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Spatio-Temporal Variation of PM-bound Heavy Metals Along a Major Motorway in Ibadan, Nigeria

Akinola, O. I.[1], Ana, G.R.E.E[1], and Adie, G. U.*[2]

[1]Department of Environmental Health Sciences, Faculty of Public Health, College of Medicine, University of Ibadan, Ibadan, Nigeria

[2]Department of Chemistry, Faculty of Sciences, University of Ibadan, Ibadan, Nigeria

E-mail: gildie1975@gmail.com

*Corresponding Author: Adie, G.U., as above

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Abstract

Traffic-related particulate matter (PM_{10}) contributes extensively to ambient air pollution and are reported to be associated with respiratory illnesses. Populations living along major urban motorways are at higher risk. This study evaluated levels of PM_{10} bound heavy metals along Idi Ape - Iwo busy motorway in Ibadan. Five sampling locations 150 metres apart were purposively selected along the road, namely: Abayomi Area (AA), Iyana Agbala (IA), Barracks Area (BA), Agbaakin Layout (AL) and Holiness Junction (HJ). Air samples were collected at each location at peak periods (7-10am and 1-4pm) for 10 weeks. Traffic density was estimated via manual counting. Filters were digested using standard methods and extracts were analysed for Pb, Fe, Cu, Cd, Zn, Cr and Ni using atomic absorption spectrophotometry technique. Mean traffic density for all locations and vehicles ranged from 992 – 21149. Daily mean PM_{10} for all sampling locations ranged from 84.6±41.7 - 122±60µg/m^3, with levels at least three times higher than WHO guideline limit of 20µg/m^3. Mean metal concentrations (mg/m^3) for all locations were: Pb- 279±149, Fe-2.20±1.97, Cu - 0.0178±0.0090, Cd - 138±37, Zn - 0.428±0.196 and Cr - 0.179±0.06 with levels higher in order 10^2 – 10^7 in magnitude. This suggests a hazardous PM_{10}. There was no regular trends in all parameters in time and space within the study period. Mean metal concentrations increased proportionally with traffic density for all sampling locations. Policies on traffic-related pollution become pertinent to reduce these slow killing pollutants associated with dust and exhaust fumes from motorways in Nigeria.

Variation spatio-temporelle des métaux lourds de PM, le long d'une autoroute majeure d'Ibadan, au Nigéria

Abstrait

Les particules liées à la circulation (PM10) contribuent largement à la pollution de l'air ambiant et seraient associées à des maladies respiratoires. Les populations vivant le long des principales autoroutes urbaines sont plus à risque. Cette étude a évalué les niveaux de métaux lourds liés aux PM10 le long de l'autoroute à grande circulation Idi Ape - Iwo à Ibadan. Le long de la route, cinq lieux d'échantillonnage distants de 150 mètres ont été choisis à dessein, à savoir: les zones Abayomi (AA), Iyana agbala (IA), les zones Barracks (BA), Agbaakin Layout

(AL) et Holiness Junction (HJ). Des échantillons d'air ont été prélevés à chaque endroit aux périodes de pointe (7h à 10h et de 13h à 16h) pendant 10 semaines. La densité de la circulation a été estimée par comptage manuel. Les filtres ont été digérés en utilisant des méthodes standard et les extraits ont été analysés pour Pb, Fe, Cu, Cd, Zn, Cr et Ni en utilisant une technique de spectrophotométrie d'absorption atomique. La densité moyenne de la circulation pour tous les lieux et véhicules variait de 992 à 21149. La moyenne journalière de PM10 pour tous les lieux d'échantillonnage variait de 84,6 ± 41,7 à 122 ± 60µg / m3, avec des niveaux au moins 3 fois supérieurs à la limite recommandée par l'OMS par 20µg / m3. Les concentrations moyennes en métaux (mg / m3) pour tous les sites étaient: Pb-279 ± 149, Fe-2,20 ± 1,97, Cu - 0,0178 ± 0,0090, Cd - 138 ± 37, Zn - 0,428 ± 0,196 et Cr - 0,179 ± 0,06 avec des niveaux plus élevé dans l'ordre 102 - 107 de magnitude. Cela suggère un PM10 dangereux. Il n'y avait pas de tendance régulière dans tous les paramètres dans le temps et l'espace au cours de la période d'étude. Les concentrations moyennes de métaux ont augmenté proportionnellement à la densité de la circulation pour tous les lieux d'échantillonnage. Les politiques en matière de pollution liée à la circulation deviennent pertinentes pour réduire ces polluants à mort lente associés à la poussière et aux gaz d'échappement des autoroutes au Nigéria.

Introduction

Every year, millions of tonnes of toxic pollutants are emitted into the air, both from natural sources and anthropogenic sources. It has been reported that air pollution poses the biggest risk factor responsible for deaths across the globe with European Union population reaching 92% in urban environment being exposed to high levels of both $PM_{2.5}$ and PM_{10} (EEA, 2014, 2016). There are four categories of emission sources: stationary (industrial processes and domestic combustions); mobile (road and stationary traffic); natural (volcanic eruptions, forest fires) and accidental (discharges, industrial fires) (US EPA,2014). In most cases, heavy metal pollution is usually associated with these pollution sources, especially in densely populated industrialised areas. Chromium, Cobalt, Nickel, Lanthanum, Zinc, and Molybdenum have been identified as the most common heavy metals emitted by vehicle traffic, totaling at least 77% of the total mass of 43 metals emitted (Chen *et al.,* 2013).

Kelly and Fussell (2015) and Adamiec (2017) indicated that high vehicular traffic from both exhaust and non-exhaust sources has proven to be one of the major contributing sources to air pollution problems. Furthermore, Wrobel *et al*. (2000) indicated that traffic associated emissions could account for over 50% of the total particulate matter in the urban areas. A study by Buckeridge *et al*. (2002) linked particulate matter especially $PM_{2.5}$ with most admissions for respiratory-related problems in the United States.

High amounts of primary (CO_2, NO_X, SO_2, particulate matter, heavy metals) and secondary pollutants (smog, acid generation radicals, etc) have been reported to be released into ambient air from traffic-related activities(Ekwumemgbo *et al*., 2016). Environmental pollution by heavy metals from automobiles has received much attention in the recent past particularly because of increasing release of traffic-related Particulate Matter (PM) as a result of increasing number of vehicles on the roads and deleterious effects on human health and the environment (Davis *et al.,* 2006).

Studies reported in literature indicate general levels of heavy metals in ambient air without detailing the phases in which these metals exist. There is paucity of data on point sources of heavy metals in ambient air. Therefore, this study assessed the spatio-temporal distribution of PM_{10} – bound heavy metals along the busy Idi-Ape – Iwo road motorway.

Materials and Methods

Overview of Ibadan City

Ibadan is the capital of Oyo State in Nigeria. It is the third largest city in Nigeria in terms of population and one of the largest cities (metropolitan area) in West Africa, with about 3.5million inhabitants. Ibadan is an old, primarily indigenous African city situated $7^{0}24$', 7.0632 "N and $3^{0}55$' 2.3268"E. Ibadan covers a large land area of about 12 kilometres at an altitude ranging from about 150 to 200 metres with isolated ridges and peaks reaching 270 metres. Ibadan presents a typical picture of many African cities each known for having the old town area (inner core) and then the transitional and peripheral areas. A greater percentage of the people are of the Yoruba tribe while other tribes like Igbo, Hausa, etc are of a smaller percentage but peacefully coexist with them (Official website of Ibadan, 2019).

Description of sampling points

Five sampling locations were purposively selected along Idi-Ape - Iwo road motorway which is one of the busiest roads in Ibadan metropolis. The distance covered was over 750 metres. The sampling locations were 150 metres apart. They included: Abayomi area (AA), Iyana-Agbala (IA), Barracks Area (BA), Agbaakin Layout (AL) and Holiness Junction (HJ). At each sampling location, PM_{10} was measured at 10metres, 20metres and 30metres away from the main road to study the distribution of PM_{10} and PM_{10}-bound metals with distance. The summary of sampling location description is presented in Table 1.

Traffic density determination

The traffic density was determined by adopting the manual counting method by Abam and Unachukwu (2009). This involved counting manually the number of cars, buses, motorcycles and trucks that passed through the sampling points for a period of 10 minutes and this was recorded on a prepared tally sheet while stopwatch was used for timing. This exercise was carried out for 10 weeks. The data obtained were used to estimate the hourly traffic density. The data were obtained with the help of field assistants.

Sample collection and preparation

Particulate Matter (PM_{10}) samples were collected using Spectro fine particulate sampler model SLE-FPS105 (ICS, India) at three sampling points (10, 20 and 30metres) along a flat plain

Table 1: General description of sampling locations

Indicator	AA	IA	BA	AL	HJ
Location of sampling points					
Residential area	+	+	+	+	+
Commercial area	+	+	+	+	+
Industrial activities	-	-	-	-	-
Nature of road network					
Tarred roads	-	+++	-	-	-
Untarred roads	+++		+++	+++	+++
Activities around sampling points					
Refuse burning	++	++	++	++	+++
Emission	+++	++	++	+++	+++
Dumpsite	++	+	+	++	+
Generator emission	+++	++	+++	+	+++
Bush burning	-	-	-	-	-
Road construction	-	-	-	-	-

Key: +++ highly present + + moderately present + present - absent
AA-Abayomi Area, IA-Iyana Agbala, BA-Barracks Area, AL-Agbaakin Layout, HJ - Holiness Junction

perpendicular to the Idi-Ape – Iwo road motorway into the community to determine how far PM_{10} can travel. The sampler was fitted with an inlet designed to provide a clean aerodynamic cut-point for particles greater than 10 µm. The sampling rate was $1m^3$/hr. Cellulose membrane filters were used as the sampling media (1.0 µm pore size, 47 mm diameter).The filters were dried to constant weight and stored in a desiccator prior to sample collection.The set up comprised fitting a filter paper to the filter holder of the sampler with the aid of a forceps for each sampling point and sampler switched on. After sampling, all filters with particulates were stored in a desiccator prior weighing. According to Begum *et al*., (2007), the filters were equilibrated in a desiccator for at least 24 hours to remove any moisture absorbed before being weighed. PM_{10} was calculated using equation 1.

$$PM_{10}(\mu m/m^3) = \frac{(Wf - W_i)}{V_{std}} \times 10^6 \quad ....(1)$$

Where: PM = mass concentration of particulate matter (TSP, fine or coarse friction), µg/m^3.

W_i = average initial weight of clean filter, g.

W_f = average final weight of exposed filter, g.

V_{std} = air volume sampled (m^3.

10^6 = conversion of g to µg

Determination of heavy metals

PM_{10}-bound heavy metals collected on the filter papers were determined by adopting the USEPA—Method IO-3.1 (US EPA, 1999). Each filter was firstly cut into tiny fragments using a stainless cutter to increase the surface area for faster digestion. The metals were extracted from the filter by digestion with 50 mL of aquaregia in a digestion vessel using a temperature controlled hot plate set at 95±5°C. The sample was heated at 95±5°C without boiling for dissolution of the PM under refluxing condition. After the filter paper was observed to be completely dissolved, the solution was cooled. The digestate was filtered through Whatman No. 41 filter paper and extract was collected in a 100mL volumetric flask. The filter paper was then washed while still in the funnel, with no more than 5 mL of hot (~95°C) HCl, then with 20 mL of hot (~95°C) deionized water. Washings were collected in the same 100mL volumetric flask. Filter with residue was removed from the funnel and inserted in the digestion vessel and 5 mL of concentrated HCl was added and the vessel was placed on the heating source again and heated at 95±5°C until the filter paper dissolved again. Digestion vessel was removed from the heating source, the cover and sides were rinsed with deionized water .The solution was again filtered and collected in the same 100mL volumetric flask to have up to 90% to 99% recovery rate. The combined extract was allowed to cool, then diluted to mark.This was analysed for Pb, Fe, Cu, Cd, Zn, Cr and Ni using Atomic Absorption Spectrophotometer (205 Buck Scientific model, Buck Scientific, Inc. East Norwalk, CT, USA). This was repeated for all samples and Blank filter papers. The concentrations of metals in the blank were subtracted from the sample during the calculation. AnalaR grade reagents were used. Work spaces were always kept clean to avoid contamination. Glassware were soaked in dilute acid solution and rinsed properly before use. The silica gel used as drying agent was well regenerated (turned blue) before use to achieve maximum performance.

Results and Discussion

Summary of vehicular density in all locations

The summary of vehicular density in all locations is indicated in Table 1. The number of vehicles in descending order followed the trend: Cars> Motorcycles>Buses>trucks. The estimated overall total number of all vehicles that plied that motorway within the estimated period of 10 weeks was 36,357. The number of vehicles counted at each sampling location ranged from 3214 – 4378 (lowest for BA and highest for HJ). The elevated number of vehicles indicated at Holiness Junction (HJ) location could have been because it was a crossroad with other vehicles joining from other routes.There is a strong

Table 2: Summary of Vehicular Density in all Locations

Locations →	AA		IA		BA		AL		HJ		Overall Total
Types ↓	AM	PM	AM	PM	AM	PM	AM	PM	AM	PM	
Buses	440 ±196	673 ±161	499 ±54	489 ±120	518 ±231	703 ±243	447 ±285	615 ±100	671 ±120	745 ±226	5800
Cars	2101 ±244	1910 ±591	1829 ±609	2175 ±806	1915 ±567	2133 ±406	2001 ±887	2219 ±253	2383 ±487	2483 ±288	21149
Motorcycles	807 ±96	764 ±89	751 ±196	805 ±12	926 ±266	762 ±193	912 ±458	895 ±104	815 ±125	1080 ±472	8516
Trucks	71.3 ±49.0	51.3 ±8.0	84 ±51	52 ±35	53 ±44	356 ±527	78.3 ±8.4	70 ±24	107 ±58	69.3 ±40	992
Total	3422	3399	3214	3260	3500	3946	3688	3793	3884	4378	36457

N = 3 (mean of 3 measurements taken at 10, 20 and 30 metres into the community)

correlation (0.64, Table 4) between vehicular density and PM_{10} concentration within the sampling locations with PM_{10} level lowest at BA and highest at HJ. This suggests that traffic density could be a major contributor to particulate matter in ambient air.

Summary of PM_{10} in ambient air

The mean levels of PM_{10} (μm/m^3) ranged from 68.6 – 111 with lowest levels observed at Barracks Area (BA) while highest levels were observed at Holiness Junction (HJ) location. One of the reasons for increased PM in ambient air could be high traffic congestion. As a result, there are usually high releases of emissions from vehicular exhausts to the ambient air.

Furthermore, two stroke engines used by motorcycles have been reported to emit more pollutants to the air due to incomplete combustion (Martini *et al*., 2009). The high number of motorcycles may not be unconnected with the elevated levels of PM_{10} within the sampling locations. BA witnessed low PM level possibly because there was free flow of traffic within the vicinity as there were always 'no parking' and 'no waiting' sign posts and heavily armed soldiers available to control traffic. Mean PM_{10} levels for all sampling locations were higher by at least three folds compared to limit of 20 μm/m^3 set by WHO.

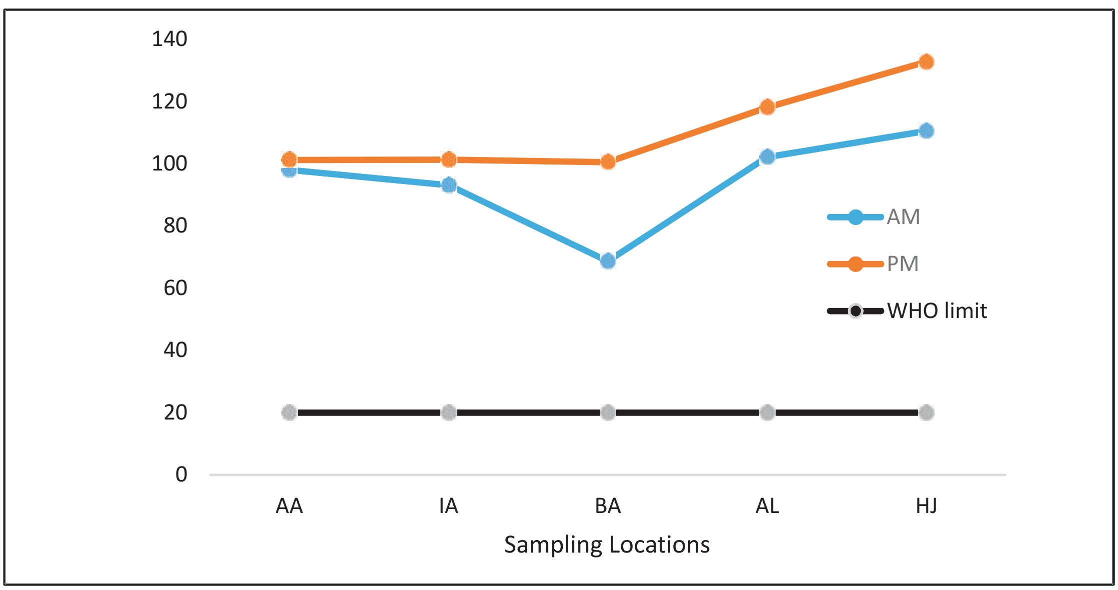

Figure 1: Mean levels of PM_{10} at peak periods at all sampling locations

Mean of PM_{10}–bound heavy metals at all sampling locations

The mean concentrations (mg/m^3) of Fe, Cu, Zn, Cr, Cd and Pb followed the trend: Pb > Cd > Fe > Zn >Cr > Cu. There was no regular trends in levels of all metals within the sampling locations in time and space (Table 3). The mean metal concentrations ranged from 0.665 – 5.00 for Fe, 0.009-0.042 for Cu, 0.215-0.718for Zn, 0.0083 – 0.0303 for Cr, 84.0 – 194 for Cd and 157 – 680 mg/m^3. These levels were higher than those reported in similar studies in literature (Singh *et al.* 2002, Martuzevicius *et al.*, 2004, Shridhar *et al.*, 2010). Most metals showed higher concentrations in HJ probably because of the 'move and halt' kind of movement at crossroads, as a result higher tear and wear of brake linings and other parts as well as higher emissions as the vehicles usually move at lower gears occur here. The source of Pb could be from leaded fuels while Cd and Fe could be from tearing and wearing of wheels and tyres which normally contain these metals. The mean concentration of Zn within the sampling locations (Table 3) were within 0.35 mg/m^3 reported in Athens (Valavanidis *et al.*, 2006), 0.46mg/m^3 in Delhi, India (Shridhar *et al.*, 2010), 0.0027mg/m^3 in Bilbao, Spain (Aranguiz *et al.*, 2002), 0.0098mg/m^3 Ponzone, Italy (Rizzio *et al.*, 2001) and 0.0046mg/m^3 , Paris, France (Ayrault *et al.*, 2010).

All metals indicated higher concentrations with varying magnitudes compared with WHO limits for ambient air quality (Table 3). This is an indication that policies on ambient air quality in Nigeria need to be made or where in existence, should be enforced.

Correlation studies

Correlation studies were conducted among the parameters at 95% confidence level. A strong correlation of 0.64 was observed between PM_{10} and vehicular density indicating direct influence of vehicular emissions on particulate matter in air. There is also a strong correlation between the following: Cu/PM_{10} (0.85), Cu/VD (0.79), Cu/Fe (0.91), Pb/PM_{10} (0.69), Pb/VD (0.78), Pb/Cu (0.93) etc as indicated in Table 4. These strong associations suggest common source of metals cum PM_{10}. There is always Pb arising from exhaust fumes of many vehicles and Fe, Cu, Cd and Cr from wear and tear of rubbing parts, tyres and brake linings.

Table 3: Mean distribution of PM_{10}-bound heavy metals (mg/m3) in air within the study locations

Location	Period	Fe	Cu	Zn	Cr	Cd	Pb
AA	AM	1.11 ±0.45	0.009 ±0.003	0.271 ±0.051	0.0123 ±0.0068	84.0 ±11.8	157 ±121
	PM	2.27 ±1.94	0.017 ±0.005	0.568 ±0.296	0.0237 ±0.007	132 ±44	221 ±145
IA	AM	0.698 ±0.126	0.013 ±0.007	0.217 ±0.060	0.0083 ±0.0061	178 ±66	255 ±56
	PM	2.37 ±2.69	0.017 ±0.006	0.403 ±0.185	0.0203 ±0.0068	151 ±15	338 ±141
BA	AM	0.665 ±0.320	0.014 ±0.010	0.345 ±0.165	0.0123 ±0.004	102 ±15	188 ±23
	PM	2.21 ±2.53	0.011 ±0.005	0.215 ±0.087	0.0163 ±0.0029	147 ±10	265 ±58
AL	AM	6.30 ±2	0.013 ±0.009	0.262 ±0.092	0.015 ±0.008	151 ±33	224 ±44
	PM	5.00 ±0.36	0.019 ±0.007	0.649 0.180±	0.0303 ±0.010	194 ±34	227 ±24
HJ	AM	0.676 ±0.082	0.023 ±0.009	0.635 ±0.319	0.0203 ±0.007	87.7 ±19.0	236 ±51
	PM	0.719 ±0.053	0.042 ±0.019	0.718 ±0.270	0.02 0.0094±	156 ±28	680 ±319
WHO Limit		0.01	0.01	0.04	0.005	0.000005	0.0005

Table 4: Pearson Correlation Studies

	PM_{10}	VD	Fe	Cu	Zn	Cr	Cd	Pb
PM_{10}	1							
VD	**0.64** (0.25)	1						
Fe	0.13 (0.82)	-0.23 (0.71)	1					
Cu	**0.85** (0.06)	**0.88** (0.048)	**0.91** (0.03)	1				
Zn	**0.95** (0.02)	**0.79** (0.11)	-0.067 (0.91)	-0.28 (0.64)	1			
Cr	**0.88** (0.47)	**0.57** (0.31)	0.47 (0.42)	**0.62** (0.26)	**0.84** (0.073)	1		
Cd	0.08 (0.89)	-0.29 (0.60)	**0.68** (0.21)	-0.15 (0.81)	-0.22 (0.72)	0.10 (0.86)	1	
Pb	**0.69** (0.19)	**0.78** (0.11)	-0.44 (0.44)	**0.94** (0.02)	**0.73** (0.15)	0.36 (0.55)	-0.07 (0.91)	1

p value in bracket $p \leq 0.05$ not significant   VD – Vehicular Density

Conclusion

Levels of PM_{10} and PM_{10}-bound heavy metals were determined in ambient air samples collected from selected locations along the busy Idi-Ape - Iwo road motorway to study the pollutants' spatio-distribution in air. Vehicular density was determined to assist in establishing the sources of these pollutants in air. There were no regular variations in the distribution both in time and space of the pollutants within the sampling locations. PM_{10} and all metals analysed were higher than WHO air quality limits.

Therefore, there is a strong need to enact air quality guidelines or where already existing their enforcement to safeguard human health and the entire ecosystem.

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