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ESTIMATION AND SOURCES OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) IN DUST AT FUEL FILLING STATIONS IN THE TAMALE METROPOLIS, GHANA

[1]Cobbina, S. J., [2]Ofori, A. S., [3]Bakobie, N.

[1] *Faculty of Natural Resources and Environment, University for Development Studies, Tamale, Ghana,*
[2] *Faculty of Science, Université Libre de Bruxelles, Brussels, Belgium.*
[3] *School of Engineering, University for Development Studies, Tamale, Ghana*

**Corresponding Author's E-mail: scobbina@uds.edu.gh/ofori496@gmail.com*

Abstract

The aim of the study was to identify and quantify the concentrations and potential sources of Polycyclic Aromatic Hydrocarbons (PAHs) present in dust particles at fuel filling stations in the Tamale Metropolis. Sixteen USEPA recognised PAHs were identified after samples were analysed and their potential sources identified using PAH isomeric ratios. Sample extraction was conducted using an Accelerated Solvent Extractor (ASE) and PAH levels in samples were further analysed using the GC-MS system. The total PAHs concentrations in the High-Vehicular Traffic Areas fuel filling stations ranged from 38.70 µg/kg to 1,423,280 µg/kg with an average concentration of 158,080 ± 102,650 µg/kg, while that of the Low-Vehicular Traffic Areas ranged from 305 µg/kg to 3,176,400 µg/kg with an average concentration of 370,220 ± 218,140 µg/kg. Amongst the 16 PAHs identified, Benzo(a)Anthracene had the highest concentration for both Traffic Areas. The average concentration of Benzo(a)Pyrene was 39,310 µg/kg and this was 200 times higher than that recorded from the US urban background soils. The PAH isomeric ratios indicated a strong influence from petroleum and combustion sources on PAH concentration in the fuel filling stations. It is therefore necessary that appropriate measures are taken with regards to pollution control and risk management at fuel filling stations.

Keywords: Benzo(a)Anthracene, Benzo(a)Pyrene, Pyrene, Source Identification, Fuel Filling Stations

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are known to be organic pollutants that are extensively found in the environment. Most PAHs are able to enter the environment by way of incomplete combustion of organic materials including coal, oil, wood, gasoline, garbage, and tobacco. They may be found in the environment as they attach to dust and soil particles, sediments and water of aquatic ecosystems, drinking or wastewater, and food (Mahgoub, 2016).

Air pollution with organic pollutants such as PAHs is a major environmental issue present in both high and middle income countries (German Federal Environment Agency [GFEA], 2012). In the year 2004, it was recorded that 530,000 tons of the 16 USEPA recognised PAHs were produced from diverse sources and released into the global atmosphere. The top global PAH producer in that period was China which recorded a total of 114,000 tons, followed by India with a record of 90,000 tons, the United States producing 32,000 tons, and Sudan recording the minimum with 5,000 tons (Zhang & Tao, 2009; Ofori et al., 2020a).

In Africa, several studies have been conducted to determine the sources and distribution of PAHs in environmental samples (Ofori et al., 2020a). PAHs have been recorded in street dust, smoked fish, smoked meat, roasted food, agricultural soils, surface water and sediments, and in urban and indoor air mixtures (Ofori et al., 2020a).
According to Zhang & Tao (2009), the contribution of Africa to the global PAH emissions was 18.8%. Moreover, the PAH emissions from Nigeria and the Democratic Republic of the Congo ranked 4th and 8th respectively on a global basis. Factors such as savanna fires, and oil exploration activities especially in the Niger Delta were paramount in

projecting Africa's global PAH contribution to such a level (Zhang & Tao, 2009; Ofori et al., 2020a).

Population growth in cities and communities of Ghana over the years has resulted in improved use of vehicles with growing traffic on the roads, and the increase in setting up of industries known for contributing to a higher level of anthropogenic PAH emissions (Safo-Adu et al., 2014). Several studies have been undertaken in urban environments of Ghana to assess the levels of PAHs present in street soils. A study by Bandowe & Nkansah (2016) concluded that PAH concentration records in the Kumasi Metropolis of Ghana were similar to mega cities in other middle-income countries. Likewise, dust samples collected from the road of the Tamale highway in the Northern Region of Ghana were reported to contain high levels of PAHs (Obiri et al., 2011). It is noted that since most of the automobiles (predominantly the heavy-duty vehicles) in the Tamale Metropolis of Ghana are not properly maintained, they release high concentrations of PAHs into the atmosphere as exhaust fumes, which find their way onto particulate matter in the environment (Obiri et al., 2011).

Over the years, studies have been conducted on the occurrence of PAHs in urban road soils (Essumang et al., 2006; Hassanien & Abdel-Latif, 2011; Mahugija, 2015; Bandowe & Nkansah, 2016) on the African continent, but very few have focused on their occurrence in fuel filling stations. Prior to the undertaking of this study, two major possible scenarios of PAH pollution occurring in the fuel filling stations were coined. First was the possible accumulation of PAH-containing particulate matter in the fuel filling stations as a result of vehicular emissions coupled with the movement of dust particles by air masses. Second was the possible occurrence of PAHs due to incidences of fuel spills and leakages happening within the vicinity of the fuel filling stations. Therefore, the goal of this research was to identify and quantify PAHs present in fuel filling stations of different vehicular traffic areas in the Tamale Metropolis of Ghana, and further identify their potential sources.

Methods

Study Area

Tamale Metropolis is located in the Northern Region of Ghana and known to be the only Metropolis in the region as at now. It is positioned in the centre of the region as it shares borders with the Sagnarigu District (West and North direction), Mion District (East direction), and East Gonja (South-West direction). It has a total land size of about 646.9 km^2 and geographically located between latitude 09°16 and 09°34 North and longitude 00°57 West (Ghana Statistical Service, 2014).

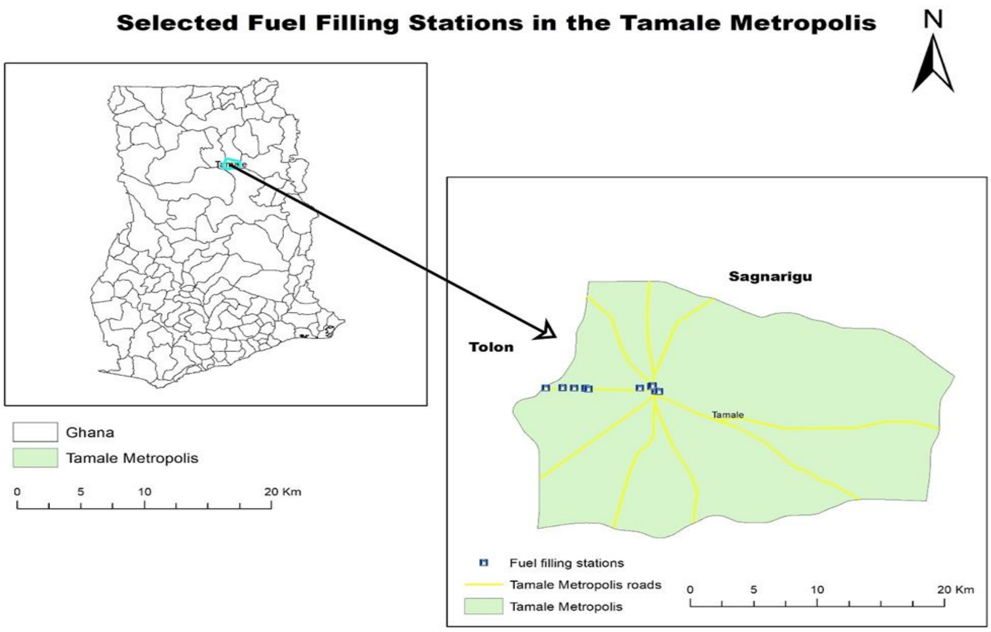

Figure 1: Study Area showing Fuel Filling Stations

Source: *Ofori et al., (2020b)*

Criteria for Site Selection

In defining the area categories of the fuel filling stations, i.e. High-Vehicular Traffic Areas (H-VTA) and Low-Vehicular Traffic Areas (L-VTA), authors followed the parameters used by Ofori et al. (2020b). On the basis of the criteria used, five (5) fuel filling stations each from the H-VTA and L-VTA in the Tamale Metropolis were selected. Fuel filling stations were represented using alphabets from A to J; where from Station A to Station E were categorised as H-VTA fuel filling stations, and those from alphabets F to J were categorised as L-VTA fuel filling stations.

Sample Collection

Sample collection took place from 19th to 21st February 2020 and was undertaken following the method provided by Ofori et al. (2020b). Two (2) samples of 20 g each (one from each fuel dispensing area) were collected from each of the ten (10) fuel filling stations, making a total of twenty (20) samples.

Sample Preparation, Extraction, Clean-up and Analysis

For the preparation, extraction, clean-up, and analysis of samples, the same methods used by Ofori et al. (2020b) was adopted. The Dionex Accelerated Solvent Extractor (ASE) 350 was used for the extraction process with a solvent mixture of 1:1 methanol and acetone. Rotary evaporation was used for the clean-up process of samples after extraction. The Agilent Technologies GC/MS system (GC 7890B and MS 7000C) was used for PAH analysis in samples (Ofori et al., 2020b).

Source Identification of PAHs

The isomeric ratios of Anthracene/(Anthracene + Phenanthrene), Benzo(a)Anthracene/(Benzo(a) Anthracene + Chrysene), Fluoranthene/(Fluoranthene + Pyrene), and Indeno[1,2,3,cd]pyrene/(Indeno[1,2,3,cd]pyrene + Benzo[g,h,i]perylene) were used to identify the different potential sources of PAHs (Yunker et al., 2002) in the study areas.

Results and Discussions

PAHs concentrations recorded from the L-VTA fuel filling stations (especially Station F and Station G) were higher with an average concentration of 370,220 µg/kg when compared to the L-VTA fuel filling stations. For H-VTA fuel filling stations, the average concentration of PAHs recorded was 158,081 µg/kg (Table 1 & 2). However, Obiri et al. (2011) in a study on PAHs in street dust sampled from the Tamale Metropolis recorded higher levels of PAHs from H-VTAs than L-VTAs. One major reason that contributed to the differences in results between the two studies was that, in the current study, sampling was limited to fuel filling stations where many factors may be responsible for PAH concentration levels. Some of these factors include the intensity and frequency of traffic jams in the area of the fuel filling stations, the type of vehicles visiting the fuel filling stations, the characteristics of neighbourhood where fuel filling station is located, fuel spill incidences within the fuel filling stations, the nature of roads and speed of vehicles close to the fuel filling stations, as well as price variations of fuel products amongst the fuel filling stations which indirectly influences their rate of patronage.

For instance, it was observed that station F which was located in the L-VTA recorded the highest concentration of PAHs (Table 1). This is because it was visited more by heavy-duty vehicles, and also provided parking space for them. Also, many vehicle users chose to purchase their fuels there because it was newly opened and the fuel products sold there were relatively cheaper for the public. Wood and grass combustion could also be a contributing factor to the higher PAH concentrations in the L-VTAs, since station F and station G in particular were located near areas where bush burning was common during the dry seasons. Station B which is located in a H-VTA recorded the second highest concentration of PAHs. It was observed that because of its location in the Tamale business District, it served as an active parking place for many vehicles and also, a convenient station to fuel vehicles.

Table 1: PAH Levels of Fuel Filling Stations

Area Category	Fuel Filling Station	Total PAH Concentration (µg/kg)
H-VTA	Station A	11,658
	Station B	1,924,546
	Station C	587,067
	Station D	5,635
	Station E	396
L-VTAs	Station F	4,147,852
	Station G	1,772,030
	Station H	1,243
	Station I	1,808
	Station J	597

Source: *Filed Data (2018)*

Amongst the 16 PAHs identified, Benzo(a)Anthracene was of highest concentration in both H-VTAs and L-VTAs with average concentrations of 284,656 µg/kg and 635,280 µg/kg respectively (Table 2). This finding is in line with studies conducted by Essumang et al. (2014) in Ghana, and Franco et al. (2017) in Brazil which concluded that Benzo(a)Anthracene is the most abundant PAH in many environments. The results therefore indicate a possible carcinogenic risk posed towards the health of the public, as Benzo(a)Anthracene is stated by the IARC as a Class 2B compound (Lerda, 2011; Ofori et al., 2020b).

Pyrene recorded the second highest mean concentration among the 16 PAHs identified in both low-vehicular and high-vehicular traffic areas (356,584 µg/kg and 189,051 µg/kg respectively). Obiri et al. (2011) confirmed the high levels of pyrene in street dusts of the Tamale Metropolis and called for concerted efforts to help reduce its levels in the environment. Research studies conducted on mice with pyrene contamination showed that mice that were fed with pyrene developed nephropathy (Agency for Toxic Substances and Disease Registry [ATSDR], 1995).

Though the U.S. EPA mentioned that there is inadequate data to categorise pyrene as a carcinogen, the high contamination levels of pyrene in this study may pose a major potential health risk to the public. Fluorene had the least concentrations in the H-VTAs and L-VTAs fuel filling stations respectively. However, it was possible that, some amounts were lost in the clean-up process during analysis because of its high volatility.

Table 2: Mean Concentrations of Individual PAHs in Low and High-Vehicular Traffic Area Fuel Filling Stations

PAHs	Low-Vehicular Traffic Area			High-Vehicular Traffic Area		
	Total Conc. (μg/kg)	Mean Conc. (μg/kg)	Standard Deviation (μg/kg)	Total Conc. (μg/kg)	Mean Conc. (μg/kg)	Standard Deviation (μg/kg)
Naphthalene	787 (191-596)	157	82	39 (1-22)	8	3
Acenaphthene	43,787 (59-35471)	8,757	4,855	5,623 (96-2912)	1,125	475
Acenaphthylene	422 (BDL-363)	84	50	144 (BDL-117)	29	16
Fluorene	305 (1-250)	61	34	62 (BDL-33)	13	5
Anthracene	8,893 (3-6,844)	1,779	938	1,658 (BDL-918)	332	144
Phenanthrene	8,955 (3-6,971)	1,791	955	1,704 (738-966)	341	150
Fluoranthene	447,822 (81-374,825)	89,564	51,392	82,482 (166-65,590)	16,496	8,953
Pyrene	1,782,923 (232-1,595,030)	356,584	220,423	945,256 (41-672,015)	189,051	93,201
Benzo(a)Anthracene	3,176,398 (1107-1,798,975)	635,280	278,811	1,423,278 (5,040-1,131,744)	284,656	154,238
Chrysene	53,533 (34-46,304)	10,707	6,364	47,904 (33-38,835)	9,581	5,304
Benzo(a)Pyrene	373,676 (39-299,718)	74,735	41,039	19,453 (9,219-10,234)	3,891	1,689
Benzo(k)Fluoranthene	1,732 (299-1,432)	346	196	143 (27-117)	28	16
Benzo(b)Fluoranthene	15,318 (BDL-12,773)	3,064	1,751	623 (BDL-558)	124	77
Dibenzo[a,h]anthracene	989 (365-625)	198	91	233 (22-211)	47	29
Indeno[1,2,3,cd]pyrene	957 (1-659)	191	92	250 (27-223)	49	31
Benzo[g,h,i]perylene	7,035 (3-6,941)	1,407	979	452 (BDL-285)	90	40
Mean concentration (μg/kg)	**370,221**			**158,081**		

Source: *Filed Data (2018)*

Comparisons of the mean concentrations of some PAHs identified in this study with those of some sampled soils, Benzo(a)Pyrene for instance was about 200 times higher than that recorded from US urban soils (Agency for Toxic Substances and Disease Registry [ATSDR], 1995) and about 20 times higher than that of the Ontario industrial and commercial area soils (Tang et al., 2005). Also, Benzo(a)Anthracene which had the highest concentration in this study was about 70, 46 and 16 times higher than that of Ontario, British Columbia and US urban soils respectively (Table 3). These differences can be attributed to the fact that a number of fuel filling stations (especially in the L-VTAs) were situated in areas where the combustion of gasoline, burning of sawmill dust, bush burning and as well, the singeing activities of abattoirs were common.

Table 3: Some Published Environmental Standards for PAHs in Soils

List of PAHs	**Mean PAH Concentrations (μg/kg)**			
	US Urban Soils (ATSDR 1995)	**British Columbia: Industrial/Commercial Area (Tang et al., 2005)**	**Ontario Industrial/Commercial Area (Tang et al., 2005)**	**This Study**
Naphthalene	-	50,000	4,600	83
Acenaphthene	-	-	100,000	4,941
Acenaphthylene	-	-	15,000	57
Fluorene	-	-	340,000	37
Anthracene	-	-	28,000	1,055
Phenanthrene	-	50,000	40,000	1,065
Fluoranthene	200–166,000	-	40,000	53,030
Pyrene	145–147,000	100,000	250,000	272,818
Benzo(a)Anthracene	169–59,000	10,000	6,600	459,968
Chrysene	251–640	-	17,000	10,144
Benzo(a)Pyrene	165–220	-	1,900	39,313
Benzo(k)Fluoranthene	15,000–62,000	-	-	188
Benzo(b)Fluoranthene	300–26,000	10,000	12,000	1,594
Dibenzo[a,h]Anthracene	-	10,000	1,900	122
Indeno[1,2,3,cd]Pyrene	8,000–61,000	10,000	19,000	121
Benzo[g,h,i]Perylene	900–47,000	-	12,000	749
Total	**25,130–509,860**	**240,000**	**888,000**	**845,283**

Source: *Filed Data (2018)*

According to Yunker et al. (2002) and Singh (2014), the interpretations for the use of isomeric ratios of PAHs to determine their potential sources in the environment are; if the mean ratio of Anth/(Anth + Phen) was found to be less than 0.10, then it indicated petroleum as the source of PAH and when greater than 0.10, the PAH concerned was coming from lubricant oils and fossil fuels. Also, the ratio of Flt/ (Flt + Pyr) when less than 0.40 implicated a petroleum source and greater than 0.50 implicated biomass and coal combustion as sources of PAH. Additionally, the ratio occurring between 0.4 and 0.5 specified liquid fossil fuel combustion as sources of PAH. A ratio of I(cd)P/(I(cd)P + B(ghi)P) less than 0.20 designated a petroleum source, and that greater than 0.50 implicated combustion of biomass and coal as sources of PAH. If the ratio is however between 0.20 and 0.50, it indicated a liquid fossil fuel combustion as source of PAH ratio of B(a)A/(B(a)A + Chr) when less than 0.2 implicated petroleum as source of PAHs, while the range of 0.2 – 0.35 suggested petroleum combustion (especially liquid fossil fuel, vehicle, and crude oil) as source of PAHs, and when greater than 0.35 indicated the combustion of coal, grass, and wood.

From Table 4, the mean value (0.60) of the Anth/(Anth + Phen) ratio suggested lubricant oils and fossil fuels as the potential source of PAH. This was expected as analysed samples were from fuel stations where these products are sold. The Flt/(Flt + Pyr) ratio was 0.47, indicative of a PAH pollution from liquid fossil fuel combustion. This could be traced to vehicular emissions. The I(cd)P/(I(cd)P + B(ghi)P) ratio with mean value of 0.28 also suggested liquid fossil fuel combustion as potential source of PAHs. This further confirmed vehicular emissions as a major contributor to PAHs in the study area. Moreover, the B(a)A/(B(a)A + Chr) ratio implicated the PAH potential sources to include combustion of coal, grass, and wood. This was supported by the fact that a number of the fuel filling stations in the L-VTA were sited close to communities where bush burning was a common practice, especially during the dry seasons. Also, fuel filling stations F and G of the L-VTA were located close to an abattoir where singeing activities for meat processing was practiced. Another contributing source of PAHs from biomass combustion was a sawmill factory located in the L-VTA. It can therefore be stated that petroleum sources (i.e. lubricant oils and fossil fuels, and liquid fossil fuel combustion) contributed the most to the concentrations of PAHs in the sampled fuel filling stations.

Table 4: Summary of Isomeric Ratios of Selected PAHs in Fuel Filling Stations

Ratio	A	B	C	D	E	F	G	H	I	J	Range	Mean
Anth/(Anth + Phe)	1.00	0.48	0.49	1.00	1.00	0.50	0.51	0.50	0.51	0.00	0.00-1.00	0.60
B(a)A/(B(a)A + Chr)	0.98	0.97	0.96	0.95	0.00	1.00	0.97	0.97	0.94	0.00	0.00-1.00	0.77
Flt/(Flt + Pyr)	0.82	0.09	0.05	0.68	0.77	0.19	0.28	1.00	0.29	0.50	0.05-1.00	0.47
I(cd)P/(I(cd)P + B(ghi)P)	0.00	0.59	0.89	0.00	0.00	0.04	0.89	0.10	0.30	0.00	0.00-0.89	0.28

Source: *Field Data (2018)*

Conclusion

On the average, it was found that the L-VTAs recorded a higher concentration of PAHs than the H-VTAs. Both areas showed a predominance of Benzo(a)Anthracene, Pyrene, and Benzo(a)Pyrene, in that order. The mean concentration of identified Benzo(a)Pyrene was higher when compared with other background soils of different countries. It was also found that petroleum sources were the most contributory sources of PAHs. It is therefore required that the regulatory body for environmental protection and its related institutions in Ghana make sure the existing laws for controlling pollution in fuel filling stations are well monitored and evaluated, as well as ensuring that drivers are using vehicles that are in good conditions or well maintained, thereby posing less harm to public health.

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