RESEARCH ARTICLE

Quantification and characterization of vehicle-based polycyclic aromatic hydrocarbons (PAHs) in street dust from the Tamale metropolis, Ghana

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Abstract

Objective Concentrations of polycyclic aromatic hydrocarbons (PAHs) in street dust in the Tamale metropolis, Ghana, have been measured in this study.

Results The concentrations of the various types of PAHs identified in street dust samples from high vehicular traffic density in the metropolis are as follows: naphthalene, 10,000 μg/kg; acenaphthylene, 13,000 μg/kg; acenaphthene, 76,000 μg/kg; fluorene, 18,900 μg/kg; phenanthrene, 40,000 μg/kg; anthracene, 21,000 μg/kg; fluoranthene, 35,200 μg/kg; pyrene, 119,000 μg/kg; benzo[a]anthracene, 17,700 μg/kg; chrysene, 10,600 μg/kg; benzo[k]fluoranthene, 18,700 μg/kg; benzo[a]pyrene, 10,900 μg/kg and benzo[g, h, i]perylene, 21,000 μg/kg. Calculation of the phenanthrene/anthracene ratio indicated that the PAHs identified in this study were from vehicular fallout as the ratio was less than 10.

Conclusion It is clear from the results of the study that road users in the Tamale metropolis, especially hawkers, are exposed to the harmful effects of PAHs, and this suggests the need for the establishment of mitigation measures by the regulatory agencies.

Keywords PAHs · Street dust · Vehicular emissions · Contamination · Risk

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1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of lipophilic anthropogenic chemicals that are ubiquitously distributed in the environment. PAHs consist of carbon and hydrogen atoms arranged in two or more fused aromatic rings. They exhibit a zero or very slight dipole moment, large molecular surface area, and no capability for hydrogen bonding. Consequently, low aqueous solubility, semi-volatility, high octanol–water partition coefficient (Kow), stability, and their potential or proven carcinogenicity make them of considerable ecotoxicological concern (Singh et al. 2008). Owing to their toxic and carcinogenic properties and widespread distributions in the global environment, PAHs have attracted the attention of sundry environmental chemists and regulatory agencies for many years. The US Environmental Protection Agency (USEPA) and European Union have listed 16 kinds of PAHs as priority pollutants (Mastral and Callean 2000; US EPA 1993).

PAHs are known to enter aquatic environments through industrial discharges, petroleum spills, urban runoffs,

atmospheric fallout, etc. (Simcik et al. 1996; Mastral and Callean 2000). Owing to their low solubility and high octanol/water partition coefficient, PAHs in water tend to be absorbed to particulate material and then accumulated in bottom sediments (Hong et al. 1995; Tolosa et al. 1996; Viguri et al. 2002). The particle size of particulates and atmospheric environment largely determine the residence time of PAHs in the atmosphere (Singh et al. 2008). For small particles (~1 μm in diameter), residence time is usually of the order of a few weeks, while, for particles with diameter of 1–10 μm, it may last for a few days (Singh et al. 2008).

Studies conducted by different scientists in developed countries have centered on the measurement of individual PAH compounds in dust, soil, air, sediment, and fishes. Among some of the PAH compounds that have been identified are benzo[a]pyrene, anthracene, pyrene, etc. According to Essumang et al. (2006), 16 different PAH compounds were identified in urban soil from streets in Kumasi metropolis, Ghana.

Another study by Liu et al. (2001) found out that benzo [k]fluoranthene and benzo[a]pyrene are among the most particulate PAHs that are found in the environment. This was due to the fact that PAHs of high molecular weights were mainly in the particulate phase. Many researchers have studied the distribution of PAHs on atmospheric particles of different sizes, and the results showed that most particulate PAHs typically were adsorbed on fine particles (Venkataraman et al. 1994; Venkataraman and Friedlander 1994; Pistikopoulos et al. 1990; Kaupp and Mclachlam 2000) or settle on road surfaces.

Automobile exhaust, industrial emission and smoke from the burning of wood, charcoal and tobacco contain high levels of PAHs. Automobiles, especially the heavy duty vehicles, run on diesel that releases large concentrations of different types of PAHs into the ambient air in Tamale, Northern Ghana, along high, low or medium vehicular densities from the combustion of fossil fuels. With the establishment of the Trans Economic Community of West African States (ECOWAS) highway policy which seeks to link all the member states by road in order to facilitate trade, investment and regional integration, the density of the vehicles that ply the Tamale metropolis is expected to rise (Armah et al. 2010). The Tamale metropolis lies along the ECOWAS international highway. Most heavy duty vehicles that run on diesel or petrol use road networks in the metropolis so as to be able to reach land-locked countries such as Burkina Faso, Niger and Mali, just to mention a few. However, very little work has been conducted in the Tamale metropolis to determine the concentrations of PAHs in street dust to serve as a baseline for future reference. Quantification and characterization of different types of PAHs in vehicular fallouts in street dust is essential to pollution control and risk management.

It was within this context that this study was designed to measure the concentrations of the various types of PAHs that are deposited in street dusts taken from different traffic points in the Tamale metropolis of Ghana.

The main objectives for this study were to:

- Identify the various types of PAHs from vehicular movement in the Tamale metropolis
- Determine the concentrations of each type of PAH identified from the fallout due to vehicular movement in the Tamale metropolis
- Compare the concentrations of the various types of PAHs obtained in this study with permissible standards

2 Methodology

2.1 Study area

Tamale (Fig. 1) is an important agricultural region for the country, producing some of the most important food crops consumed in the country like maize, rice sorghum, millet, yam, cassava and groundnuts. Consequently, the already high vehicular movement within the area is expected to intensify as the Trans ECOWAS becomes more operational.

2.2 Sampling techniques

A random sampling technique was adopted in obtaining samples from road dusts from high traffic (500 vehicles by hour) and low traffic (20 vehicles by hour) points, respectively, in the metropolis. Each traffic point was zoned into four different categories. In all, 128 street dust samples were taken from different streets in each zone in the Tamale metropolis in 2009 as shown as in Table 1. The samples were put together, after which a representative sample (laboratory sample) was obtained from the composite sample.

2.3 Sample collection and preparation

The samples were obtained from street dust from each of the four zones. The samples were put into amber glass containers and sealed with an aluminum foil. The samples were stored in an ice chest at 4°C and conveyed to the laboratory. In the laboratory, the samples were freed from stones and other foreign materials and air dried to a constant weight, ground with mortar and pestle and then sieved through a 200 μm mesh.
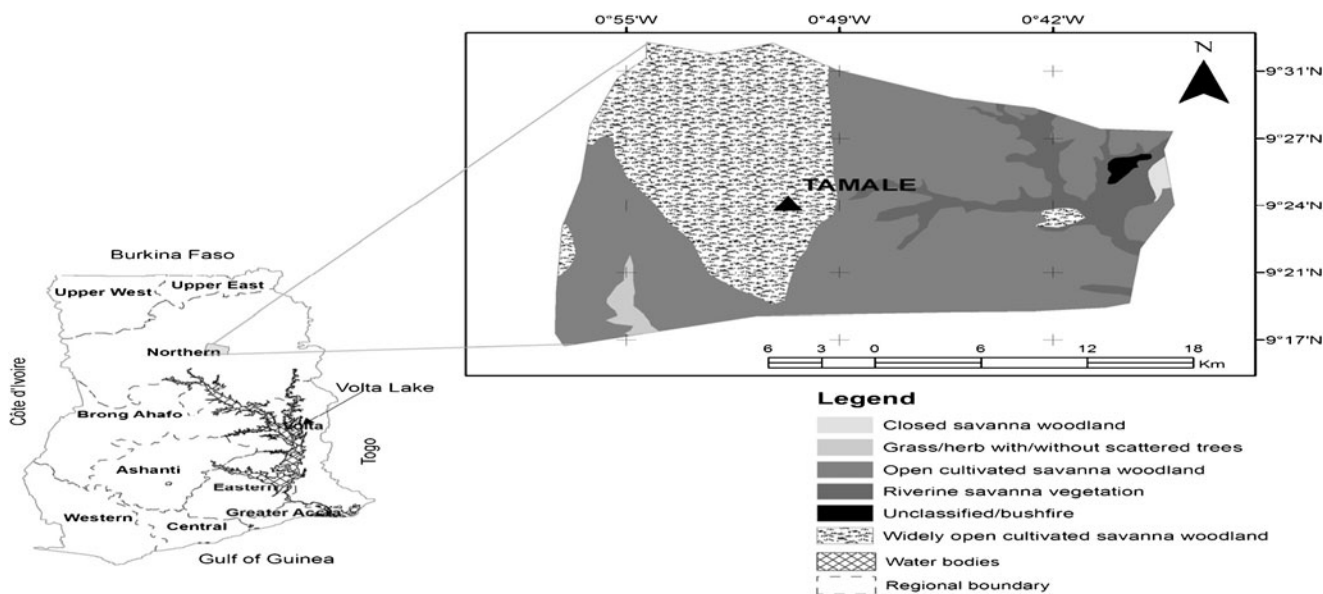
Fig. 1 Map of the northern region of Ghana showing Tamale

2.4 Analysis of PAHs

Ten grams of a crushed, air-dried and homogenized sample was put into a soxhlet thimble. Dichloromethane (120 mL) was put into a round-bottom flask. The soxhlet apparatus was assembled, and the spiked sample was extracted for the PAHs for 6 h. The soxhlet apparatus was cooled to room temperature before removing the solvent.

For high-level contaminated samples, the solvent was carefully and quantitatively transferred from the round-bottom flask into a stoppered measuring cylinder. The flask was rinsed with 2 mL dichloromethane and added to the content of the measuring cylinder. The contents of the measuring cylinder were thoroughly mixed. Five milliliters of this solution was pipetted into a 50 mL beaker, and 0.5 g of activated alumina was added to it. The content of the beaker was swirled and then allowed to evaporate. A glass-fritted chromatographic column was set up containing activated silica gel to a depth of 60 mm, covered with 0.5 g of activated alumina containing 5 mL of the PAH extract to a depth of 30 mm. The column was conditioned by passing 20 mL of pentane through the column. The pentane eluate was discarded, after which 25 mL of dichloromethane was added to the silica gel column. The

eluate was collected and quantitatively transferred into a rotary evaporation apparatus. The flask was rinsed with 10 mL of dichloromethane and was then added to the soxhlet apparatus. The volume was reduced to 1.0 mL; it was quantitatively transferred to a GC–MS vial. Working deuterated p-terphenyl PAH internal standard (200 μL) was added to the GC–MS vial. The vial was sealed tightly with a crimp top for the chromatographic determination of various types of PAHs.

For low-level contaminated samples, the solvent in the round-bottom flask was quantitatively transferred into a rotary evaporation apparatus. The flask was rinsed with 2 mL of dichloromethane, and the solvent was added to the rotary evaporation apparatus. The contents of the rotary evaporator were rinsed with 10 mL of dichloromethane, and the solvent was added to the beaker. The alumina residue was transferred from the beaker to the top of the column containing the alumina and silica gel and eluted with 20 mL of pentane; the pentane eluate was discarded. A clean rotary evaporation apparatus was placed beneath the column, and the PAHs were eluted from the column with 25 mL of dichloromethane. The eluate was collected. The extract was rinsed with 2 mL of dichloromethane, and then, the solvent was added to the rotary evaporator apparatus.

Table 1 Total number of samples collected for the study

Sample category	Zones				Total
	Zone 1	Zone 2	Zone 3	Zone 4	
High vehicular Traffic	16	16	16	16	64
Low vehicular Traffic	16	16	16	16	64
Total	32	32	32	32	128

Table 2 Mean concentrations of PAHs in street dusts from high and low vehicular traffic densities in Tamale metropolis, Ghana

Compound	M/E	High traffic area		Low traffic area	
		Mean conc. (µg/kg)	Percentage of the PAH in the sample area (%)	Mean conc. (µg/kg)	Percentage of the PAH in the sample area (%)
Naphthalene	128	10,000	2.43	3,860	3.69
Acenaphthylene	152	13,000	3.16	3,400	3.25
Acenaphthene	154	76,000	18.45	5,000	4.78
Fluorene	166	18,900	4.59	3,600	3.44
Phenathrene	178	40,000	9.71	8,100	7.74
Anthracene	178	21,000	5.10	3,000	2.87
Fluoranthene	202	35,200	8.54	3,000	2.87
Pyrene	202	119,000	28.9	32,000	30.58
Benzo[a]anthracene	228	17,700	4.30	10,000	9.55
Chrysene	228	10,600	2.57	10,000	9.55
Benzo[k]fluoranthene	252	18,700	4.54	10,000	9.55
Benzo[a]pyrene	252	10,900	2.65	2,700	2.58
Benzo[g, h, i]perylene	276	21,000	5.10	10,000	9.55

The volume was reduced to 1.0 mL, after which the solution was quantitatively transferred to the GC–MS vial. The working deuterated p-terphenyl standard (200 µL) was used as an internal standard and was added to the GC–MS vial. The vial was sealed tightly with a crimp top for the chromatographic determination using the GC–MS chromatogram. The efficiency of the solvent extraction process was determined as 87.54%. Recovery and reproducibility studies were conducted. During the recovery and reproducibility studies, the sample was cautiously spiked by adding 1.00 mL of working deuterated surrogate standard solution (i.e. 100 g of deuterated p-terphenyl) to the sample in the thimble. The thimble was then placed in a clean soxhlet funnel. A recovery of 98.6% was recorded in the recovery and reproducibility studies.

3 Results and discussion

The results of the various types of PAHs identified in street dust from vehicular fallout in the Tamale metropolis and their concentrations are presented in Table 2.

From Table 2, 13 different types of PAHs were identified in street dusts from vehicular movement in the Tamale metropolis. The concentrations of the various types of PAHs identified in street dust from high vehicular traffic in the Tamale metropolis ranged from 10,600 µg/kg (chrysene) to 119,000 µg/kg (pyrene), while in the case of low vehicular traffic, it ranged from 2,700 µg/kg (benzo[a]pyrene) to 32,000 µg/kg (pyrene).

The concentration of pyrene was the highest for both high vehicular traffic (i.e. 119,000 µg/kg) and low

Table 3 Descriptive statistics of data from high and low traffic densities in Tamale metropolis

Parameter	Mean concentration (µg/kg)	Standard deviation	Variance	Kurtosis
Naphthalene	6,930	3,100.86	9.615E6	−1.988
Acenaphthylene	8,200	4,821.72	2.325E7	−2.027
Acenaphthene	40,500	35,639.60	1.270E9	−2.032
Fluorene	1.12E4	7.685E3	5.906E7	−2.027
Phenathrene	24,050	16,014.0	2.564E8	−2.031
Anthracene	12,000	9,038.18	8.169E7	−2.029
Fluoranthene	19,100	16,164.39	2.613E8	−2.031
Pyrene	7.55E4	4.357E4	1.907E7	−2.032
Benzo[a]anthracene	13,850	3,868.83	1.497E7	2.032
Chrysene	10,300	359.57	1.293E5	−0.297
Benzo[k]fluoranthene	14,350	4,370.75	1.910E7	−2.025
Benzo[a]pyrene	6,800	4,122.609	1.700E7	−2.018
Benzo[g, h, i]perylene	40,500	35,639.66	1.270E9	−2.032

vehicular traffic density (i.e. 32,000 μg/kg). This suggests that there is high persistence of this type of PAH in the environment. Though the health effects of breathing high concentrations of pyrene is not known, contact with the skin can cause several diseases such as blistering or redness of the skin which may lead to peeling of the skin. Much concerted effort is required to reduce the levels of pyrene in the Tamale environment.

Descriptive statistics involving measures of central tendency (mean, median, mode), distribution (skewness, kurtosis) and dispersion (standard deviation, range, minimum, maximum, variance) of the levels of PAHs in the Tamale metropolis are shown in Table 3.

Again, from Table 2, the concentration of benzo[a]pyrene was 10,900 μg/kg. Benzo[a]pyrene is a common PAH and is known to cause lung and skin cancers in laboratory animals. USEPA has classified benzo[a]pyrene as a class B2 human carcinogen (Mastral et al. 2000). This implies that exposure to this carcinogenic chemical in the Tamale metropolis by street hawkers will pose a health hazard to them.

Comparing the results of concentrations of the various types of PAHs recorded in both high and low vehicular sampling points, it can be inferred that the presence of PAHs in street dust is due to the emission of PAHs from the combustion of fossil fuels.

Out of the 13 PAHs identified in street dusts from high vehicular densities in the study area, acenaphthene, phenanthrene, fluoranthene and pyrene were present at the highest concentrations, in agreement with data in the literature (Savinov et al. 2003; Motelay-Massei et al. 2004; Wang et al. 2004); as it can be observed in Table 2, the percentages of these three PAHs with respect to the total PAH content in the dust samples are 18.45%, 9.71%, 8.54% and 28.9%, respectively. From low vehicular densities in the study area, out of the 13 PAHs identified in street dusts, phenanthrene and pyrene were present at the highest concentrations, in agreement with data in the literature (Savinov et al. 2003; Motelay-Massei et al. 2004; Wang et al. 2004), as seen from Table 2. The percentages of phenanthrene and pyrene with respect to the total PAH content in the street dust are 7.74% and 30.58%, respectively. However, constant highest concentrations of benzo[a]anthracene, benzo[a]pyrene, chrysene, benzo[k]fluoranthene and benzo[g, h, i]perylene were recorded in this study out of the 13 PAHs identified in street dust from low vehicular traffic density. This result is in agreement with data in the literature presented by Motelay-Massei et al. (2004). It therefore calls for further studies so as to be able to understand the fate and transport mechanism of these compounds in low vehicular traffic points.

The background soil concentrations of PAHs in US urban soils set by the Agency for Toxic Substance and Disease Registry are presented in Table 4 (ATSDR 1995).

Table 4 Background soil concentrations of PAHs in urban soils

Compound	Concentration range in μg/kg
Acenaphthene	–
Acenaphthylene	–
Anthracene	–
Benzo[a]anthracene	169–59,000
Benzo[a]pyrene	165–220
Benzo[b]fluoranthene	15,000–62,000
Benzo[e]pyrene	60–14,000
Benzo[g,h,i]perylene	900–47,000
Benzo[k]fluoranthene	300–26,000
Chrysene	251–640
Fluoranthene	200–166,000
Fluorene	–
Indeno(1,2,3-c)pyrene	8,000–61,000
Phenanthrene	–
Pyrene	154–147,000

Comparing the concentrations of PAHs in street dust from both high and low vehicular traffic densities in the Tamale metropolis in Table 2 with the background soil concentrations of PAHs in urban soils in Table 3, it was found out that the concentrations of acenaphthene, acenaphthylene, anthracene, fluorene and phenanthrene were not major PAH pollutants in US urban soils and, as such, their concentrations cannot be compared with the levels recorded in the Tamale metropolis.

From Table 4, it can be seen that concentrations of benzo[a]pyrene and chrysene in US urban soils were lower than their corresponding concentrations recorded in the Tamale metropolis. The elevated levels of benzo[a]pyrene, a known carcinogen in street dust in the Tamale metropolis, could pose a significant health hazard to residents in the central business district of the metropolis as well as to hawkers who sell by the road side.

Similarly, comparing the results of concentrations of PAHs in street dust from high vehicular traffic density in Tamale (Table 2) with the concentrations of PAHs in urban soils in Kumasi metropolis, Ghana, by Essumang et al. (2006) revealed that concentrations of naphthalene (41,700 μg/kg), acenaphthylene (99,300 μg/kg), acenaphthene (111,200 μg/kg), chrysene (33,600 μg/kg), benzo[k]fluoranthene (45,700 μg/kg), benzo[a]pyrene (27,900 μg/kg) and benzo[g,h,i]perylene (47,000 μg/kg) were found to be higher than what was obtained in the Tamale metropolis. However, concentrations of fluorene (8,900 μg/kg), phenanthrene (12,900 μg/kg), anthracene (5,400 μg/kg), fluoranthene (16,200 μg/kg), pyrene (15,000 μg/kg) and benzo[a]anthracene (13,800 μg/kg) in Essumang et al. (2006) were found to be lower than that recorded in Table 2.

Table 5 Ratios of concentrations of phenanthrene/anthrene and fluoranthene/pyrene in Tamale Metropolis

Sampling Location	Phenanthrene/ Anthracene	Fluoranthene/ Pyrene
High Vehicular Traffic	1.905	0.296
Low Vehicular Traffic	2.70	0.219

According to Aichner et al. (2007), similarities and differences between the composition of the PAH components can be used as chemical fingerprints to identify potential sources. For example, fluoranthene is a universal product of combustion of organic matter and is present in fossil fuel products while phenanthrene has petroleum combustion and a diagenetic origin (Savinov et al. 2003).

Similarly, phenanthrene/anthracene and fluoranthene/pyrene ratios, respectively, have been used to distinguish between PAHs of diverse origin. According to Wang et al. (2004), phenanthrene or anthracene is temperature dependent; hence, low ratios of phenanthrene/anthracene show that the PAHs originated from pyrolytic processes such as fuel combustion in automobiles usually <10 (Wang et al. 2004). On the other hand, petrogenic PAHs, formed by the slow maturation of organic matter, typically show higher phenanthrene/anthracene ratios usually >10. Also the ratio of the concentration of fluoranthene to pyrene can also be used to indicate whether the PAH is of pyrolytic or petrogenic origin. A high concentration of fluoranthene over pyrene is classically related to the pyrolitic origin.

The ratios of concentrations of phenanthrene/anthracene and fluoranthene/pyrene, respectively, are presented in Table 5.

It can be seen from Table 5 that the value corresponding to the phenanthrene/anthracene ratio for both samples from high vehicular traffic and low traffic points in the Tamale metropolis is less than 10. This indicates that exhausts from motor vehicles in the metropolis are the major sources of PAHs measured in this study. Also, the fluoranthene/pyrene ratios as shown in Table 5 are 0.296 and 0.219, respectively, for both high and low vehicular traffic densities in the metropolis, which indicate that PAHs measured in this study are from exhausts of automobiles that ply the Tamale metropolis.

To assess the source of PAHs, a principal component analysis (PCA) was employed, which provides a means of reducing the complexity of the total PAH data set. The results of the PCA are provided in Tables 6 and 7.

Principal component analysis is essentially a method of data reduction that aims to produce a small number of derived variables that can be used in place of the larger number of original variables to simplify subsequent analysis of the data. The underlying principle behind the technique is an effort to reduce the complexity of the data by decreasing the number of variables that need to be considered. From Table 6, the first principal component (scaled eigenvector), by definition the one that explains the largest part of the total variance, has a variance (eigenvalue) of approximately 12.7; this amounts to 97.6% of the total variance. The "Cumulative %" column of Table 6 tells us how much of the total variance can be accounted for by the first k components together. Consequently, to simplify

Table 6 Total variance explained

Component	Initial eigenvalues			Extraction sums of squared loadings		
	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %
1	12.699	97.687	97.687	12.699	97.687	97.687
2	.282	2.168	99.855	.282	2.168	99.855
3	.011	.088	99.943	.011	.088	99.943
4	.003	.020	99.963	.003	.020	99.963
5	.002	.015	99.978	.002	.015	99.978
6	.001	.007	99.985	.001	.007	99.985
7	.001	.007	99.992	.001	.007	99.992
8	.000	.003	99.995	.000	.003	99.995
9	.000	.002	99.997	.000	.002	99.997
10	.000	.001	99.999	.000	.001	99.999
11	.000	.001	99.999	.000	.001	99.999
12	6.250E-5	.000	100.000	6.250E-5	.000	100.000
13	1.280E-5	9.845E-5	100.000	1.280E-5	9.845E-5	100.000

Extraction method: principal component analysis

Table 7 Component matrix[a]

| | Component | | | | | | | | | | | | |
	1	2	3	4	5	6	7	8	9	10	11	12	13
Naphthalene	.993	−.056	.101	.005	−.002	−.001	−.002	.000	.000	.000	5.421E-5	7.507E-5	1.563E-5
Acenaphthylene	.999	−.042	−.013	.008	.014	−.027	.001	.000	.002	.000	7.276E-5	.000	−4.543E-5
Fluorene	.999	−.040	−.018	.013	.006	.005	−.025	−.002	−.001	.001	.000	.000	−8.821E-5
Phenanthrene	.999	−.040	−.007	−.004	.009	.004	.004	.009	−.004	.001	.008	.001	.000
Anthracene	.999	−.040	−.005	.004	.016	.010	.009	−.012	.004	−.001	.002	−.002	−2.866E-6
Fluoranthrene	.999	−.041	−.007	−.003	.008	.007	.003	.000	.003	.000	−.004	.006	.000
Pyrene	.999	−.039	−.006	−.004	.007	.003	.004	.006	−.001	.001	−.004	−.003	−.002
Benzo[a]anthracene	1.000	−.005	−.008	−.014	−.008	−.003	.001	−.007	−.013	−.003	−.001	.001	−1.698E-5
Chrysene	.858	.513	.003	.004	.001	.000	.000	.001	.000	−5.102E-6	7.029E-6	1.113E-5	4.765E-6
Benzo[k]fluoranthene	.999	−.015	−.008	−.021	−.017	−.002	−4.985E-5	−.004	.004	.009	.001	6.704E-5	−2.209E-5
Benzo[a]pyrene	.997	−.059	−.018	.036	−.025	.002	.008	.001	.000	−1.043E-5	7.796E-5	4.594E-5	5.352E-5
Benzo[g, h, i]perylene	.999	−.025	−.008	−.018	−.017	−.001	−.005	.003	.007	−.008	.001	.000	−1.571E-5
Acenaphthene	.999	−.039	−.007	−.004	.007	.003	.002	.006	−.001	.001	−.003	−.002	.003

Extraction method: principal component analysis

[a] 13 components extracted

matters, it can be assumed that a one-component solution is adequate in this case.

From the component matrix (Table 7), the first component has a high positive correlation with each of the PAH variables and is simply a weighted average of the PAH pollution rates and so provides a measure of the overall level of PAH pollution in the Tamale metropolis.

4 Conclusion

The concentrations of the 13 PAHs were studied in urban roadside street dust from regions of high and low vehicular traffic in the Tamale metropolis, Ghana, and ranged from 10,600–119,000 μg/kg for the high vehicular traffic region and 2,700–32,000 μg/kg for low vehicular traffic regions.

The concentration of the phenanthrene/anthracene ratio is below 10, indicating that motor vehicle exhaust appears to be the major source of PAHs found in urban street dust of this city. On the other hand, the fluoranthene to pyrene ratio was generally between 0.8 and 1.2, which is consistent with PAH data from automobile exhaust, according to literature.

It is clear from the results of the study that road users in the Tamale metropolis, Ghana, are exposed to the harmful PAHs in street dust. This suggests the need to conduct a health-based risk assessment of resident adults and children in the metropolis from exposure to PAHs. It is interesting to note that most of the vehicles imported into Africa and, for that matter, Ghana are over-aged vehicles from Europe and other developed countries; their engines might have run down and therefore release large amounts of toxic pollutants such as PAHs. Much work should be conducted in Ghana to determine the health effects from exposure to PAHs from vehicular movement, as this is central to environmental pollution control and risk management.

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