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Characterization of Size-Fractionated Particulate Matter and Deposition Fractions in Human Respiratory System in a Typical African City: Nairobi, Kenya

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ABSTRACT

Information from elemental and mass composition of size-fractionated airborne particle matter (PM) provides insightful knowledge about their impact on human health, meteorology and climate. To attain insight into the nature of size-fractionated PM from a typical African city, samples were collected from an urban background site in Nairobi, Kenya, during the months of August and September in 2007. PM samples ranging in size from 0.06 to 16 μm aerodynamic diameter were collected on pre-weighed polycarbonate filters with 0.4 μm pore size using a nine-stage cascade impactor. Particles less than 0.06 μm were collected on a backup filter. A total of 170 samples were collected and analysed for trace elements using the Proton Induced X-Ray Emission (PIXE) technique. The analysis showed that Si, Fe and S dominated in all size ranges and displayed unimodal mass-size distribution whereas K, Cu, Zn and Pb, depicted bimodal mass-size distribution highlighting the multiplicity of their sources. To estimate human exposure to PM, deposition fractions of both the coarse and fine PM in the human respiratory system were calculated. The deposited concentration was found to be highest in the head airways region compared to the tracheobronchial and pulmonary regions.

Keywords: Urban air quality; Human health; Particulate pollution.

INTRODUCTION

The impact of airborne particulate matter (PM) on human health, climate and meteorological phenomena is poorly understood in Africa (Petkova *et al.*, 2013). Specifically, lack of air pollution information from African countries, limits the knowledge on how exposure to air pollutants affects public health, especially with regard to PM (Gulis *et al.*, 2004, Ngo *et al.*, 2015). This is particularly relevant as the exposure risk and eventual negative effects of PM on human health is influenced by particle sizes and chemical composition (Milford and Davidson, 1985, Dockery and Pope, 1994, Montoya *et al.*, 2004, Pope and Dockery, 2006, Heal *et al.*, 2012). The majority of the available studies from Africa have reported on both $PM_{2.5}$ (particles less than or equal to 2.5 μm in aerodynamic diameter) and PM_{10} (particles less than 10 μm in aerodynamic diameter). However, cascade impactors can be utilized to give a better size fractionation of particles depending on the number of stages being used (Adams *et al.*, 1983; Maenhaut *et al.*, 1996). This finite fractionation leads to obtaining reliable size distribution data of the sampled PM. The obtained knowledge on particle size distribution has been useful in understanding the deposition pattern of pollution in the human respiratory system (Haber *et al.*, 2003; Behera *et al.*, 2015). The urban centres in Africa are growing at exceedingly high rates whereas the provision of prerequisite social services and amenities are not commensurate with this growth (UN-HABITAT, 2006; UNDESA, 2010; Karanja and Makau, 2012). The urban population is reported to be exposed to wide range of air pollutants (Gulis *et al.*, 2004; Ngo *et al.*, 2015) whose sources have been identified as vehicular emissions, biomass burning and mineral dust (Formenti *et al.*, 2003; Gatari and Boman, 2003; Boman *et al.*, 2009; Kinney *et al.*, 2011; Petkova *et al.*, 2013; Gaita *et al.*, 2014). Therefore, the objectives of this study were to obtain a size distribution of PM and elemental concentrations in a typical African city and to determine the deposition

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fractions of measured PM and trace elements in the respiratory system. The results are expected to contribute to furthering our understanding of risks associated with human exposure to air pollutants as well as public health policies.

MATERIALS AND METHODS

Description of the Sampling Area

Although Nairobi city is located about 200 km south of the Equator, the city has a subtropical highland climate as opposed to the expected tropical climate (Peel *et al.*, 2007; Wikipedia, 2016). This is due to its location at a high altitude of 1795 m above sea level. The regional climate of Nairobi and East Africa in general, is influenced by the seasonal movement of the intertropical convergence zone (ITCZ) (Henne *et al.*, 2008). As a result of this ITCZ displacement, the East Africa region has two rain seasons with dry spell intermediates. The first rainy season starts during mid-March and ends in May and the second rainy season occur between mid-October and mid-December. The average annual rainfall is approximately 900 mm but varies from less than 500 mm to more than 1500 mm. The average daily temperature varies from about 17°C in months of July and August to about 20°C in the month of March (Wikipedia, 2016). Nairobi has a population of about 4 million people (KODP, 2012) in an area of approximately 684 km^2 and this population is projected to grow to about 12 million by 2030 (MoNMD, 2008). The fast growing population has given rise to increased controlled and uncontrolled infrastructural development, increased vehicular traffic activities and poor social services delivery (van Vliet and Kinney, 2007; Karanja and Makau, 2012; Egondi *et al.*, 2013). The majority of the industries within the Nairobi metropolitan area are located to the east and south east of the city centre. They include food processing, power generation, chemical processing, battery manufacturing and scrap metal recycling industries.

Sampling and Filter Analysis

Twenty-four hour samples were collected from an urban background site in Nairobi, Kenya, during the months of August and September in 2007. The sample site was at the rooftop of the Engineering building, University of Nairobi (1.279°S, 36.817°E), approximately 17 m above the ground level. PM samples in the size range from 0.06 to 16 μm were collected on pre-weighed polycarbonate filters (0.4 μm pore sizes) using a PIXE cascade impactor (PIXE International Corporation, USA) that had nine stages and a backup filter. Samples were collected at an air flowrate of 1 L min^{-1} for a period of 24 h starting at about 1800 local time (UTC+3). A total of 170 samples were collected and they were analysed for trace elements at the PIXE analytical facility, Lund University, Sweden (Johansson *et al.*, 1981). The samples were irradiated with a 2.55 MeV proton beam, which covered a circular area with a 10 mm diameter. The beam current was adjusted according to the filter loading to assure non-destructive analysis and in order to optimize the analysis, while also avoiding pileup spectral artefacts and dead time corrections that are associated with high count rates. The PIXE analytical technique is preferred over traditional Energy Dispersive X-Ray Fluorescence (EDXRF) as it has lower detection limits, shorter analytical time and better sensitivities (Maenhaut *et al.*, 1996; Maenhaut and Malmqvist, 2001).

Data Analysis

PM Mass Distributions

In order to generate mass distributions, PM and elemental composition data from this study were used to calculate lognormal mass distributions (LNMD) by using Eq. (1) (Hinds, 1998).

$$LNMD = \frac{\Delta C / C_t}{\Delta \log D_p} \quad (1)$$

where ΔC is the airborne mass concentration of the element in a given size range, C_t is the total concentration of the given element in all size ranges, D_p is the aerodynamic diameter of the sampled particles and $\Delta \log D_p$ is the difference between the log of the lower and upper D_p in each size range.

Deposition Fractions in Human Respiratory System

The obtained PM and elemental composition data was used to estimate the deposition patterns along the human respiratory system, including the head airways, tracheobronchial and pulmonary regions. The deposited fractions along the airways were calculated using particle statistics and a Multiple Path Particle Dosimetry model (MPPD V2.1). This model is developed by Chemical Industry Institute of Toxicology (CIIT) and the National Institute of Public Health and the Environment (RIVM), based on the work of Raabe *et al.* (1977) and Anjilvel and Asgharian (1995). The model is described in detail by Winter-Sorkina and Cassee (2002) and its robustness as well as its usefulness have been demonstrated in a number of studies as highlighted by Behera *et al.* (2015). The model is utilized in calculating deposition fractions and exposure doses for humans and rats to airborne PM. The model requires input parameters that relate to particle properties namely; mass median aerodynamic diameter (MMAD) and geometric standard deviations (GSD), which were calculated from the sampled PM size ranges. The MMAD and the GSD of the sampled PM were calculated using a "Probit method" described by O'Shaughnessy and Raabe (2003). To simulate the deposition in the respiratory system of an average human adult, the following assumptions were made regarding human symmetry, breathing parameters and particle properties; spherical particles of density 1 g cm^{-3}, a tidal volume of 625 mL, nasal breathing route, breathing frequency of 12 breaths per minute, functional residual capacity of 5563.3 mL and inspiration fraction of 0.50. The robustness and usefulness of the model have been demonstrated in a number of studies as highlighted by Behera *et al.* (2015).

Meteorology during the Sampling Period

As shown in Fig. 1, the local wind direction during the sampling period was mainly easterly and the wind speed was

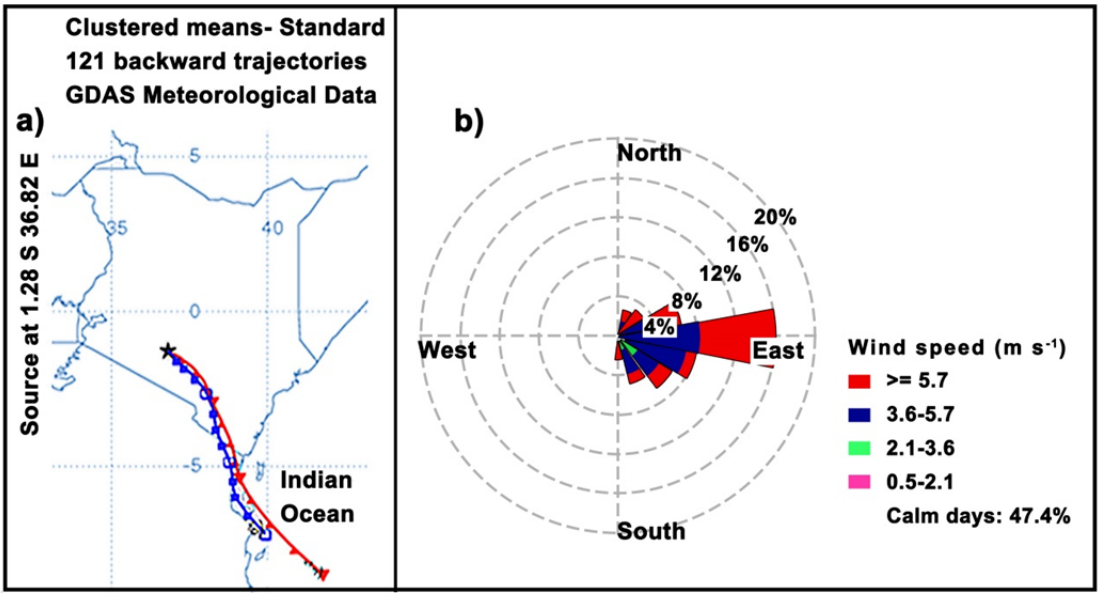

Fig. 1. The clustered backward trajectories (a) showing the origin of air mass at 250 m above the ground, arriving in Nairobi city and local wind direction (b) during the months of August and September, 2007.

between 2 and 6 m s^{-1}. In addition, clustered back trajectories generated using the HYSPLIT *(HYbrid* Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Hess, 1998; Draxler and Rolph, 2015) during the sampling period, showed that the air mass parcels arriving in Nairobi originated from the southeast. The back trajectories for the sampling period can be clustered into two groups, marked by the red and blue lines. The blue line represents the air masses (34%) that originated and travelled over the continental landmass, while the red line represents the air masses (66%) that originated from the Indian Ocean during the measurement period. In addition to the southeasterly back trajectories, Gatebe *et al.* (1999) reported that air masses reaching Kenya between 500 and 700 hPa originate from the Saharan region, which may influence airborne PM in East Africa.

RESULTS

Airborne Concentrations of Particulate Matter

Table 1 lists the summed concentration values of measured elements, total mass of PM and the detection limits (DL) from the measured elements after subtraction of the blank values. A total of eleven elements; Si, S, K, Ca, Ti, Mn, Fe, Cu, Zn, Br and Pb were quantified. From the statistical analysis of measured elemental concentrations, Si was the dominant element followed by Fe and S. These elements were detected in almost all of the 170 filter samples indicating the strong influence of their sources on PM of all size ranges present in the ambient air of Nairobi city.

Distribution of Mass Concentrations of PM and Elements

The concentration values from PM and elemental constituents were grouped into three size categories; coarse (2–16 µm), fine (0.12–2 µm) and ultrafine (< 0.12 µm). Fig. 2 shows the distribution of each measured element concentration in respective size range. Si, Fe, Ca, Ti and Mn had approximately 70% of their respective concentrations in the coarse mode, highlighting their commonality as crustal elements (Towett *et al.*, 2013). S, Zn and Br had approximately 50% of their concentrations in the fine size range compared to Pb and Zn, which had approximately 75% of their mass concentrations in the same size range. K had approximately equal proportions in both coarse and fine size mode pointing to its sources as mineral dust and biomass burning as reported by Gaita *et al.* (2014). S, K, Cu and Br had between 10 and 31% of their mass concentration in the ultrafine size range.

Normalized Mass-Size Distributions of PM and Elements

Due to the diversity of the PM sizes measured in this study, the elements' concentrations are presented in lognormal distribution. The normalized mass-size distributions of measured PM and elemental constituents as a function of their respective particle size ranges are depicted in Figs. 3(A)–3(D). The PM concentrations displayed a bimodal distribution pattern (Fig. 3(B)) with the prominent peak around 200 nm and the other around 2 µm. Si, Fe, Ca, Mn and Ti depicted unimodal patterns with peaks at around 2 µm size range (Fig. 3(A)) confirming their crustal and mineral dust origin (Towett *et al.*, 2013). Similarly, K and Cu had peaks in the same size range in addition to second peaks around 0.25 µm (Fig. 3(B)). For K, the size distribution was an indication of multiple sources of particles containing K such as microscale particles from biomass burning emissions (Cachier *et al.*, 1998; Andreae and Merlet, 2001) and dust particles from local soil (Gatari *et al.*, 2005; Towett *et al.*, 2013). As for Cu, the bimodal distribution was an indication of multiple sources probably from direct traffic emissions and from re-suspended road dust. The mass-size distributions of both S and Br displayed unimodal patterns with the highest concentrations around 0.25 µm (Fig. 3(C)). S is attributed to the emission from diesel and

Table 1. Summary of the results from PIXE analysis of filter samples collected using a cascade impactor in Nairobi, Kenya, during the month of August and September 2007. All concentrations are in ng m^{-3}.

Elements and PM	Detection limit	Minimum	Maximum	Mean ± Stdev	Percentage of PM	Number of valid samples
Si	11	8.3	1400	260 ± 270	2.7	170
S	3.1	4.4	410	59 ± 68	0.6	170
K	1.8	< DL	270	42 ± 41	0.4	170
Ca	1.5	< DL	170	30 ± 38	0.3	168
Ti	1.1	< DL	56	5.5 ± 8.2	0.1	113
Mn	0.5	< DL	33	4.7 ± 6.2	< 0.1	118
Fe	0.4	< DL	510	69 ± 100	0.7	169
Cu	0.3	< DL	29	0.7 ± 2.3	< 0.1	93
Zn	0.2	< DL	270	22 ± 38	0.2	167
Br	0.2	0.2	13	1.6 ± 1.6	< 0.1	170
Pb	0.4	< DL	32	3.6 ± 5.5	< 0.1	133
PM		1000	78000	9800 ± 8500		170

DL – detection limit.
Note. The concentrations listed in the table have been obtained after the subtraction of the blank values.

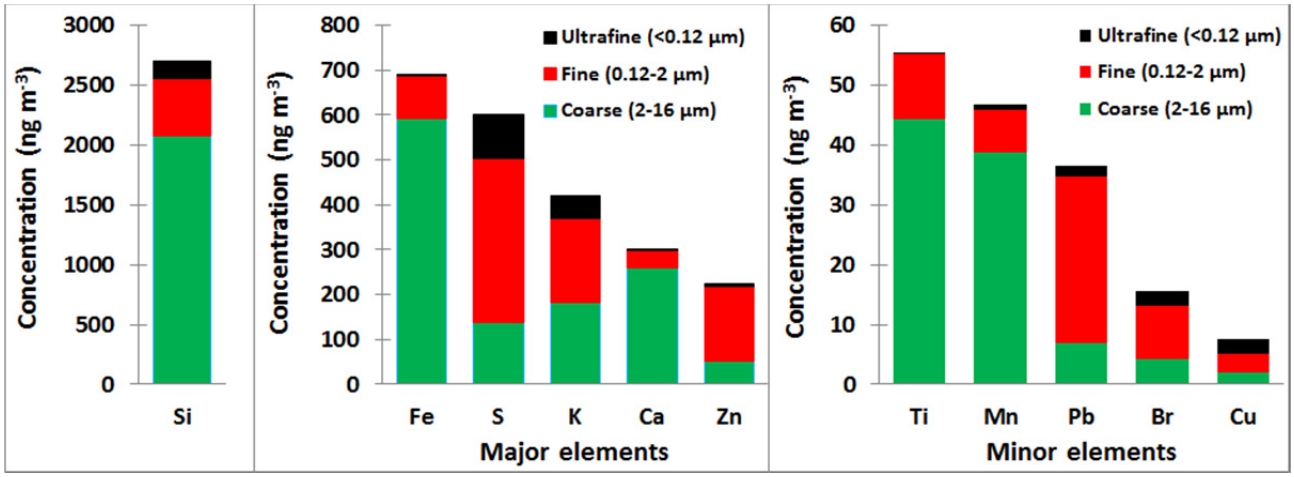

Fig. 2. Distribution of major and minor elements in different size ranges. The size ranges have been defined as coarse, fine and ultrafine.

heavy fuel oil combustion (Tan *et al.*, 2009) whereas Br is attributed to both biomass burning (Cicerone, 1994) and exhaust emissions (Maenhaut and Akilimali, 1987). In Kenya, the diesel fuel for both vehicular and industrial consumption contains 5000 parts per million of sulphur (UNEP, 2014). Both Zn and Pb displayed bimodal patterns with two peaks at size ranges 0.25 and 1 µm (Fig. 3(D)). This similarity was an indication of contribution from multiple sources, namely metal smelting industries and traffic (Chueinta *et al.*, 2000; Chow *et al.*, 2004; Were *et al.*, 2012; Gaita *et al.*, 2014). One noteworthy observation in this study was the pattern of Cl concentrations in the different size ranges. Cl concentration was below detection limit in almost all size ranges except in the 2–4 µm range whereby the average concentration was 54 ± 42 ng m^{-3}. The higher Cl concentrations coincided with the days when the 72 h back trajectories originated from the Indian Ocean (Fig. 1(a), the red line) thus implying marine influence over Nairobi city.

Enrichment Factors

Table 2 shows the aerosol-crust enrichment factors (EF), which were calculated for all the elements as per Eq. (2),

$$EF_{crustal,x} = \frac{(C_x / C_{Fe})_{aerosol}}{(C_x / C_{Fe})_{SSAsoil}} \quad (2)$$

where C_x was the concentration of respective element and C_{Fe} was the concentration of Fe which was used as the reference element. The denominator consisted of soil elements sampled from the Sub-Sahara African region by Towett *et al.* (2013) which were deemed to be representative of the regional concentration of crustal elements. Calculated EF values near unity suggest crustal weathering as the main source of the element in question while much greater EFs indicate other non-crustal sources (Milford and Davidson, 1985). The calculated EFs for Ti and Ca were close to unity indicating that they originated from the same source i.e., mineral dust. The EFs of K were almost equal to unity for particle size above 1 µm and this was a confirmation of crustal dust dispersion source for one portion of K in the filter samples. For particle sizes below 1 µm, the EFs of K increased by a

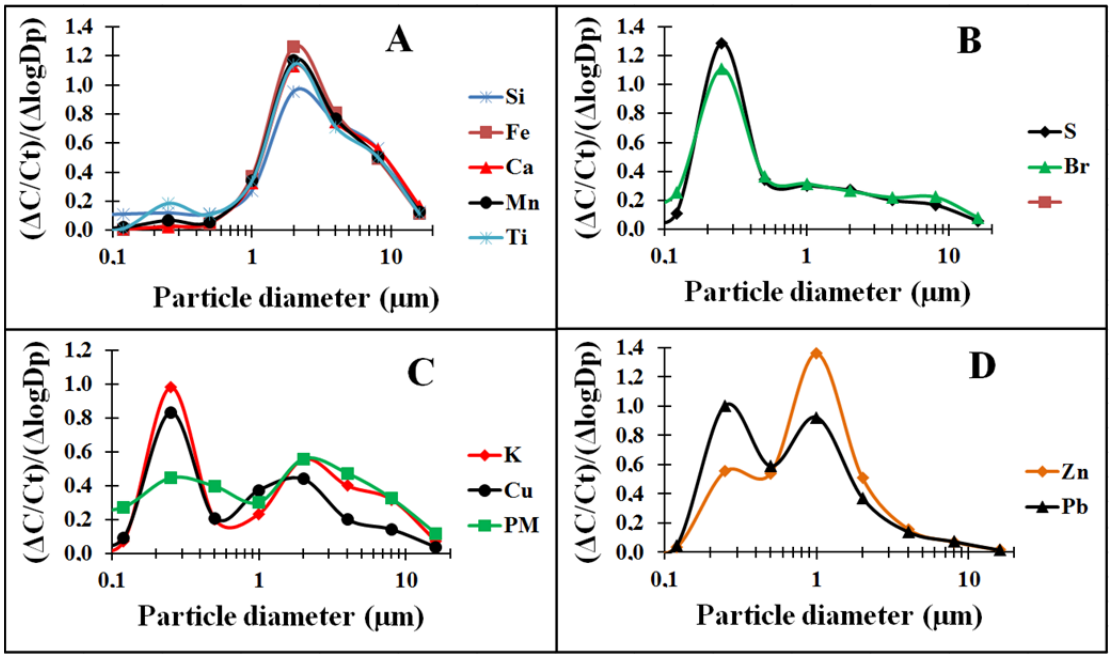

Fig. 3. Normalized mass-size distributions of PM and selected elements highlighting their respective unimodal and bimodal patterns.

Table 2. Enrichment factors calculated using elements from soil samples collected from Sub-Sahara African countries by (Towett *et al.*, 2013).

PM size ranges (µm)	S	K	Ca	Ti	Mn	Fe	Cu	Zn	Br	Pb
> 16	10	0.9	0.3	0.6	3	1	6	30	30	6
16–8	7	0.9	0.3	0.6	3	1	6	30	20	7
8–4	5	0.7	0.2	0.5	2	1	5	50	10	8
4–2	4	0.6	0.2	0.5	2	1	7	100	8	10
2–1	20	0.9	0.2	0.5	2	1	20	900	30	100
1.0–0.5	200	6	0.3	1	3	1	90	3000	300	7000
0.5–0.25	800	40	0.2	3	5	1	500	4000	1000	1000
0.25–0.12	700	30	1	3	20	1	600	3000	3000	700
0.12–0.06	1000	40	2	3	30	1	1000	2000	5000	800
< 0.06	3000	200	0.8	3	30	1	5000	4000	4000	2000

factor of 5 to 40 apart from the particle size < 0.06 µm which increased by a factor of 200. This observation was attributed to contribution of K from biomass burning (Maenhaut and Akilimali. 1987). The EF values for S indicated that the contribution of sulphur in the sampled PM was non-crustal and possibly from high sulphur containing diesel and heavy fuel oil. The observed pattern of the calculated EFs in this study was generally in agreement with values from a study carried out in Kenya by Gatari *et al.* (2009). In that study, the PM was segregated in two size fractions; $PM_{2.5}$ and PM_{10}. Si was not included in the EFs since it was missing in the study by Towett *et al.* (2013)

Deposition Fractions of PM in the Human Respiratory System

The health effects of PM are related to the exposure dose which in turn is a function of the deposited fractions in the human respiratory system as well as the clearance mechanisms of the human body (Winter-Sorkina and Cassee, 2002). Figs. 4(A) and 4(B) show deposition fractions of both coarse and fine PM calculated using MPPD model v 2.11 as described in the method section. Deposition fractions of both coarse and fine PM are highest in the head airways region accounting for 87% and 84% of the total deposited mass respectively. This significant deposition in the head airways region can be attributed to a combination of both sedimentation and inertial impaction of PM on larynx and airways (Zhang and Yu, 1993). Deposition fractions in the tracheobronchial and pulmonary regions were approximately 15% of the total deposited fraction for both coarse and fine PM.

DISCUSSION

Given the increase of severe droughts as a consequence of climate change in Africa, the mineral dust component will continue to play a key role in the air quality issues as highlighted by the dominance of the crustal elements; Si,

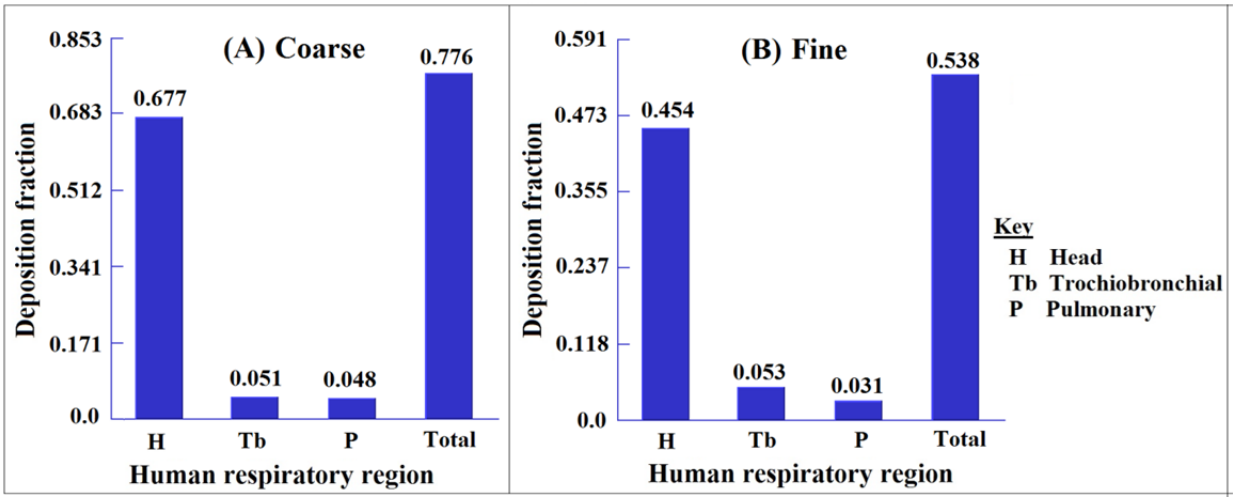

Fig. 4. Deposition fractions of coarse and fine PM in the human head airways, tracheobronchial and pulmonary regions calculated using Multiple Path Particle Dosimetry model, MPPD V2.1

Fe and Ca (WMO, 2013; Gaita *et al.*, 2014). From the size distribution data, the majority of the elements are in PM less than 2.5 µm and thus have higher probability of penetrating deeper into the respiratory system (Anderson *et al.*, 2012). The impact of PM on human health in Nairobi city, as a representative of a typical African urban centre, can be discussed in the context of estimating deposition fractions of both coarse and fine PM. A study in Nairobi city by Gaita *et al.* (2014) identified sources of airborne PM to include traffic, mineral dust, industrial activities and combustion. The said study reported 24 h average $PM_{2.5}$ concentration of 21 µg m^{-3} from an urban background site. Using the deposition fractions from fine PM, the estimated deposited masses are 9.5 µg in the head airways region, 1.1 µg in the tracheobronchial region and 0.7 µg in the pulmonary for every 1000 L of air breathed by an average urbanite in Nairobi city. However, Kinney *et al.* (2011) reported $PM_{2.5}$ average concentration of 98.1 µg m^{-3} from a sidewalk in the Central Business District which translates to a deposition of 45 µg (head airways), 5.2 µg (tracheobronchial) and 3.0 µg (pulmonary). A study on occupational exposure by Ngo *et al.* (2015) reported that bus drivers in Nairobi city were exposed to about 103 µg m^{-3} which translates to a deposition of 46 µg to the head airways, 5.5 µg to the tracheobronchial and 3.2 µg to the pulmonary tract. The deposited PM mass from the last two studies highlights the grave implication of PM pollution which may be linked to increased cases of respiratory related illnesses in Nairobi city (Gulis *et al.*, 2004). The exponential growth of motor vehicles in African urban centres will continue to pose a greater health risk due to lack of regulatory and control measures on the exhaust emissions and type of fuel consumed (KNBS, 2013a, b). A study conducted in Budapest, Hungary by Salma *et al.* (2002) reported highest deposition values in the extrathoracic airways compared with other parts of the respiratory system as is the case in this study. Comparing with a study from Singapore by Behera *et al.* (2015) conducted during smoke haze and non-haze seasons, the reported deposition fractions in the head airways region from that study for both coarse and fine PM were lower than fractions in this study. The probable explanation was the dominance of mineral dust component for PM sampled in Nairobi city compared to the combustion related PM in Singapore.

The obtained deposition fractions in this study were utilized to estimate deposited mass of toxic trace elements measured in the fine PM size range. Of interest, Zn has been reported as toxic in inhalable particulate form as zinc oxide (Cooper, 2008). The slum dwellers in Nairobi city and Accra burn old car tires to extract wires and also use the fires for cooking (Obiri-Danso *et al.*, 2008; Egondi *et al.*, 2013). The slum dwellers are thus exposed to Zn from the tires (Councell *et al.*, 2004) and with high levels of PM from burning tires, the risk of toxicity increases. The exposure to exhaust and non-exhaust traffic emissions as reported by Ngo *et al.* (2015) and industrial pollution, points to the risk of toxicity to Pb and Cu (Pasanen *et al.*, 2012).

SUMMARY

In summary, the PM concentration data from this study has been utilized in calculation of deposition fractions of PM. The obtained fractions give estimated mass deposited in the head airways, tracheobronchial and pulmonary regions of human respiratory system. Although the human body is equipped with removal mechanism, during severe pollution episodes the absorbed PM can be redistributed in the body leading to high risk of overloading and eventual negative health effects such as cardio-related illnesses (Villeneuve *et al.*, 2002). This study has provided information on deposition fractions of pollutant PM in human respiratory in Nairobi city. The information from this study can be used together with the hospital records on respiratory illness to establish an in-depth understanding of the health effects of air pollution in Nairobi city as well as in other African cities.

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