Science of the Total Environment 663 (2019) 265–274

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Respiratory tract deposition of inhaled roadside ultrafine refractory particles in a polluted megacity of South-East Asia

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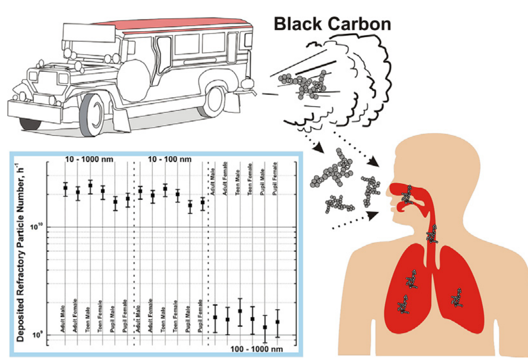
HIGHLIGHTS

- Population from developing regions faces upraise in health related problems.
- Refractory fine particle deposition dose was determined in a developing mega-city.
- The observed deposition dose of refractory fine particles was unprecedentedly high.
- The ultrafine refractory particles dominated deposited particle dose.
- Decisive actions have to be taken to reduce immense black carbon emissions.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 November 2018
Received in revised form 25 January 2019
Accepted 25 January 2019
Available online 26 January 2019

Editor: Pavlos Kassomenos

Keywords:
Air pollution
Black carbon
Exposure
Lung-particle interaction
Respiratory tract deposition

ABSTRACT

Recent studies demonstrate that Black Carbon (BC) pollution in economically developing megacities remain higher than the values, which the World Health Organization considers to be safe. Despite the scientific evidence of the degrees of BC exposure, there is still a lack of understanding on how the severe levels of BC pollution affect human health in these regions. We consider information on the respiratory tract deposition dose (*DD*) of BC to be essential in understanding the link between personal exposure to air pollutants and corresponding health effects. In this work, we combine data on fine and ultrafine refractory particle number concentrations (BC proxy), and activity patterns to derive the respiratory tract deposited amounts of BC particles for the population of the highly polluted metropolitan area of Manila, Philippines. We calculated the total *DD* of refractory particles based on three metrics: refractory particle number, surface area, and mass concentrations. The calculated *DD* of total refractory particle number in Metro Manila was found to be 1.6 to 17 times higher than average values reported from Europe and the U.S. In the case of Manila, ultrafine particles smaller than 100 nm accounted for more than 90% of the total deposited refractory particle dose in terms of particle number.
This work is a first attempt to quantitatively evaluate the *DD* of refractory particles and raise awareness in assessing pollution-related health effects in developing megacities. We demonstrate that the majority of the population may be highly affected by BC pollution, which is known to have negative health outcomes if no actions are

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https://doi.org/10.1016/j.scitotenv.2019.01.338

taken to mitigate its emission. For the governments of such metropolitan areas, we suggest to revise currently existing environmental legislation, raise public awareness, and to establish supplementary monitoring of black carbon in parallel to already existing PM_{10} and $PM_{2.5}$ measures.

1. Introduction

Developing megacities of South-East Asia are not only undergoing rapid socio-economic transformation, but also suffer from high levels of air pollution. The main contributors to poor air quality are the transport sector and residential energy use, which exert negative effects on health, climate, and economics locally, regionally, and globally (Apte et al., 2011; Anenberg et al., 2013; Baklanov et al., 2016). As of 2010, outdoor air pollution is estimated to lead to over 3 million annual premature deaths worldwide with the highest numbers of deaths occurring in Asia (Lelieveld et al., 2015). These numbers are projected to double by 2050, because of an expected massive urbanization (Hugo et al., 2012). Calculations of the burden of disease have shown that, for the first time, air pollution has entered into the 10 most relevant factors leading to premature deaths (Cohen et al., 2017).

One of the most prominent sources of air pollution in developing megacities is private and public transit vehicles, especially by diesel-driven cars and trucks, which often emit high concentrations of Black Carbon (BC) particles (Folberth et al., 2015; Gurjar et al., 2016). The use of diesel engines might have earned its popularity due to its benefits over gasoline engines, e.g. better durability, fuel efficiency (Jung, 2017), and lower cost of the fuel. The emission of carbonaceous particles from diesel engines is however much higher than from gasoline-driven vehicles (e.g. Agarwal et al., 2015). In less affluent economies, this fact is often ignored due to non-existing or less stringent emission regulations. Worse, it can be a common practice there to salvage second hand diesel engines from heavy-duty vehicles for use in public transport (e.g. Baker et al., 1993; Bacero and Vergel, 2009).

Black carbon pollution was found to have a particularly negative effect on human health compared to average outdoor particles (Janssen et al., 2011; Ostro et al., 2014). Cardiovascular and respiratory mortality was associated with exposure to BC in southern Europe (Ostro et al., 2014). In the U.S., 14,000 deaths (approx. 0.6% of total deaths) were estimated to be linked with the exposure to BC (Li et al., 2016). In the South and East Asia, the expansion of the vehicular fleet, and thus, the increase of diesel soot pollution, was estimated to lead to more than 2 million additional premature deaths in 2010 (Lim et al., 2012; Stephen et al., 2017). Results of epidemiological studies were further supported by toxicological evidence that diesel and gasoline exhaust particles negatively affect airway epithelia and lung cells, as well as increase blood pressure (Künzi et al., 2013; Louwies et al., 2015).

To understand the relationship between levels of atmospheric pollution and health related effects, epidemiologists have relied on personal exposure data (Dons et al., 2011; Williams and Knibbs, 2016). Buonanno et al. (2012) have highlighted the need for personal exposure studies as an essential tool for identifying health risks, establishing and reviewing air quality standards and evaluating effective policy interventions. On the other hand, the respiratory tract deposition dose of airborne particles can be an equally (or even more) valuable measure, as it includes the nuances of personal exposure, activity patterns, and physical parameters into one descriptive quantity.

The respiratory tract deposition dose of BC particles was extensively investigated both in situ and in laboratory studies by scientists mostly from the U.S., Australia, and Sweden (e.g. Daigle et al., 2003; Morawska et al., 2005; Löndahl et al., 2012; Rissler et al., 2012; Kristensson et al., 2013). It was found that patients with chronic obstructive pulmonary disease (COPD), the third most common cause of death globally, had an increased deposited dose of carbonaceous particles (Löndahl et al., 2012). According to that study special attention should be paid to vulnerable subgroups and individual dose, especially in the regions where the ambient concentrations of BC are exceptionally high. Such environments are commonly associated with developing countries and rapidly urbanizing megacities (e.g. Ozdemir et al., 2014). Yet, no studies exist on the respiratory tract deposited dose of BC particles in less affluent economies, where air pollution is dominated by the primary emission of traffic-related BC particles.

This work aims at estimating the respiratory deposition of airborne BC particles in the metropolitan area of the megacity Manila, Philippines, where atmospheric composition is strongly affected by particulate diesel emissions. Metro Manila has a population of about 12 million (as of 2010, Philippine Statistics Authority, https://psa.gov.ph, retrieved November 2, 2018). Close to 58% (or 6.8 million) of these were adults between 20 and 64 years old. The adult population comprises 48% male and 52% female subjects. The teenage population (age 10–19 years) in Manila counts about 2.3 million (19% of the total Metro Manila population) with a balanced gender distribution. With respect to road transport, the total number of registered vehicles was 8 million in 2014 (Land Transportation Office, http://openstat.psa.gov.ph/dataset/road-transport, retrieved November 2, 2018). About 24% of these vehicles use diesel, and belong mostly to a pre-Euro4 standard. More than quarter of the vehicles in the Philippines are based in Metro Manila. In Metro Manila, 34% and 66% of the vehicles used gas and diesel, respectively.

To evaluate the respiratory tract deposited dose of refractory particles (a proxy for BC) we consolidated two sets of data: a) information about urban aerosol particle mixing state and refractory particle number size distributions (Kecorius et al., 2017; Alas et al., 2018), and b) time spent in micro-environments by university occupants and their family members (Kecorius et al., 2018). We then identified the sub-groups of individuals mostly affected by particulate pollution and quantified the personal dose of refractory particles in one of the world's most densely populated region.

2. Materials and methods

This study expands on previous efforts of determining the refractory particle number size distributions in the atmosphere of Metro Manila (Kecorius et al., 2017), and analyzing the personal use of a daytime (Kecorius et al., 2018). We refer the reader to these two works for the detailed methodologies of experimentally determined aerosol particle mixing state and the personal daytime use. In the following sections, we will briefly introduce the corresponding data and methods.

The data on atmospheric particle number size distribution, the state of mixing, equivalent black carbon (eBC) and other physical-chemical properties were collected during an intensive research project called the “Manila Aerosol Characterization Experiment 2015” (MACE-2015). MACE-15 was designed to raise awareness on the urgency of monitoring and mitigating the air quality crises in megacities. The observed ambient concentrations of BC were extraordinary high, even for megacity standards. The hourly mass concentration of eBC at the roadside was measured to be up to 138 μg m^{-3}. The results of mobile measurements allowed us to indicate the hot-spots, where concentrations of 80 μg m^{-3} and more were recorded on regular basis (Alas et al., 2018).

2.1. Refractory particle number size distribution

A TROPOS-type volatility tandem differential mobility analyzer (V-TDMA, Philippin et al., 2004) was used to measure size segregated mixing state of aerosol particles. After the data analysis using Gysel et al. (2009) inversion routine, the number fraction of refractory species at 300 °C and geometric mean diameter of approx. 80 nm was shown to represent soot particles (Kecorius et al., 2017). The refractory particle number size distribution was then reconstructed by multiplying the ambient particle number size distribution (PNSD, measured using mobility particle size spectrometer) with the number fraction of low volatility particles in a measured PNSD size range. As the particle volatility was measured only in 6 size bins, we have fitted the retrieved refractory PNSDs with log-normal distributions. To convert between particle number and mass distributions, a bulk three-dimensionally-ordered graphite density of 2.19 g/cm^3 was used (Tesner, 1973). Please note that before applying the number to mass conversion, particle mobility diameter was converted to volume-equivalent diameter using empirical, size-dependent aerodynamic shape factor (Park et al., 2004).

2.2. Activity patterns

In our previous work (Kecorius et al., 2018), we have shown the daily activity patterns of 258 respondents in five different age groups using specifically designed questionnaires. The school/university occupants and their family members were asked to record their daily activity for 1 week, with a specific focus on time spent outdoors and commuting. For this work, adult, teen, and pupil groups were chosen due to low participant numbers in the age groups of toddlers and elderly. Because we did not measure personal exposure to refractory particles in environments other than roadside, this work focuses on time periods when people spent their time either outdoors or in public transport. Simple indoor aerosol models (e.g. Hussein and Kulmala, 2008) do exist and were used to simulate indoor aerosol particle number size distributions based on a mass-balance equation (Hussein et al., 2013). Such models require the outdoor particle number concentration, indoor domain geometries, ventilation rate, penetration efficiency, and dry deposition as input parameters. Unfortunately, none of these parameters are currently available for Filipino households, which made it hard to implement such models in the present study.

Based on the questionnaire results, we divided previously reported activity patterns into three different categories – outdoor city, outdoor work, and transit transport (Fig. 1). We have assumed that during transit transport, the majority of commuting people are subjected to

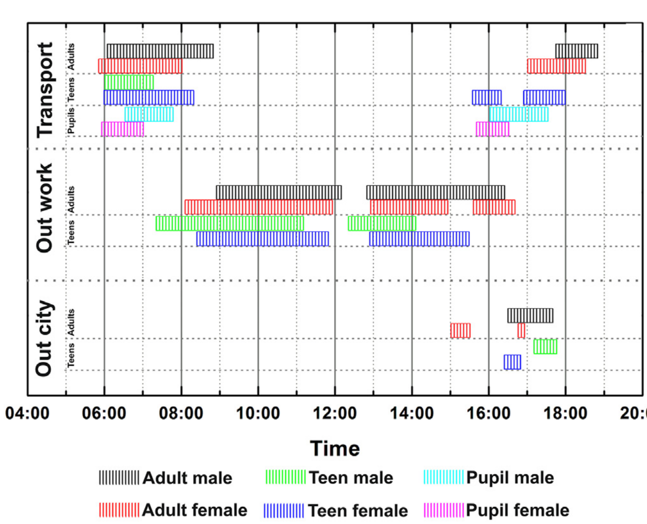

Fig. 1. Daily activity patterns for adult, teen, and pupil age groups in outdoor environments.

roadside pollutant concentrations. This assumption is supported by the fact that popular means of transportation in South-East Asia and the Pacific still remains the public utility *Jeepneys*, scooters, tricycles, busses, and other open-type vehicles (Chin, 2013; Boquet, 2017). In many cases, individuals using such kind of transport would be subjected to much higher (direct tailpipe) pollutant concentrations than those measured at a roadside (e.g. Betancourt et al., 2017). Also note the distinction between two categories of adults and teens working indoors and outdoors. Later in the text, we will discern the deposited dose for people working exclusively outdoors.

2.3. Deposition dose

To calculate the *DD* of refractory particles we have used a mathematical formulation provided by Hussein et al. (2013):

$$DD = \int_{t_1}^{t_2} \int_{D_{p1}}^{D_{p2}} V_E \cdot \xi \cdot d\ \log D_p \cdot dt \qquad (1)$$

The V_E is minute ventilation (m^3/min), ξ - deposited refractory particle number size distribution (particles/cm^3), which can be calculated as $\xi =$ *deposition fraction* x *particle number size distribution*, and D_p is a volume equivalent particle diameter (nm). The time for exposure was obtained from activity tables. The refractory particle dose was calculated in fine (10–1000 nm), ultrafine (10–100 nm) and accumulation (100–1000 nm) modes by integrating the deposited refractory particle number size distributions in the corresponding size ranges. The different metrics (e.g. surface area, mass) *DD* were calculated by introducing a πD_p^2 (in case of surface area) or $\frac{\pi}{6} D_p^3 \rho_p$ (in case of mass) term in Eq. (1).

Calculation of the *DD* requires information about a) volume of air breathed; b) deposition fraction of particles in the respiratory system, and c) refractory particle number size distribution measured in an environment, where the subjects spent their time. Hussein et al. (2013) have also used hygroscopic particle parameterization in his *DD* calculations, which we will not consider in our study. The neglecting of the hygroscopic growth factor in our *DD* calculation is based on the fact that freshly emitted soot particles in an urban environment are nearly hydrophobic (Weingartner et al., 1997; Löndahl et al., 2009).

2.4. Deposition fraction (DF)

There are two ways to obtain the deposition fraction (DF) of particles in the respiratory system: 1) experimentally, or 2) using particle deposition models, e.g. Multiple-Path Particle Dosimetry Model (MPPD, Chemical Industry Institute of Toxicology, Research Triangle Park, NC). Although both methods have widely been used, each suffers some intrinsic limitations. For example, the experimental determination of DF must be ethically approved, requires a carefully prepared set-up, up-to-date equipment, and a controlled environment, all of which increase the complexity of such experiments. On the other hand, commonly used particle dosimetry models do not account for particle hygroscopic growth and are generally poor at predicting individual differences in deposition, especially for vulnerable subgroups.

In this work, we use the averaged DF calculated from previous studies (Fig. 2), which include DFs from wood combustion particles (Löndahl et al., 2007; Kristensson et al., 2013) as well as busy roadside and diesel combustion particles (Löndahl et al., 2009, 2012; Rissler et al., 2012). These are the environments abundant in refractory particles and thus shall well represent Metro Manila. We have also discerned the highest and lowest reported DFs, which we used to calculate the higher and lower ranges of deposited doses. We chose averaged DF, because of two reasons. Firstly, at the time of this study, the experimentally determined DFs for Filipinos were not available. Secondly, to model the DF, information about physiological parameters, such as functional residual capacity, upper respiratory tract volume, breathing frequency,

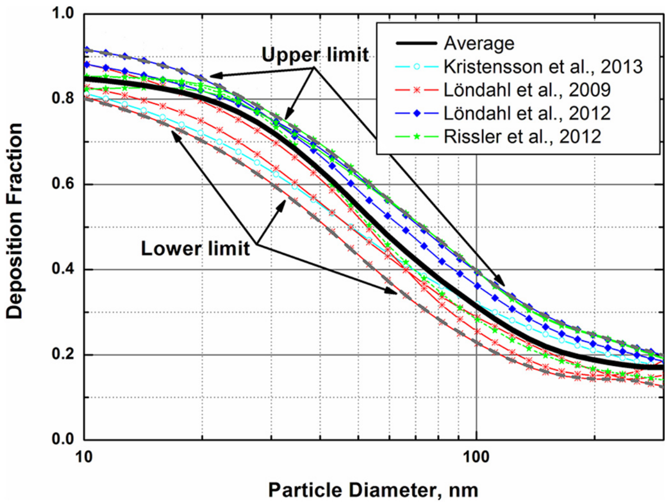

Fig. 2. Size-dependent deposition curves reported in previous studies (experimental and model) and averaged DF used in this study, which is the mean of previously reported curves for wood and diesel combustion particles. Note: In the study by Kristensson et al. (2013), the authors used deposition fraction from Löndahl et al. (2007).

tidal volume etc. is needed. Unfortunately, such information was also not available at the time of this study. Some physical parameters could be used from research notes prepared by the California Environmental Protection Agency. However, it is known that the lung volumes among various ethnic or racial groups may vary (Donnelly et al., 1991), which may increase the uncertainties in final DF calculations.

Although some studies on pulmonary and ventilatory functions in adult Filipinos do exist (Lin and Kelso, 1999; Roa et al., 2013), the desired parameters to model the DF remain unknown. For the reasons mentioned above, we have chosen to use previously reported deposition fractions of mostly hydrophobic combustion particles.

2.5. Volume of breathed air

Another important parameter in evaluating the deposited particle dose is the breathed air volume (minute ventilation, V_E, in Eq. (1)). We have used V_E values reported by Natera et al. (1998) for adult Filipino males and females during light physical activity. In parallel, because the minute ventilation for Filipino teens and pupils are not reported, adult V_E values were adapted for teenagers, and the V_E values for pupils were taken from Smith (1994). These values are shown in Table 1. The V_E value for adult males was around 11% lower and the values for adult females – 4% higher than those reported by Smith (1994). When comparing V_E values for teens, our used values were 2% and 0.2% lower than the values reported for Caucasian teen males and females, respectively.

The authors are aware that by using the values reported for Caucasians and not Filipinos may increase the calculation uncertainties, however, values for Filipino teenagers and pupils were not available at the time of this study. Despite this, in the work by Hussein et al. (2013) and Rissler et al. (2017), it was noted that the breathing pattern and deposition fraction for teens (and children) are expected to be similar to that of adults. If the information on V_E will become available in the future, it will be possible to recalculate the deposition dose according to the methodology provided in this study.

Table 1
Volume of air breathed reported for adult Filipinos by Natera et al. (1998) and Caucasian pupils by Smith (1994). Volume of air breathed is in m^3/h.

	Age (±stdev)		Number of people		Volume of air breathed	
	Male	Female	Male	Female	Male	Female
Adult	33 ± 14	35 ± 14	71	92	1.4	1.3
Teen	16 ± 2	16 ± 2	20	43	1.4	1.3
Pupil	9 ± 1	9 ± 1	10	10	1.1	1.1

2.6. Deposited refractory particle concentrations

The final parameter needed to calculate the respiratory tract deposited dose is the deposited refractory particle number size distribution (parameter ξ in Eq. (1)). As already mentioned, ξ is a product of deposition fraction and refractory particle number size distribution (r-PNSD). In this study, we have used previously derived r-PNSD with 1-h time resolution (Kecorius et al., 2017), which can also be found in a supplementary material (SP1–3). To simplify the calculation of Eq. (1) and to provide the deposited dose of refractory particles in three size ranges we have firstly fitted ξ with a log-normal function, and then integrated ξ between 10 to 1000, 10 to 100 and 100 to 1000 nm to get deposited refractory particle number concentrations in fine (FP), ultrafine (UFP) and accumulation (ACCUM) modes. Deposited refractory particle surface area and mass concentrations in three size ranges were calculated from ξ (assuming bulk particle density of 2.19 g/cm^3). The deposited particle size distributions and the integrated concentrations can be found in Fig. 3. Because observed r-PNSDs during working days and weekends were rather similar, in this work we have used only working day r-PNSD and time activity information to determine refractory particle deposition dose.

2.7. A conversion between total and refractory particle number

In this study, our main goal is to assess the respiratory tract deposition dose of refractory particles, which is a proxy for BC. To highlight the magnitude of the deposition dose in Metro Manila, we also aim to compare the results observed in this study to previous investigations. Unfortunately, no similar studies exist to this date, making a direct comparison rather difficult. Several attempts were made to estimate the deposited dose of traffic generated combustion particles (e.g. Löndahl et al., 2009; Kumar et al., 2012; Manigrasso et al., 2017). However, the deposited dose of refractory particle was not investigated. Instead, generic particle number, surface area and mass deposition in the airways were reported. Further in this paragraph, we would like to explore the possibility to evaluate the refractory particle number concentration. The relation between generic and refractory particles can be drawn if information on the refractory particle number fraction is known. In this work, we use the refractory particle number fractions from an urban street canyon in Germany, reported by Rose et al. (2006). Combining the information about refractory particle number fraction with particle number size distributions measured at the same location during the two year period (2014 to 2016, excluding winter months because of bias to refractory fractions due to domestic heating) we determined the roadside total refractory particle number ($f_N = \frac{N_{total}}{N_{refractory}} = 0.25$), surface area ($f_{SA} = \frac{SA_{total}}{SA_{refractory}} = 0.35$) and mass ($f_M = \frac{M_{total}}{M_{refractory}} = 0.34$) factors, which we then used to transform between total generic and refractory particle (number, surface area and mass) concentrations. It has to be noted that the refractory particle number, surface area or mass concentrations retrieved using f_N, f_{SA} and f_M can result in an over-/underestimation of the corresponding refractory particle values. This is because our used factors resemble the environment, where the diesel vehicles constitute 22% of the total vehicular fleet (https://www.kba.de, accessed December 12, 2017). For example, in case of Greece, the average diesel car fraction is approximately 10%, while in Luxemburg this fraction is above 70% (as of 2011, Cames and Helmers, 2013). Although this method is rather qualitative, it allowed us to put our study into the perspective of other works, enabling the comparison between our observed and previously reported results.

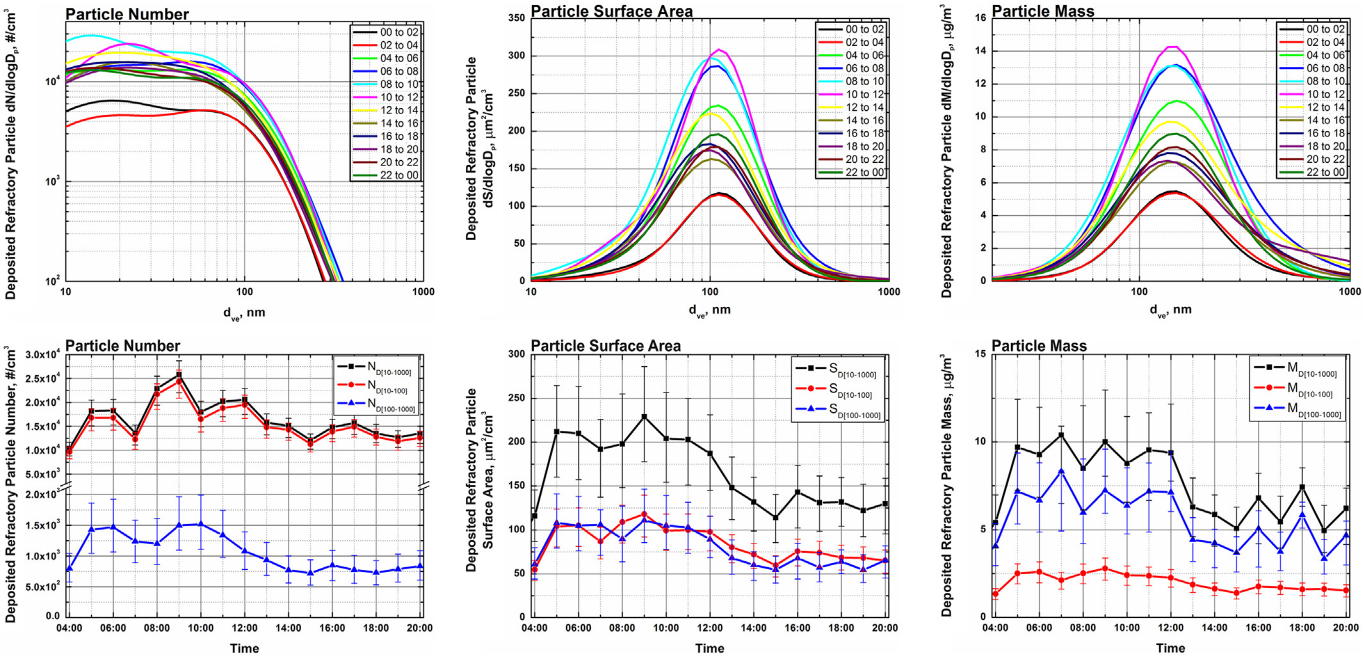

Fig. 3. Daily variation of deposited refractory particle number, surface area and mass size distributions (upper row, as $dN/dlogD_p$) and integrated deposited concentrations (bottom row) in different size ranges. Error bars show minima and maxima values resulted from upper and lower limits of the deposition fraction used (see Section 2.1).

3. Results and discussion

3.1. Deposited refractory particle concentrations

Daily variation of deposited refractory particle size distributions and integrated concentrations are shown in Fig. 3. In the upper row of the Fig. 3, the averaged deposited particle number, surface area and mass size distributions (DPNSD, DPSASD, and DPMSD, respectively) in 2-hour time intervals are presented. Two-mode structure can be noticed in the DPNSD. In our previous work (Kecorius et al., 2017), we have identified two modes of refractory particles. The smaller, nucleation mode (approx. 20 nm) was associated with refractory non-soot particles (most likely ash, tiny carbonaceous and/or nucleated and oxidized organic particles), while the larger ultrafine particle mode (at around 80 nm) were shown to be soot agglomerates. Nucleation mode refractory particles contributed to more than 50% of the total deposited particle number concentration and together with ultrafine particles dominated the deposited fine particle mode. It indicates that approximately half of deposited refractory particle number in the respiratory system derives from nucleation mode particle size range. The DPSASD peaked at ~110 nm with nucleation mode input to the total DPSA concentration decreasing to only 6%. This contribution of nucleation mode particles decreases even further (to 1%) in the case of DPMSD, which had a maximum at a geometric mean diameter of 150 nm.

The deposited refractory particle size distributions were integrated to obtain deposited refractory particle concentrations (number – DPN; surface area – DPSA; and mass – DPM) in fine, ultrafine, and accumulation modes (Fig. 3 lower row and Table 2). The average DPN concentration of fine mode particles (integrated from 10 to 1000 nm) ranged between $5.4 \cdot 10^3$ and $2.6 \cdot 10^4$ with a mean value of $1.6 \cdot 10^4$ refractory particles per cubic centimeter (rp/cm^3). The lowest values were observed during evening and night hours, and the highest – during the rush hours. This is because the deposited refractory particle concentration is directly proportional to observed refractory particle concentration at a measurement site; the deposited concentrations of refractory particles follow the same daily pattern as the roadside particle concentration. That is, the highest deposited refractory particle concentration was observed when the traffic intensity was at its highest and the meteorological conditions favored the accumulation of the pollutants (Kecorius et al., 2017). It can also be seen that the ultrafine refractory particles exclusively (more than 90%) dominated the DPN. When considering the higher and lower limits of the deposition fraction (see Section 2.1), the calculated values of DPN, DPSA, and DPM concentrations were on average 16% higher and 22% lower than the average values, respectively.

Table 2
Average deposited refractory particle (rp) number, surface area and mass concentrations in different size ranges.

Particle mode	Number $(\cdot 10^4)$	Surface area	Mass
	rp·cm^{-3}	μm^2·cm^{-3}	μg·m^{-3}
Fine [10–1000 nm]	1.6	160.0	7.4
Ultrafine [10–100 nm]	1.5	82.0	1.9
Accum. [100–1000 nm]	0.1	78.0	5.5

The average DPSA and DPM concentrations of fine mode particles were 160 μm^2·cm^{-3} and 7.4 μg·m^{-3}, respectively. The DPSA concentration between 5 a.m. and 1 p.m. varied within a range of 150 and 250 μm^2·cm^{-3}. In afternoon the values slightly decreased, however, remained as high as 100 to 150 μm^2·cm^{-3} during the rest of the day. Ultrafine and accumulation mode DPSA concentrations were somewhat comparable and were approximately 100 μm^2·cm^{-3} during the daytime and slightly lower, between 50 and 75 μm^2·cm^{-3} in the afternoon and at night. Same pattern can also be seen for the DPM concentration - in the first hours of the day it was as high as 10 μg·m^{-3}, and decreased to just above 5 μg·m^{-3} in the afternoon. The DPM concentration of ultrafine particles did not change much and remained at around 1.9 μg·m^{-3} throughout the day. The refractory ultrafine particle contribution to DPSA and DPM concentrations were correspondingly 50 and 25%.

In general, it is also useful to compare our observed values with previously literature-reported values. Such a comparison can provide information whether the results summarized in this study are comparable with other investigations. Unfortunately, a direct comparison between deposited refractory particle number, surface area, and mass concentrations is not possible due to the absence of such studies at the time of this work. The most adjacent parameter to compare is the particle lung deposited surface area (LDSA). The LDSA was identified to be linked to negative health effects of aerosol particles (e.g. Oberdörster, 2000). Unfortunately, all studies focusing on the LDSA do not yet distinguish between generic and refractory particles (e.g. Kuuluvainen et al., 2016; Hama et al., 2017). We have used the alveolar deposition fraction of the hydrophobic particles reported by Kristensson et al. (2013) to calculate the total LDSA concentration of refractory particles for an alveolar region (see supplementary material SP4). The average total LDSA concentration of refractory particles in Metro Manila was found to be 90 $\mu m^2 \cdot cm^{-3}$. The highest and the lowest total LDSA concentrations of approx. 130 and 62 $\mu m^2 \cdot cm^{-3}$ were observed during traffic rush hour (9 a.m.) and afternoon/night time, respectively. In a similar study by Hama et al. (2017), LDSA concentration in Leicester, UK, varied between 20 and 60, with an average of 37 $\mu m^2 \cdot cm^{-3}$. It can be seen that the average total LDSA of refractory particle concentration in Metro Manila, Philippines is twofold higher than those reported from Europe.

It must be noted that here we compare total refractory particle LDSA (in case of Metro Manila) to a generic particle LDSA concentration. The values become noticeably higher if the generic-to-refractory particle conversion factor is used (see Section 2.5). In this case, the total refractory particle LDSA concentration at a roadside in Metro Manila is up to 7 times higher than in European cities. The same studies have also reported peak LDSA concentrations as high as 200 $\mu m^2 \cdot cm^{-3}$ (70 $\mu m^2 \cdot cm^{-3}$ if converted to refractory LDSA using $f_{SA} = 0.35$). In the case of Metro Manila the total LDSA concentration of above the 100 $\mu m^2 \cdot cm^{-3}$ was not a momentary deviation from the average value, but a sustained occasion of extremely high refractory particle LDSA concentrations over an extended period of time (from 5 to 12 a.m.).

3.2. The deposited dose of refractory particles

Combining the information about the deposited refractory particle concentrations, the daily activity patterns and minute ventilation, allowed us to determine the total deposited dose of refractory particles (DDRP) in the human respiratory tract. The main results are shown in Fig. 4 and are summarized in Table 3. Here, we present the results as a cumulative DDRP, meaning that the exposure time and the minute ventilation are the main factors determining the DDRP. The daily activity pattern is likewise important; however, in our case, did not influence the deposition dose significantly. This is because more than half of the DDRP was already received before midday, when the exposure and the deposited refractory particle concentrations were the highest (Fig. 3). Further, we would like to distinguish between individuals working outdoors and indoors. Street vendors, public utility *Jeepney* drivers, traffic enforcers etc., (presented as black circles in Fig. 4) spend approx. 10 h of their daytime at a roadside. Naturally, their cumulative DDRP number was found to be on average 3 times higher than those spending their time indoors, $2.46 \cdot 10^{11}$ versus $7.1 \cdot 10^{10}$, respectively. The same is also valid for DDRP surface area (24.4 versus 7.5 cm^2) and mass (110.5 versus 35.0 μg). As for the deposited refractory particle number concentration, the ultrafine refractory particles constitute more than 90% of the total deposited particle number.

Adult males, who reported to be working outdoors, were found to receive the highest deposition dose of refractory particle number ($2.83 \cdot 10^{11}$), surface area (27.9 cm^2) and mass (127.0 μg). This is because their exposure time to roadside pollution was the highest. And vice versa, the pupil, spending least time on the roadside have received the lowest DDRP number ($4.1 \cdot 10^{10}$), surface area (4.5 cm^2) and mass (20.7 μg). In general, the variation in deposited dose between different age groups can be explained by the discrepancies in minute ventilation and the exposure time as the deposition fraction used in this study was identical for all age groups.

Strikingly, when compared to other cities, people from Metro Manila receive, in 3 to 10 h of their daytime spent outdoors, on average 1.5 to 5.1 times more particles (in number) than people in e.g. Europe during the whole 24 h time period (e.g. Hussein et al., 2013). When comparing DDRP surface area and mass, our observed and previously reported values did not differ more than 10%. Please recall that the values reported by Hussein et al. (2013) are for generic particles. If we use the conversion factor from generic-to-refractory particles (described in Section 2.5) and assume that the individuals spend 3 h of their daytime at a traffic influenced roadside, we find that the cumulative DDRP number, surface area and mass in Metro Manila are on average 6, 3, and 2 times higher than the values reported from western countries, respectively; even when compared to a cumulative 24 h deposition dose (see Aleksandropoulou and Lazaridis, 2013; Hussein et al., 2013; Hussein et al., 2015).

The more general way to present the deposited dose of particles is to normalize dose to the time of exposure. The average roadside exposure times of individuals, who reported to be working outdoors and indoors were 10.5 and 3.5 h, respectively. Dividing the DDRP number, surface area, and mass by the exposure times, we get corresponding averages of DDRP$\cdot h^{-1}$ to be $2.1 \cdot 10^{10}$, 2.2 cm^2 and 10.2 μg. The highest average number, surface area, and the mass DDRP$\cdot h^{-1}$ was received by adults ($2.2 \cdot 10^{10}$, 2.2 cm^2 and 10.3 μg, respectively) and teens ($2.3 \cdot 10^{10}$, 2.4 cm^2 and 10.7 μg, respectively), while the lowest DDRP$\cdot h^{-1}$ was received by pupils ($1.7 \cdot 10^{10}$, 1.9 cm^2 and 8.9 μg, respectively). It can be seen that the teen age group received slightly higher (on average 4% more) DDRP$\cdot h^{-1}$ than the adults. The difference can be explained by the daytime activity.

In contrast to other studies, Kumar et al. (2012) have provided a comprehensive overview of nanoparticles in European cities, which also included the respiratory tract deposition doses received by the public exposed to roadside aerosol particles. The values were in a range from 1.17 to 7.56 with an average of $2.9 \cdot 10^{10}$ particles h^{-1} (excluding data from Zürich, Lahti, Birmingham, and Leicester due to unusually high particle concentrations). However, if we convert these values to refractory particles, we find that in Metro Manila the number of

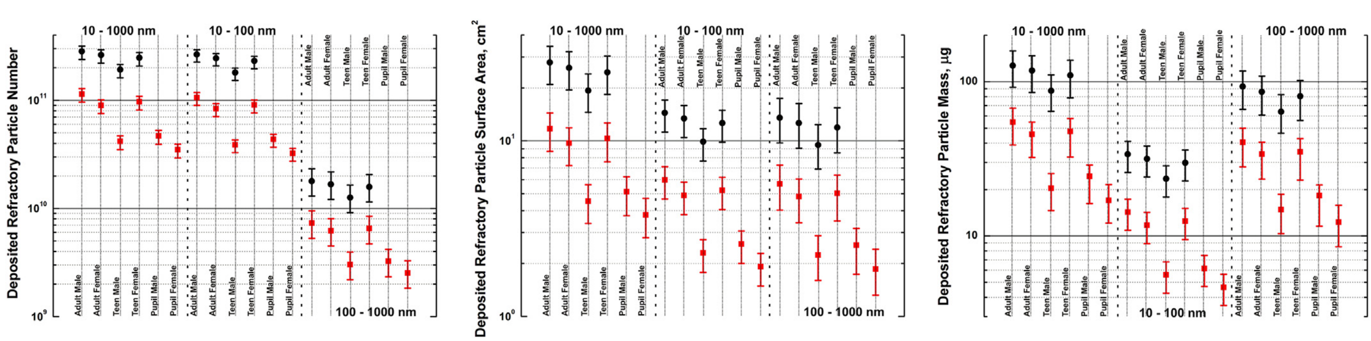

Fig. 4. Cumulative, size segregated deposited refractory particle number, surface area and mass dose received by different age groups when the individuals spent their daytime outdoors. Black and red markers represent the deposition dose of the subjects working outdoors and indoors, respectively. Error bars show minima and maxima values resulted from upper and lower limits of the deposition fraction used (see Section 2.1).

Table 3
Deposited dose of particles in three different size ranges (FP – fine particles, UFP – ultrafine particles, ACCUM – accumulation mode particles), and refractory particle deposition dose observed in this study. Please note that here we present only the results on the refractory particle deposition dose due to outdoor exposure. Information regarding indoor microenvironments was not available at the time of this study.

	Exposure time	Particle number ($\cdot 10^{10}$)			Particle surface (cm^2)			Particle mass (μg)		
		FP	UFP	ACCUM	FP	UFP	ACCUM	FP	UFP	ACCUM
Subjects working outdoor (this study)										
Adult male	12:05	28.3	26.5	1.8	27.9	14.4	13.5	127.0	33.9	93.1
Adult female	11:40	26.2	24.5	1.7	26.0	13.4	12.6	117.7	31.6	86.1
Teen male	07:30	19.2	18.0	1.3	19.3	9.9	9.5	87.4	23.5	63.9
Teen female	10:55	24.7	23.1	1.6	24.5	12.6	11.9	110.1	29.8	80.4
Subjects working indoor (spending less time outdoor; this study)										
Adult male	05:05	11.4	10.6	0.7	11.7	6.0	5.7	54.8	14.3	40.5
Adult female	04:35	9.0	8.4	0.6	9.7	4.9	4.8	45.6	11.7	33.9
Teen male	01:50	4.2	3.9	0.3	4.5	2.3	2.2	20.4	5.6	14.8
Teen female	04:45	9.7	9.1	0.7	10.3	5.2	5.0	47.6	12.5	35.1
Pupil male	02:45	4.7	4.4	0.3	5.1	2.6	2.6	24.4	6.2	18.3
Pupil female	01:55	3.5	3.2	0.3	3.8	1.9	1.9	17.0	4.6	12.3
Hussein et al. (2013)[a]		FP	UFP	ACCUM	FP	UFP	ACCUM	FP	UFP	ACCUM
Adult male	24 h dose	5.7	5.3	0.3	8.0	1.8	6.3	39.5	1.6	38.0
Adult female	24 h dose	4.0	3.8	0.2	5.5	1.3	4.3	27.2	1.1	26.1
Teen male	24 h dose	5.5	5.2	0.3	7.7	1.8	6.0	38.1	1.5	36.6
Teen female	24 h dose	4.0	3.8	0.2	5.6	1.3	4.0	27.6	1.1	26.5
Hussein et al. (2015)[a]		FP	UFP	ACCUM	FP	UFP	ACCUM	FP	UFP	ACCUM
Unspecified	24 h dose	32.8	32.4	0.3	–	–	–	45.8	5.0	40.8
Kumar et al. (2012)[b]										
Adult male	01:00	2.9	–	–	–	–	–	–	–	–
Aleksandropoulou and Lazaridis (2013)										
Unspecified	24 h dose	–	–	–	–	–	–	99.1	–	–
Löndahl et al. (2009)[a]										
Unspecified	01:00	0.63	–	–	0.49	–	–	4.32	–	–
Manigrasso et al. (2015)[a]										
Unspecified	01:00	6.6	–	–	–	–	–	–	–	–
Martins et al. (2015a)[c]								$PM_{2.5}$		
Unspecified	01:00	–	–	–	–	–	–		1.8, 3.9, 54.8, 1.5	

[a] Values reported as a total particle number, not segregating between less/more volatile and refractory species.
[b] Values in a size range from 10 to 1000 nm, not segregated between less/more volatile and refractory species. Zurich, Lahti, Birmingham and Leicester was excluded from the comparison due to unusually high particle concentrations.
[c] Total deposition dose of $PM_{2.5}$ particles at home (1.8 μg), outdoor (3.9 μg), subway (54.8 μg) and workplace (1.5 μg).

DDRP$\cdot h^{-1}$ is on average 3 times higher than in Europe. Compared to studies by Löndahl et al. (2009) and Manigrasso et al. (2015), we find that DDRP$\cdot h^{-1}$ observed in Metro Manila is more than 10 times higher than the values reported from the studies in Europe.

The instances when the reported particle deposition dose in terms of number and mass was higher than our observed values were in the studies by Manigrasso et al. (2017) and Martins et al. (2015a). In the case of Manigrasso et al. (2017), the cumulative deposition dose of particle number per hour was reported to be more than $1.4 \cdot 10^{14}$. That is approx. 1600 times higher than the values observed in our study, even after the refractory particles conversion factor was applied. One possible explanation might be that instrumentation used in the work by Manigrasso et al. (2017) heavily overestimates the exposure concentrations of the particles. In a study by Martins et al. (2015a), the generic particle deposition dose of $PM_{2.5}$ in the subway was found to be 54.8 μg. Here, the majority of aerosol particles must have come from non-combustion related processes – the abrasion and wear of rail tracks, wheels and braking pads, and the resuspension of dust (e.g. Martins et al., 2015b). Thus, the composition of deposited particles coming from a subway system must be much different, which hinders the direct comparison to our observed values.

3.3. Limitations of the study

Particle number/surface area/mass size distributions, respiratory tract deposition fraction, individuals' physical parameters (e.g. minute ventilation, tidal volume), and daytime activity patterns are the decisive measures in characterizing the deposited dose of aerosol particles. Ambiguities in these parameters determine the deposited dose itself. Further in this section, we will discuss the limitations of this work focusing on previously mentioned parameters.

The refractory particle number size distribution can be considered as one of the most robust parameters in this study. Determined from the combined measurements of the mobility particle size spectrometer and the volatility differential mobility analyzer, it proved to be a suitable proxy for soot (Kecorius et al., 2017). To ensure that our measured quantities precisely characterize the properties of particles, we have used the closure between measured equivalent black carbon mass concentrations (using Multiple Angle Absorption Photometer (MAAP, Model 5012, Thermo, Inc., Waltham, MA USA) and retrieved refractory particle number size distributions. The refractory particle mass concentration derived from the number fraction of low-volatility particles and equivalent black carbon mass (as soot proxy) directly measured with MAAP was in a reasonable agreement (slope of 0.992 and $R^2 = 0.85$).

The respiratory tract deposition fraction and individuals' physical parameters, on the other hand, are one of the weaker aspects of this work. The rationale behind deposition fraction calculation is given in Section 2.1. In general, we do not expect deposition fraction used in this work to differ significantly from the real, yet unknown one. By using the lower and higher ranges of deposition fractions, derived from previous studies, we see that the calculated values of deposited particle number, surface area, and mass concentrations differ on average by ±20%. In other words, our determined average deposited dose of refractory particles may have been either 20% lower or 20% higher depending on the specific situation. Even if the real deposited dose was 20% lower, it still does not diminish the magnitude of the deposited refractory particle dose in the case of Filipinos.

The information about the use of daytime, retrieved from time activity tables, may also carry some level of uncertainty. In this study, we have used the data from 258 reports on daytime use (Kecorius et al., 2018). More statistically robust data is needed, especially from developing countries. The activity logbooks, albeit useful, were also recognized as only partly reliable source of information. Use of sensors to identify the operation of stoves, ovens, door, and window opening together with a combination of global positioning system was suggested as more accurate ways to determine personal location and activities (Isaxon et al., 2015).

Last but not least, the conversion between generic and refractory particles must be taken critically, likewise, the comparison of the results to other studies. As it was already noted before, only limited number of studies exist on determining the refractory particle deposition dose. It makes this study unique, yet, hard to put into context of other works. We have tried to solve this shortcoming by introducing the generic-to-refractory particle conversion factor, which can be applied for Europe (under assumption that vehicular fleet is similar to that of Germany). It allowed us to scale the reported values as if the measurements were done for refractory particles. By converting the values reported from Europe, we expect a slight overestimation in the refractory particle concentrations (see more details in Section 2.5). That is, the differences reported in this study are expected to be higher if the refractory particles are compared.

To summarize, in this study, the respiratory tract deposited dose of refractory particles obtained per hour ($DDRP \cdot h^{-1}$) represents the urban roadside environment and is a higher estimate in comparison to other environments (urban background, parks, and residential areas). Indoor exposure to BC particles, e.g. while cooking, was also recognized to contribute significantly to daily deposition dose of particulate pollutants (e.g. Buonanno et al., 2013). In this work, however, we did not have concrete information available, neither on indoor refractory particle number concentrations nor detailed information about the relevant indoor environment (e.g. geometry, ventilation rate, building shell penetration coefficients, etc.) to model the indoor BC concentrations, as described in Hussein et al. (2015). As a consequence, we needed to limit our present study to the estimation of the refractory particle deposition dose in outdoor exposure scenarios. In future, we assume that the door/outdoor relationships of BC in developing South-East Asia regions will receive growing scientific attention. Another aspect of the study is that we have calculated the $DDRP \cdot h^{-1}$ for healthy subjects. Previously, it was shown that the deposited dose of particles is higher for subjects with asthma (Chalupa et al., 2004) and COPD due to higher ventilation rates and deviating deposition fractions (Löndahl et al., 2012). Dose also increases with level of exercise due to higher ventilation rates. None of which was sufficiently investigated in developing regions. In other words, there are still considerable knowledge gaps in determining the real-world respiratory tract deposition of air pollution in the lower income countries. Despite this, the presented results adequately show that the deposition dose of refractory particles may be exceptionally high in the regions with immense BC pollution.

4. Summary and conclusions

In developing regions, the strongest contributor to the degrading air quality is the road transport sector. Here, in many instances, missing, obsolete, and uncontrolled exhaust after-treatment technologies cause the increase in unprecedented concentrations of black carbon (BC) particles. This particular type of pollution was recognized by the World Health Organization as a major risk factor for human health. Despite the scientific evidence of extraordinary high concentrations of carbonaceous particles in poorer economies, no studies exist, which concretely estimate the respiratory tract deposition dose of refractory particles.

In this work, we combine our two previous studies on refractory particle concentrations and activity patterns in a highly polluted metropolitan area, the megacity of Metro Manila, Philippines, in South-East Asia, to demonstrate how uncontrolled urban BC pollution affects the personal deposition dose. We calculated the total deposited dose of refractory particles (a proxy for soot) based on three concentration metrics: refractory particle number, surface area, and mass. The average total deposited dose in adults, teens, and pupils in the vicinity of traffic was $2.2 \cdot 10^{10}$ (surface: 2.2 cm^2; mass: 10.3 μg), $2.3 \cdot 10^{10}$ (surface: 2.4 cm^2; mass: 10.7 μg), and $1.8 \cdot 10^{10}$ (surface: 1.9 cm^2; mass: 8.9 μg) refractory particles per hour, respectively. The variation in deposited dose between different age groups can be explained by the discrepancies in minute ventilation and time activity patterns. Compared to previous studies, calculated deposition dose of total refractory particle number in Metro Manila was from 1.6 to 17 times higher than values reported from Europe and U.S.

In the course of this work, we identified the following knowledge gaps: a) information about respiratory functions and physical parameters (e.g. minute ventilation, tidal volume, etc.) of the population from different socio-economic backgrounds is far from complete; b) information about residential exposure to particulate pollution and the relation between indoor/outdoor particle concentrations is lacking; c) information about daily activity patterns is limited; d) air pollution may be monitored using inadequate instrumentation (we recommend supplementary black carbon measurements to already existing $PM_{2.5}$ and PM_{10} measurements).

This study is the first attempt to quantitatively evaluate the respiratory tract deposition dose of refractory aerosol particles, and provide the information needed to assess the health related effects based on individual dose in polluted megacities. As the declined heart rate variability (indicator for distress) was directly linked to an increased aerosol particle deposition in the respiratory system, we suspect that the population of the developing megacities may face increased cardiovascular morbidity and mortality, if the emissions of black carbon remain uncontrolled and no concrete mitigation strategies are in place. The review of air quality guideline values with the scope to reduce traffic-related BC emission may have a potential to substantially improve public health in developing countries.

Acknowledgements

This study was supported by the Partnership for Clean Air Inc., the Researchers for Clean Air, Inc., the Metro Manila Development Authority, the City Office of Manila, De La Salle University (through URCO project number 35FU2TAY11-3TAY12 and the visiting professor grants of the Vice-Chancellor for Academics), CHED (COE research grant of Dr. Vallar and Dr. Galvez) and Leibniz-Institut für Troposphärenforschung e.V. (TROPOS). Mylene Cayetano acknowledges the support of the Ministry of Science, ICT and Future Planning in South Korea through the International Environmental Research Center and the UNU & GIST Joint Programme on Science and Technology for Sustainability in 2015.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.01.338.

References

Agarwal, A.K., Gupta, T., Bothra, P., Shukla, P.C., 2015. Emission profiling of diesel and gasoline cars at a city traffic junction. Particuology 18, 186–193.

Alas, H.D., Müller, T., Birmili, W., Kecorius, S., Cambaliza, M.O., Simpas, J.B.B., Cayetano, M., Weinhold, K., Vallar, E., Galvez, M.C., Wiedensohler, A., 2018. Spatial characterization of black carbon mass concentration in the atmosphere of a Southeast Asian megacity:

an air quality case study for Metro Manila, Philippines. Aerosol Air Qual. Res. 18, 2301–2317.

Aleksandropoulou, V., Lazaridis, M., 2013. Development and application of a model (ExDoM) for calculating the respiratory tract dose and retention of particles under variable exposure conditions. Air Qual. Atmos. Health 6 (1), 13–26.

Anenberg, S., Balakrishnan, K., Jetter, J., Masera, O., Mehta, S., Moss, J., Ramanathan, V., 2013. Cleaner cooking solutions to achieve health, climate, and economic cobenefits. Environ. Sci. Technol. 47, 3944–3952.

Apte, J.S., Kirchstetter, T.W., Reich, A.H., Deshpande, S.J., Kaushik, G., Chel, A., Marshall, J.D., Nazaroff, W.W., 2011. Concentrations of fine, ultrafine, and black carbon particles in auto-rickshaws in New Delhi, India. Atmos. Environ. 45, 4470–4480.

Bacero, R., Vergel, K., 2009. Assessment of Jeepney's components, systems and separate technical units for the development of standards. The Proceedings of the 17th Annual Conference of the Transportation Science Society of the Philippines.

Baker, J., Santiago, R., Villareal, T., Walsh, M., 1993. Vehicular Emissions Control in Metro Manila. Asian Development Bank. PPTA. 1723.

Baklanov, A., Molina, L.T., Gauss, M., 2016. Megacities, air quality and climate. Atmos. Environ. 126, 235–249.

Betancourt, R.M., Galvis, B., Balachandran, S., Ramos-Bonilla, J.P., Sarmiento, O.L., Gallo-Murcia, S.M., Contreras, Y., 2017. Exposure to fine particulate, black carbon, and particle number concentration in transportation microenvironments. Atmos. Environ. 157, 135–145.

Boquet, Y., 2017. The Philippine Archipelago. Springer, pp. 496–510.

Buonanno, G., Marini, S., Morawska, L., Fuoco, F.C., 2012. Individual dose and exposure of Italian children to ultrafine particles. Sci. Total Environ. 438, 271–277.

Buonanno, G., Stabile, L., Morawska, L., Russi, A., 2013. Children exposure assessment to ultrafine particles and black carbon: the role of transport and cooking activities. Atmos. Environ. 79, 53–58.

Cames, M., Helmers, E., 2013. Critical evaluation of the European diesel car boom-global comparison, environmental effects and various national strategies. Environ. Sci. Eur. 25 (1), 15.

Chalupa, D.C., Morrow, P.E., Oberdörster, G., Utell, M.J., Frampton, M.W., 2004. Ultrafine particle deposition in subjects with asthma. Environ. Health Perspect. 112 (8), 879.

Chin, H., 2013. Sustainable Urban Mobility in South-eastern Asia and the Pacific. Regional Study prepared for Global Report on Human Settlements. p. 73.

Cohen, A.J., Brauer, M., Burnett, R., Anderson, H.R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R., Feigin, V., 2017. Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the global burden of diseases study 2015. Lancet 389 (10082), 1907–1918.

Daigle, C.C., Chalupa, D.C., Gibb, F.R., Morrow, P.E., Oberdörster, G., Utell, M.J., Frampton, M.W., 2003. Ultrafine particle deposition in humans during rest and exercise. Inhal. Toxicol. 15 (6), 539–552.

Donnelly, P.M., Yang, T.S., Peat, J.K., Woolcock, A.J., 1991. What factors explain racial differences in lung volumes? Eur. Respir. J. 4 (7), 829–838.

Dons, E., Panis, L.I., Van Poppel, M., Theunis, J., Willems, H., Torfs, R., Wets, G., 2011. Impact of time–activity patterns on personal exposure to black carbon. Atmos. Environ. 45 (21), 3594–3602.

Folberth, G.A., Butler, T.M., Collins, W.J., Rumbold, S.T., 2015. Megacities and climate change–a brief overview. Environ. Pollut. 203, 235–242.

Gurjar, B.R., Ravindra, K., Nagpure, A.S., 2016. Air pollution trends over Indian megacities and their local-to-global implications. Atmos. Environ. 142, 475–495.

Gysel, M., McFiggans, G.B., Coe, H., 2009. Inversion of tandem differential mobility analyser (TDMA) measurements. J. Aerosol Sci. 40 (2), 134–151.

Hama, S.M.L., Ma, N., Cordell, R.L., Kos, G.P.A., Wiedensohler, A., Monks, P.S., 2017. Lung deposited surface area in Leicester urban background site/UK: sources and contribution of new particle formation. Atmos. Environ. 151, 94–107.

Hugo, G., Bardsley, D., Sharma, V., Tan, Y., Williams, M., Bedford, R., 2012. Addressing Climate Change and Migration in Asia and the Pacific. Asian Development Bank.

Hussein, T., Kulmala, M., 2008. Indoor aerosol modeling: basic principles and practical applications. Water Air Soil Pollut. Focus 8 (1), 23–34.

Hussein, T., Löndahl, J., Paasonen, P., Koivisto, A.J., Petäjä, T., Hämeri, K., Kulmala, M., 2013. Modeling regional deposited dose of submicron aerosol particles. Sci. Total Environ. 458, 140–149.

Hussein, T., Wierzbicka, A., Löndahl, J., Lazaridis, M., Hänninen, O., 2015. Indoor aerosol modeling for assessment of exposure and respiratory tract deposited dose. Atmos. Environ. 106, 402–411.

Isaxon, C., Gudmundsson, A., Nordin, E.Z., Lönnblad, L., Dahl, A., Wieslander, G., Bohgard, M., Wierzbicka, A., 2015. Contribution of indoor-generated particles to residential exposure. Atmos. Environ. 106, 458–466.

Janssen, N.A.H., Hoek, G., Simic-Lawson, M., Fischer, P., van Bree, L., ten Brink, H., Keuken, M., Atkinson, R.W., Anderson, H.R., Brunekreef, B., Cassee, F.R., 2011. Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM10 and PM2.5. Environ. Health Perspect. 119, 1691–1699.

Jung, J.C., 2017. Case study: Volkswagen's diesel emissions scandal. Thunderbird Int. Bus. Rev. 59 (1), 127–137.

Kecorius, S., Madueño, L., Vallar, E., Alas, H., Betito, G., Birmili, W., Cambaliza, M.O., Catipay, G., Gonzaga-Cayetano, M., Galvez, M.C., Lorenzo, G., 2017. Aerosol particle mixing state, refractory particle number size distributions and emission factors in a polluted urban environment: case study of Metro Manila, Philippines. Atmos. Environ. 170, 169–183.

Kecorius, S., Tamayo, E.G., Galvez, M.C., Madueño, L., Betito, G., Gonzaga-Cayetano, M., Vallar, E., Wiedensohler, A., 2018. Activity pattern of school/university tenants and their family members in Metro Manila–Philippines. Aerosol Air Qual. Res. 18, 2412–2419.

Kristensson, A., Rissler, J., Löndahl, J., Johansson, C., Swietlicki, E., 2013. Size-resolved respiratory tract deposition of sub-micrometer aerosol particles in a residential area with wintertime wood combustion. Aerosol Air Qual. Res. 13, 24–35.

Kumar, P., Morawska, L., Harrison, R.M., 2012. Nanoparticles in European cities and associated health impacts. Urban Air Quality in Europe. Springer Berlin Heidelberg, pp. 339–365.

Künzi, L., Mertes, P., Schneider, S., Jeannet, N., Menzi, C., Dommen, J., Baltensperger, U., Prévôt, A.S., Salathe, M., Kalberer, M., Geiser, M., 2013. Responses of lung cells to realistic exposure of primary and aged carbonaceous aerosols. Atmos. Environ. 68, 143–150.

Kuuluvainen, H., Rönkkö, T., Järvinen, A., Saari, S., Karjalainen, P., Lähde, T., Pirjola, L., Niemi, J.V., Hillamo, R., Keskinen, J., 2016. Lung deposited surface area size distributions of particulate matter in different urban areas. Atmos. Environ. 136, 105–113.

Lelieveld, J., Evans, J.S., Fnais, M., Giannadaki, D., Pozzer, A., 2015. The contribution of outdoor air pollution sources to premature mortality on a global scale. Nature 525 (7569), 367–371.

Li, Y., Henze, D.K., Jack, D., Henderson, B.H., Kinney, P.L., 2016. Assessing public health burden associated with exposure to ambient black carbon in the United States. Sci. Total Environ. 539, 515–525.

Lim, S., Vos, T., Flaxman, A., Danaei, G., Shibuya, K., Adair-Rohani, H., ... Ezzati, M., 2012. A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: a systematic analysis for the global burden of disease study 2010. Lancet 380 (9859), 2224–2260.

Lin, F.L., Kelso, J.M., 1999. Pulmonary function studies in healthy Filipino adults residing in the United States. J. Allergy Clin. Immunol. 104 (2), 338–340.

Löndahl, J., Massling, A., Pagels, J., Swietlicki, E., Vaclavik, E., Loft, S., 2007. Size-resolved respiratory-tract deposition of fine and ultrafine hydrophobic and hygroscopic aerosol particles during rest and exercise. Inhal. Toxicol. 19, 109–116.

Löndahl, J., Massling, A., Swietlicki, E., Bräuner, E.V., Ketzel, M., Pagels, J., Loft, S., 2009. Experimentally determined human respiratory tract deposition of airborne particles at a busy street. Environ. Sci. Technol. 43 (13), 4659–4664.

Löndahl, J., Swietlicki, E., Rissler, J., Bengtsson, A., Boman, C., Blomberg, A., Sandström, T., 2012. Experimental determination of the respiratory tract deposition of diesel combustion particles in patients with chronic obstructive pulmonary disease. Part. Fibre Toxicol. 9 (1), 30.

Louwies, T., Nawrot, T., Cox, B., Dons, E., Penders, J., Provost, E., Panis, L.I., De Boever, P., 2015. Blood pressure changes in association with black carbon exposure in a panel of healthy adults are independent of retinal microcirculation. Environ. Int. 75, 81–86.

Manigrasso, M., Vernale, C., Avino, P., 2015. Traffic aerosol lobar doses deposited in the human respiratory system. Environ. Sci. Pollut. Res. 24 (16), 13866–13873.

Manigrasso, M., Natale, C., Vitali, M., Protano, C., Avino, P., 2017. Pedestrians in traffic environments: ultrafine particle respiratory doses. Int. J. Environ. Res. Public Health 14 (3), 288.

Martins, V., Minguillón, M.C., Moreno, T., Querol, X., de Miguel, E., Capdevila, M., Centelles, S., Lazaridis, M., 2015a. Deposition of aerosol particles from a subway microenvironment in the human respiratory tract. J. Aerosol Sci. 90, 103–113.

Martins, V., Moreno, T., Minguillón, M.C., Amato, F., de Miguel, E., Capdevila, M., Querol, X., 2015b. Exposure to airborne particulate matter in the subway system. Sci. Total Environ. 511, 711–722.

Morawska, L., Hofmann, W., Hitchins-Loveday, J., Swanson, C., Mengersen, K., 2005. Experimental study of the deposition of combustion aerosols in the human respiratory tract. J. Aerosol Sci. 36 (8), 939–957.

Natera, E.S., Cuevas, C.D., Azanon, E.M., Palattao, M.B., Espiritu, R.T., Cobar, M.C., Palad, L.H., Torres, B., Shiraishi, K., 1998. Compilation of Anatomical, Physiological and Dietary Characteristics for a Filipino Reference Man (No. IAEA-TECDOC–1005).

Oberdörster, G., 2000. Toxicology of ultrafine particles: in vivo studies. Philos. Trans. R. Soc. Lond. A 358, 2719–2740.

Ostro, B., Tobias, A., Karanasiou, A., Samoli, E., Querol, X., Rodopoulou, S., Basagaña, X., Eleftheriadis, K., Diapouli, E., Vratolis, S., Jacquemin, B., 2014. The risks of acute exposure to black carbon in Southern Europe: results from the MED-PARTICLES project. Occup. Environ. Med. 72 (2), 123–129.

Ozdemir, H., Pozzoli, L., Kindap, T., Demir, G., Mertoglu, B., Mihalopoulos, N., Theodosi, C., Kanakidou, M., Im, U., Unal, A., 2014. Spatial and temporal analysis of black carbon aerosols in Istanbul megacity. Sci. Total Environ. 473, 451–458.

Park, K., Kittelson, D.B., McMurry, P.H., 2004. Structural properties of diesel exhaust particles measured by transmission electron microscopy (TEM): relationships to particle mass and mobility. Aerosol Sci. Technol. 38 (9), 881–889.

Philippin, S., Wiedensohler, A., Stratmann, F., 2004. Measurements of non-volatile fractions of pollution aerosols with an eight-tube volatility tandem differential mobility analyzer (VTDMA-8). J. Aerosol Sci. 35, 185–203.

Rissler, J., Swietlicki, E., Bengtsson, A., Boman, C., Pagels, J., Sandström, T., Blomberg, A., Löndahl, J., 2012. Experimental determination of deposition of diesel exhaust particles in the human respiratory tract. J. Aerosol Sci. 48, 18–33.

Rissler, J., Gudmundsson, A., Nicklasson, H., Swietlicki, E., Wollmer, P., Löndahl, J., 2017. Deposition efficiency of inhaled particles (15-5000 nm) related to breathing pattern and lung function: an experimental study in healthy children and adults. Part. Fibre Toxicol. 14 (1), 10.

Roa, C.C., Zaldivar, C.A., Salonga, R.C., Bobadilla, J., Lansang, M.A., Reodica, R., Balgos, A., Blanco, J., Tanchuco, J.Q., 2013. Normal standards for ventilatory function in adult Filipinos. Philipp. J. Intern. Med. 51 (1), 1–6.

Rose, D., Wehner, B., Ketzel, M., Engler, C., Voigtländer, J., Tuch, T., Wiedensohler, A., 2006. Atmospheric number size distributions of soot particles and estimation of emission factors. Atmos. Chem. Phys. 6 (4), 1021–1031.

Smith, H., 1994. Human Respiratory Tract Model for Radiological Protection. 66. ICRP Publication.

Stephen, S., Vos, T., Flaxman, A.D., Danaei, G., Shibuya, K., Adair-Rohani, H., AlMazroa, M.A., Amann, M., Anderson, H.R., Andrews, K.G., Aryee, M., 2017. A Comparative

Risk Assessment of Burden of Disease and Injury Attributable to 67 Risk Factors and Risk Factor Clusters in 21 Regions, 1990–2010: A Systematic Analysis for the Global Burden of Disease Study 2010.
Tesner, P.A., 1973. Formation of soot particles. Faraday Symposia of the Chemical Society, Royal Society of Chemistry. Vol. 7, pp. 104–108.
Weingartner, E., Burtscher, H., Baltensperger, U., 1997. Hygroscopic properties of carbon and diesel soot particles. Atmos. Environ. 31 (15), 2311–2327.
Williams, R.D., Knibbs, L.D., 2016. Daily personal exposure to black carbon: a pilot study. Atmos. Environ. 132, 296–299.