHs were done using silica-alumina column and the characterization of these individual components were carried out using GCMS. Priority PAHs (PAH₁₆ as defined by the U.S. Environmental Protection Agency) were predominantly represented by four to six ring compounds with concentration ranging between 0.022 $\mu\text{g g}^{-1}$ to 24.06 $\mu\text{g g}^{-1}$. Based on ratios of selected molecular markers, it can be concluded that PAHs in soil were generally derived from combustion sources. A strong correlation between benzo (g,h,i)perylene with total identified PAHs strongly suggest that vehicular emission is the major constituent of this combustion sources.

Keywords: PAHs, ,Kota Bahru ,soils, combustion sources, vehicular emission.

Introduction

Pollution of urban area by carboneous compound had received considerable attention over the last decades, mainly because of the public health risk associated with fine particles and carcinogenic and mutagenic effects of PAHs [1]. Polycyclic aromatic hydrocarbons (PAHs) consist of two or more fused benzene rings. These substances are produced by maturation of organic matter, hence the relative abundance of an extremely complex mixture of these compounds in ancient organic-rich sediments and petroleum. Incomplete combustion or pyrolysis is also a known source of PAHs in the environment [2]. Natural source of PAHs include volcanic eruptions, natural vegetation fires and diagenetic processes. However, in urban areas, their occurrences are normally associated with anthropogenic activities such as domestic waste and fossil fuels burning. Other sources are from oil spillage and waste discharge from domestic or industrial activities [3]. In certain study, they also correlate formation of PAHs with biological orientation in soils [4].

Soils are one of the known sink of PAHs, whereby they are deposited either in gaseous state or associated with particulate matter in the air. Therefore it is a good indicator of pollution and environmental risk as it is continuously subjected to pollution due to its open system nature and capability to accumulate various pollutants. Previous research showed that there was a significant level of PAHs found in soils all over the earth including remote areas which originated from forest fires or airborne pollution [5]. Soils from urban industrial area usually consist of high concentration of PAHs, sometimes be 10 to 100 times higher than those in less populated and undeveloped areas. Similarly, urban area is also reported to have higher soil concentration of PAHs than forest or agricultural soils, mainly because of direct exposure to vehicular emissions [6]. A few studies have been carried out in Malaysia [7,8], but none has been reported for PAHs level in soils of Kota Bharu, which is one of the busiest state capital in the east coast of Peninsular Malaysia. Consequently, this study has been carried out to determine the concentration and spatial distribution of PAHs compound in soils from the town of Kota Bharu, Kelantan.

Experimental

Site description

Kota Bharu is one of the rapid growing towns in the east coast of Peninsular Malaysia. Kota Bharu serves as the state capital of Kelantan and is located about 455 km north east of capital Malaysia, Kuala Lumpur. This town acts

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as a center of business activities, industrial areas, and public agencies. It lies near the mouth of Kelantan River at 6°8'N 102°15'E. Kota Bharu has the population of approximately 425 294, making it the largest town on the east coast of Peninsular Malaysia [9]. The dispersal capability of the atmosphere in Kota Bharu is very restricted. The pattern of air flow over the town at night is from the inland towards Kota Bharu. During the day, the pattern is reversed; the wind blows from the sea towards the inland. Such pattern causes limited movement of the pollutants produced from within the town area.

Sampling and preparation

Sampling was done around the urban and suburb area of Kota Bharu district (during June 2001). Twenty sampling sites had been chosen and they were classified according to the zones. The coordinate of the sampling sites are given in Table 1. Samples were collected using metal spade from a 0 to 6 cm of topsoil. At each station, 10 soil samples were taken randomly within a 0.5 meter quadrat and mixed into a composite sample representative of that particular site. The samples were then transported in sealed aluminium foil to the laboratory and once in the lab, they were freeze dried. Only soil fractions of < 500 µm were used for further analysis.

Extraction and analysis of PAHs

Four gram from each soil samples was extracted for 30 min with 20 ml of dichloromethane in an ultrasonic bath extractor. The extracts were filtered and then concentrated to 1 ml prior to silica-alumina column fractionation. PAHs compounds were eluted using 20 ml mixture of DCM: Hexane (1:1). The fraction was then evaporated using nitrogen blowdown until 1 ml and ready for GCMS analysis.

Table 1: Location of sampling site.

Zone	Station	Position	Location
Zone A (Town Center Area)	1	102°14'29 E 06°07'48 N	Kota Bharu Central Market
	2	102°14'09 E 06°07'45 N	Lido Cinema
	3	102°14'18 E 06°07'56 N	State Mosque
	4	102°14'31 E 06°07'54 N	Pantai Timur Shopping Center
	5	102°14'56 E 06°07'40 N	Berek 12, Shell Gas Station
	6	102°14'44 E 06°07'24 N	State Stadium
	7	102°14'53 E 06°07'34 N	Kota Bharu General Hospital
	8	102°14'24 E 06°08'10 N	Kampung Pak Nik Ya
Zone B (Town Outskirt Area)	9	102°14'47 E 06°08'43 N	Jalan Pengkalan Chepa
	10	102°14'53 E 06°07'11 N	Kampung Merbau
	11	102°14'33 E 06°06'11 N	Kampung Wakaf Siku
	12	102°15'02 E 06°07'33 N	Kampung Kweng Dollah
Zone C (Residential Area)	13	102°15'03 E 06°07'59 N	Jalan Dusun Raja
	14	102°15'09 E 06°08'11 N	Taman Bahagia
	15	102°14'21 E 06°06'46 N	Taman Patehjai
	16	102°11'54 E 06°02'26 N	Kampung Seberang Pasir Mas
Zone D (Agricultural)	17	102°16'27 E 06°08'02 N	Kampung Paya Rambai
	18	102°17'15E 06°07'00 N	Kampung Tapang
	19	102°14'15 E 06°08'30 N	Kampung Raja Seksyen Dua
Zone E (Industrial Area)	20	102°15'53 E 06°06'23 N	Lundang Industrial Estate

Sixteen USEPA priority PAHs were selected and they are abbreviated as naphthalene (NAP), acenaphthylene (ANY), acenaphthene (ACP), fluorine (FLU), phenanthrene (PHEN), anthracene (ANT), fluoranthene (FLT), pyrene (PYR), benz(a)anthracene (BaA), Chrysene (CHR), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno(1,2,3-cd)pyrene (IPY), dibenz(a,h)anthracene (DBA), benzo(g,h,i)perylene (BgP). These PAHs were quantitatively analyzed by Shimadzu GCMS (GC17AAF) using a 30 m x 0.25 mm fused silica capillary column (SUPELCO PTE) with helium as the carrier gas at flow rate of 1.2 ml min⁻¹. 1 µl of aliquot from a known volume of analyte solution was injected manually. Verification of peaks was carried out based on key fragment ions, retention times compared to those of external PAHs standards, and/or mass spectra. Only those peaks located within the proper range (2%) of retention time were integrated for qualification and quantification. Quantitations were performed using external standards of a mixture of PAHs.

Quality control

One laboratory procedural blank was included for each batch of analysis containing 5 different soil samples. To each of this 5 samples and 1 procedural blank, n-dotriacontane (C₃₂) and 9,10 – dihydroanthracene internal standards (obtained from Polyscience Division of Preston Industries Inc., Illinois USA) were added to assess the recoveries and performance of the whole analytical procedures. None of the procedural blanks were found to be contaminated with the PAHs compounds of interest, whilst mean recoveries for PAHs internal standards were >85; no correction was made to the concentration of PAHs reported in this paper.

Results and discussion

OC concentration in Kota Bharu soils

Generally, with exception of a few stations, majority of stations studied yielded average organic carbon (OC) content of less than 3 % (Figure 1). Station 12 showed the highest OC with a value of 7.8% whilst lowest percentage was shown by station 2 and 4 with a value of 0.6%. The average value of OC content in this study was 2.86%. No significant correlation was observed between OC and the concentrations of PAHs.

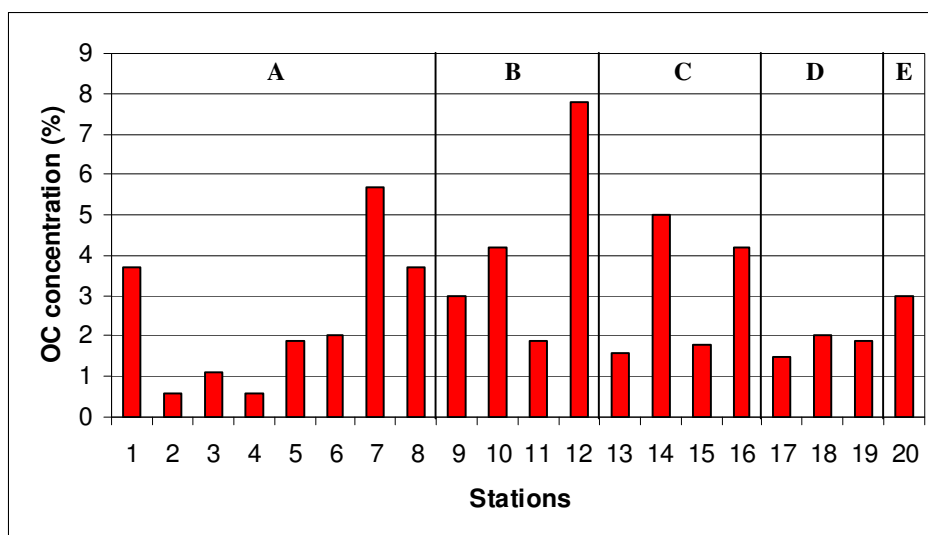


Fig. 1: Organic carbon percentage according to the stations

PAH concentration in Kota Bharu soils

The level of 16 individual priorities PAH analysed in soils and their sums (TIP) are shown in Table 2. BaP appeared to be the dominant and most abundant component detected in all stations. The highest BaP was detected in station 12 (Zone B) with the concentration of $3.95 \mu\text{gg}^{-1}$, while the lowest in station 4 and 17 with both sharing the amount of $0.0047 \mu\text{gg}^{-1}$. Likewise, most soil samples in this study exhibited the presence of elevated concentration of individual 4 and 5 rings aromatic compounds. NAPH on the other hand was not detected in any station whereas presence of 2 and 3 ring PAHs in soils were low in most soils with highly uneven distribution. It is known that low molecular weight PAHs are relatively more volatile and could likely be evaporated by the high temperature and solar radiation of tropical soils.

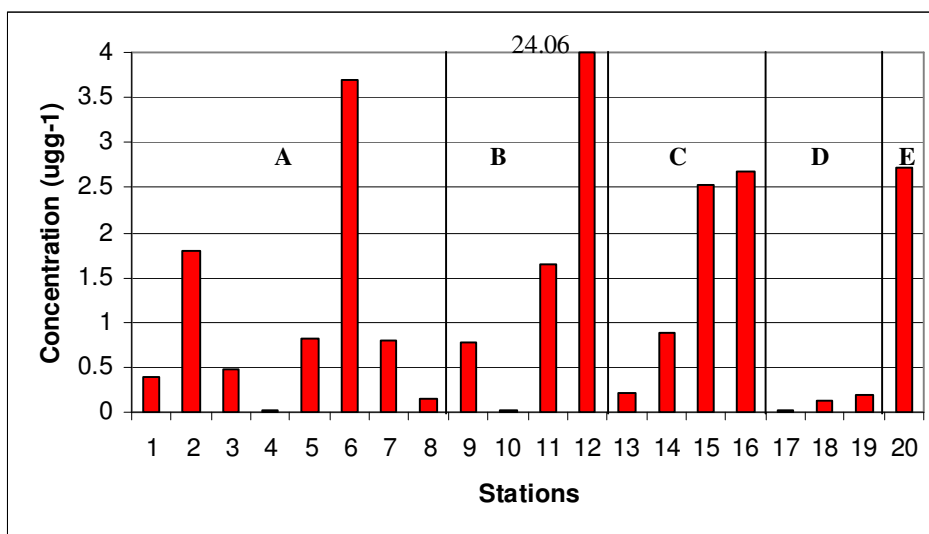


Fig. 2: Distribution of total identified PAHs in stations

With exception of station 12 (zone C), which exhibited significantly higher TIP ($24.06 \mu\text{gg}^{-1} \text{ dw}$), other remaining stations showed TIP concentration ranging from $0.022 \mu\text{gg}^{-1} \text{ dw}$ (Station 17, Zone D) to $3.69 \mu\text{gg}^{-1} \text{ dw}$ (Station 6, Zone A) with a mean value of $2.20 \mu\text{gg}^{-1} \text{ dw}$ (Figure 2). By zone, town area (Zone A) gives a value ranging from $0.025 \mu\text{gg}^{-1} \text{ dw}$ to $3.69 \mu\text{gg}^{-1} \text{ dw}$, giving a mean value of $1.14 \mu\text{gg}^{-1} \text{ dw}$. Zone B shows a range of $0.022 \mu\text{gg}^{-1}$ to $0.78 \mu\text{gg}^{-1}$ with the mean value of $0.31 \mu\text{gg}^{-1} \text{ dw}$. The residential area (zone C), gives a range of $0.22 \mu\text{gg}^{-1}$ to $2.53 \mu\text{gg}^{-1} \text{ dw}$ with a mean value of $1.32 \mu\text{gg}^{-1} \text{ dw}$. Station 12 from zone C shows an anomalously high concentration of TIPs ($24.06 \mu\text{gg}^{-1} \text{ dw}$) evaluation of the sampling data revealed that this site is in the vicinity of a motor vehicle workshop, consequently this station has been excluded for the statistical calculation to minimize biasness of the presented data. The agricultural area in Zone D gives a value ranging between $0.022 \mu\text{gg}^{-1}$ (station 17) to $2.68 \mu\text{gg}^{-1}$ (station 16) with a mean value of $0.76 \mu\text{gg}^{-1}$. The station located in the industrial area in Kota Bharu (Zone E) gave a value of $2.73 \mu\text{gg}^{-1}$. Out of 20 stations analysed, only 7 stations recorded high level of PAHs which exceeded the guideline value prescribe ($1 \mu\text{gg}^{-1}$) for 16 PAHs [10]. These stations were located in the town area (zone A and B), residential area (zone C) and industrial area (Zone E).

Table 2: Concentration of PAHs ($\mu\text{g g}^{-1}$ d/wt) in Kota Bharu soils

Zone	Station	ANY	ANA	FLU	PHEN	ANT	FLT	PYR	BaA	CHR	BbF	BkF	BaP	IPY	DBA	BgP	TIP
	1	0.0022	0.0033	-	-	-	0.0129	0.0262	-	0.0416	-	-	0.2619	0.0058	0.0129	0.017	0.3838
	2	0.0191	-	-	0.0313	0.0635	-	0.0437	-	-	-	-	1.3725	0.0014	0.0841	0.1686	1.7842
	3	0.0051	-	-	0.0095	-	0.0923	0.0758	0.0389	0.0503	0.0499	0.017	0.0586	0.0434	0.008	0.0372	0.486
A	4	0.0011	0.0023	-	0.0014	0.0025	-	-	-	-	-	-	0.0047	0.0044	0.0067	0.0024	0.0255
	5	-	-	-	0.0053	0.0295	-	-	-	0.0288	0.1589	-	0.4191	0.0801	0.0237	0.0748	0.8202
	6	0.1017	-	-	0.0538	0.1107	0.5248	0.4536	0.3446	0.4395	0.2257	-	0.1231	0.4552	0.4283	0.4283	3.6893
	7	0.0051	0.0053	0.0208	-	-	-	-	-	-	-	-	0.6701	0.0204	0.017	0.0537	0.7924
	8	-	-	0.0086	0.0252	0.0217	-	-	-	0.0164	0.0028	0.0104	0.0059	0.0279	0.02	0.0051	0.144
B	9	0.1157	0.2088	-	0.019	0.1087	-	-	-	-	-	-	0.0252	-	0.0874	0.2099	0.7747
	10	-	-	-	0.0033	0.0027	-	0.0004	-	0.003	0.001	0.0006	0.0112	-	-	-	0.0222
	11	0.0657	-	-	0.1341	0.1123	-	0.0015	-	-	0.1818	-	0.4904	0.1789	0.2563	0.2201	1.6411
	12	0.6204	-	-	0.5763	0.7093	3.1421	2.57	1.877	2.341	1.3443	-	3.9539	2.9462	0.9473	3.0272	24.055
C	13	-	0.089	0.0068	0.0301	0.0333	-	-	-	-	-	-	0.0627	-	-	-	0.2219
	14	-	0.0204	-	-	0.0793	0.044	0.0287	0.0293	-	0.0531	-	0.0452	0.3788	0.0429	0.1672	0.8889
	15	0.0772	-	-	0.0477	0.0948	0.4176	0.3534	0.2306	0.2783	0.1369	-	0.3841	0.2478	-	0.2632	2.5316
	16	0.5071	0.7918	-	-	-	-	-	-	-	0.4411	-	0.8834	-	-	0.0522	2.6756
	17	-	-	-	-	-	-	0.0037	-	-	-	0.0062	0.0047	0.003	-	0.0045	0.0221
D	18	-	-	-	0.0252	0.0409	0.0275	0.0037	-	-	-	-	0.0369	-	-	-	0.1342
	19	-	-	-	0.0277	0.0158	-	-	-	-	-	-	0.123	-	-	0.0349	0.2014
E	20	0.0469	-	-	0.0075	0.0185	0.3516	0.2986	0.2107	0.2935	0.2735	0.146	0.4389	0.3009	0.0653	0.2772	2.7291

-: not detected; TIP: Total identified PAHs; Zone A: town center, Zone B: outskirts of town center, Zone C: residential area, Zone D: agricultural area, Zone E: industrial area

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Table 3 shows a comparison of PAHs concentration obtained in this study with those reported in literature [11]. Only residential and agricultural areas show a slight difference with other countries but the values are still within similar range. These comparisons suggest that generally PAHs concentrations in Kota Bharu soils are still relatively low and within the prescribed guideline for the 16 priority PAHs compound.

Table 3: Comparison of PAHs concentration in Kota Bharu soils with selected literatures

Study Area	PAHs concentration ($\mu\text{g g}^{-1}$)	Number of PAHs
<i>Agricultural (rural)</i>		
Brazil	0.096	20
UK	0.19	12
Germany	1.90	6
India	6.7	11
Kota Bharu ^a	0.76	16
<i>Residential (urban)</i>		
Bangkok	0.38	20
Brazil	0.39	20
Germany	1.80	6
UK	4.20	12
India	9.3	11
Kota Bharu ^a	1.32	16
<i>Roadside (urban)</i>		
Australia	3.30	14
USA	58.68	14
India	12.9	14
Kota Bharu ^a	1.45*	16
<i>Industrial (urban)</i>		
UK	4.50	12
Germany	16.00	6
Austria	79.00	18
India	13.7	11
Kota Bharu ^a	2.73	16

Source: [13]; * mix values for zone A and zone B; ^a: this study.

Variations of PAHs distribution in many areas could be attributed to several factors such as the meteorological and geographical aspects, land use, site history and traffic density. This is supported by Zaini [12], which indicated that the characteristics of the surrounding areas can also give a great impact to the pollutants composition especially hydrocarbon pollutions. In the case of Kota Bharu, it is most likely attributed to traffic density and relevant activities in the sampling area. Evaluation of some diagnostic ratio (described in the following section) seems to support this contention. In addition, sampling site characteristic is also in agreement with this argument; for instance, station 12 located at Kampung Kweng Dollah have the highest density of population in Kota Bharu with many shops, government premises and community activity center located in the area. Furthermore, the location of station 12 which is located about 5 meters away from a motorcycle workshop and 2 meters from Jalan Sultanah Zainab, a major road in Kota Bharu, could well contributed to the anomalously high concentration of TIP found at this station, BgP which is widely known to be associated with vehicular emission is rather high at this station ($3.03 \mu\text{g g}^{-1} \text{ dw}$).

In the case of zone D, main activities in the area are cow husbandry and other agricultural activities. Most of stations in this zone were dominated by bushes and secondary forest, thus it is not surprising that these sampling stations

exhibited relatively lower TIP concentrations. In addition, these stations exhibited relatively lower BgP concentration, which could be a reflection of much lower traffic density plying the area.

The concentration of PAHs obtained in zone E is not high as expected from an industrial area. This could be due to the types of industries located in the area which were mainly electronic based, thus potential source of PAHs in this area would be from vehicles or atmospheric transportation.

Source Diagnosis of PAHs in Kota Bharu soils

PAHs ratio have been widely used for source apportionment studies. It is essential to identify the origin and potential sources of PAHs in soil in order to assess the environmental risk. Researchers have prescribed individual PAHs ratio for source diagnosis of these compounds in environment [14]. Table 4 summarizes some of ratios quoted in literature which had been used to distinguish whether PAHs compounds found in the environment were derived from petroleum/ petrogenic or combustion sources.

Table 4: PAHs ratios and values for source diagnosis

Ratio		Values and indicative of		References
		Petrogenic sources (P)	Combustion sources (C)	
R1	ANT/(ANT&PHEN)	< 0.1	> 0.1	[14]
R2	FLT/(FLT&PYR)	< 0.4	≥ 0.4 – 0.5	[14]
R3	BaA/ (BaA&CHR)	< 0.2	> 0.35	[14]
R4	IPY/ (IPY&BgP)	< 0.2	≥ 0.2 – 0.5	[14]
R5	CombPAH/ 16PAH	0.3	0.7	[15]

CombPAH: sum of FLT, PYR, BaA, CHR, BbF, BkF, BeP, BaP, IPY and BgP

Table 5: Calculated ratios for source diagnosis

Zone	Station	R1		R2		R3		R4		R5	
		P	C	P	C	P	C	P	C	P	C
	1	-	-	0.33	-	-	-	-	0.25	-	-
	2	-	0.67	-	-	-	-	0.01	-	-	-
	3	-	-	-	-	-	0.44	-	0.54	-	-
A	4	-	0.64	-	-	-	-	-	-	-	0.71
	5	-	0.85	-	-	-	-	-	0.52	-	-
	6	-	0.67	-	0.54	-	0.44	-	0.52	-	-
	7	-	-	-	-	-	-	-	0.28	-	-
	8	-	0.46	-	-	-	-	-	-	-	-
	9	-	0.85	-	-	-	-	-	-	-	-
B	10	-	0.45	-	-	-	-	-	-	-	0.73
	11	-	0.46	-	-	-	-	-	0.45	-	-
	12	-	0.55	-	-	-	0.44	-	0.49	-	-
	13	-	0.53	-	-	-	-	-	-	-	-
C	14	-	1.0	-	-	-	1.00	-	-	-	-
	15	-	0.67	-	0.54	-	0.45	-	0.48	-	-
	16	-	-	-	-	-	-	-	-	-	-
	17	-	-	-	-	-	-	-	0.40	-	-
D	18	-	0.62	-	-	-	-	-	-	-	-
	19	-	0.36	-	-	-	-	-	-	-	-
E	20	-	0.71	-	-	-	0.42	-	0.52	-	-

R1: ANT/(ANT&PHEN); R2: FLT/ (FLT&PYR); R3: BaA/ (BaA&CHR); R4: IPY/ (IPY&BgP); R5: CombPAH/ 16PAH; P: petrogenic sources; C combustion sources; -: not available.

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Using these diagnostic ratios, values were calculated and applied in this study for source apportionment of the PAHs found in the soils of Kota Bharu (Table 5). Based on the ratios calculated, it can be concluded that PAHs compounds found at all station studied were generally derived from combustion sources. According to the literature, differentiation of these combustion sources could be carried out further by evaluating selected diagnostic ratios as shown in Table 6.

Table 6: Ratio of individual PAHs

Ratio	Values	Indicative of	Ref
FLT/ (FLT&PYR)	> 0.5	Grass, wood, coal combustion sources	[14]
IPY/ (IPY&BgP)	> 0.5	Grass, wood, coal combustion sources	[14]
BaP/BgP	< 0.6	Non-traffic sources	[16]
	> 0.6	Traffic sources	

Table 7: Results of calculated ratios

Zone	Station	FLT/(FLT&PYR)	IPY/(IPY&BgP)	BaP/BgP	
		Wood, grass and coal combustion*	Traffic*	Traffic*	non-traffic*
	1	-	-	15.41	-
	2	-	-	8.14	-
	3	0.55	-	1.58	-
A	4	-	0.65	1.96	-
	5	-	-	5.60	-
	6	-	-	-	0.29
	7	-	-	12.48	-
	8	-	0.85	1.16	-
	9	-	-	-	0.12
B	10	-	-	-	-
	11	-	-	2.23	-
	12	0.55	-	1.31	-
	13	-	-	-	-
C	14	-	-	-	0.27
	15	-	-	1.46	-
	16	-	-	16.92	-
	17	-	-	1.04	-
D	18	0.88	-	-	-
	19	-	-	3.52	-
E	20	-	-	1.58	-

- : not available

Using similar diagnostic ratios, attempt was made to evaluate the type of combustion sources which is responsible for the presence of PAHs compound in these soils (Table 7). Evaluation of the ratios calculated suggests that traffic or vehicular emission is the main contributor of PAHs for most stations. However, it must be concluded that a few stations (viz station 3, 4, 6, 8, 9, 12, 14 and 18) could well received PAHs loading from mixed sources of vehicular and non vehicular emission such as wood/ grass/ coal combustion. Similar pattern could be seen in studies done in Dalian, China [17].

An association of BgP with vehicular emission source has long been established [18]; in this study, almost all stations showed the presence of BgP in soils. In this study, a strong correlation exist between BgP with TIP (Figure 3) ($r = 0.91$), which provide further support to the contentation that vehicular emission is a major source of PAHs in the soil of Kota Bharu.

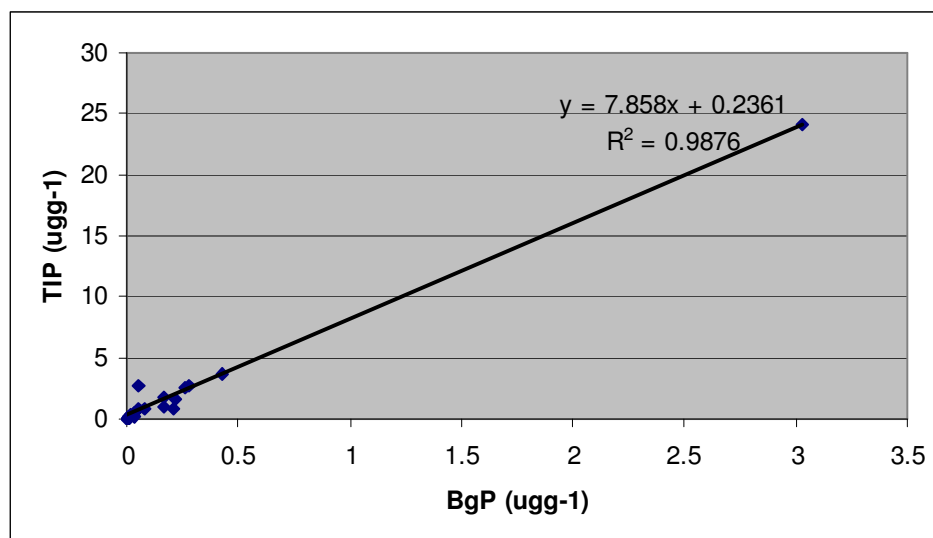


Figure 3: Correlation between BgP and total identified PAHs

Conclusion

The highest concentration of total identified PAHs was obtained in station 12 with the concentration of $24.06 \mu\text{g g}^{-1}$ and the lowest were detected in station 10 and 17 with a value of $0.22 \mu\text{g g}^{-1}$. In addition, it was noted that most stations exhibited the presence of predominantly four to six rings PAHs. Evaluation of several diagnostic ratios suggests that these PAHs were mainly derived from combustion rather than petrogenic sources and vehicular emission seems to be the dominant contributor of these combustion sources. Furthermore, a strong correlation between BgP and TIP provide further support to the contention that vehicular emission is the main source of PAHs in Kota Bharu soil.

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