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Original Research Article

Assessment of Residential Indoor - Outdoor Airborne Particulate Matter in Ibadan, Southwestern Nigeria

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Quantitative information on mass and elemental concentrations and sources of indoor aerosols, examining the relationship between indoor and outdoor particulate matter (PM) in Ibadan City were provided. Mass concentrations of PM$_{2.5}$ and PM$_{2.5-10}$ and elemental composition were determined using standard weight and Instrumental Neutron Activation Analysis ($k_0 - INAA$) method. Nine elements viz: As, Br, Cu, K, La, Na, Sb, Sm and Zn were detected in the fractionated PM samples. The mean mass concentrations of PM loadings obtained for firewood, kerosene and cooking gas environments were 50.0, 22.2 and 22.7 μg/m^3 for PM$_{2.5}$ and 20.1, 24.3 and 9.0 μg/m^3 for PM$_{2.5-10}$ respectively. It was observed that the mean elemental concentration of potassium (K) in the fine fraction is approximately twice the values in the coarse fractions for kerosene residence and four times firewood residence. The high concentrations of Potassium in both fractions indicate that firewood is predominantly present in the study area. This has detrimental implications on the health of the populace.

Keywords: Particulate matter (PM), Indoor air pollution, Cooking fuel.

INTRODUCTION

Indoor airborne particulate matter has generally gained great attention in the recent years due to its large societal burden and various adverse effects (Zhang and Smith, 2007). Recent comparative risk study by the World Health Organization (WHO) revealed that 28 percent of the overall disease and deaths is caused by indoor air particulates in developing countries. This may be attributed to the fact that urban people spend on the average 87 percent indoors and only a mere 6 percent outdoors (Jerkins *et al.*, 1992). Apart from death, inhalation of particulate matter in the indoor environment can cause a range of adverse respiratory effects (Bai *et al.*, 2007; Dockery *et al.*, 1993; Dominici *et al.*, 2003).

Particulate matters in the indoor environment occur from a wide variety of sources, depending on the type of activities and processes taking place. Some activities such as sweeping and cleaning of floors have potential to generate dust through mechanical attrition of solid materials in the indoor environment. Also, combustion of fuels such as biomass, cooking gas and liquid fuels (Kerosene) for domestic heating, cooking and lighting purposes are considered as anthropogenic sources of indoor air pollution. Poppendieck *et al* (2010) and Pudpong *et al.*, (2011) confirmed in their studies that in an indoor environment without smoking, cooking and fuel- based lighting were the most significant sources of indoor

PM$_{10}$ concentrations while Siddiqui *et al* (2009) and Dionisio *et al* (2008) studies showed that the smoke generated from wood burning and biomass cook stoves produced indoor PM$_{2.5}$ concentrations that range from 101 to 8200 I g/m^3.

In any city, the choice of the fuel usage is greatly determined by the level of income of the people. Apart from these sources, an important process that contributes to the indoor air particulate concentration is the infiltration of the particulate from the outside environments and is greatly determined by the penetration of outdoor particles into the indoor environment. The behavior of indoor particulate is also affected by the structural system of a building, material characteristic, outdoor-indoor transport of particulates and activities of inhabitants. (Diapouli *et al.*, 2010).

In Nigeria, there have been several studies on airborne particulate most especially the outdoor in residential, industrial, workplace environment and vehicular traffic prone area. But studies of indoor PM have not been thoroughly studied compared to outdoors. However, few studies have characterized indoor particle size distributions (Ideriah *et al.*, 2001; Ajimotokan *et al.*, 2009; Adah *et al.*, 2010). Like in outdoor PM studies, the investigation of indoor PM in this work focused on both the physical characteristic based on size

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segregation (fine: $PM_{2.5}$ and coarse: $PM_{2.5-10}$) and elemental composition and concentration.

This study provides the mass and elemental concentrations of the indoor PM of some selected residential houses in the Ibadan metropolis with different fuels usage pattern for domestic purposes as well as source apportionment of sources that contribute to the indoor PM. The results of this study will also provide baseline information on indoor air PM in Ibadan and may help government officials in their efforts to improve policies regarding air pollution control in the city.

MATERIALS AND METHODS

The study was carried out in two densely populated residential sites viz; Olorunsogo and Alakia residential areas of Ibadan, Nigeria, from October, 2010 to January, 2011. Four receptor sites were chosen to cover various classes of people; sources of fuel for domestic cooking, traffic density and proximity to industries were avoided to minimize the domination of industrial emission.

Olorunsogo represent the areas where the low/average income citizens live and these classes of citizens are often prone to indoor and outdoor pollution associated problems due to combination of types of energy used, its usual high population density and poor indoor air management practices.

The four sites selected are located in densely and less densely populated residential areas of Ibadan city, as shown on the plate 1 below. The first and the second sites were kerosene using stove residence and outdoor using firewood in an open space. The third and fourth sites were cooking gas residence and open space where there is little or no activity. The sites were selected for particle monitoring based on particle generating activities, including cooking with different cooking fuels. The integrated indoor $PM_{2.5}$ and $PM_{2.5-10}$ were collected for 10 hours daytime period due to nature of activities.

Ten samples each of $PM_{2.5}$ and $PM_{2.5-10}$ were collected at three sites where there are major activities such as cooking with kerosene stove, outdoor cooking with firewood and cooking with gas and five samples each of $PM_{2.5}$ and $PM_{2.5-10}$ in one site where there is little or no activities.

The Gent Stacked Filter Unit Sampler (Hopke et al., 1997) was used for the collection of fine (< 2.5 μm) and coarse (2.5 - 10 μm) particles on two grades of Whatmann polycarbonate nucleopore filters of 0.4 and 0.8 μm pore size respectively was placed in a single indoor location adjacent to area of the home where the majority of cooking activities occurred, and where there was free air flow exchange. The samples were collected at nose-level height of about 1.6 m above ground level and at such places where free air circulation was not hindered.

Elemental analysis of the filter samples, the analytical procedure and counting (Almeida et al., 2010; Owoade et al., 2013) was performed using k_0 - Instrumental Neutron Activation Analysis (k_0-INAA) technique available at Portuguese Research Reactor.

Elemental masses were calculated using the k_0-INAA method and the National Institute of Standards and Technology (NIST) standard 1633a-Coal Fly Ash, which was co-irradiated with the samples, was used as quality control. Blank Nucleopore filters were treated as regular samples. All measured species in the blank were very homogeneously distributed; therefore loaded filters concentrations were corrected by subtracting the filter blank contents. The INAA technique has been optimized for the irradiation and quantification of As, Br, Cd, Cr, Fe, Na, Sb, Sn and Zn. Elemental composition of both $PM_{2.5}$ and $PM_{2.5-10}$ were inputed

to chemical mass balance (CMB 8.2) software for source identification and apportionment.

RESULTS

Mass Concentration of Particulate Matter (PM)

Figure 1 shows the summary of the descriptive statistics of the $PM_{2.5}$ and $PM_{2.5-10}$ concentrations during the study period. The minimum and maximum mean mass concentrations of $PM_{2.5}$ for indoor are 22.20 μg/m^3 and 50.00 μg/m^3 while that of $PM_{2.5-10}$ are 9.00 μg/m^3 and 24.29 μg/m^3 respectively. The overall mean mass concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ in the indoor micro-environments were 31.64 μg/m^3 and 17.80 μg/m^3 respectively. It was observed that PM mass concentration of wood was the highest with a value of 50.00 μg/m^3 and kerosene has the lowest mass concentration of 22.20 μg/m^3 of $PM_{2.5}$. Gas had the lowest mass concentration of 9.00 μg/m^3 and kerosene was the highest concentration with 24.29 μg/m^3 of $PM_{2.5-10}$. The mean mass concentration of $PM_{2.5}$ and $PM_{2.5-10}$ for outdoor was 53.61 μg/m^3 and 20.20 μg/m^3 respectively.

The chart reveals that the mass concentration of PM from firewood is 2.25 times that of kerosene while cooking gas is 0.37 times more than that of kerosene and the ratio of $PM_{2.5-10}$ to $PM_{2.5}$ outdoor is 0.4. The highest daily mean PM_{10} was observed at the firewood site with a value of 70.10 μg/m^3 while cooking gas site has the lowest value of 31.71 μg/m^3.

Figures 2 and 3 show the daily mass concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ for each site. For $PM_{2.5}$, the least concentration (12.11 μg/m^3) was observed in the kerosene while the highest (74.83 μg/m^3) was observed in firewood cooking sites for indoor environment. For $PM_{2.5-10}$, the lowest mass concentration of 2.03 μg/m^3 was observed for the cooking gas household users while the highest of (59.56 μg/m^3) was observed from kerosene cooking. The increased indoor PM mass concentrations recorded in fine for firewood and course for kerosene may be attributed to the high penetration of outdoor particles as shown in the line graph. The penetration of outdoor- generated particles into the indoor environment greatly influenced the indoor PM loading (Diapouli et al 2010). It was observed that the mean PM_{10} concentration of cooking gas was considerably less compared to other sampling sites. The concentrations in the $PM_{2.5-10}$ were much lesser than $PM_{2.5}$. This is because Particles in cooking emission are mainly in the fine fraction range (Man-Pun et al., 2011).

Elemental Concentration

Nine elements (Br, Cu, K, La, Na, Sb, Zn, Sm and As) were detected and analyzed for their elemental concentrations for both fractions of the PM. Nine elements were sought for and used as input to the CMB receptor modeling for source contribution estimates. Tables 1 and 2 show the total elemental concentrations and standard errors of indoor and outdoor at each site. From the results of fine fractions ($PM_{2.5}$), the highest mean value was recorded for Copper (Cu) (17.65 μg/m^3, 22.44 μg/m^3) and Potassium (K) (63.88 μg/m^3) in kerosene, cooking gas and firewood site respectively, while the lowest mean value was recorded for Arsenic (0.002 μg/m^3, 0.004 μg/m^3 and Samarium (Sm) (0.002 μg/m^3) in kerosene, firewood and cooking gas site respectively.
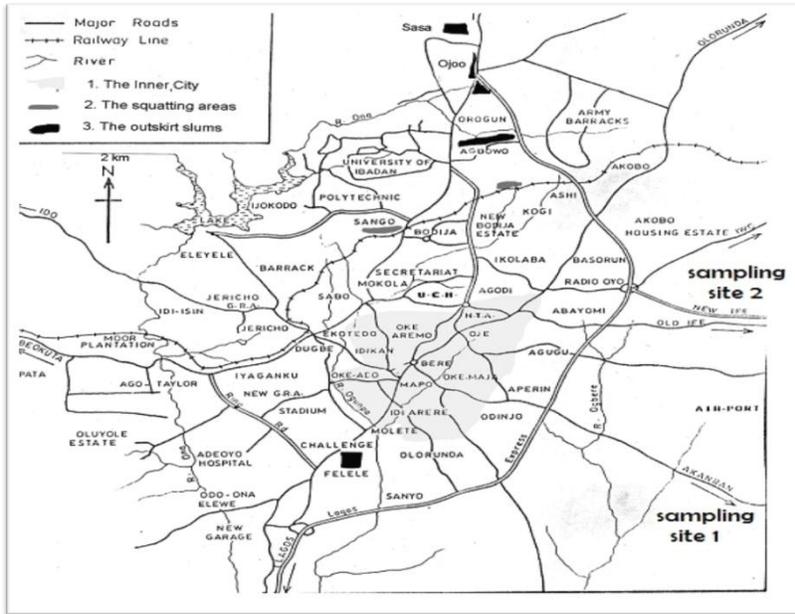
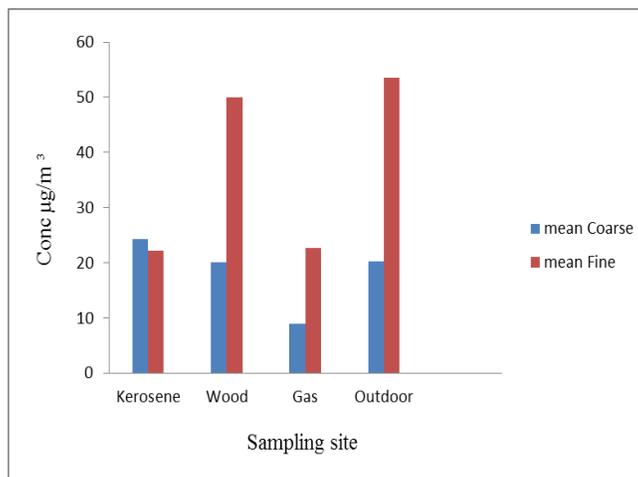Plate1: Map of Ibadan showing the study area

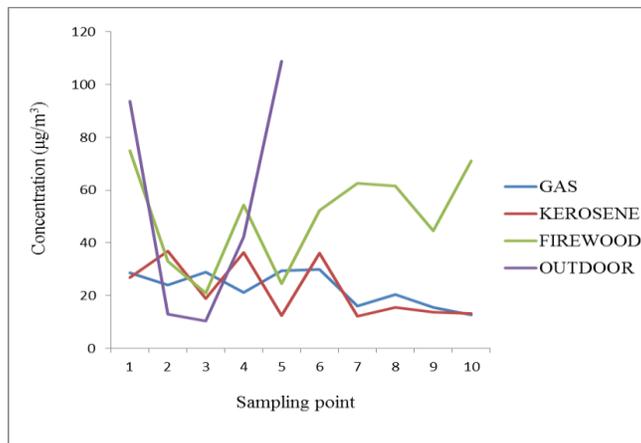**Fig. 1.** The daily variation of fine and coarse for the four sites

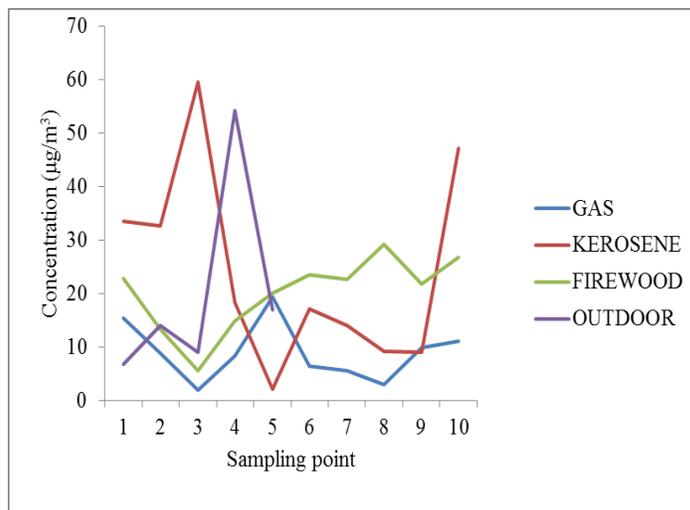**Fig. 2.** Graph of the mass concentrations at each site for fine fraction of PM

Fig. 3. Graph of the mass concentrations at each site for coarse fraction of PM

Table 1. Mean and standard error of elemental concentrations in fine fractions

Element	MEAN (μg/m^3)			
	Kerosene(Std error)	Firewood (Std error)	Gas (Std error)	Outdoor (Std error)
Br	1.04 (0.03)	2.04 (0.06)	1.20 (0.03)	3.31 (0.09)
Cu	17.65 (2.82)	0.07 (0.01)	22.44 (3.59)	0.68 (0.06)
K	6.02 (0.69)	63.88 (7.35)	11.65 (1.34)	26.61 (0.11)
La	0.002 (0.00)	0.02 (0.002)	0.02 (0.002)	0.05 (0.004)
Na	2.06 (0.05)	7.27 (0.07)	4.59 (0.12)	12.19 (0.05)
Sb	0.01 (0.001)	0.01 (0.001)	0.03 (0.002)	0.04 (0.003)
Zn	0.40 (0.04)	0.12 (0.01)	0.42 (0.04)	1.78 (0.19)
As	0.002 (0.001)	0.004 (0.004)	0.002 (0.00)	0.001 (0.00)
Sm	ND	ND	0.002 (0.00)	0.01 (0.002)
Total mean	27.18 (3.62)	73.41 (7.51)	40.35 (5.12)	44.67 (0.51)
Max	17.65 (2.82)	63.88 (7.35)	22.44 (3.59)	26.61 (0.11)
Min	0.002 (0.001)	0.004 (0.004)	0.002 (0.00)	0.001(0.00)

ND (NOT DETECTED)

Table 2. Mean and standard error of elemental concentrations in coarse fractions

Element	MEAN (μg/m^3)			
	Kerosene (Std error)	Firewood (Std error)	Gas (Std error)	Outdoor (Std error)
Br	0.44 (0.01)	0.32 (0.01)	0.59 (0.02)	0.23 (0.01)
Cu	0.13 (0.02)	0.03 (0.01)	0.10 (0.02)	0.35 (0.06)
K	2.72 (0.31)	15.38 (1.77)	11.09 (1.28)	3.89 (0.45)
La	0.002 (0.00)	0.01 (0.001)	0.02 (0.002)	0.01 (0.001)
Na	2.78 (0.07)	2.64 (0.07)	4.56 (0.114)	2.11 (0.05)
Sb	0.01 (0.001)	0.01 (0.001)	0.02 (0.001)	0.003 (0.00)
Zn	0.39 (0.04)	0.28 (0.03)	0.49 (0.05)	0.12 (0.012)
As	0.001(0.00)	0.002 (0.00)	0.003 (0.001)	0.001 (0.00)
Sm	ND	ND	ND	0.001 (0.00)
Total mean	6.47 (0.45)	18.67 (1.89)	16.87 (1.48)	6.72 (0.13)
Max	2.78 (0.07)	15.38 (1.77)	11.09 (1.28)	3.89 (0.45)
Min	0.001 (0.00))	0.002 (0.00)	0.003 (0.001)	0.001 (0.00)

The high value of Copper may likely be due to tyre wear, refuse incineration and other anthropogenic sources (Oluyemi and Asubiojo, 2001).

In the coarse fractions ($PM_{2.5-10}$), the highest mean value was recorded for Sodium (Na) (2.78 $\mu g/m^3$) and Potassium (K) (15.38 $\mu g/m^3$, 11.09 $\mu g/m^3$) kerosene, firewood and cooking gas site respectively and the lowest mean value was recorded for Arsenic in all the sites between 0.001 and 0.003 $\mu g/m^3$.

The ground total mean concentrations in fine fractions were 27.18 $\mu g/m^3$, 73.41 $\mu g/m^3$, 40.35 $\mu g/m^3$ and 44.67 $\mu g/m^3$ for kerosene, firewood, cooking gas and outdoor driven environment sites respectively, while 6.47 $\mu g/m^3$, 18.64 $\mu g/m^3$, 18.87 $\mu g/m^3$ and 6.72 $\mu g/m^3$ were the ground total mean concentrations of kerosene, firewood, cooking gas and outdoor site respectively for coarse fractions. The non-detection of Sm in most of both fractions of the indoor environment strongly suggested that Samarium (Sm) is predominantly from outdoor sources.

The results of $PM_{2.5}$ over the sampling period in the study area show a maximum elemental concentration of 39.55% from firewood site. The elemental concentration for kerosene site was 14.64%, while in cooking gas site was found to be 21.60% and the outdoor site has 24.07%. Also in $PM_{2.5-10}$, the maximum elemental concentration was 38.31% from firewood site while 13.28%, 34.62% and 13.79% were from kerosene, cooking gas and outdoor site respectively. In the fine fractions, Potassium (K) is found to be 2.2 (kerosene residence) and 4.2 (firewood residence) times greater than the values recorded for coarse fraction. The high concentrations of Potassium in both fractions are believed to be an indication that firewood is predominantly dominant in the study area.

In the firewood site, potassium has the highest values of 63.88 $\mu g/m^3$ and 15.38 $\mu g/m^3$ for fine and coarse fractions respectively. The high concentration of Potassium in the fine fraction suggests that it is most probably from vegetative burning, residential wood combustion, cooking or incineration and clearly confirms the elemental characteristics of such location.

The mean concentrations obtained for As (0.003 $\mu g/m^3$) in this study were lower than the 0.013 $\mu g/m^3$ WHO limit value fixed for urban areas and less by half the 0.006 $\mu g/m^3$ value for European country (Aldabe *et al.*, 2011). WHO reported that even at low concentration values, they can have severe health impacts if inhaled (WHO, 2005).

Air Pollutants Sources and Markers

Several studies have shown that some chemical elements are good indicators for many air pollutant sources. Such elements are regarded as elemental markers. In this study, the extent or the strength at which these elements serve these purposes has been adequately utilized in the identification of possible sources of air pollutants (elemental concentration) and apportionment (CMB). Table 3 shows some marker elements and their possible sources with references, this provided references for marker elements that were used in this study.

Species Concentration and Apportionment

The observed elemental concentrations apportioned by CMB in the Tables 4 and 5 show the estimated elemental source contribution for sampling locations. The results for fine particulate matter collected over the sampling period in Ibadan shows a higher contribution of 39.9±13.6% from firewood. The kerosene source contributed 14.6±7.1%, while the contribution

from gas was found to be 21.9±9.0% and the outdoor source contributed 23.6±70.2%. It was observed that the elements (Br, K, La Zn and As) in kerosene, (Br and As) in Firewood and (Br, Zn and As) in Cooking gas were overestimated while also, (Sb and Sm) in kerosene, (Cu, K, Na, Sb and Zn) in Firewood, (Sb) in Gas and (Br, K, Cu and Sm) in outdoor were underestimated, this was as a result of the fact that accurate source profiles of these sources were not readily available to be included in the CMB and many other activities that dominate the area. The underestimation of Cu, K, Zn, Br, As, Na, Sb, Sm and La could be because their actual concentration in the sampling site is much higher than their concentration in the source signature input of the CMB programmed.

The overestimation might be from various activities taking place around the location, such as cooking with the pot open, painting, and construction e.t.c. The underestimation of both Cu and Zn could be due to their concentrations in the source profile input into CMB calculation for tyre and wear which might be less than the actual concentration of these elements present in the tyre and wears in the ambient air of the sampling site (Almeida et al., 2005). Cu can be from diesel fuel coming from the fuel filling station located few meters away from the sites. The source profiles used for these calculations were obtained from EPA – CMB 8.2 as it contains standard source profile for sources such as soil, tire and brake wear, vehicle exhausts, refuse and bush and wood burning (Gertler *et al.*, 2003). The underestimation of K, Na, Br, La, As and Sb suggests additional contributing sources not accounted for in the CMB calculation. This might be due to the fact that accurate source profiles of these sources were not readily available to be included in the CMB.

CONCLUSIONS

The results of this study have shown that the total mean mass concentrations of the particulate fractions; ($PM_{2.5}$ and $PM_{2.5-10}$) are 31.64 $\mu g/m^3$ and 17.80 $\mu g/m^3$ respectively in the indoor micro-environments. The outdoors mean mass concentrations are 53.61 $\mu g/m^3$ and 20.10 $\mu g/m^3$ respectively. This shows that the fine ($PM_{2.5}$) fraction is more dominant in all the environments.

Elemental source apportionment accounted for $14.6 \pm 7.1\%$, $39.9 \pm 13.6\%$, $21.9 \pm 9.0\%$ and $23.6 \pm 70.2\%$ in Kerosene, Firewood, Cooking Gas and Outdoors respectively. Potassium (K) had highest elemental concentration of 34%. The use of firewood is associated with the highest concentrations of PM loadings at our study sites. This has serious detrimental implications for health of the populace as 74% of Nigerians cook with firewood. The mean concentrations obtained for Arsenic (As) in this study were slightly higher than the WHO values (0.003 $\mu g/m^3$) for Urban areas, but less by half of the European values (0.006 $\mu g/m^3$).

The result of the CMB indicated that firewood source is the most prominent source of pollution in the study area with Potassium (K) accounting for over 34 % of the pollutant. A continuous trend like this is predicted because wood is widely used as a means of energy generation for cooking in the study area. A national indoor air quality monitoring network to determine the trend of daily, weekly and monthly mass load of indoor PM concentrations in all the cities in Nigeria should be established and encouraged by regulatory bodies. Also, educate the public on potential health risks of indoor air pollution from firewood combustion and ways to reduce their exposures.

Table 3. Possible Sources and their Marker Elements

Source Categories	Trace Elements / Compounds	References
Soil	Ca, Mn, Fe	Lucarelli *et al.* (2004); Huang *et al.* (1994); Janssen *et al.* (1997)
Brake and tire wears.	Zn, Cu, Pb, Cd, Ni, Fe, Mn, Ca, Mg. Cr, and Ni.	
Cu, Zn engine oil and brakes;		Harrison *et al. (*2003); Harrison *et al.* (1996);
Zn tire; Ba brakes and diesel fuel.	Cu, Zn, Ba, and Pb	Ogunsola *et al.* (1994)
Vehicle Exhaust, Vehicle Wear	Cd, Pb, As, Br, Zn.	
	Cu, Zn, Pb, Ba, Mo.	Chellam *et al.* (2005)
Limestone/ concrete	Ca, Mg	Ogunsola *et al.* (1994); Chan *et al.* (1991)
Wood/Bush Burning Biomass	K, C, elemental C, Ca, Zn, Mn, Cu and phosphorus.	Harrison *et al.* (2003)
burning/Refuse/ waste burning	Non- soil K, Cu, Zn, Pb,	Huang *et al.* (1994)
		Huang *et al.* (1994); Janssen *et al.* (1997) ;
Traffic		Ojanen *et al.* (1998); Molnar *et al.* (2005);
		Chan *et al.* (1991); Van Born *et al.* (1990);
Residual Oil, Oil, and Heavy Fuel Oil	K. Zn, Ba, AI, Cr, Fe, Hg, La, Mg. Mn.	Harrison *et al.* (1996); Sweet *et al.* (1993);
Combustion	Na. Sb, Sc, V, OC, BC, Pb, Br, Cu, C	Allen *et al.* (2001); Ojanen, (1998)
	V, Ni	
		Van Born *et al.* (1990); Janssen *et al.* (1997); Lee *et al.* (1994)

Table 4. PM$_{2.5}$ Chemical source profiles applied in the Ibadan source apportionment study

	KROSENE			FIREWOOD		
Element	Measured	Calculated	Calculated/Measured	Measured	Calculated	Calculated/Measured
Br	1.03±0.00	13.34±13.03	12.95±0.00	2.04±0.06	106.46±4.50	52.19±75.00
Cu	17.65±2.82	18.05±1.87	1.02±0.66	0.07± 0.01	*** ***	0.00±0.00
K	6.02±0.69	20.43±16.47	3.39±23.87	63.88±7.35	*** ***	0.00±0.00
La	0.001±0.002	0.003±0.00	3.00±0.00	0.02±0.002	0.02±0.002	1.00±1.00
Na	2.06±0.05	2.21 ± 3.11	1.07±62.20	7.27±0.18	*** ***	0.00±0.00
Sb	0.009±0.00	0.00±0.00	0.00±0.00	0.01±0.001	0.00±0.001	0.00±1.00
Zn	0.04±0.40	15.06±2.69	376.50±6.73	0.12±0.013	*** ***	0.00±0.00
Sm	0.00±0.00	0.00±0.00	0.00±0.00	0.004±0.001	0.002±0.001	0.50±1.00
As	0.002±0.05	0.04±1.810	20.00±35.49	0.007±0.10	6.67±1.05	952.86±10.50

*** Means numerical overflow

Table 5. PM$_{2.5}$ Chemical source profiles applied in the Ibadan source apportionment study

	GAS			OUTDOOR		
Element	Measured	Calculated	Calculated/ Measured	Measured	Calculated	Calculated/ Measured
Br	1.20±0.03	15.54±15.90	12.95±5.30	3.31±9.28	0.24± 0.24	0.07±0.03
Cu	22.44±3.59	21.54±2.22	0.960±0.62	0.68±10.85	0.42±0.10	0.62±0.01
K	11.65±1.34	24.66±19.64	2.12±14.66	26.61±0.00	2.25±2.51	0.09±0.00
La	0.02±0.002	0.02±0.004	1.00±2.00	0.05±0.45	0.05±0.01	1.00±0.02
Na	4.59±0.11	4.83±6.78	1.05±61.64	0.01±0.17	0.00±0.00	0.00±0.00
Sb	0.03±0.002	0.00±0.01	0.00±4.50	0.04±0.25	0.00±0.02	0.00±0.08
Zn	0.42±0.04	179.67±4.58	427.79±114.50	1.78±18.69	2.98±2.54	1.67±0.14
Sm	0.002±0.00	0.001±0.001	0.50±0.00	*** ***	1.14±0.91	0.00±0.00
As	0.002±0.00	0.06±2.15	30.00±0.00	0.001±0.01	0.001±0.10	1.00±10.00

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