IJST EXPLORE International Journal of Science and Technology
CENTRE OF PROFESSIONAL RESEARCH PUBLICATIONS
ISSN 2049-7318

International Journal of Science and Technology Volume 3 No. 8, August, 2014

Heavy metals and black carbon assessment of PM_{10} particulates along Accra–Tema highway in Ghana

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ABSTRACT

Inhalable particulate matter, Black Carbon(BC) and heavy metal levels in the ambient air of the Accra – Tema highway in Ghana was assessed. Using IVL sampler and Sierra Anderson (GAST) pump, airborne particulates were sampled on Teflon filters from November to December, 2011. Elemental and black carbon concentrations were determined using ICP – AES and Smokestain Reflectometer respectively. The average concentrations of PM_{10} and BC were 86.97µg/m^3 and 17.82µg/m^3 respectively showing significant contribution of carbonaceous compounds to the ambient air particulates levels. The PM_{10} and BC levels varied significantly with the days of the week, with the weekends recording relatively lower levels. Elements of crustal origin were identified as the major elements while those of anthropogenic origins were present in trace quantities. Enrichment Factor model (EF) was used to identify species of crustal and non–crustal origin in the ambient air particulates. The Enrichment Factor values obtained for the elements V, Cr, Cu, Ni, Zn, As, Cd and Pb showed that they are mostly from anthropogenic sources.

Keywords: *PM_{10}, IVL sampler, ICP – AES, Smokestain Reflectometer, Black Carbon, Enrichment Factors.*

1. INTRODUCTION

The release of gaseous pollutants and air particulate matter into the atmosphere at concentrations high enough above their normal ambient levels have increased considerably as a result of increasing human activities [1]. This eventually has a measurable effect on humans, animals and plants. Particulate Matter (PM) which can be inhaled into the human respiratory system is related to most serious health effects including pulmonary and cardiovascular illness [2,3]. Particulate Matter (PM) with aerodynamic diameter less than 10µm (PM_{10}) also known to be inhalable particulates influence many atmospheric processes including cloud formation, visibility, solar radiation, precipitation and plays a major role in acidification of clouds, rain and fog [4]. Inhalable particulate pollution in the atmosphere originates from sources of both natural and anthropogenic processes. Soil dust, biomass burning, industrial emissions and vehicular exhaust emissions are the major sources of the PM_{10} particulate. Incomplete combustion processes from motor vehicle, fossil fuel and biomass burning are some of the anthropogenic sources of PM_{10}.

PM_{10} particulate pollution is one of the major problems in major cities of developed world, and has also now become a serious and worsening situation in rapid growing cities in the developing world, especially in Africa due to urbanization and industrialization[5]. Accra is highly populated and proliferated with vehicles of all kinds causing congestion, vehicular traffic and increasing anthropogenic emissions, affecting ambient air particulate concentration. The largest harbour in Ghana is located at Tema, a coastal town at a distance of about 30 km due east from central Accra. Apart from the harbour, Tema is also home to many industries including, oil, steel, aluminium, textiles, cement, pharmaceuticals, food manufacturing etc. Accra and Tema are linked by an 18 km four-lane highway which forms part of the Trans West Africa Highway with very heavy vehicular density. The majority of these vehicles are either old or used imported vehicles, with weak and less efficient engines that release huge amounts of smoke (exhaust emissions) [6]. Nitrogen oxides (NOx), sulphur oxides (SOx), Carbon monoxide (CO), Black Carbon (soot), Organic Carbon, polycyclic Aromatic Hydrocarbons (PAH) and heavy metal elements are the common chemical contaminants in the vehicular exhaust fumes, which can cause serious environmental and health threats [1].

The composition of inhalable particulate matter is complex and differs depending on the source and location. The occurrence of Black Carbon (BC) and toxic heavy metals such as Pb, Zn, Cu, Fe and Ni in the inhalable particulates contributes to substantial health effects [3]. Some of the heavy metals in the particulates are strong triggers of carcinogenesis, teratogenesis and mutagenesis [7]. Toxic metal elements present in air particulate matter are usually transported through long distances, causing widespread distribution of their environmental and health effects [8]. They accumulate in organisms as a result of direct intake from surroundings through respiration and ingestion. Black Carbon (BC) is a climate forcing agent (causes global warming) formed through incomplete combustion [9].

Some aerosol studies in Ghana have looked at the emissions from industry and domestic biomass burning[10] and their impact on the ambient air quality. So far, not much has been done in aerosol studies. This work assesses the levels of heavy metals and Black Carbon (BC) content in PM_{10} and the possible sources along the Accra-Tema highway.

2. MATERIALS AND METHODS

2.1 Site Selection and Sample Collection

The study was carried out at the Accra-Tema motorway which is a four lane highway and the main link road between the port city of Tema and Accra the capital city. The highway is about 18 km link road and forms part of the Trans-West Africa Highway. This road is characterized by both light and heavy-duty vehicles for most part of the day. It is noted for high-speed vehicular movements with toll booths at both ends, which serve as the only means of slowing down vehicular movements. This road was chosen because of the large vehicular traffic with residence and peasant farming spread alongside. The sampling site (toll booth closer to Accra) labeled in Fig 1 as A is about 3 km from the Tetteh Quarshie interchange.

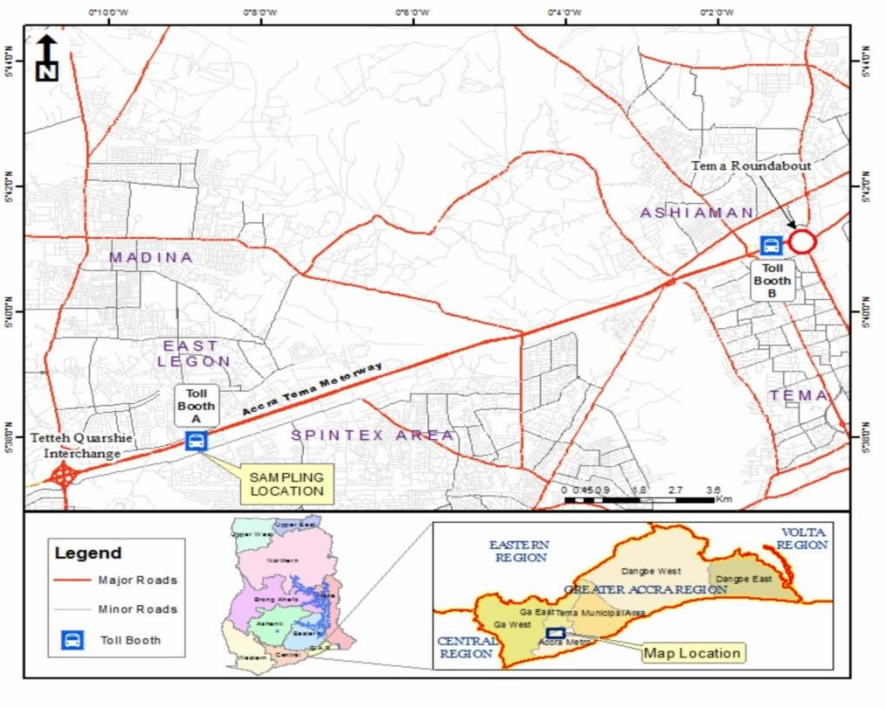

Figure 1: Geographical map of the sampling area showing the sampling site

There are very little variations in temperature at the study area throughout the year. The mean monthly temperatures range from 24.7°C to 28°C with an annual average of 26.8°C. The relative humidity levels are generally high, varying from 65% in mid-afternoon to 95% at night. The predominant wind direction at the study area is towards the NNE sector. Wind speeds normally range between 2.2 to 4.4 m/s.

Teflon filters of 47 mm diameter were used to sample the PM_{10} particulates. Teflon filters are well noted for their low chemical background, low affinity for water vapour and not affected by most acidic and alkaline media. Both field and blank filters were equilibrated to constant temperature and relative humidity in a desiccator for a period of not less than 24 hours. The filters were weighed using a Sartorius MC-micro-gram sensitive balance with a weighing accuracy of ± 5 µg in a temperature and relative humidity controlled environment.

The ambient air PM_{10} particulate sampling was carried out using IVL sampler and Sierra Anderson (GAST) pump. The sampler was mounted at the toll booth on Accra-Tema highway (shown in figure 1 and indicated as **A**). The sampler was operated at a flow rate of 16.7 L/min. Daily sampling was carried out for 24 hours for 28 days, from November to December, 2011. The loaded filters were also subjected to the same constant temperature and humidity conditioning for 24 hours before weighing for the gravimetric analysis.

2.2 Sample Analysis

Gravimetric analysis was performed to determine the mass concentration of the sample aerosol in micrograms per cubic meter (µg/m^3). The mass concentration of the aerosol samples were calculated by dividing the weight of the particulates captured on the filters by the volume of air (at ambient conditions) that passed through the sample filters [11] as shown in the equation below;

$$C_{PM} = M_p / V_a$$

Where:

C = mass concentration of particulate matter (µg/m^3)
M_p = Total mass of PM_{10} collected on the sample filter (µg)
V_a = Total air sample volume determined from the total volumetric flow rate (L/min) and sampling time (min).

2.3 Black Carbon Analysis

Black carbon is the light absorbing part of the carbonaceous aerosols. The reflectometer (Model 43D, Diffusion Systems Ltd, London) was used for the black carbon analysis in this work [12]. The operating principle of a reflectometer is known as the 'black smoke method'. A light source shines its light on an aerosol filter, and the reflected light is measured by photodiodes located in a black housing. The light source is a high-performance LED with maximum emission at 650 nm. The reflector reading is first converted to output voltage. The output voltage is converted to a measure of blackness, the 'black smoke number', which is uniquely determined from the three output voltages obtained from (1) the aerosol filter to be evaluated, (2) a filter which is totally black and (3) another filter which is totally white. Provided that thin layers of aerosol particles are collected on the filter (a single dust layer), there is a simple relationship between the concentration of black particles collected on the filter and the three output voltages. The 'black smoke number' can be related to the concentration of black aerosol by use of the Lambert–Beer law [12]. Five Reflectance readings (output voltage readings) were taken from each filter and the average used to compute the black carbon concentration. Provided that thin layers of aerosol particles are collected on the filter (''a single dust layer''), the equation relating the output voltages to the "black smoke number" is:

$$RZ = RZ_{max}(U_{RZ0} - U_{RZ})/(U_{RZ0} - U_{RZmax}),$$

where, U_{RZ0}, U_{RZ}, and U_{RZmax} are the output voltages of the total white filter spot, the actual filter spot, and the total black filter spot respectively. RZ0, RZ, and RZmax are ''black smoke numbers'' for these three types of filters, respectively[13]. The equation used for converting ''black smoke number'' to concentration of black particles (C) is:

$$C = -(RM/V) \ln(1- (RZ- RZ0)/kRZmax),$$

where V is the sampled air volume, *k* and RM are calibration constants. Here, *k* = 0.95,

RM = 11.2 µg, U_{RZmax} = 0.5, URZ0 = 8, RZmax = 9 were used. Finally, C is adjusted by a multiplication constant (the ratio of the filter area to the black spot area) to get the total BC concentration[13]. Regular linearity checks were performed using a white/grey standard which comes with the instrument.

2.4 Elemental Analysis

The rings on all the Teflon filters were first carefully detached from the loaded area. The rings were weighed and the mass subtracted from that of the initially loaded filter to obtain the mass of the particulates together with the filter. The loaded Teflon filters and one blank filter, all detached from their rings were each digested with 4 ml HNO_3 (65%) and 1 ml H_2SO_4 for 30 minutes using Ethos 900 Milestone Microwave Digester.
Three point calibration standard solutions were prepared from a 250 ml CPI-International multi-element standard containing 21 elements in 1% HNO_3. The concentration of each element in the standard was (1000±3) mg/ml. The calibration solutions were prepared by measuring 1ml, 0.6 ml and 0.3 ml of the standard and dissolving in 100 ml distilled water each to obtain solutions of 10 mg/ml, 6 mg/ml and 3 mg/ml concentrations respectively. To obtain optimum analytical signal, the following plasma settings or optimum analytical conditions were made: RF Power-1150 W, Peristaltic Pump Rate–100 rpm, Auxiliary Gas Flow–0.5 L/min and nebulizer flow–30 psi.
The solutions obtained after acid digestion were analyzed using ICP-AES. Fourteen (14) elements being As, Zn, Cu, Co, Ni, Ca, Mg, Mn, Na, Pb, V, Cd, Cr and Fe were identified and quantified.

2.5 Enrichment Factor (EF) analysis

Enrichment Factor (EF) model was used to identify species (elements) of natural and anthropogenic sources in PM_{10} particulates on the highway.
The Enrichment Factor (EF) was calculated using the following equation:

$$\mathbf{EF_{crust} = (Tr/Ref)_{pm} \quad (Tr/Ref)_{crust}}$$

Where $(Tr/Ref)_{pm}$ is the concentration ratio of elements and reference elements in the PM_{10} particulates. $(Tr/Ref)_{crust}$ is the concentration ratio of elements to reference elements in crustal materials. For this work Fe was used as the reference element and EF calculation was based on the average upper continental crust composition given by Taylor and McLennan (1985)[14]. Elements were categorized as "non-enriched," "moderately enriched," and "enriched" if their average enrichment values are less than 3, between 3 and 30, and above 30, respectively [15]. By convention, an arbitrary average EF value of <10 indicates that a trace element in an aerosol has a significant crustal source, and in contrast, an EF value of >10 is considered as a significant proportion of an element with a non–crustal source [16, 17].

3. RESULTS AND DISCUSSION

3.1 Particulate Levels

The average concentration of PM_{10} particulates was found to be (86.97±18.65) µg/m^3. The highest and the lowest concentration levels were 120.64 µg/m^3 and 54.86 µg/m^3 respectively. The average PM_{10} particulate mass concentration of 86.97 µg/m^3 exceeded the WHO air quality guidelines value of 50 µg/m^3.

The mean PM_{10} Particulate levels for the days of the week are shown in figure 2. The weekends (Saturday and Sunday) recorded less particulate levels and vehicular traffic as compared with the working days. Variation in vehicular density on the highway could be the major contributing factor to the observed differences in particulate levels. Variations in emission from the

other sources and meteorological conditions (humidity, wind speed and wind direction) could also contribute to the observed differences.

Aboh *et. al.*, 2009 reported a daily mean PM_{10} concentration of 179 µg/m^3 from February 2006 to February 2007 at Kwabenya in Accra. Dotse *et al.*, 2012[5] reported a daily mean of 96.56 µg/m^3 at Ashaiman near Tema from February to May, 2008 (dry season). The mean levels obtained in these works are seen to be higher than that of this work because the sampling time included the Harmattan period when there is high deposition of dust from the Sahara desert. The high deposition rate of fugitive dust and combustion emissions mostly from bush burnings during the Harmatten season could account for the relatively high levels[18]. Rozos and Christides, 2010[19] reported a daily average PM_{10} concentration of 67 µg/m^3 in an industrial area of Elefsis in Greece. This was much lower than the average PM_{10} level in the ambient air on the Accra-Tema highway. The great number of unpaved link roads within Accra and Tema resulting in significant re-suspended dust particulates could also contribute to the difference. Construction works on roads and buildings, which are common in this area, may have also contributed to the average PM_{10} particulate level. Other possible anthropogenic contributions are refuse and biomass burnings close to the sampling site.

Table 1: Mean concentrations of PM_{10}, BC and % BC in the ambient air

	Mass concentration			
Particulate	Mean	SD	Max	Min
PM_{10} (µg/m^3)	86.97	18.65	120.64	54.86
BC (µg/m^3)	17.82	2.94	21.82	10.63
% BC	21.08	3.48	30.69	15.72

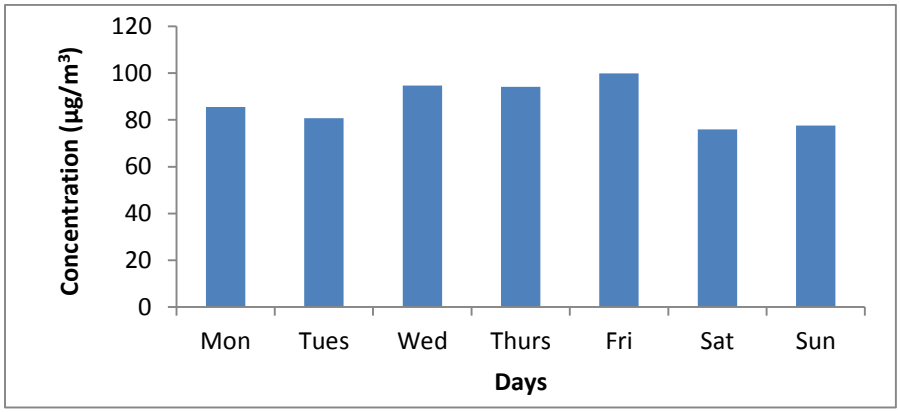

Figure 2: Mean PM_{10} Particulate levels for the days of the week.

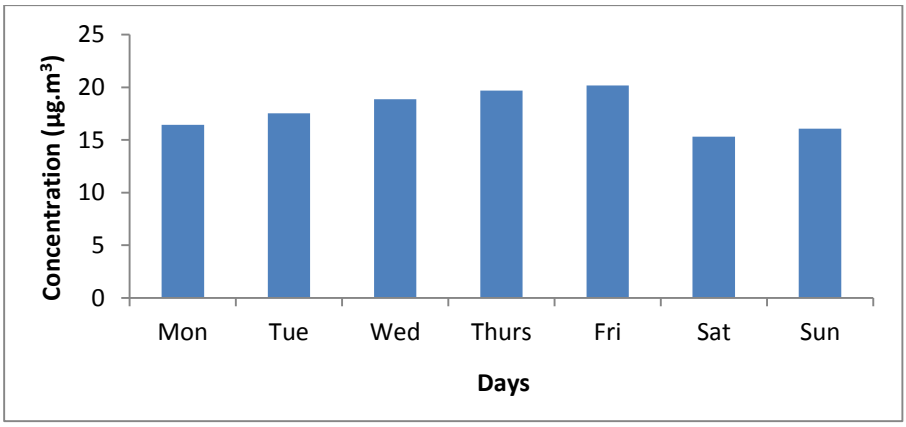

Figure 3: Mean Black Carbon levels for the days of the week

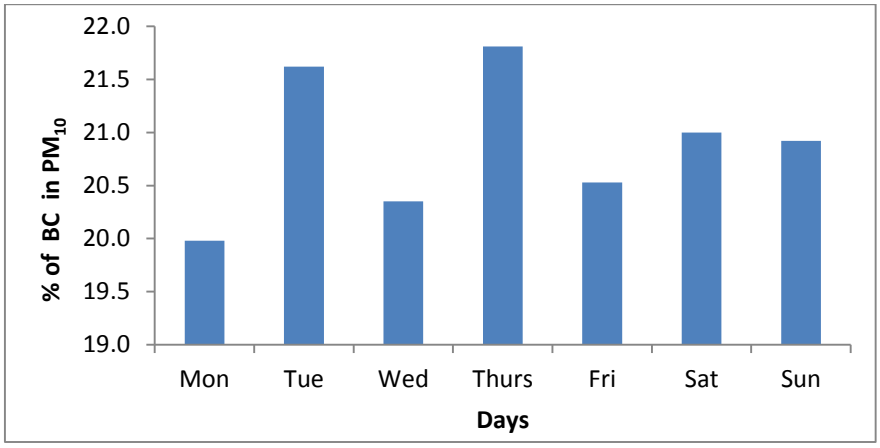

Figure 4: Percentage BC in PM_{10} particulate for the days of the week

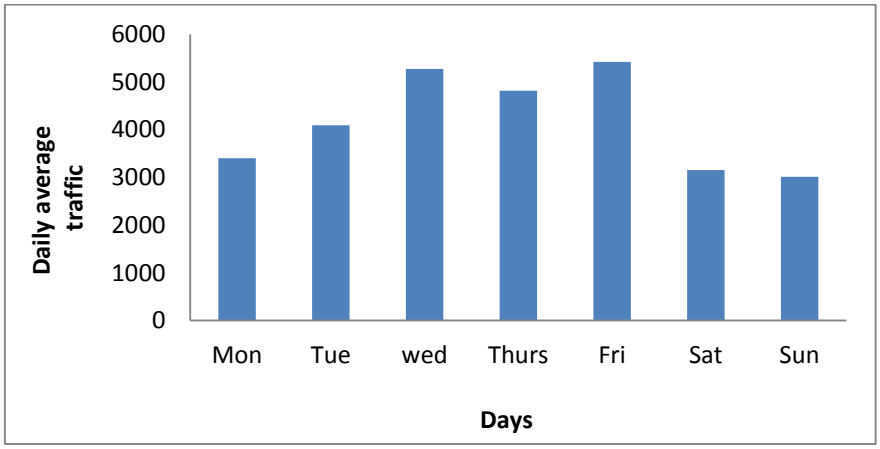

Figure 5: Mean vehicular traffic for the days of the week

Table 2: Daily average vehicle size count as percentage of total traffic count on the highway for the days of the week.

Days	Vehicle size fraction (%)		
	Small	Medium	Big
Monday	42.3	56.4	1.4
Tuesday	69.7	28.5	1.8
Wednesday	63.0	35.6	1.3
Thursday	67.8	30.6	1.5
Friday	64.1	35.0	0.8
Saturday	60.2	38.0	1.7
Sunday	49.7	48.7	1.6

Where, Small vehicles = vehicles with 1.3-2.4 litre or 100-173 horsepower engine.
Medium Vehicles = vehicle with 2.5-4.6 litre or 180-300 horsepower engine.
Big Vehicles = vehicles with engine capacity and power above 4.6 litre and 310 horsepower respectively[20].

The levels of BC and percentage of BC in PM_{10} obtained are presented in figures 3 and 4 respectively. The Black Carbon (BC) particulate concentration levels and percentage BC in PM_{10} also varied for the different days of the week. Similarly, the weekends recorded lower BC levels as compared with the working days. It can be seen from figures 2, 3, and 5 that Friday recorded the highest levels of PM_{10}, BC and vehicular traffic. However, it had the least value of percentage BC in PM_{10} (Fig.4). The least value in size fractions for the big vehicles as seen in Table 2 was recorded on Friday. This shows that diesel engines which mostly power big vehicles contribute greatly to BC emissions. The average Black Carbon (BC) concentration level was 17.82 $\mu g/m^3$. Aboh *et. al.*, 2009[21] reported a daily mean BC concentration of 4 $\mu g/m^3$. This value was relatively low because the sampling site was not close to any major road. On the average, 21% of the PM_{10} particulates were found to be made up of Black Carbon. This shows the significant contribution of exhaust emissions from heavy trucks, tractors and large buses (diesel engine vehicles) to PM_{10} particulates on the highway as the sampling site is about 18 km from the heavy industrial site at Tema. The 21% BC contents in the PM_{10} particulates reveals the high influence of carbonaceous compounds (Organic Carbon and black Carbon) on the ambient air quality.

Fourteen elements: As, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mn, Na, Ni, Pb, V and Zn were determined from the elemental analysis. Table 3 presents the summary of the average concentrations in $\mu g/m^3$ of the elements. Cd and Co recorded the least concentrations ($0.007 \mu g/m^3$) while Fe gave the highest ($5.596 \mu g/m^3$). The overall order of decreasing elemental concentration is Fe>Ca>Na>Mg>Ni>Cr>V>Zn>Mn>Cu>As>Pb>Cd and Co. Fe and Ca which are mostly of crustal origin were present in relatively high in concentration. Na, which could be traced from both crustal and sea spray also had relatively high concentration. These elements could also have some anthropogenic origins. Fe, aside from crustal origin could partly be coming from brake wear and muffler ablation[10]. The use of Pb containing anti-knocking gasoline additives played a dominant role in the buildup of atmospheric lead levels before the change over to methylcyclopentadienyl manganese tricarbonyl (MMT) in Ghana [22,23]. The mean level of Pb observed was lower than the WHO guidance level of 0.5 $\mu g/m^3$ (annual). The mean level of Pb obtained in this work (11 ng/m^3) compares well with the 10.2 ng/m^3 obtained by Aboh *et. al.*, 2009[22]. These low levels of Pb in the ambient air have shown the positive impact of the ban of leaded gasoline in Ghana in the year 2004 on air quality. Cd and Ni could be coming largely from vehicular emissions [24,25]. Zinc in the form of zinc diethyldithiophosphate and Ca are used in lubricant additives (motor oil)[10,26]. These are likely to be released into the air from old and weak vehicle engines, particularly those with leaky piston rings that burn more oil. Crustal source and industrial emissions however could also contribute to the level of Zn.

Table 2: Mean elemental concentrations in PM_{10}.

Elements	Mass concentration ($\mu g/m^3$)			
	Mean	SD	Max	Min
As	0.059	0.031	0.147	0.012
Ca	4.249	2.729	8.502	1.975
Cd	0.007	0.009	0.029	0.001
Co	0.007	0.019	0.071	0.001
Cr	0.115	0.016	0.157	0.092
Cu	0.075	0.091	0.377	0.018
Fe	5.596	1.305	9.094	3.754
Mg	1.34	1.041	2.903	0.011
Mn	0.085	0.012	0.119	0.066
Na	4.172	3.386	9.613	1.185
Ni	0.218	0.071	0.423	0.137
Pb	0.011	0.017	0.051	0 .005
V	0.102	0.081	0.263	0.028
Zn	0.091	0.043	0.209	0.036

The mean annual threshold standards set by the European Union for As, Cd and Ni are 6 ng/m^3, 5 ng/m^3 and 20 ng/m^3 respectively[24]. The levels of As, Ni and Cd in this work were higher than the mean concentrations threshold limits set by the European Commission. This could be due to the high levels of vehicular emissions mostly from old and less efficient engines, industrial processes and biomass burning. Break wear could also contribute to Ni in ambient air. Additionally, the average Mn level of 84 ng/m^3 was found to be lower than the World Health Organization air quality guideline of 150 ng/m^3[27]. This shows that the buildup of manganese from vehicular emissions as a result of the use of MMT as a gasoline octane enhancer in Ghana since its inception in 2004 has not yet had any significant effect on the natural background level[10,22]. Manganese is known to be an essential element for human survival, but it is also toxic when high concentrations are present in the body[28].

3.2 Enrichment Factor (EF)

The separation of natural and anthropogenic source components is one of the basic tasks of aerosol measurements. Enrichment Factor (EF) analysis can be used to identify soil derived elements from anthropogenic components. To verify the crustal and anthropogenic source contributions associated with PM_{10} particulates, EF was calculated for each element. EF calculations were based on the average upper continental crust composition given by Taylor and McLennan (1985)[14] with Fe being the reference element. Fe was used because of its high crustal origin. The results of the Enrichment Factors (EF) of metal elements are shown in table 3. The EF of the metal elements ranged from 0.57(Mg) to 329.76(As). Mg and Ca which are known to be

mostly of crustal origin were found to be non-enriched. The non-enrichment of Mn confirms that the Mn concentration is within the background levels. The moderate enrichment of V and Pb show the significant anthropogenic contributions. Some toxic and harmful elements, such as Cr, Cu, Ni, Zn, As and Cd are enriched thus showing their anthropogenic origin[29].

Table 3: Enrichment Factors (EF) of metal elements

Elements	Mg	Mn	Fe	Ca	Na	Co	V	Pb	Cr	Zn	Cu	Ni	As	Cd
EF	0.57	0.9	1.0	1.03	1.77	2.82	7.6	8.85	11.56	13.07	13.72	29.24	325.76	325.12

4. CONCLUSION

This study has looked at the ambient air PM_{10} particulate levels on a major highway linking Accra and Tema which also forms part of the Trans West Africa Highway. The average mass concentrations of PM_{10} and BC particulates were found to be 86.97µg/m^3 and 17.82 µg/m^3 respectively. The levels of both PM_{10} and Black Carbon (BC) were found to vary significantly for the different days of the week. PM_{10} mean value exceeded the WHO guideline values of 50 µg/m^3. Variation in vehicular density on the highway contributed greatly to the observed differences in particulate levels on different days. Black Carbon was found to make up approximately 21% of the PM_{10} particulate in the ambient air showing the high contribution of carbonaceous compounds (Black Carbon and Organic Carbon) to air quality along the highway. Results of the enrichment factors showed that the elements V, Cr, Cu, Ni, Zn, As, Cd and Pb present in the ambient air are mostly of anthropogenic origins.

Acknowledgement

Our profound gratitude goes to the Chief Executive Officer of Ghana Highway Authority and the Managing Director of AD & TS for allowing us to mount the sampler at the Accra – Tema toll booth. Our sincere thanks also go to the staff of Tema Oil Refinery (TOR) for their permission to use their facilities at the Quality Control Laboratory.

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