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Spatial and temporal variations of the particulate size distribution and chemical composition over Ibadan, Nigeria

Grace O. Akinlade · Hezekiah B. Olaniyi · Felix S. Olise · Oyediran K. Owoade · Susana M. Almeida · Marina Almeida-Silva · Philip K. Hopke

Abstract Spatial and temporal variation of suspended particulate matter was measured in Ibadan, Nigeria during the raining and dry months of 2013 and 2014, respectively. Six different locations were considered, reflecting city-spread, population density, lifestyle, and vehicular- and industrial-related activities. Elemental characterisation of the samples were carried out using energy dispersive x-ray fluorescence (ED-XRF) spectroscopy, while the black carbon content was determined using an optical transmissometer. Backward trajectory analysis indicated that most air mass was of maritime origin except on few occasions that suggested the Sahara desert origin. Average $PM_{2.5}/PM_{10}$ ratio for the entire sampling period was 0.32 ± 0.04. This is slightly lower than the reported values in the coastal city of Lagos, partly because of more adequately paved and better road networks resulting in lower dust re-suspension and/or there are relatively finer fraction particulates from other sources, which might include marine, industrial, and secondary emission-related sources. Black carbon showed predominance in the fine fractions and was, majorly, of traffic origin. Major enriched elements of concern are S, Zn, As, and Pb aside from those of sea origin. Refuse burning was observed to be the principal source of Pb and Zn in all the sites except one, which showed major industrial-related activities source. This study will provide data for future measurement and modelling works as well as providing a benchmark for relevant agencies for policy making on setting emission standards for the country.

Keywords Transmissometer · Black carbon · ED-XRF · Spectroscopy · Secondary sources · Nigeria

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Introduction

Air quality has received a lot of attention in the last few decades due to its impact on human health (Pope et al. 2002). This has led to environmental protection agencies setting emission standards or guidelines in many nations. Nigeria is listed as one of the fastest growing economies in the world (Taiwo 2005), with many small scale industries springing up. In spite of likely environmental hazard from pollutants from these industries, there are no national policies on urban planning to demarcate industrial from residential sectors and to set emission guidelines or standards. Particulate matter is an important component of pollutants contributing to the

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degradation of air quality, especially the respirable and inhalable fractions. Several epidemiological studies have established that the fine or respirable fraction ($PM_{2.5}$) is more important when considering the health implications of the particulate matter (Almeida et al. 2014; Dockery et al. 1993; HEI 2000; Jerrett 2005; Pope et al. 1995, 2002).

To set emission standards for Nigeria, thorough investigations need to be carried out on the number and nature of sources of the particulate matter. This would form a benchmark for relevant agencies for policy making. Apart from industries, there are other potential sources of air pollution in Nigeria. One of the prominent sources is traffic-related re-entrainment of road dust due to poor road infrastructure and vehicular emission from ubiquitous poorly maintained vehicles plying the roads. Another important source of pollutants is related to electric power generation. The epileptic nature of electricity has encouraged the use of an enormous number of both petrol and diesel engine generators for both residential and industrial power needs. Emission from these sources has led to increased level of both gaseous and particulate pollutants, including black carbon. In rural and semi-urban areas, solid fuels such as coal and fire woods are still largely used. There are also some medium-scale industries springing up, such as secondary iron- and steel-smelting companies and foundry workshops, without any known installed emission abating device.

Earlier works on air particulate measurement in Nigeria, especially total suspended particulate matter were those of Oluwole et al. (1988); Asubiojo et al. (1993); Obioh et al. (1994); and Oluyemi et al. (1994). Measurements involving size-fractionated particulates in some traffic and industrial-related corridors, both in Ile-Ife and Lagos, Southwest Nigeria have been reported (Obioh et al. 2005; Owoade 2006; Owoade et al. 2013). Other measurement involving PM_{10} and $PM_{2.5}$ in some other regions of the country was that of Obioh et al. (2013), where six megacities in Nigeria were considered, providing a sort of baseline measurement of daily PM values for the six geopolitical zones of the country. Ibadan, one of the largest cities in West Africa, has not been captured in any of these studies. Ibadan, the Oyo State capital has a land size of 3080 km^2 and with a human population of 2.949 million according to Nigeria Demographics Profile, 2014 (http://www.indexmundi.com/). The only study conducted by Godson and Zainab (2013) in the city, employed the real-time measurement of the particulate matter without information on the size distribution of the particulate matter. It, therefore, becomes imperative that detailed measurement cutting across different site classes during both the dry and wet seasons be carried out for Ibadan. The objectives of this work are to provide information on the seasonal variation of various particulate matter sizes over Ibadan; determine elemental and black carbon (BC) concentrations across the megacity; and investigate the extent to which heavy metals are enriched by anthropogenic activities.

Experimental

Site description

The receptor sites, chosen to reflect city-spread, population density, lifestyle, and vehicular and industrial-related activities, are shown in Fig. 1. These sites were Iwo road (7.402° N, 3.945° E), Oyo State Housing corporation premises, Bodija (7.418° N, 3.902° E), University of Ibadan (UI) Campus (7.444° N, 3.895° E), Oyo State Dental clinic premises, Dugbe (7.388° N, 3.879° E), Oluyole industrial estate (7.360° N, 3.846° E), and Olubadan High School premises, Aperin (7.372° N, 3.919° E).

The Iwo road-sampling site was along the busy Lagos-Ibadan expressway. It is a high-traffic-density area, with frequent traffic jams during peak periods (early morning and late afternoon). Bodija is a low density residential area with moderate volume of traffic. The Dugbe site, adjacent to the monumental cocoa house is a commercial area. This location is also about 100 m from the popular Ogunpa market and experiences heavy human traffic and moderate vehicular traffic flow. Oluyole estate is the home to most industries in Ibadan, and it is therefore an Industrial area with adequate mix of residential activities. Aperin is a densely populated residential semi-urban community with no commercial or industrial activities. It is one of the ancient areas of the city. While the UI sampling site witnessed a very low volume of traffic to moderate influx of vehicular traffic at peak periods, it was chosen to represent areas of relative passive vehicular, commercial, and industrial activities.

Sampling and gravimetric analysis

Air particulate sampling was carried out with the aid of a low volume Gent air sampler (Hopke et al. 1997),

Fig. 1 Map of Ibadan showing the six sampling locations

equipped with the Norwegian institute for air research (NILU) stacked filter unit (SFU). The SFU carried two 47-mm nuclepore filters, one for the collection of coarse particles with aerodynamic diameter greater than 2.5 but less than 10 μm and the other for the collection of fine particles with aerodynamic diameter less than 2.5 μm. The sampling campaign was from June 2013 to February 2014 (2013 wet months to 2014 dry months) with one measurement per site per month, and each measurement lasted for, at least, 8 h. On the average, sampling was performed between 8:00 am and 4:00 pm, and the samplers operated at flow rates between 16 and 18 L/min.

The filters were pre-conditioned before and after exposure in a desiccator at about 25° C and 50 % humidity for 24 h before weighing. Initial and final weights of the filters were obtained using a micro weighing balance (Sartorius, Gottingen, Germany) of 1-μg sensitivity. Filter weight before and after sampling was obtained as the average of three measurements, when the observed variations were less than 1 %.

Elemental and black carbon analyses

The size-fractionated particulates were characterised for their elemental contents using x-ray fluorescence (XRF) spectroscopy while the black carbon analysis was carried

out using a dual-wavelength optical transmissometer data acquisition system from Magee scientific, respectively. Both facilities were accessed at the Department of Chemical and Bimolecular Engineering and Centre for Air Resources Engineering and Science, Clarkson University, Potsdam, NY, USA. The XRF system provided high sensitivity in the chemical characterisation of the following elements in both the fine and coarse fractions, with the detection limits (in μg/g) in parenthesis: Na(2000), Mg(1000), Al(600), Si(300), P(120), S(30), Cl(10), K(150), Ca(80), Ti(25), Mn(0.5), Fe(15), Zn(0.8), As(0.6), Br(0.6), Rb(0.5), Sr(0.5), and Pb(1.7).

The Magee scientific optical transmissometer data acquisition system (OTDAQ) measured and recorded filter absorbance data relative to a blank reference filter. The results were interpreted in terms of the surface loading of particulate matter collected on the filter, as a means of providing an analysis for the composition of the sample. As a stand-alone system, the OTDAQ was used to collect absorbance data at both ultraviolet (370 nm) and infrared (880 nm) wavelengths.

Crustal enrichment factor analysis

To investigate the contribution of crustal and non-crustal sources to the detected elements, the crustal enrichment factor analysis was carried out. Using aluminium as a

reference (usually, Sc or Al is used because they rarely enter the atmosphere from anthropogenic sources) (Klos et al. 2011; Taylor 1964) crustal composition, the enrichment factor, EF was calculated, thus:

$$EF_{Al} = \frac{(C_x / C_{Al})_{sample}}{(C_x / C_{Al})_{crust}} \qquad (1)$$

where EF_{Al} is the enrichment factor with Al as the normalisation element, C_X and C_{Al} are the concentrations of the considered elemental species and aluminium, respectively.

To account for the local variation in soil composition in the EF calculations, elements with EF values greater than 10 were taken to be enriched by other sources rather than the crust (Farinha et al. 2004).

Backward trajectory analysis

In order to have adequate information on the origin of the particulate matter impacting the receptor sites, back-trajectories were simulated for each sampling day using the NOAA/ARL HYSPLIT version 4.9 model (Draxler and Rolph 2012). The backward trajectory timescale was set at 96 h as an acceptable compromise between the accuracy, which strongly decreases with time and the time needed to investigate the secondary aerosol formation processes. Since studies have observed some limitations in accuracy of trajectories calculations (Stohl 1998), the confidence in the results from back-trajectory simulations were tested using different starting heights and times.

Results and discussion

Particulate matter concentration

The spatial and temporal variations of both PM_{10} and $PM_{2.5}$ and the $PM_{2.5}/PM_{10}$ ratio are presented in Fig. 2. Generally, the $PM_{2.5}/PM_{10}$ ratios were higher during the
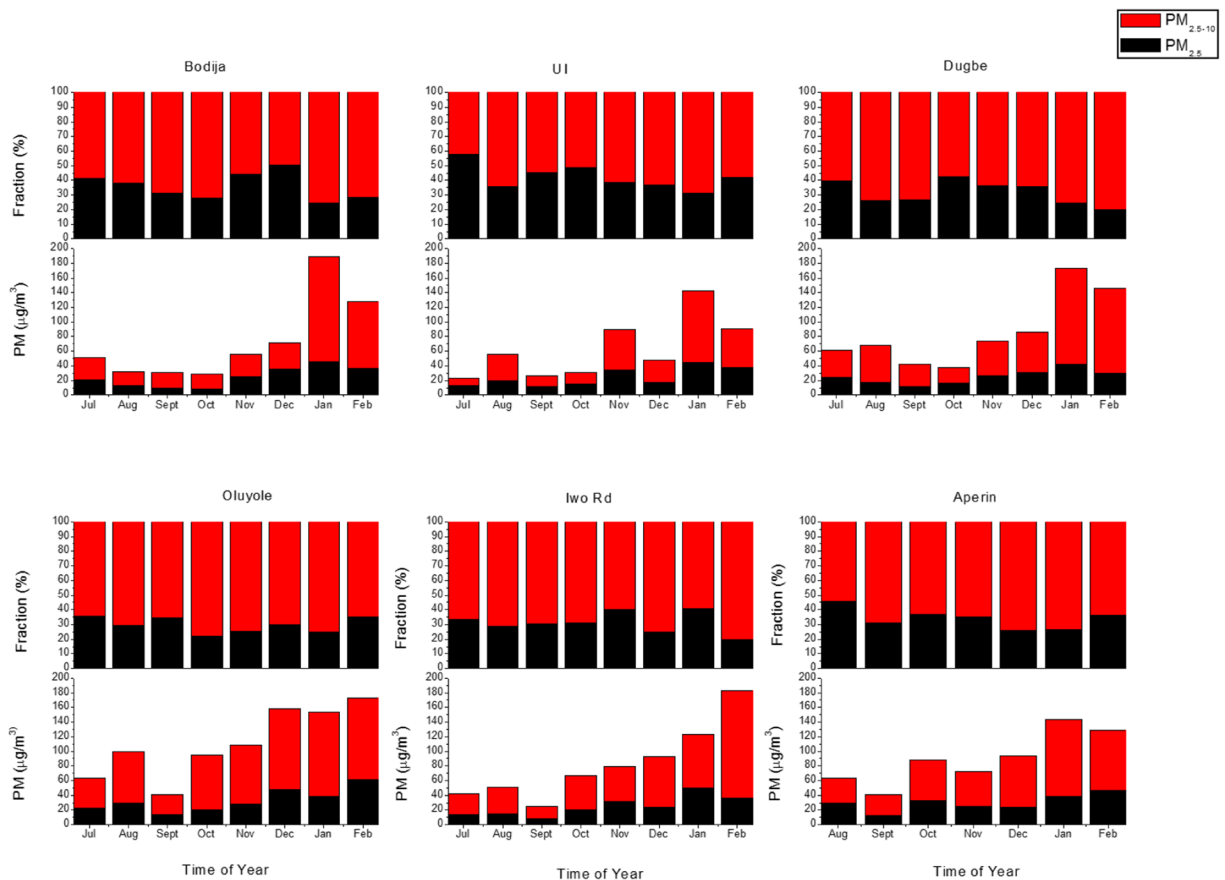

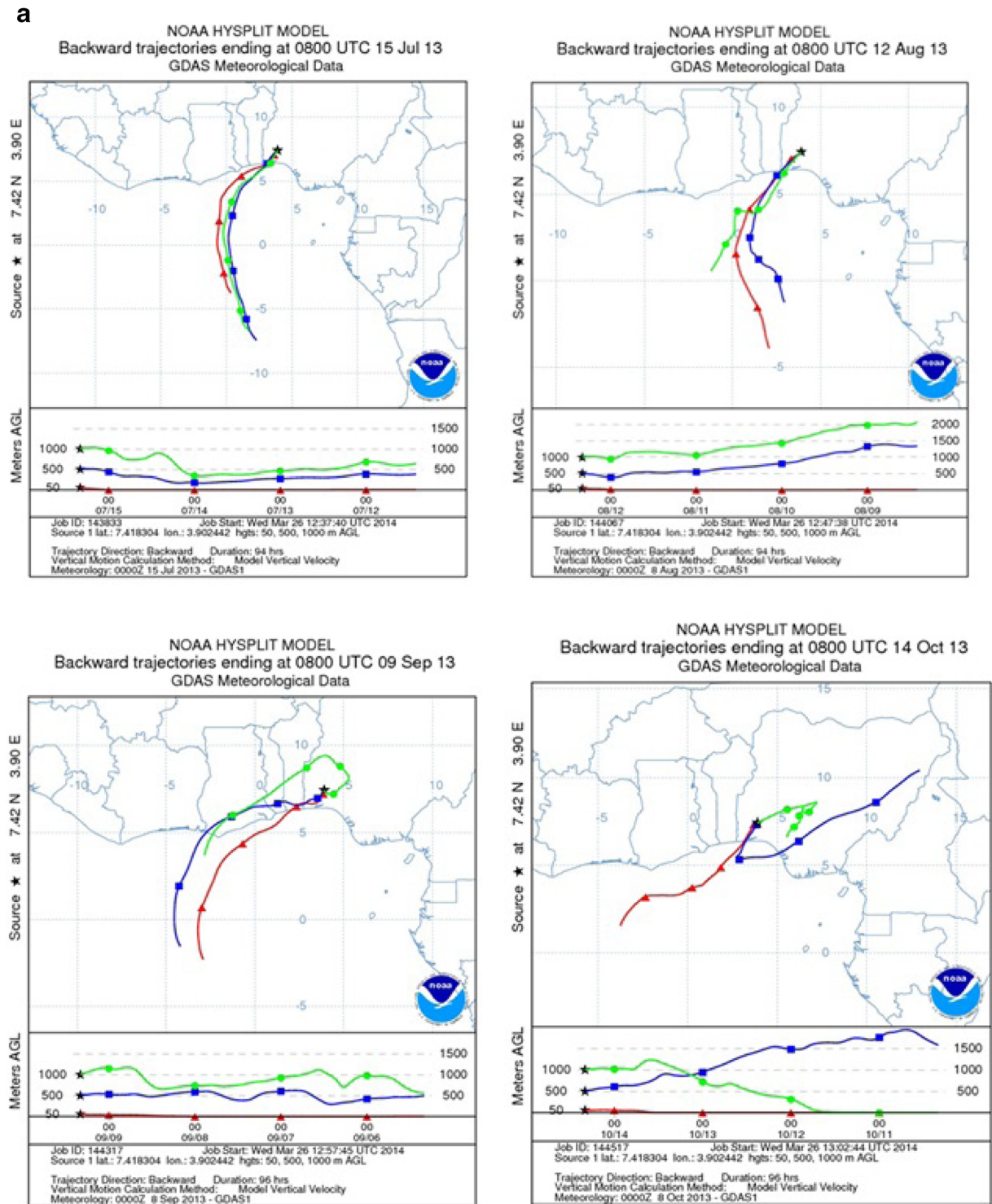Fig. 2 Monthly PM concentrations ($\mu g\ m^{-3}$) at the various sites

a

Fig. 3 a Four-day backward trajectory of air mass ending in Ibadan for the sampling period between July 2013 and October 2013 (Bodi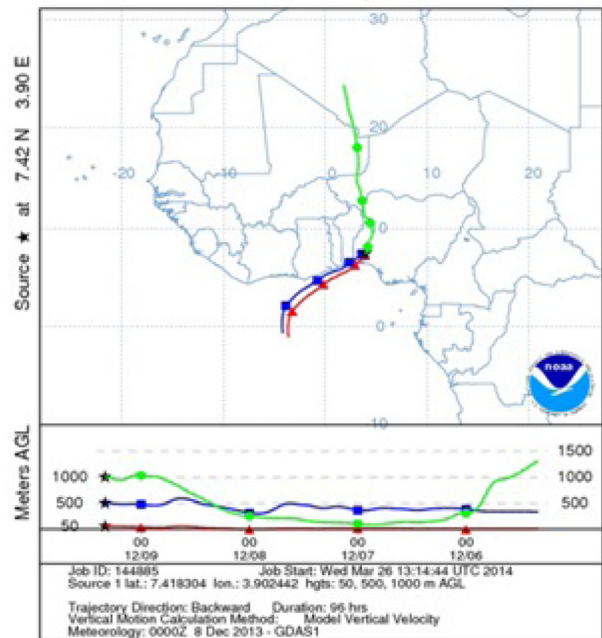ja was chosen to represent other locations). **b** Four-day backward trajectory of air mass ending in Ibadan for the sampling period between November 2013 and February 2014 (Bodija was chosen to represent other locations)
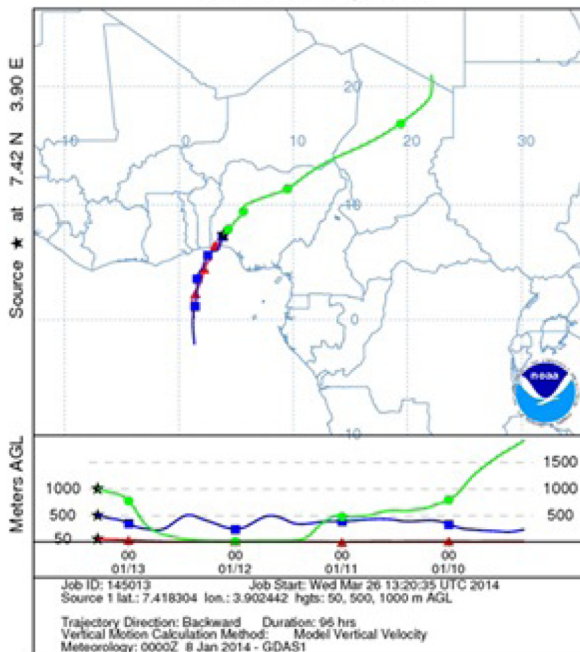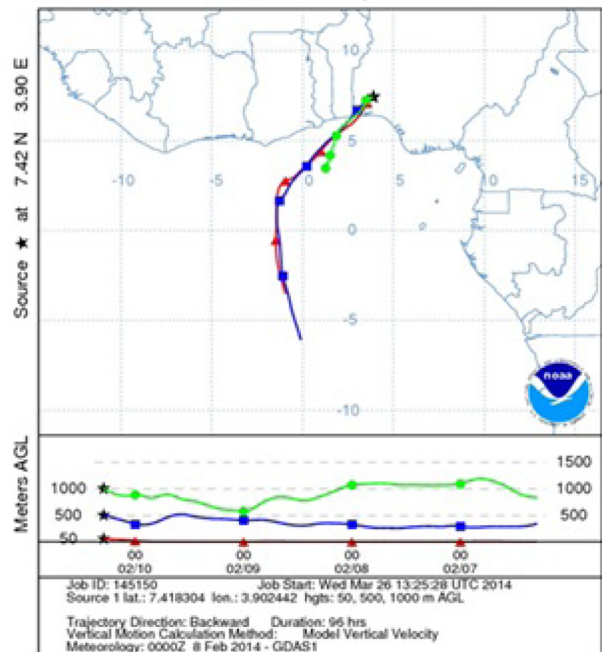
b
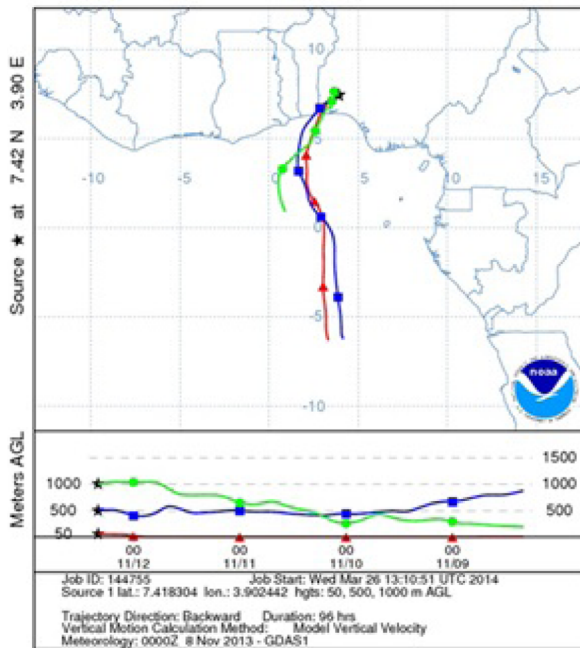
Fig. 3 continued.

wet months than the dry months, in all the sampling locations. This is an indication that dust re-suspension is a major contribution to PM_{10}, which reduces the ratio during the dry season. Average $PM_{2.5}/PM_{10}$ ratio for the entire sampling period was 0.32 ± 0.04. This is slightly lower than the values obtained in an earlier study in Lagos (Owoade et al. 2013). This is either because Lagos has more adequately paved and better road networks in the studied areas resulting in lower dust re-suspension or there were relatively more fine particulates from other sources, which might include industrial and secondary emission-related sources.

The least $PM_{2.5}/PM_{10}$ ratio of 0.29 was observed for the commercial and industrial areas. This implies that over 70 % of the PM was in the coarse fraction. This can be attributed, majorly, to dust re-suspension, as most of the roads are unpaved. The highest $PM_{2.5}/PM_{10}$ ratio was observed at the control site. This is expected as the site was far from the road and any obvious source of pollutants, except vehicular traffic. However, this high value is still lower than the mean values recorded in urban and rural European areas (Van Dingenen et al. 2004) and some rural and semi-rural areas of the USA (USEPA 2001; Vega et al. 2002).

The residential areas (Bodija and Aperin sites) recorded $PM_{2.5}/PM_{10}$ ratios less than that of the control site but higher than that of the commercial and industrial sites. The Aperin site, located inside the expansive Olubadan High School, is far from the road. This might have accounted for such ratio since the re-suspended particulates would have settled appreciably before impacting the receptor. Bodija, on the other hand, has paved roads with less vehicular traffic compared to the other locations.

As expected, the concentrations of $PM_{2.5}$ and PM_{10} fractions of the particulate matter were generally lower during the wet months than during the dry months. In all the site classes, the mean value of $PM_{2.5}$ was less than 25 $\mu g.m^{-3}$ except for the Aperin and Oluyole sites. We noted that Aperin, a densely populated semi-rural community did not have electric power supply for upward of two years, including the sampling period. We therefore concluded that the major impacts resulted from activities from homes running on the small generators for electricity supply and cooking from kerosene stoves, charcoal, and wood. Other regular activity noted throughout the sampling period was refuse burning. Although Oluyole site also presented high $PM_{2.5}$ concentrations just like Aperin, its own contribution was attributed to the presence of many industries in the area.

Oluyole presented the highest levels of PM_{10}, attributed to road dust re-suspension. The roads were unpaved, and the combination of both industrial and residential related activities resulted in an increased level of small and heavy-duty vehicular movement. Except for the Iwo road and Oluyole sites, all the other sites recorded their highest PM_{10} levels in January. UI recorded a very large variation between December 2013 and January 2014. The reason for this could only be explained with air mass back-trajectory analysis (Fig. 3a, b). Normally, air mass of the Sahara desert

Table 1 Annual mean particulate matter concentration ($\mu g\ m^{-3}$)

| Site-class	Parameter	$PM_{2.5}$	$PM_{2.5-10}$
Low residential	Mean	24.29	49.20
	Standard deviation	14.04	44.44
	Minimum	8.08	20.14
	Maximum	45.89	142.83
	Median	22.87	30.64
Control	Mean	24.30	38.99
	Standard deviation	12.76	29.35
	Minimum	11.83	9.65
	Maximum	44.81	98.00
	Median	18.72	33.03
Commercial	Mean	24.89	61.44
	Standard deviation	9.88	40.25
	Minimum	11.35	21.74
	Maximum	42.52	130.93
	Median	25.70	48.99
Industrial	Mean	32.68	78.64
	Standard deviation	15.43	33.17
	Minimum	14.14	26.64
	Maximum	60.82	115.23
	Median	28.62	77.35
High-density traffic	Mean	24.87	58.01
	Standard deviation	13.97	40.48
	Minimum	7.62	17.14
	Maximum	50.10	146.48
	Median	21.92	46.74
High-density residential	Mean	29.78	60.28
	Standard deviation	10.84	27.64
	Minimum	12.55	27.76
	Maximum	46.64	105.70
	Median	29.24	55.48
WHO standard	Annual mean	10	
EPA	Annual mean	12	
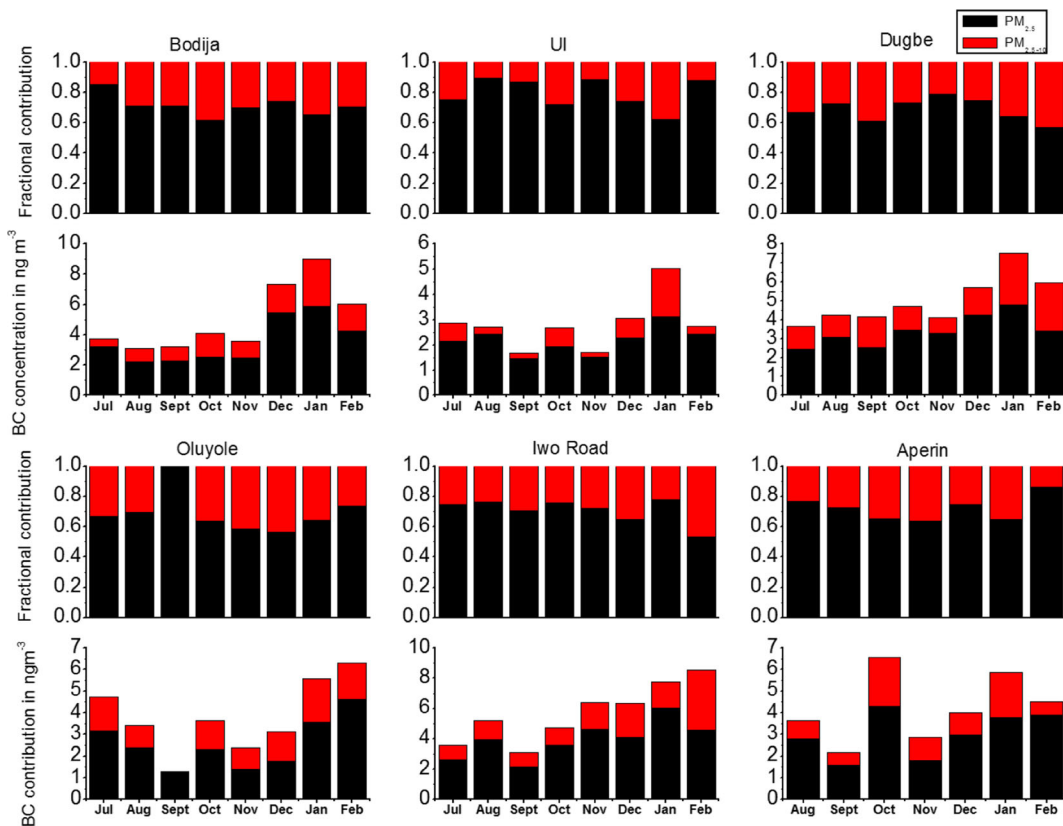
Fig. 4 Monthly black carbon concentrations at the various sites

(North-East trade wind) origin was expected in the region during the period, but 4 days back-trajectory, ending in Ibadan, for all the periods revealed that all the air mass reaching the region were of maritime origin. But in January 2014, air mass at height 1500 m reaching Ibadan originated from the Saharan desert (Fig. 3g). The trajectory was brought to the surface, a day prior to sampling, at Bodija and UI. We concluded this to be the main reason why UI and the other locations had a very high PM_{10} concentration in January 2014.
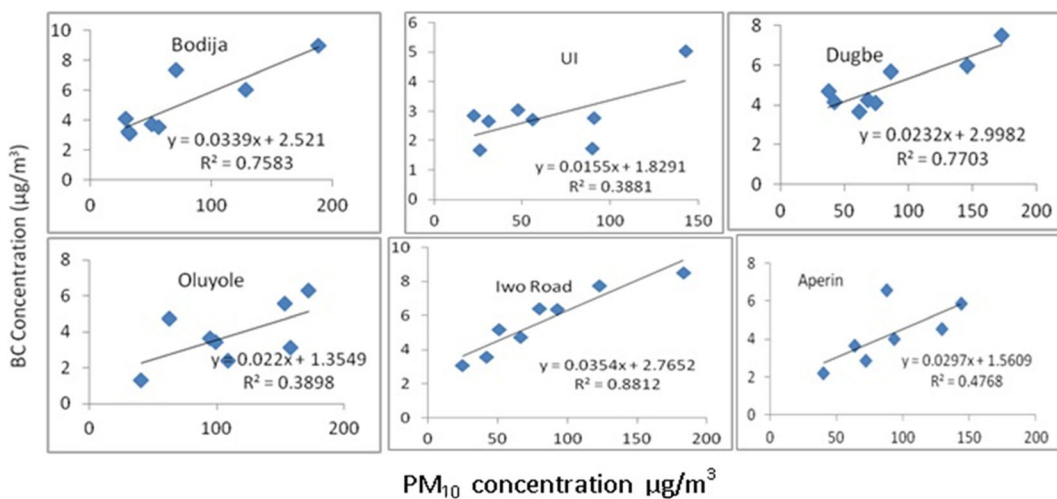
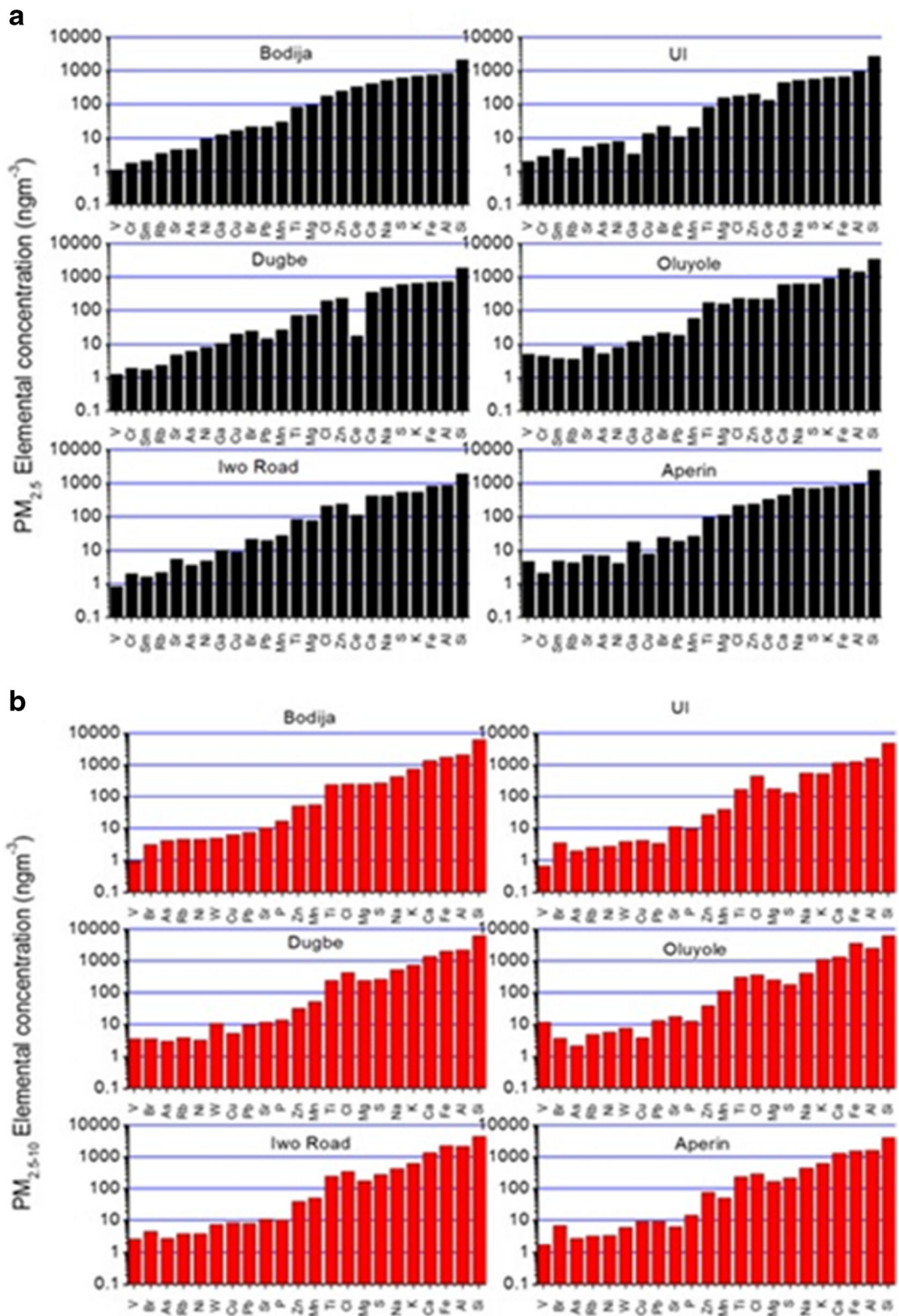

Fig. 5 Relating the total BC to the PM10 concentration

Fig. 6 Mean elemental concentration at the various sampling sites in Ibadan, Nigeria **a** for fine fraction and **b** for coarse fraction

PM_{10} values averaged over 8 h were higher than the daily limit of 50 $\mu g.m^{-3}$ set by the World Health Organisation (WHO) or the European Union (EU), especially during the dry months. The PM_{10} values were lower than the daily US national ambient air quality standard (NAAQS) limit of 150 $\mu g.m^{-3}$ set by USEPA (2013) except on few cases during the dry months. However, at UI, Bodija, and Iwo Rd, the daily limits were not exceeded during the wet months. In all the locations, the daily limit of 25 $\mu g.m^{-3}$ (WHO) and 35 $\mu g.m^{-3}$ (USEPA 2013) for $PM_{2.5}$ was exceeded throughout the dry months but were within the acceptable limits during the wet months, except for the Oluyole and Aperin sites.

Table 1 presents the statistics of PM_{10} and $PM_{2.5}$ across Ibadan for the period under investigation. The average value of $PM_{2.5}$ in $\mu g\ m^{-3}$ for Bodija, UI, Dugbe, Oluyole, Iwo road, and Aperin were 24.29, 24.30, 24.89, 32.68, 24.87, and 29.78 $\mu g\ m^{-3}$, respectively; while the average values for PM_{10} in $\mu g\ m^{-3}$ were 73.4, 63.3, 86.3, 111.3, 82.9, and 90.1 for Bodija, UI, Dugbe, Oluyole, Iwo Rd, and Aperin, respectively. On the Average, it was

observed that the UI location, which was expected to be the control, was in no way cleaner than the other parts of the city in terms of $PM_{2.5}$ concentrations. This is because the fine particles are expected to have longer resident time in the atmosphere and can be transported over long distances. However, there was a marked reduction in the concentration of PM_{10} measured in the area. In all the locations, the annual limit of 20 $\mu g\ m^{-3}$ and 10 $\mu g\ m^{-3}$ for PM_{10} and $PM_{2.5}$, defined by WHO (2005), were exceeded. The total annual $PM_{2.5}$ average for Ibadan was 27 $\mu g\ m^{-3}$ while that of PM_{10} was 85 $\mu g\ m^{-3}$.

Black carbon distribution

Figure 4 presents the black carbon concentrations across the sampling sites for the period under study. Relatively, higher concentrations were found at Bodija, Dugbe, and Iwo Road, where vehicular activities were more. This implied traffic-related source dominance in the emission of BC in Ibadan. Low BC concentrations recorded in UI further established the location as a less polluted place.
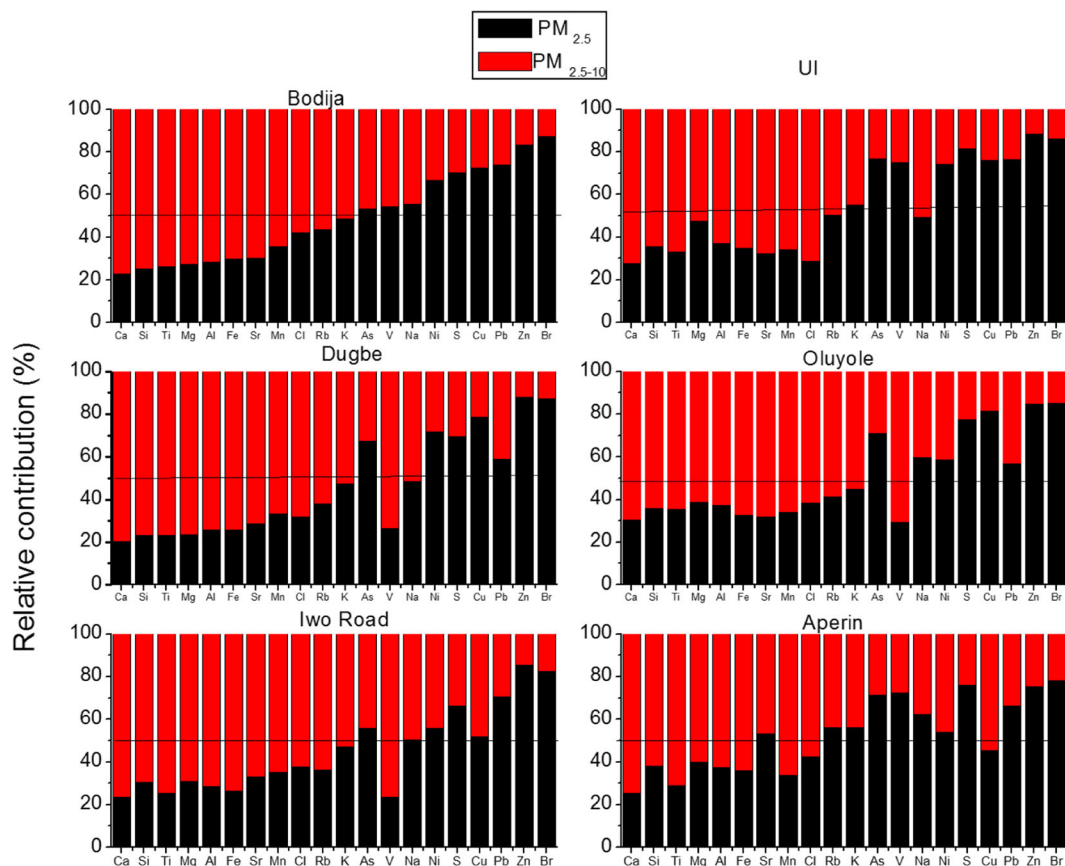

Fig. 7 Fractional contributions to the measured elemental concentrations
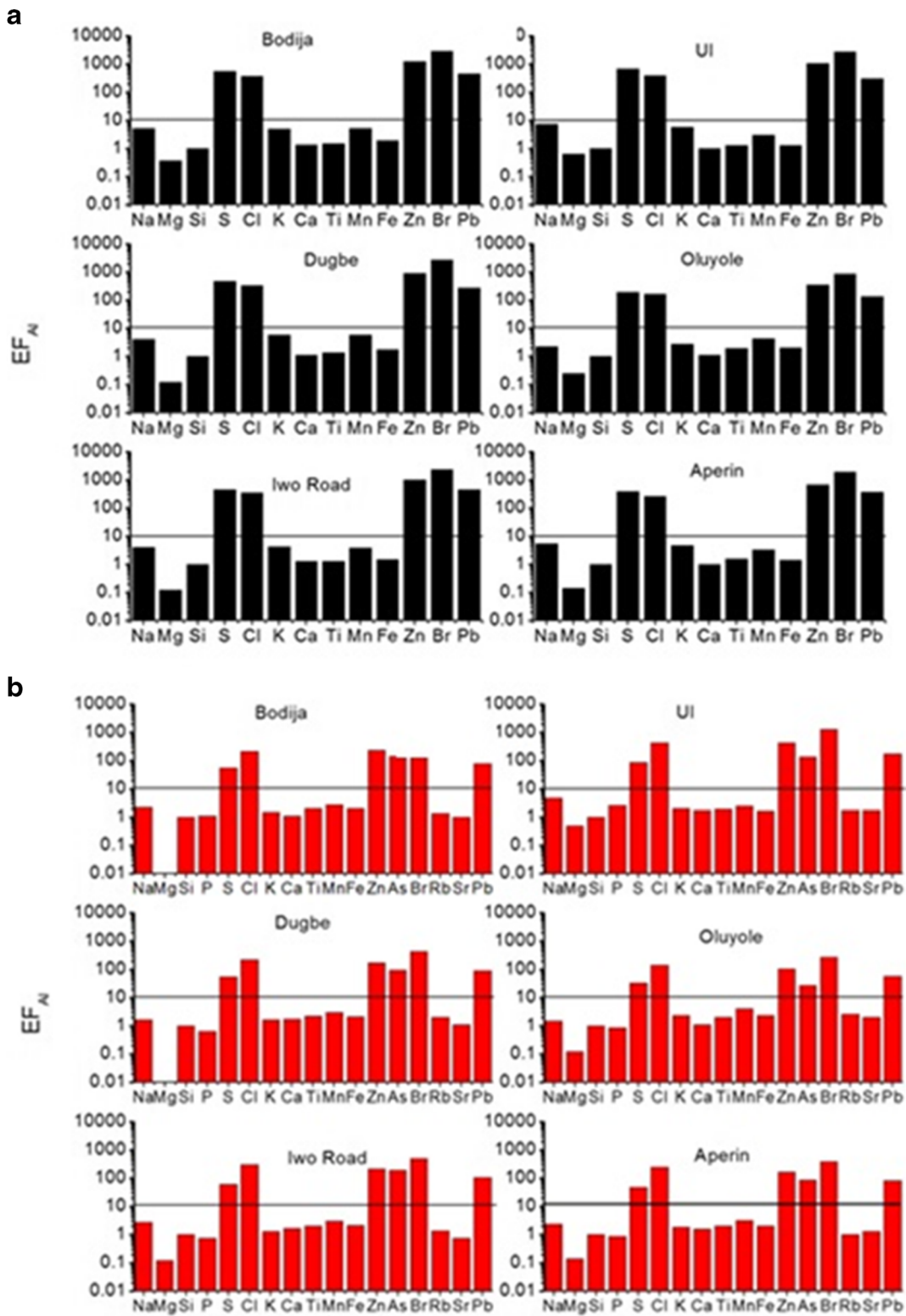
Fig. 8 Crustal enrichment factor analysis for some of the elements **a** for fine fraction and **b** for coarse fraction

The contribution of BC to $PM_{2.5}$ was also higher at the traffic corridors (14–16 %) compared to the other sites (7–10 %). The contribution of total BC to PM_{10} ranged from 3–7 % across all the sites. When these figures are compared with what was obtained in Europe, where BC only contributes 5–10 % to $PM_{2.5}$ and less to PM_{10} (Putaud et al. 2004), it can be established that the emission of black carbon in Nigeria especially from traffic, is still very high. In all the locations, about 70 % of the BC emitted was in the fine fraction as shown in Fig. 4.

Figure 5 shows that BC fairly correlated well with PM_{10} in all the locations except in UI and Oluyole, for the reasons earlier stated. Thus, traffic was the major source of suspended particulate matter, either in the form of vehicular emission or road dust re-suspension.

Elemental concentrations and crustal enrichment factor analysis

Figure 6a, b shows the elemental concentrations at the various sites for the fine and coarse fractions, respectively. Generally, the elements with concentrations higher than 100 ng m^{-3} for the fine fractions were Cl, Zn, Ce, Ca, Na, S, K, Fe, Al, and Si except at Dugbe, where Ce presented concentrations lower than 100 ng m^{-3} and at Oluyole, where Ti concentrations were higher than 100 ng m^{-3}. In the coarse fractions, elements with concentrations higher than 100 ng m^{-3} were Ti, Cl, Mg, S, Na, K, Ca, Fe, Al, and S except at Oluyole, where Mn presented concentrations higher than 100 ng m^{-3}.

In order to distinguish elements of anthropogenic from those of natural origins, the fine/coarse ratio was calculated. Elements with higher fine/coarse ratio are generally regarded as being of anthropogenic origin. Figure 7 shows the fractional contributions of the fine and coarse fractions to the measured elemental concentrations. Elements with higher fine/coarse ratio, which resulted predominantly from anthropogenic sources were As, Na, Ni, S, Pb, Cu, Zn, and Br in all the locations while the fine/coarse ratio for Vanadium was only higher at Bodija and Aperin (residential areas). Vanadium is a fingerprint element for oil combustion from vehicular sources, in the case of Bodija and from domestic- and plastic-related burnings, at Aperin. Crustal elements such as Ca, Si, Al, Fe, Ti, and Mn (Mason and Moore 1982) presented low fine/coarse ratios.

Figure 8a, b shows that elements that were enriched in relation to the soil in both fractions were S, Cl, Zn, Br,

and Pb while As was only enriched in the coarse fraction. Cl and Br have been identified as sea salt composition (Almeida et al. 2013). Therefore, elements of major concern are S, Zn, As, and Pb. Zinc, K, and Pb are associated with refuse burning (Calvo et al. 2013); however significant levels of correlation existed between Pb and Zn in the Bodija and Dugbe sites, while poor correlation between the elements were observed in other locations. Strong correlation between Zn and K were observed for the Bodija, Dugbe, and Iwo road sites while no significant correlation was observed for the other sites. In the same vein, Pb correlated well with K at the Bodija, Dugbe, and Iwo road while poor correlations between the said elements were observed at the other sites. From the relationships between Pb, Zn, and K, we concluded that refuse burning was the major contributing factor to Pb and Zn in the Bodija, UI, Dugbe, and Iwo road sites while that of Oluyole site was most likely due to the presence of industries. However, the source of Pb in Aperin could not be directly linked with refuse burning, as no significant correlations existed between the relevant fingerprint elements.

Conclusion

Suspended particulate matter was measured for 8 months in Ibadan, the capital city of Oyo State, Nigeria. The measurement period spanned the raining months of 2013 and the dry months of 2014. Backward trajectory analysis indicated that most air mass was of maritime origin except on few occasions where the wind aloft (height 1500 m) originated from the Sahara desert. The particulate matter loading was higher during the dry months partly due to the transport of the Saharan dust at the height 1500 m and road dust re-suspension. Average $PM_{2.5}/PM_{10}$ ratio for the entire sampling period was 0.32 ± 0.04. This was slightly lower than the values obtained in an earlier study in Lagos. This was partly because of more adequately paved and better road networks in the studied areas, resulting in lower dust re-suspension or because there were relatively finer fraction particulates from other sources, which might include marine, industrial, and secondary emission-related sources.

Black carbon content of the suspended particulate matter showed predominance in more than 70 % of the fine fractions and was, majorly, of traffic source. Elemental characterisation of samples revealed that most of the elements with high $PM_{2.5}/PM_{10}$ ratio were not

enriched in relation to the soil composition. Major enriched elements of concern were S, Zn, As, and Pb aside from those of sea origin. Correlations between the elements revealed that refuse burning was the major source of Pb and Zn in the Bodija, UI, Dugbe and Iwo road. At Oluyole, the enrichment of Pb and Zn were mostly due to industrial activities while the source of Pb and Zn in Aperin could not be concluded.

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