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Contamination of Environment with Polycyclic Aromatic Hydrocarbons in India

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Abstract

Environment in India is contaminated with polycyclic aromatic hydrocarbons (PAHs) due to occurring of large anthropogenic activities *i.e.* fuel combustion, mineral roasting and biomass burning. Hence, contamination of 13 toxic PAHs: phenanthrene, anthracene, fluoranthene, pyrene, benz (a) anthracene, ben-zo (b) fluoranthene, benzo (k) fluoranthene, benzo (a) pyrene, benzo (ghi) perylene, dibenz (ah) anthracene, indeno1,2,3-(cd) pyrene, coronene and coronene in the environment (*i.e.* ambient particulate matter, road dust, sludge and sewage) of the most industrialized area: Raipur city, India is described. The $\sum PAH_{13}$ concentration in the 16 environment materials was ranged from 7980 - 1,051,300 µg/kg with mean value of 172,613 ± 154,726 µg/kg. The concentration variations, toxicities and sources of the PAHs in various environmental compartments are discussed.

Keywords

Polycyclic Aromatic Hydrocarbons, Dust, Sewage Sludge

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a large group of chemical compounds, I, with a similar structure comprising two or more joined aromatic carbon rings [1]. The compounds are formed by combustion of fuels, biomass and waste materials [2]. Polycyclic aromatic compounds are carcinogenic and mutagenic compounds, causing irreversible changes in the structure and functioning of living organisms [3]. There are thousands of PAH compounds in the environment but 13 compounds *i.e.* naphthalene, phenanthrene, anthracene, fluoranthene,

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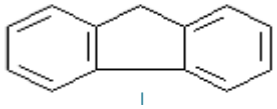
pyrene, chrysene, benz (a) anthracene, benzo (b) fluoranthene, benzo (k) fluoranthene, benzo (a) pyrene, benzo (ghi) perylene, dibenz (ah) anthracene, indeno1,2,3-(cd) pyrene and coronene of the increased environmental and health interests. These compounds differ substantially in their physical, chemical and toxicological properties and therefore, their quantification in the environment is needed. The most potent carcinogens have been shown to be benzo [a] anthracene, benzo [a] pyrene and dibenz [ah] anthracene. They are multimedia contaminants, reported at elevated levels in several environmental samples *i.e.* dust, particulate matter, sludge and sewage of various region of the World [4]-[26].



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2. Methods and Materials

2.1. Selection of Sampling Sites

Raipur (21˚23'N, 81˚63'E) is the capital city of the Chhattisgarh state with population of ≈2 million. The Raipur city is now becoming an important regional commercial and industrial destination for the coal, power, steel and aluminum industries. Several steel rolling mills, sponge iron plants, steel plants, agro-industries, thermal power plants and vehicles ($>1.0 \times 10^5$) are emitting effluents in and around the city.

2.2. Collection of Samples

The road dust, sludge and sewage samples were collected using a stainless-steel scoop from 13 locations of Raipur city in February 2010, **Figure 1**. They were kept in 250-mL glass bottle and dried at 30˚C in an oven for overnight. The samples were crushed into fine particles by mortar and sieved out the particles of mesh size < 0.1 mm. The samples were stored in aluminum foil.

The coarse particulate matter (PM_{10}) and fine particulate matter ($PM_{2.5}$) were collected by using Partisol Model 2300 Sequential speciation air sampler. The sampler was installed at the roof of the building, ≈10 m above from the ground level at residential site: Dagania, Raipur. Both $PM_{2.5}$ and PM_{10} were collected simultaneously over 47 mm quartz fiber filters housed in molded filter cassette. The sampler was run for 24 hrs (6.00 am - 6.00 am) at flow rate of 10 L/min. One sample blank was used for collection of both PM_{10} and $PM_{2.5}$. The loaded filters were dismounted, brought to laboratory, and heated up to 30˚C for 6 hrs to remove the moisture contents. The filters were transferred into the desiccator, and finally weighted to record the particulate contents.

2.3. Analysis of Carbons

The CHNSO-IRMS Analyzer by SV Instruments Analytica Pvt Ltd. was used for analysis of the total carbon (TC). Three carbons *i.e.* elemental carbon (EC), organic carbon (OC) and carbonate carbon (CC) were analyzed in the samples. The total carbn (TC) sample was oxidized with O_2 at 1020˚C with constant helium flow by measuring the resulting CO_2 with thermal conductivity detector. The CC content was analyzed by treating the sample with HCl acid in the CO_2 free atmosphere. The resulting CO_2 was measured by coulometric titration method. The OC content was analyzed by titration method using $K_2Cr_2O_7$ as oxidant, and the excess of $K_2Cr_2O_7$ was determined by titration with the $FeSO_4 \cdot 7H_2O$ solution. The EC content was evaluated by using following equation.

$$EC = TC - (OC + CC)$$

2.4. Analysis of PAHs

The PAH samples were analyzed by capillary gas chromatography (Varian STAR 3400 CX) using temperature programmable splitless injection, a fused silica RTX5-MS column and ion trap mass spectrometric detection [27].

3. Results and Discussion

3.1. Carbon Concentration

All samples are colored, ranging from brown to black, depending on the EC content. The content of EC, OC and

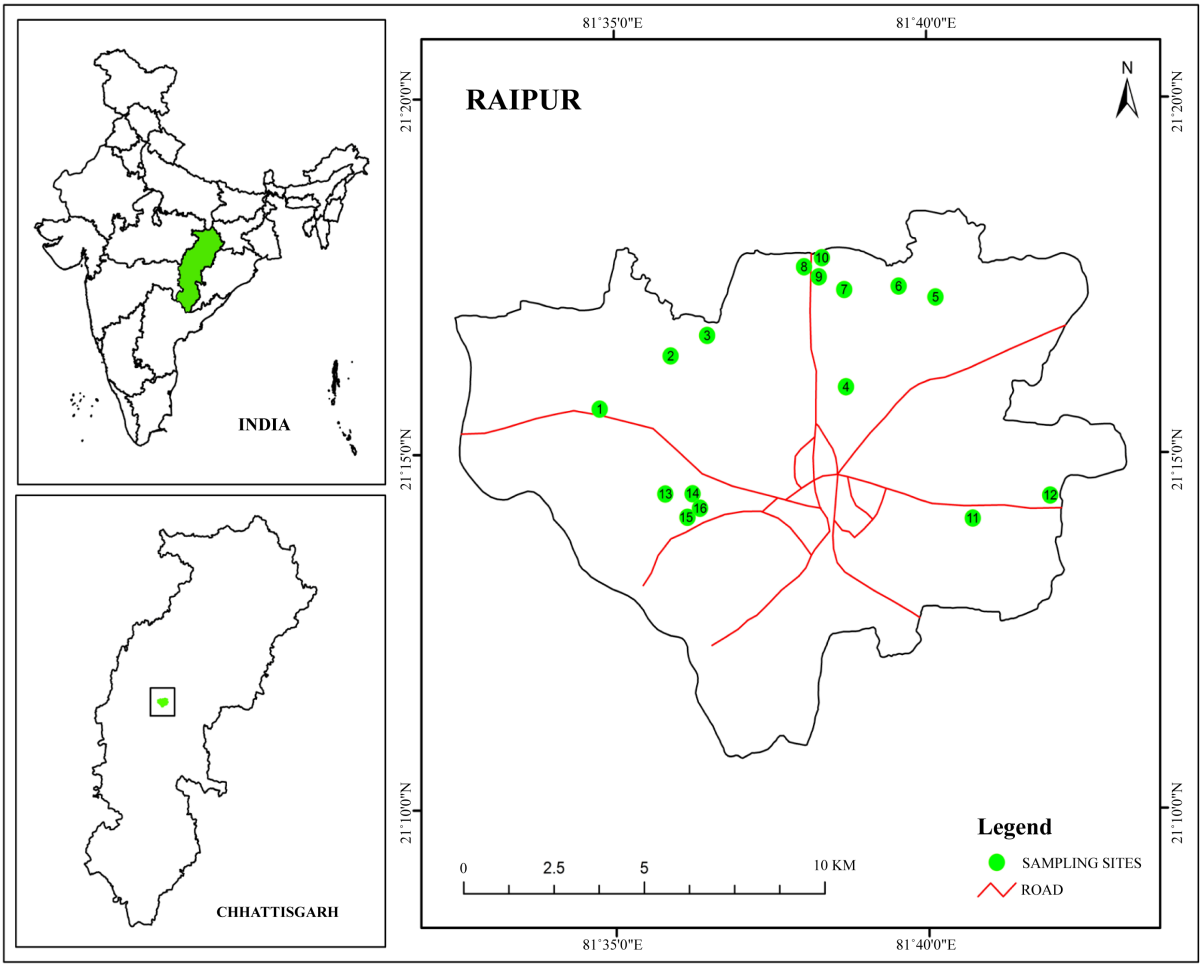

Figure 1. Representation of sampling locations.

CC in the 16 environmental shown in **Table 1**. Relatively high content of EC in all samples was achieved, ranging from 6.5% - 13.5% with mean value of 8.4% ± 1.1%. Very low content of OC and CC was observed in the dust, sludge and sewage samples unlikely to PM samples may be due to their degradation and water solubility, **Figure 1**. The EC content with the OC and CC had good relation (r = 0.94 - 0.96), indicating origin from the similar sources.

3.2. PAHs Concentration

The chemical characteristics of 13 PAHs *i.e.* phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benz [a] anthracene (Baa), chrysene (Cry), benzo [b] fluoranthene (Bbf), benzo [k] fluoranthene (Bkf), benzo [a] pyrene (Bap), benzo [ghi] perylene (Bgh), dibenz [a,h] anthracene (Dba), indeno [1,2,3-cd] pyrene (Ind) and coronene is summarized in **Table 2**. The content of 13 PAHs in 16 environmental samples is presented in **Table 3**. The sum of total concentration of PAHs (ΣPAH_{13}) in the road dust of Raipur city (n = 8) was ranged from 10,427 - 26,031 µg/kg with mean value of 15,282 ± 3377 µg/kg. The highest concentration of the ΣPAH_{13} was observed at site no 5 (*i.e.* Birgaon) due to higher industrial and traffic emissions, **Figure 2**. Similarly, the concentration of ΣPAH_{13} in the SL, MW, AW and TPPW was found to be 7980, 9669, 10,570 and 8326 µg/kg, respectively. No signal for Cor was detected in the environmental samples *i.e.* RD, SL, MW, AW and TPPW samples. The major fraction of PAHs in the RD, AW and TPPW samples was contributed by three compounds *i.e.* Phe, Fla and Pyr, **Figure 3**. A different distribution pattern of PAHs in the SL and MW samples was observed, dominated by Pyr and Bgh contents, **Figure 3**. The concentration of ΣPAHs in the PM_{10} and $PM_{2.5}$ was strongly enriched, >25-folds higher than the road dust with appearing of strong Cor signal. The $PM_{2.5}$ sample

Table 1. Chemical characteristics of PAHs.

S. No.	PAHs	No. of rings	Abbreviation	Formula
1	Phenanthrene	3	Phe	$C_{14}H_{10}$
2	Anthracene	3	Ant	$C_{14}H_{10}$
3	Fluoranthene	4	Fla	$C_{16}H_{10}$
4	Pyrene	4	Pyr	$C_{16}H_{10}$
5	Benz [a] anthracene	4	Baa	$C_{18}H_{12}$
6	Chrysene	4	Cry	$C_{18}H_{12}$
7	Benzo [b] fluoranthene	5	Bbf	$C_{20}H_{12}$
8	Benzo [k] fluoranthene	5	Bkf	$C_{20}H_{12}$
9	Benzo [a] pyrene	5	Bap	$C_{20}H_{12}$
10	Dibenz [ah] anthracene	5	Dba	$C_{22}H_{14}$
11	Benzo [ghi] perylene	6	Bgh	$C_{22}H_{12}$
12	Indeno [1,2,3-cd] pyrene	6	Ind	$C_{22}H_{12}$
13	Coronene	6	Cor	$C_{24}H_{12}$

Table 2. Concentration of carbons and polycyclic aromatic hydrocarbons in environmental samples.

S. No.	Sample type	Location	BC, %	OC, %	CC, %	∑PAHs, mg/kg
1	RD1	Tatibandh	6.5	0.32	0.45	10.4
2	RD2	Hirapur	6.7	0.38	0.47	11.2
3	RD3	Sarora	6.9	0.34	0.41	13.1
4	RD4	Khamtarai	6.8	0.36	0.47	14.1
5	RD5	Birgaon	10.4	0.53	0.68	26.0
6	RD6	Urla	7.2	0.39	0.52	14.2
7	RD7	Sankra	8.2	0.42	0.56	16.7
8	RD8	Siltara	8.2	0.44	0.52	16.5
9	SL	Siltara	7.5	0.03	0.12	8.0
10	MW	Siltara	7.6	0.03	0.11	9.7
11	TPPW	Monate, Urla	8.9	0.04	0.17	8.3
12	AW	IGAU	9.0	0.05	0.19	10.6
13	$(PM_{10})1$	Dagania	6.8	5.8	4.6	505
14	$(PM_{10})2$	Dagania	6.7	5.6	4.2	347
15	$(PM_{2.5})1$	Dagania	13.5	8.3	5.1	1051
16	$(PM_{2.5})2$	Dagania	13.2	8.1	5.0	700

RD, SL, MW, TPPW, AW, PM_{10} and $PM_{2.5}$ represent road dust, sludge, municipal/sewage waste, thermal power plant waste, agricultural waste, coarse particulate matter and fine particulate matter, respectively.

was dominated by higher PAHs *i.e.* Bbf, Bgh and Ind, **Figure 4**. Whereas, the PM_{10} sample was dominated by PAHs *i.e.* Fla, Pyr, Bgh and Ind, **Figure 4**. The PAHs content in the dust was negatively and fairly correlated with particle size ($r = -0.89$), **Figure 5**. The concentration of the PAHs in the environmental samples of studied area was found to be comparable to the other parts of the country and World [4]-[25].

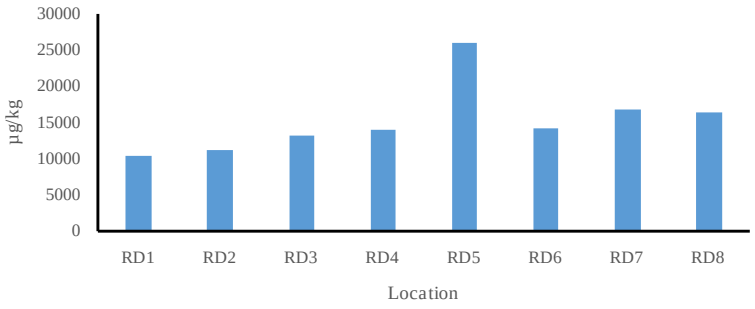

Figure 2. Spatial distribution of PAHs in the road dust.

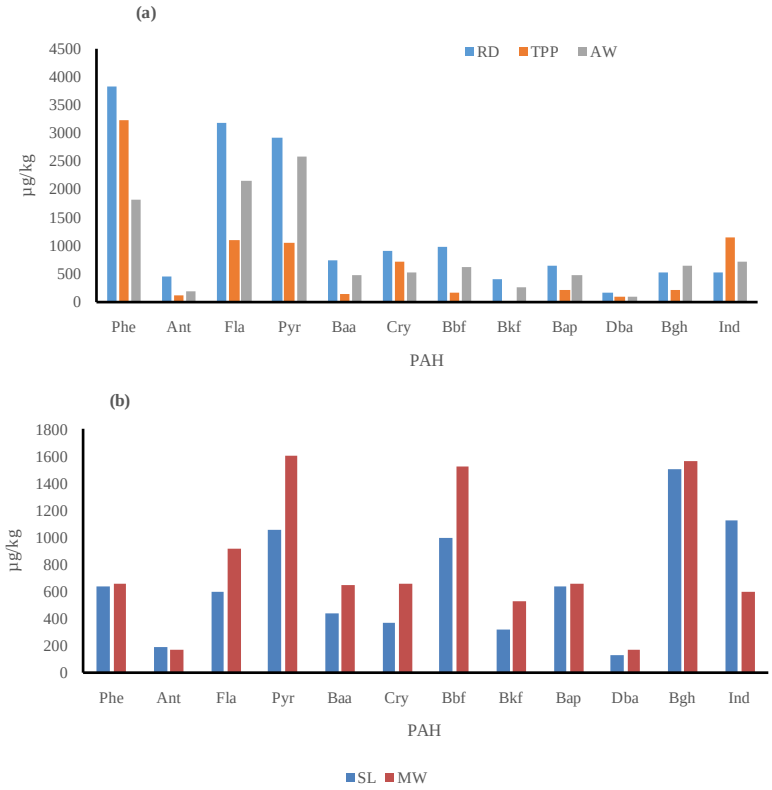

Figure 3. Distribution of PAHs in dust (RD), thermal power plant ash (TPPA), agricultural waste (AW), sludge (SL) and municipal waste (MW).

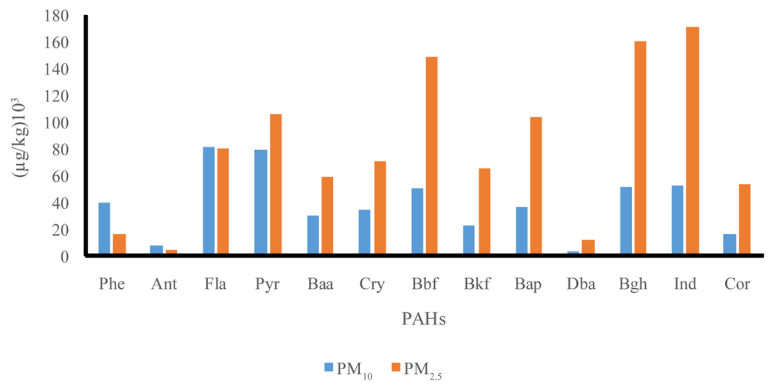

Figure 4. Distribution of PAHs in the PM_{10} and $PM_{2.5}$.

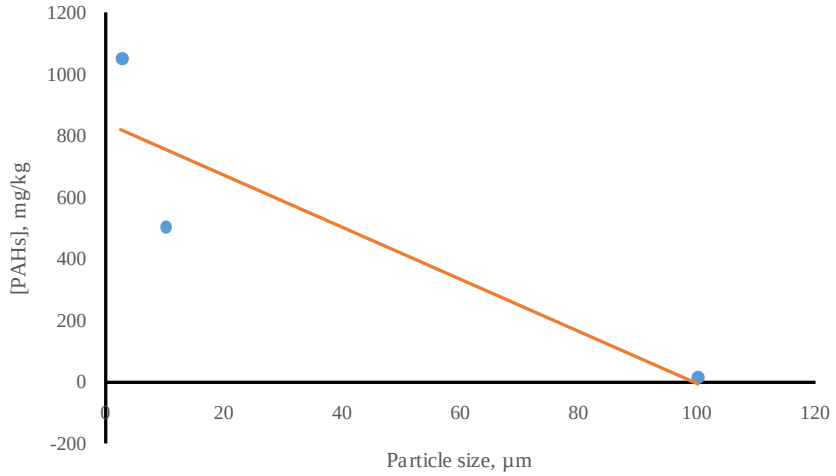

Figure 5. Correlation of PAHs content in the sample with particle size.

Table 3. Concentration of PAHs in environmental samples, µg/kg.

S. No.	Phe	Ant	Fla	Pyr	Baa	Cry	Bbf	Bkf	Bap	Dba	Bgh	Ind	Cor
RD1	3168	301	1921	2222	394	622	586	227	341	18	373	254	0
RD2	2549	218	2669	2494	408	869	629	295	358	151	262	334	0
RD3	3402	320	2874	2240	600	902	866	343	439	97	602	430	0
RD4	3484	252	2208	2978	797	895	1078	442	710	28	582	632	0
RD5	5022	763	4759	5258	1886	1595	1986	880	1636	103	1019	1124	0
RD6	2636	178	3073	2254	781	1135	1358	480	732	200	670	677	0
RD7	5688	797	4655	2928	414	490	468	175	275	496	156	194	0
RD8	4622	732	3240	2974	713	826	931	398	720	226	521	550	0
SL	632	186	598	1056	439	365	990	318	637	132	1505	1122	0
MW	653	168	914	1598	642	658	1525	526	653	167	1564	601	0
TPPW	3240	120	1104	1056	154	708	168	145	204	84	204	1139	0
AW	1829	191	2149	2574	475	515	630	256	486	101	644	720	0
(PM10)1*	39.3	7.7	81.7	79.8	29.6	34.6	50.5	22.1	35.9	3.5	51.5	52.7	15.9
(PM10)2*	24.7	5.2	39.9	39.6	18.5	23.3	43.5	19.0	22.2	2.7	45.4	47.8	15.6
(PM2.5)1*	16.1	3.8	79.9	106.1	59.1	70.6	149.1	65.4	104.4	11.3	160.3	171.1	54.1
(PM2.5)2*	15.9	4.4	37.7	47.3	30.7	42.4	115.6	51.1	53.3	6.8	119.5	129.2	46.3

* = 10^3.

3.3. Vertical Distribution of PAHs

The vertical distribution of the PAHs from 0 - 30 cm in the sludge samples was studied, and presented in **Figure 6**. The ∑PAHs content was strongly increased with increase of the sludge depth profile from 0 - 30 cm, may be due to their poor adsorption with the geo-media. Among them, extremely high vertical distribution of compounds *i.e.* Fla, Pyr, Bbf and Bap was observed.

3.4. Toxicities

The toxicities of PAHs increases as the mass number increases, and seven PAHs (*i.e.* Pyr, Baa, Bbf, Bkf, Bap, Dba and Ind) are considered to be more toxic, may bedue to higher thermal stability and delocalization of

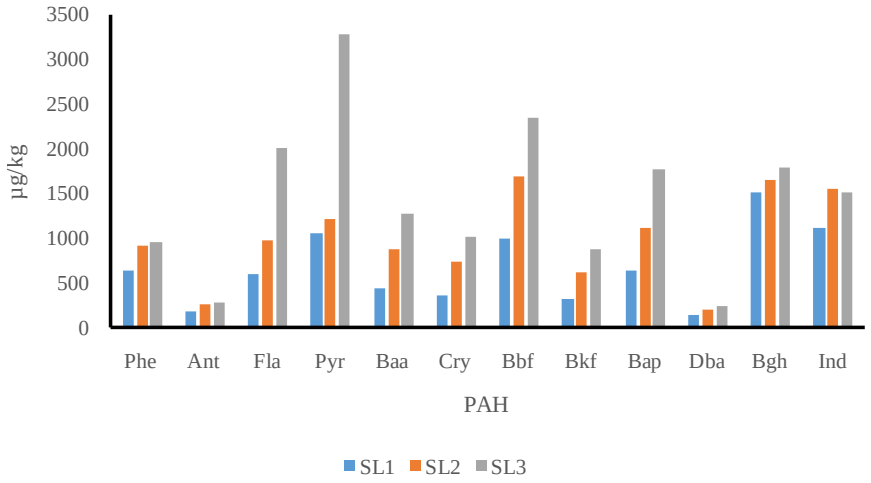

Figure 6. Vertical distribution of PAHs in the sludge, SL1 = 0 - 10 cm, SL2 = 10 - 20 cm, SL3 = 20 - 30 cm.

π-electrons. The carcinogenic potentiality of Pyr, Baa, Bbf, Bkf, Bpa, Dba and Ind reported was 0.01, 0.1, 0.1, 0.1, 0.1, 1.0 and 0.1, respectively [28]. The benzo [a] pyrene equivalent (BapE) value was computed by using the following equation:

$$\text{Total BapE} = \left[\sum_i C_i\right] \times \left[TEF_i\right]$$

where, C_i and TEF_i are the concentration and the corresponding toxic equivalent factor (TEF) value of PAHs.

The BapE value for RD, SL, MW, AW, TPPW, PM_{10} and $PM_{2.5}$ was found to be 1108, 1135, 1237, 846, 475, 52,000 and 138,500 µg/kg in the term of Bap. The carcinogenic fraction of PAHs in RD, SL, MW, AW, TPPW, PM_{10} and $PM_{2.5}$ samples was ranged from 5.7% - 15.7% with significantly higher value for SL, MW and PM samples, **Figure 7**. The concentration of PAHs in the environmental samples was found to be several folds higher than recommended value of 1000 µg/kg [29].

3.5. Correlation and Sources

The correlation matrix of the carbons and PAHs are summarized in **Table 4**. The PAHs had fair correlation with the BC, OC and CC contents (r = 0.70 - 0.96), indicating their origin from the burning processes. The lower PAHs (*i.e.* Phe, Ant, Fla and Pyr) among themselves had fair correlation, may be due to existence of their larger fractions in the gaseous forms, **Table 4**. The higher PAHs (*i.e.* Baa, Cry, Bbf, Bkf, Bap, Bgh and Ind except Dba) among themselves had good correlation, indicating origin from the burning processes, **Table 4**.

The diagnosis ratios: Phe/Antand [Fla]/[Fla + Pyr] were used to find out the sources of PAHs in the studied samples [30] [31]. The Phe/Ant ratio for TPPW, RD, AW, SL, MW, PM_{10} and $PM_{2.5}$ was found to be 27, 10.2, 9.6, 3.4, 3.9, 5.0 and 3.9, respectively, suggesting the domination of petrogenic PAHs in the TPPW, RD and AW samples, **Figure 8**. The [Fla]/[Fla + Pyr] ratio of >0.5, 0.5 - >0.4 and <0.4 was used as signature for PAHs emission from combustion of grass, wood/ coal, petroleum and diesel, respectively [31]. The [Fla]/[Fla + Pyr] ratio was ranged from 0.36 - 0.52, indicating domination of biomass or coal origin PAHs in the RD, TPPW and PM_{10} samples, **Figure 8**.

4. Conclusion

The light PAHs (3 - 4 ring) was found to be dominated in the RD, TPPW, AW and PM10 samples unlikely to SL, MW and PM2.5 samples. Their origins were largely pyrogenic, emitted by combustion of biomass, coal and diesel. The higher PAHs (5 - 6 ring) was found to extremely enrich in the $PM_{2.5}$ sample due to origin by the combustion processes. The PHAs concentration was remarkably increased vertically and might be due to poor adsorption by the geo-media. The PAHs content in the environmental samples of the studied area was found to be several folds higher than recommended value of 1000 µg/kg.

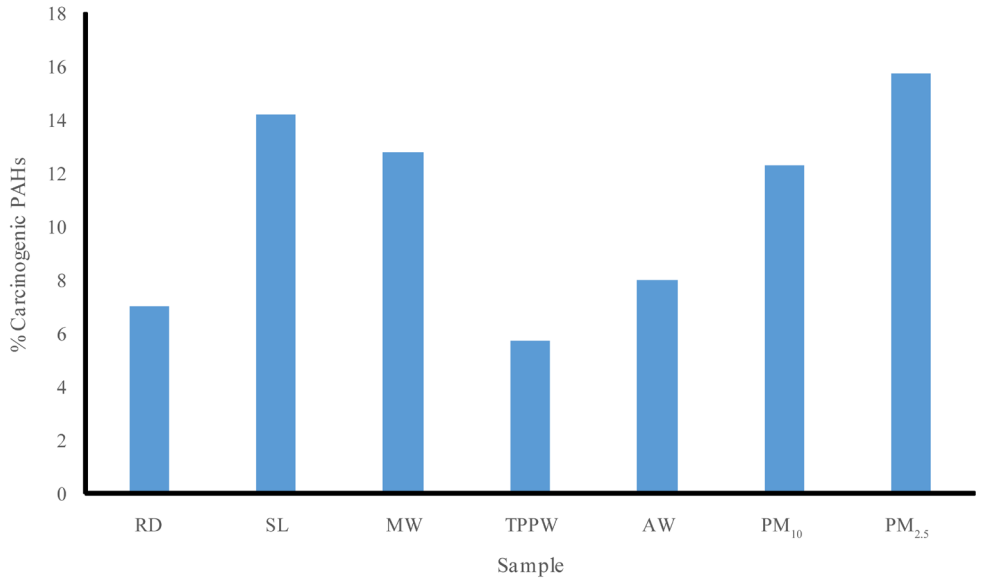

Figure 7. Percentage of carcinogenic PAHs in environmental samples.

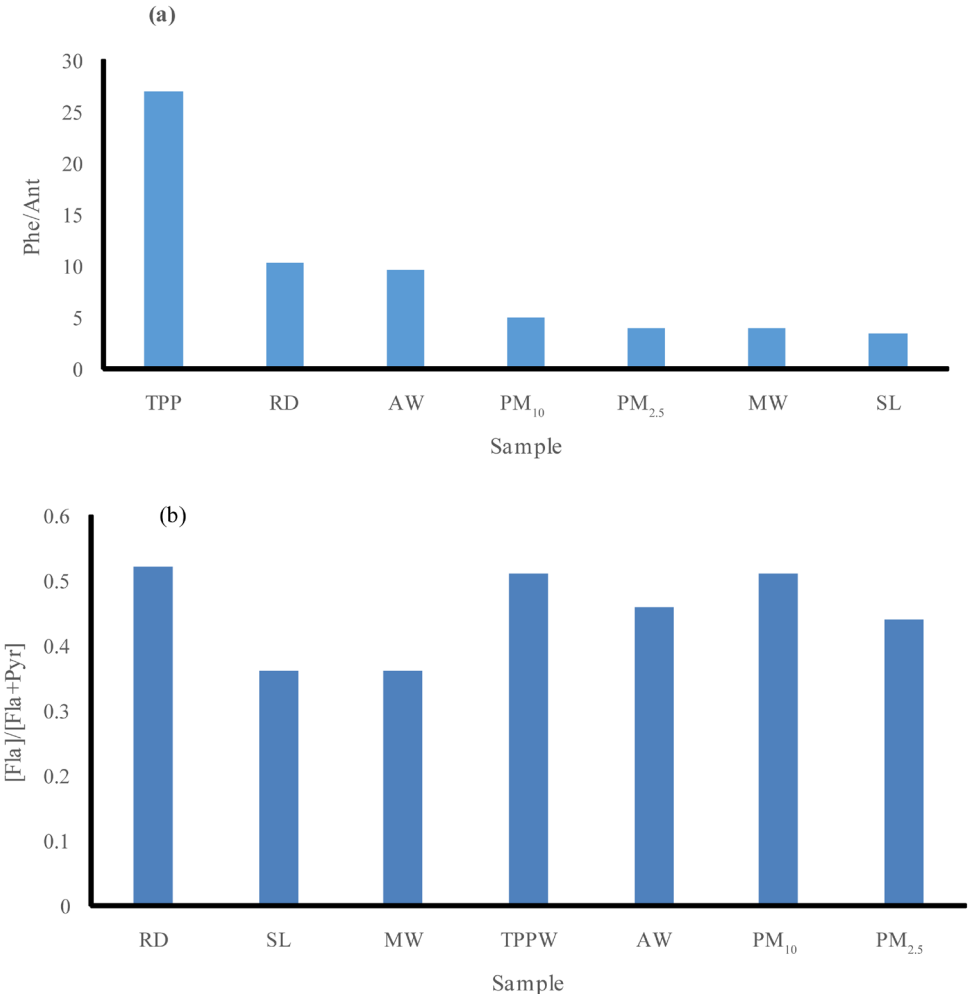

Figure 8. Diagnostic ratio for source determination of PAHs.

Table 4. Correlation matrix of PAHs in the road dust.

	Phe	Ant	Fla	Pyr	Baa	Cry	Bbf	Bkf	Bap	Dba	Bgh	Ind
Phe	1											
Ant	0.96	1										
Fla	0.80	0.81	1									
Pyr	0.61	0.64	0.69	1								
Baa	0.35	0.39	0.54	0.91	1							
Cry	−0.02	0.06	0.34	0.71	0.91	1						
Bbf	0.12	0.17	0.39	0.75	0.95	0.96	1					
Bkf	0.18	0.24	0.42	0.83	0.98	0.97	0.98	1				
Bap	0.31	0.37	0.48	0.89	0.99	0.92	0.96	0.99	1			
Dba	0.60	0.57	0.66	−0.01	−0.23	−0.37	−0.31	−0.33	−0.26	1		
Bgh	0.05	0.10	0.24	0.65	0.90	0.92	0.96	0.94	0.91	−0.47	1	
Ind	0.15	0.20	0.37	0.78	0.96	0.95	0.99	0.99	0.98	−0.33	0.95	1

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