RESEARCH ARTICLE



Individual exposure of graduate students to PM_{2.5} and black carbon in Shanghai, China

Xiaoning Lei^{1,2} · Guangli Xiu^{1,2} · Bo Li^{1,2} · Kun Zhang^{1,2} · Mengfei Zhao^{1,2}

Received: 13 October 2015 / Accepted: 3 March 2016 / Published online: 11 March 2016 © Springer-Verlag Berlin Heidelberg 2016

Abstract The personal exposure of 51 graduate students to PM_{2.5} and BC in Shanghai was evaluated using portable PM_{2.5} and BC samplers. Global positioning systems and activity diaries were used to evaluate the microenvironments/ activities that contributed to the total daily exposure of the participants, as well as to accurately estimate the PM2.5 and BC doses received by each participant. The mean PM2.5 and BC exposure concentrations were 110.0 and 5.3 μ g m⁻³, respectively. These exposure levels were considered extremely high and comparable to those measured in a busy urban street canyon for 24 h. High daily inhaled doses in terms of both PM_{2.5} and BC concentrations were measured, obtaining 1062.2 and 45.9 µg day⁻¹, respectively. Indoor activities significantly contributed to the students' daily dose. Eating contributed the least to PM2.5 and BC exposure, whereas outdoor activities contributed the most. PM2.5 dose intensity from indoor eating slightly exceeded BC dose intensity; conversely, transportation showed higher exposure for BC than PM_{2.5}.

Keywords $PM_{2.5} \cdot Black carbon \cdot Personal monitoring \cdot Individual exposure \cdot Daily dose \cdot Dose intensity$

Responsible editor: Philippe Garrigues

Guangli Xiu xiugl@ecust.edu.cn

² Australia-China Centre for Air Quality Science and Management, Shanghai 200237, China

Introduction

Air pollution in China and other parts of Asia poses significant health risks and substantially contributes to global climate change (Baumgartner et al. 2014). Short-term exposure to high concentrations of particulate matter (PM) may exacerbate bronchitis and asthma, as well as changes in heart rate variability (Sørensen et al. 2003). $PM_{2.5}$ (particulate matter less than 2.5 µm in aerodynamic diameter), known as fine particle, has been used as a PM measurement in most studies to date (Pope and Dockery 2006).

Black carbon (BC), a short-lived constituent of fine particles (PM_{2.5}), is defined as the fraction of carbonaceous aerosol absorbing light. BC is emitted from combustion processes, and its primary sources include combustion engines (especially diesel), household use of biomass and coal fuels, and heavy oil- and coal-fired power stations (Baumgartner et al. 2014). BC affects the regional and global climates by absorbing solar radiation and heating the atmosphere. This fine particle ranks second to carbon dioxide as the most important climateforcing human emission (Bond et al. 2013). In addition to climate warming, other regional climate impacts of BC may include increased glacial retreat and changes in precipitation patterns in Asia (Menon et al. 2002; Qiu and Yang 2000). BC typically comprises only a fraction of PM_{2.5}; however, recent studies have suggested that certain adverse health effects can be more strongly associated with BC than PM_{2.5} (Janssen et al. 2011a, b; Bell et al. 2009; Patel et al. 2009; Spira-Cohen et al. 2011) because of its large surface carrying potentially toxic compounds (e.g., heavy metals, persistent organic pollutants, etc.), as well as ability to induce inflammation, penetrate into the deepest regions of the lung (Braniš et al. 2010; Weichenthal 2012), and deposit in secondary organs (Semmler et al. 2004). A study in China examined the blood pressure levels of 260 women living in a rural area in

¹ State Environmental Protection Key Laboratory of Risk Assessment and Control on Chemical processes, East China University of Science and Technology (ECUST), Shanghai 200237, China

northwestern Yunnan where biomass fuels are commonly used. BC was significantly associated with changes in systolic blood pressure (SBP) (4.3 mmHg; P < 0.001) (Baumgartner et al. 2014).

To date, both indoor (Stabile et al. 2012) and outdoor measurements of BC concentration have been performed (Janssen et al. 2011a, b; Vanderstraeten et al. 2011). Personal monitoring is widely viewed as the gold standard for exposure assessment of air pollutants (Cai et al. 2013); however, most exposure assessment studies have used data based on an outdoor fixed sampling point (FSP) measurements from a few air quality monitors and reflects the mean concentrations in the entire urban area or community (Buonanno et al. 2013a). This approach may result in unreliable estimates of individual exposure to BC (Manigrasso et al. 2013), because BC has a smaller spatial and temporal variation in a given location, which rapidly decrease with increasing distance to the source (Buonanno et al. 2011a).

Toxicological and epidemiological studies have focused on the dose-response relationship (Sayes et al. 2007); however, previous studies focused on the exposure-response relationship or the absolute dose values, which can be misleading in the evaluation of a particle dose-response relationship (Buonanno et al. 2012b). In particular, the exposure of each person to pollutant concentrations varies, which is strongly related to the daily activity patterns, lifestyle, and the different microenvironments in which they frequently occur (Buonanno et al. 2011b, 2012a, b). Personal sampling can measure particle concentrations received by people in every microenvironment they visit during a typical day and by estimating the corresponding doses (Buonanno et al. 2013b). The majority of studies on individual exposure to PM_{2.5} and BC were conducted in North American and Europe (Kingham et al. 2013). However, these population activity patterns in China completely differ from those in Western countries (Yang et al. 2011; Jim and Chen 2009).

Few studies have been conducted on personal exposure to $PM_{2.5}$, BC, and individual dose–response relationship in China. Thus, we examined the daily activity patterns of 51 graduate students on the basis of the Global Positioning Systems (GPS) and diaries carried by each participant in Shanghai, China. Meanwhile, we used the daily $PM_{2.5}$ and BC exposure of graduate students to identify their contribution of microenvironments/activities to the average daily dose of the students.

Materials and methods

Study design, sampling sites, and population

We recruited 51 graduate students aged 22 to 28 years at a university in Xuhui District in Shanghai (31° 14' N, 121° 29'

E), a typical busy city (surface area 6340.5 km²) in China, from December 2014 to March 2015. None previously or currently smoked tobacco. The experimental campaigns were conducted in Xuhui District located southwest of the downtown area of Shanghai. Channels, highways, and railways make Xuhui District an important transport corridor connecting the downtown area of Shanghai with other districts and provinces such as Zhejiang and Jiangsu Provinces (Li et al. 2015). The university where the participants stayed is situated close to Shanghai South Railway Station, Shanghai Nanzhan Long-distance Passenger Transportation Station, and Shanghai Nanzhan Bus Station. In addition, the university is located on an urban street where traffic is mostly dominated by light vehicles (e.g., taxis, buses, and cars).

All participants were individually instructed on the aim of the study and how to use the devices. Written informed consent was obtained from every graduate student prior to the study. A questionnaire was developed to evaluate the household demographics, socioeconomic status, secondhandsmoking status, and medical history of the participants. Nork was installed on a small handheld computer, to record the activities (such as studying, eating, transportation, and sleeping) of the participants, including the amount of time spent performing each activity, without any significant data loss.

Instrumentation and quality assurance

We measure the PM_{2.5} concentrations with personal PM_{2.5} samplers (v3.2 MicroPEM, RTI, USA), which provides exposure data at the personal level and can be worn by individuals to significantly enhance studies in public and occupational health. The MicroPEM provides fully representative personal exposure characterization, simultaneously defining both the integrated exposure (filter-based) and the patterns of exposure real-time in a wearable low-burden package weighing less than 240 g. Data collected at the personal level will represent the breathing zone exposure of a person if the unit is worn nominally within 30 cm of the oral/nasal cavity. The MicroPEM has an extremely low noise level due in part to the low flow rate and to an innovative noise damping system. Real-time concentration data collected must represent the likely exposures of a person in typical residential settings but must allow for occasional strong source excursions setting a nominal response range for real-time sensing from nominally 5 μ g m⁻³ to 10,000 μ g m⁻³. The very low face velocity at the nominal 0.5 L min⁻¹ flow rate through the 25 mm outside diameter of the Teflon filter allows substantial loadings (up to $200 \mu g$) before filter blinding is observed.

Light and compact, the MicroAeth BC Monitor (model AE-51, Magee Scientific, US) can be used as a personal air monitor in epidemiological studies. As a real-time personal monitor, MicroAeth provides additional information on the

patterns and peak levels of exposure (Cai et al. 2013). A diffusion dryer was set in line with the inlet tubing at the breathing zone to address false positive and negative peaks in realtime BC concentrations resulting from changes in temperature and humidity. The BC data sets obtained using the Aethalometer instruments also include negative values when sampling is performed at low concentrations or at a high time resolution (short sampling/logging time interval) (Cheng and Lin 2013). In the present study, optical noise reduction averaging, a post-processing method introduced by Hagler et al. (2011), was adopted to reduce noise while preserving high time resolution in Aethalometer real-time BC data.

BC concentrations were measured by gauging the 880-nm wavelength light absorbed (attenuation) by optically absorbing particles (Buonanno et al. 2013a, b). This filter-based light absorption method for measuring BC exhibits a "loading effect," in which the instrument proportionally underestimates BC levels, with increasing aerosol loading on the filter (Kirchstetter and Novakov 2007; Weingartner et al. 2003). Thus, a new Teflon-coated borosilicate glass fiber filter was replaced before each sampling event. The pump speed was set at a 100 mL min⁻¹.

Methodology description

Both $PM_{2.5}$ and BC measures were conducted sequentially for a 3-day period with an interval 1-min for every individual. A time–activity electronic diary recording the main activities of the participants was used to identify their most timeconsuming activities. The accuracy of the diary was also checked against the Global Positioning Systems (GPS) logs. All mobile experimental apparatus, including PM_{2.5} samplers, BC monitor, and GPS tracking device, were synchronized at the start of each sampling period.

In addition to personal exposure concentrations, the amount of pollutants inhaled into the body significantly varies because of variable inhalation rates, duration in a specific microenvironment, and other biological factors such as age and body mass (Rodes et al. 2012). The inhaled dose received by volunteers was also determined for each microenvironment or activity in the current study. We considered 100 % efficiency of aerosol aspiration during inhalation. We added the partial doses measured for each microenvironment/activity to estimate the daily total inhaled dose, as reported in Eq. (1).

$$D = \sum_{i=1}^{n} \{ Et \cdot IR \ activity \cdot T \} i$$
(1)

where *D* represents the average inhalation dose (μ g), *Et* is the average PM_{2.5} or BC concentration for the time spent (*T*) in the *i*th microenvironment (μ g m⁻³), and *T* is the duration of exposure to the *i*th microenvironment (h). *IR* is the inhalation rate (m³ h⁻¹); inhalation rates for the different activities were adopted on the basis of the Wang approach (Wang et al. 2009), ranging from 0.35 (0.48) during sleep to 1.42 m³ h⁻¹ (1.90) during physical movement for female (male). These quantities represented the mean values of the respiratory rates for the age group between 18 and 60 years old. Meanwhile, the exposure–dose intensity described in Eq. (2) (Wang et al. 2011) was calculated for each activity to further analyze the contributions of different microenvironments/activities to the total daily dose.

Exposure dose intensity = Daily exposure dose fraction (%) / Daily time fraction (%) (2)

Study characteristics and daily time-activity patterns

The main characteristics based on the questionnaires are shown in Table 1.

Approximately 35 % went to school by taxi, 24 % by bus, 23 % by subway, and 18 % by means requiring transfer among these transport vehicles. All graduate students were not required to cook, and cooking by gas was the only method used in the restaurant where they go to eat. Natural ventilation was more commonly used than air-conditioning. About 24 % of the participants reported allergic rhinitis, and none of them previously or currently smoked tobacco.

A typical daily pattern in this study is shown in Fig. 1. A first peak (0700–0900 hours) for the graduate student was while having his breakfast in the restaurant near home and commuting by public transportation (subway, bus and taxi),

the second peak (1130–1230 hours) was observed during lunch in the school cafeteria, and in the evening, the last peak (1700–1900 hours) was while having his dinner at school and returning home. After returning home, he did some usual activities (washing clothes, watching TV, and playing games). From 2330 hours onwards, he was sleeping at home.

Daily time–activity patterns based on the time–activity diary and GPS log of each student showed that they spent most of the day in general indoor activities (49 %) and sleeping (32 %). The amount of time spent daily for both transportation and outdoor activities was 7 %, followed by indoor eating, with 5 %. The time spent indoors (including eating, sleeping, and general activities) was 86 % in this study (Table 2), which was in agreement with the result of an analysis of how time is spent in Hong Kong (Chau et al. 2002), which showed an average time equal to 86 % spent in indoor activities, 3 to

Table 1 Demographic and health behaviors of study participants $(n=51, \text{ age}=25\pm3 \text{ years})$

Characteristic	Percent
Boys/girls	59:41
Living in urban area	100
Exposure to gas cooking	100
Go to school by taxi/subway/bus/mix	35:23:24:18
Heavy traffic during transport	21
Exposure to second-hand tobacco smoke	0
Ventilate: air-conditioning/natural ventilation	35:65
Air purifiers	24
Allergic rhinitis	24
Asthma	0

7 % in enclosed transit, and 3 to 7 % in outdoor activities. Indeed, the daily activity patterns in Korea and China are substantially different from those in Western countries (Yang et al. 2011; Jim and Chen 2009). The values obtained in the current study were higher than the values obtained for the time spent indoors in Germany (65 %), the USA (65 %), and Canada (66 %) (Brasche and Bischof 2005; Leech et al. 2002), but similar with Belgium (89 %) (Dons et al. 2011).

Individual daily exposure to PM_{2.5} and BC

The personal concentrations of PM_{2.5} and BC during the overall experimental campaign are shown in Table 3. Fine particle concentrations measured on a personal scale of five microenvironments/activities indicated a significant difference, with an average of 110.0 μ g m⁻³, ranging from 3.0 μ g m⁻³ for indoor general activities to 574.0 μ g m⁻³ for transportation as a direct effect of primary exhaust emissions. These levels were higher than the ambient PM_{2.5} concentrations (median 94.5 μ g m⁻³) in Shanghai over the same time



Fig. 1 The personal concentrations of $PM_{2.5}$ (solid gray) and BC (dashed black) on December 29, 2014

and greatly exceeded the daily 24-h $PM_{2.5}$ Ambient Air Quality Standards both in China (75.0 µg m⁻³) (GB 3095-2012) and in America (35.0 µg m⁻³) (US EPA 2006).

The indoor microenvironments sampled in this study are located in city districts close to the station, near the overhead road, and have no industry. Specifically, the teaching buildings, office buildings, and homes nearby public transportation station and restaurants, entertainment venues, and restaurants are often sited close to the roads, and around almost every shopping mall are subway and bus stations. Thereby, it cannot be ignored that ambient pollutants made certain contribution to indoor air quality by ventilation, though both PM_{2.5} and BC concentrations in indoor microenvironment were lower than outdoor in this study. While outdoor microenvironments and indoor eating activities were equally responsible for 27 % of total exposure to PM_{2.5} according to the median values, other statistics (including 75th quartile, 25th quartile, minimum, and mean) for PM_{2.5} concentrations were higher in the case of indoor eating in comparison with all other microenvironments. The results showed that the effect of indoor eating exposure on air quality (PM_{25}) is greater than that of other microenvironments. More broadly, our findings might be useful in drafting policies aiming at reducing health impact of air pollution in China. The strategies and mitigation efforts that focus on control of indoor cooking emissions might have the larger benefits for human health. Studies on outdoor microenvironments were reported in the literature. Geng et al. (2013) conducted experimental campaigns that evaluated the association between mortality outcomes and exposure to BC and PM_{2.5} in nine selected urban air quality monitoring sites in Shanghai; the mean daily concentration of PM2.5 was 53.9 μ g m⁻³ (standard deviation 31.4 μ g m⁻³, inter-quartile range 41.8 μ g m⁻³). Nearly half of the Chinese population still cook and heat their homes with highly polluting biomass and coal fuels (Bonjour et al. 2013). Baumgartner et al. (2011) reported that the average 24-h PM2.5 exposure of 280 women living in a rural area of northwestern Yunnan, where biomass fuels are commonly used, ranged from 9.0 to 634.0 μ g m⁻³. This value slightly exceeds that of our current study. This difference may be attributed to the dining place where graduate students normally go where gas, rather than coal or biomass, is mostly used for cooking.

Pollutant concentrations are often extremely high in the traffic microenvironment. Individuals may receive a significant contribution to their daily exposure during a commute even though such individuals usually travel for no more than 6 to 8 % of their time daily (Kaur et al. 2007). In the current study, time spent in transportation microenvironments (21 %) also contributed to personal exposure. The PM_{2.5} level in transportation microenvironments ranged from 25.0 to 574.0 μ g m⁻³, with a median of 109.0 μ g m⁻³. Among the daily activities, transportation is considered one of the high-exposure microenvironments, especially in high vehicle-

Table 2	The percentage of time
spent on	different
microenv	vironments in the
exposure	measurement campaign

Indooring (eating, sleeping, and general activity)	Outdoor	Transportation	Region
86 %	7 %	7 %	Present study
86 %	3-7 %	3-7 %	Hong Kong
65 %	_	_	Germany
65 %	_	_	USA
66 %	_	_	Canada
89 %	5 %	6 %	Belgium

density metropolitan areas (Dons et al. 2011; Panis et al. 2010). The growth of motorized transport growth in China is the fastest worldwide and is predicted to exceed 380 million by 2030 (Huo and Wang 2012). Shanghai is a crowded city with a population of 24.2 million and an annual traffic volume of over 6.3 billion passengers within the Shanghai public transport system in 2013 (SMBS 2014). Exposure to PM2.5 and BC among commutes using different transport modes (taxi, bus, subway, cycling, and walking) in Xuhui District was included in our previous study (Li et al. 2015). The daily average personal exposure to PM_{2.5}, measured with a personal PM_{2.5} sampler, was $240.0 \pm 30.0 \ \mu g \ m^{-3}$. Compared with other microenvironments/activities (outdoor, transportation, and indoor eating), indoor general activity (16 %) and indoor sleeping (9 %) contributed less to daily personal exposure to $PM_{2.5}$.

With regard to BC exposure, the daily median concentration in various microenvironments/activities for volunteers ranged from 2.2 μ g m⁻³ for sleeping to 6.6 μ g m⁻³ for transportation. Levels of BC were highest during transportation, with a maximum of 21.8 μ g m⁻³. This finding indicates that BC can more consistently trace the effect of road traffic on exposure. The daily average personal exposure to BC reached 5.3 μ g m⁻³ (other statistics for BC concentration are reported in Table 3). The BC concentrations were similar to those reported in Cassino, Italy (daily average 5.1 μ g m⁻³) (Buonanno et al. 2013a, b) and in Yunnan (Baumgartner et al. 2014), with a geometric mean BC exposure of 5.2 μ g m⁻³ (4.0 μ g m⁻³ in summer and 6.0 μ g m⁻³ in winter; range 2.0 to 44.0 μ g m⁻³). However, the values surpass the daytime ambient BC in Beijing, Mexico City, and in several cities in Brazil (range 1.9 to 4.8 μ g m⁻³) (Salcedo et al. 2006; Westerdahl et al. 2009; de Miranda et al. 2012). The annual average BC concentration ranged from 3.5 to 7.8 μ g m⁻³ near busy roads across different European cities (Reche et al. 2011). Li et al. (2015) reported that the average exposure levels to BC when walking on three busy roads in Shanghai were 5.8, 5.5, and 5.5 μ g m⁻³. The findings confirmed that the daily exposure of monitored graduate students typically represents the exposure they would receive in a busy urban street canyon for 24 h.

PM_{2.5} and BC inhaled doses and exposure-dose intensities of graduate students

Inhaled doses were evaluated based on the daily activity pattern reported in the time–activity diaries and exposure concentration data, following the methodology in "Methodology description" section. Table 4 presents the dose and dose intensity of the different microenvironments/activities for PM_{2.5} and BC among graduate students. Overall mean daily inhaled dose in PM_{2.5} and BC concentrations was equal to 1062.2 and 45.9 μ g day⁻¹, respectively. These inhaled doses were similar to those of children aged 8 to 11 years in Cassino, Italy (39.2 μ g day⁻¹ for BC) (Buonanno et al. 2013a). The inhaled dose of BC in the present study was comparable to the exposure of the participants standing in transportation microenvironments (ranging from 21.2 μ g day⁻¹ for bus to 142.6 μ g day⁻¹ for taxi) or walking (ranging from 15.5 to

Table 3 Statistics (maximum, 75th quartile, median, 25th quartile, minimum, and mean \pm standard deviations) of PM_{2.5} and BC concentrations towhich population studies were exposed during the overall experimental campaign

Microenvironment or activity	PM _{2.5} (μg m ⁻³)					BC ($\mu g m^{-3}$)						
	Max	75th	Med	25th	Min	Mean ± SD	Max	75th	Med	25th	Min	Mean±SD
Indoor (eating)	256.0	187.0	145.5	115.0	64.0	149.0 ± 43.8	11.0	6.7	5.0	2.9	1.4	7.4±1.9
Indoor (sleeping)	136.0	55.0	48.0	41.0	10.0	48.0 ± 13.0	6.5	4.3	2.2	0.7	0.0	2.3 ± 1.7
Indoor (general activity)	192.0	99.0	84.0	71.0	3.0	84.0 ± 25.0	7.4	4.4	3.1	2.2	0.1	3.3 ± 1.4
Outdoor	329.0	177.8	146.0	108.0	14.0	144.0 ± 53.2	16.2	7.8	4.9	3.4	0.9	5.7 ± 3.4
Transportation	574.0	156.0	109.0	77.0	25.0	124.0 ± 68.4	21.8	9.3	6.6	5.0	1.1	7.6 ± 3.0
Ambient environment	249.0	126.0	94.5	55.5	18.0	96.6 ± 52.8	-	-	-	-	-	-

 Table 4
 Dose and dose intensity

 of PM_{2.5} and BC in different

 microenvironments/activities

	Microenvironment or activity	Dose (µg day ⁻¹)						
		Max	75th	Med	25th	Min	Mean \pm SD	intensity
PM _{2.5}	Indoor (eating)	143.2	104.6	81.4	64.3	35.8	83.5 ± 24.5	1.7
	Indoor (sleeping)	420.4	170.0	148.4	126.7	30.9	148.4 ± 40.1	0.5
	Indoor (general activity)	1179.1	608.0	515.9	436.0	18.4	516.0 ± 153.5	1.0
	Outdoor	516.0	278.8	229.0	169.4	22.0	225.4 ± 84.4	2.8
	Transportation	410.3	111.5	77.9	55.0	17.9	88.6 ± 48.8	1.3
BC	Indoor (eating)	6.2	3.8	2.8	1.6	0.8	4.2 ± 1.1	1.5
	Indoor (sleeping)	15.5	15.3	6.7	2.2	0.1	7.1 ± 5.2	0.5
	Indoor (general activity)	45.2	26.8	18.8	13.4	0.7	20.2 ± 8.9	0.9
	Outdoor	25.4	12.2	7.6	5.3	1.5	8.9 ± 5.4	2.5
	Transportation	15.6	6.7	4.7	3.6	0.8	5.2 ± 2.2	2.0

12125

99.7 μ g day⁻¹) along busy urban street canyons for 24 h. These findings are in accordance with the inhalation doses and exposure time reported in our previous study (Li et al. 2015) and those evaluated by Dons et al. (2012) in transport microenvironments (ranging from 14.1 to 77.7 μ g day⁻¹).

Figure 2 presents the relative median contributions of each microenvironment/activity to daily time-activity patterns, BC exposure, and PM_{2.5} exposure. The time-activity pattern data are discussed in "Study characteristics and daily time-activity patterns" section. With regard to PM2.5 and BC exposure, a graduate student's indoor during general activity time contributed the most (49 and 46 % for PM2.5 and BC exposure, respectively), followed by outdoor time (22 and 19 % for PM_{2.5} and BC exposure, respectively). Despite lower concentrations and inhalation rates (0.35 and 0.48 for female and male, respectively), the microenvironment with the higher contribution (14 and 16 % for PM2.5 and BC exposure, respectively) was indoor during sleeping; however, this percentage was mainly attributed to the substantial amount of time (32 % of the whole day) graduate students spend in sleeping. In the current study, eating contributed the least to BC exposure (7 %) and $PM_{2.5}$ exposure (8 %), and this could perhaps be driven by the amount of time spent in indoor eating (5%)over a 24-h period. The contributions were lower than the relative contributions from cooking/eating to daily time-activity pattern (15 % to PM exposure and 8 % to BC exposure) in the study performed by Buonanno et al. (2013a).

These results may arise from the fact that all participants eat in restaurants outside of the school or in canteens on the campus daily, i.e., the participants are not required to cook at home, thereby avoiding the production of a mass of particulates during combustion and reducing the exposure time. Although people spent equal time between outdoor and transportation, even BC concentrations in transportation were higher than those during outdoor, transportation still made less contributions to exposure dose than outdoor, mainly caused by higher inhalation rate for outdoor (0.71 and 0.95 for female and male, respectively) than transportation (0.43 and 0.57 for female and male, respectively). Transportation microenvironments contributed 12 % to BC exposure and 7 % to PM_{2.5}, almost half of BC. These results are similar to those obtained in the study by Dons et al. (2011, 2012), thus indicating that BC can more consistently trace the effect of road traffic on exposure.

The exposure–dose intensities were evaluated, as defined in "Methodology description" section. Outdoor activities presented the highest dose intensity for $PM_{2.5}$ (2.8) and BC (2.5), highlighting the extremely high BC dose received outdoors because participants were exposed to multisource pollution environments. These environments included transportation on a busy road, cooking in sidewalk food stalls, coal-fired power plants, and crop straw burning, among others. Indoor sleeping presented the lowest dose intensity (0.5) for both $PM_{2.5}$ and BC. Transportation yielded a lower exposure



Fig. 2 Relative median contributions of each microenvironment/activity to daily time–activity patterns (*orange column plots*), BC exposure dose (*green column plots*), and PM_{2.5} exposure dose (*blue column plots*)

intensity for PM_{2.5} (1.3) than BC (2.0); the values are slightly lower than the values (1.5 for PM_{2.5} and 2.8 for BC) obtained in the study involving children in Cassino (Buonanno, et al. 2013a). These results confirm that BC is more highly correlated to traffic pollution than PM and is more suitable than PM in identifying the traffic pollution mentioned in a number of studies (Heal et al. 2012; Invernizzi et al. 2011; Janssen et al. 2012). By contrast, the BC dose intensity (1.5) is slightly below PM_{2.5} (1.7) during indoor eating. The indoor general activity presented nearly equal for PM_{2.5} (1) and BC (0.9), which may be attributed to the lack of special combustion source (e.g., cooking, incense, and smoking) in the room.

Conclusion

A population-based study was performed to identify the contributions of microenvironments/activities to overall daily $PM_{2.5}$ and BC exposures. The results show the importance of individual exposure measurements and particle dose–response relationship evaluation for assessing particulate exposure. Indoor activities significantly contributed to the students' daily dose because indoor time in the present study was substantially longer than that in many Western countries. Outdoor activities presented the highest dose intensity for $PM_{2.5}$ and BC. In particular, transportation presented a higher exposure– dose intensity for BC than $PM_{2.5}$, which confirmed that BC can more efficiently identify traffic pollution than $PM_{2.5}$. Outdoor $PM_{2.5}$ and BC pollution should be emphasized in future studies, and BC should be used as a pollution indicator to form new vehicle-emission standards in China.

Acknowledgments This work was jointly supported by Shanghai Pujiang Program (13PJD013), Ph.D. Programs Foundation of Ministry of Education of China (No. 20120074140001) and the Fundamental Research Funds for the Central Universities (WB1113005). The authors are grateful to Prof. Hai-dong Kan for providing us with MicroPEM $PM_{2.5}$ samplers. The authors would like to express their special gratitude to the participants, because without their help and assistance, this project could not have been conducted successfully.

References

- Baumgartner J, Schauer JJ, Ezzati M, Lu L, Cheng C, Patz JA, Bautista LE (2011) Indoor air pollution and blood pressure in adult women living in rural China. Environ Health Persp 119:1390
- Baumgartner J, Zhang Y, Schauer JJ, Huang W, Wang Y, Ezzati M (2014) Highway proximity and black carbon from cookstoves as a risk factor for higher blood pressure in rural China. Proc Natl Acad Sci U S A 111:13229–13234
- Bell ML, Ebisu K, Peng RD, Samet JM, Dominici F (2009) Hospital admissions and chemical composition of fine particle air pollution. Am J Respir Crit Care 179:1115–1120
- Bond TC, Doherty SJ, Fahey DW, Forster PM, Berntsen T, DeAngelo BJ, Flanner MG, Ghan S, Kärcher B, Koch D, Kinne S, Kondo Y, Quinn

PK, Sarofim MC, Schultz MG, Schulz M, Venkataraman C, Zhang H, Zhang S, Bellouin N, Guttikunda SK, Hopke PK, Jacobson MZ, Kaiser JW, Klimont Z, Lohmann U, Schwarz JP, Shindell D, Storelvmo T, Warren SG, Zender CS (2013) Bounding the role of black carbon in the climate system: a scientific assessment. J Geophys Res: Atmos 118:5380–5552

- Bonjour S, Adair-Rohani H, Wolf J, Bruce NG, Mehta S, Prüss-Ustün A, Lahiff M, Rehfuess EA, Mishra V, Smith KR (2013) Solid fuel use for household cooking: country and regional estimates for 1980-2010. Environ Health Perspect 121:784–790
- Braniš M, Vyškovská J, Malý M, Hovorka J (2010) Association of sizeresolved number concentrations of particulate matter with cardiovascular and respiratory hospital admissions and mortality in Prague, Czech Republic. Inhal Toxicol 22:21–28
- Brasche S, Bischof W (2005) Daily time spent indoors in German homes–baseline data for the assessment of indoor exposure of German occupants. Int J Hyg Environ Health 208:247–253
- Buonanno G, Fuoco FC, Stabile L (2011a) Influential parameters on particle exposure of pedestrians in urban microenvironments. Atmos Environ 45:1434–1443
- Buonanno G, Giovinco G, Morawska L, Stabile L (2011b) Tracheobronchial and alveolar dose of submicrometer particles for different population age groups in Italy. Atmos Environ 45: 6216–6224
- Buonanno G, Marini S, Morawska L, Fuoco FC (2012a) Individual dose and exposure of Italian children to ultrafine particles. Sci Total Environ 438:271–277
- Buonanno G, Morawska L, Stabile L, Wang L, Giovinco G (2012b) A comparison of submicrometer particle dose between Australian and Italian people. Environ Pollut 169:183–189
- Buonanno G, Stabile L, Morawska L, Russi A (2013a) Children exposure assessment to ultrafine particles and black carbon: the role of transport and cooking activities. Atmos Environ 79:53–58
- Buonanno G, Marks GB, Morawska L (2013b) Health effects of daily airborne particle dose in children: direct association between personal dose and respiratory health effects. Environ Pollut 180:246–250
- Cai J, Yan B, Kinney PL, Perzanowski MS, Jung KH, Li T, Xiu GL, Zhang DN, Olivo C, Ross J, Miller RL, Chillrud SN (2013) Optimization approaches to ameliorate humidity and vibration related issues using the MicroAeth black carbon monitor for personal exposure measurement. Aerosol Sci Tech 47:1196–1204
- Chau CK, Tu EY, Chan DWT, Burnett J (2002) Estimating the total exposure to air pollutants for different population age groups in Hong Kong. Environ Int 27:617–630
- Cheng YH, Lin MH (2013) Real-time performance of the microAeth[®] AE51 and the effects of aerosol loading on its measurement results at a traffic site. Aerosol Air Qual Res 13:1853–1863
- China MEP (2012) Ambient air quality standards. GB 3095-2012. China Environmental Science Press, Beijing
- de Miranda RM, de Fatima AM, Fornaro A, Astolfo R, de Andre PA, Saldiva P (2012) Urban air pollution: a representative survey of PM2.5 mass concentrations in six Brazilian cities. Air Quality Atmosphere Health 5:63–77
- Dons E, Panis LI, 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 45:3594–3602
- Dons E, Panis LI, Van Poppel M, Theunis J, Wets G (2012) Personal exposure to black carbon in transport microenvironments. Atmos Environ 55:392–398
- Geng F, Hua J, Mu Z, Peng L, Xu X, Chen R, Kan H (2013) Differentiating the associations of black carbon and fine particle with daily mortality in a Chinese city. Environ Res 120:27–32
- Hagler GSW, Yelverton TLB, Vedantham R, Hansen ADA, Turner JR (2011) Post-processing method to reduce noise while preserving high time resolution in aethalometer real-time black carbon data. Aerosol Air Qual Res 11:539–546

- Heal MR, Kumar P, Harrison RM (2012) Particles, air quality, policy and health. Chem Soc Rev 41:6606–6630
- Huo H, Wang M (2012) Modeling future vehicle sales and stock in China. Energy Policy 43:17–29
- Invernizzi G, Ruprecht A, Mazza R, De Marco C, Močnik G, Sioutas C, Westerdahl D (2011) Measurement of black carbon concentration as an indicator of air quality benefits of traffic restriction policies within the ecopass zone in Milan, Italy. Atmos Environ 45:3522–3527
- Janssen NA, Hoek G, Simic-Lawson M, Fischer P, Van Bree L, Ten Brink H, Cassee FR (2011a) Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM₁₀ and PM_{2.5}. Environ Health Perspect 119:1691–1699
- Janssen N, Simic-Lawson M, Hoek G, Fischer P, Brunekreef B, Atkinson R, Cassee F (2011b) Black smoke as an additional indicator to evaluate the health benefits of traffic-related policy measures: a systematic review of the health effects of black smoke compared to PM mass. Epidemiology 22:S199–S200
- Janssen NAH, Gerlofs-Nijland ME, Lanki T, Salonen RO, Cassee F, Hoek G, Fischer P, Brunekreef B, Krzyzanowski M (2012) Health effects of black carbon. WHO Regional Office for Europe, Copenhagen, pp 1–86
- Jim CY, Chen WY (2009) Leisure participation pattern of residents in a new Chinese city. Ann Assoc Am Geogr 99:657–673
- Kaur S, Nieuwenhuijsen MJ, Colvile RN (2007) Fine particulate matter and carbon monoxide exposure concentrations in urban street transport microenvironments. Atmos Environ 41:4781–4810
- Kingham S, Longley I, Salmond J, Pattinson W, Shrestha K (2013) Variations in exposure to traffic pollution while travelling by different modes in a low density, less congested city. Environ Pollut 181: 211–218
- Kirchstetter TW, Novakov T (2007) Controlled generation of black carbon particles from a diffusion flame and applications in evaluating black carbon measurement methods. Atmos Environ 41:1874–1888
- Leech JA, Nelson WC, Burnett RT, Aaron S, Raizenne ME (2002) It's about time: a comparison of Canadian and American time-activity patterns. J Expo Anal Environ Epidemiol 12:427–432
- Li B, Lei XN, Xiu GL, Gao CY, Gao S, Qian NS (2015) Personal exposure to black carbon during commuting in peak and off-peak hours in Shanghai. Sci Total Environ 524:237–245
- Manigrasso M, Stabile L, Avino P, Buonanno G (2013) Influence of measurement frequency on the evaluation of short-term dose of sub-micrometric particles during indoor and outdoor generation events. Atmos Environ 67:130–142
- Menon S, Hansen J, Nazarenko L, Luo Y (2002) Climate effects of black carbon aerosols in China and India. Science 297:2250–2253
- Panis LI, De Geus B, Vandenbulcke G, Willems H, Degraeuwe B, Bleux N, Mishra V, Thomas I, Meeusen R (2010) Exposure to particulate matter in traffic: a comparison of cyclists and car passengers. Atmos Environ 44:2263–2270
- Patel MM, Chillrud SN, Correa JC, Feinberg M, Hazi Y, Deepti KC, Prakash S, Ross JM, Levy D, Kinney PL (2009) Spatial and temporal variations in traffic-related particulate matter at New York City high schools. Atmos Environ 43:4975–4981
- Pope CA III, Dockery DW (2006) Health effects of fine particulate air pollution: lines that connect. J Air Waste Manage 56:709–742
- Qiu JH, Yang LQ (2000) Variation characteristics of atmospheric aerosol optical depths and visibility in North China during 1980-1994. Atmos Environ 34:603–609
- Reche C, Querol X, Alastuey A, Viana M, Pey J, Moreno T, Rodríguez S, González Y, Fernández-Camacho R, de la Rosa J, Dall'Osto M, Prévôt ASH, Hueglin C, Harrison RM, Quincey P (2011) New considerations for PM, Black Carbon and particle number concentration

for air quality monitoring across different European cities. Atmos Chem Phys 11:6207–6227

- Rodes CE, Chillrud SN, Haskell WL, Intille SS, Albinali F, Rosenberger ME (2012) Predicting adult pulmonary ventilation volume and wearing complianceby on-board accelerometry during personal level exposure assessments. Atmos Environ 57:126–137
- Salcedo D, Onasch TB, Dzepina K, Canagaratna MR, Zhang Q, Huffman JA, DeCarlo PF, Jayne JT, Mortimer P, Worsnop DR, Kolb CE, Johnson KS, Zuberi B, Marr LC, Volkamer R, Molina LT, Molina MJ, Cardenas B, Bernabé RM, Márquez C, Gaffney JS, Marley NA, Laskin A, Shutthanandan V, Xie Y, Brune W, Lesher R, Shirley T, Jimenez JL (2006) Characterization of ambient aerosols in Mexico City during the MCMA-2003 campaign with Aerosol Mass Spectrometry: results from the CENICA Supersite. Atmos Chem Phys 6:925–946
- Sayes CM, Reed KL, Warheit DB (2007) Assessing toxicity of fine and nanoparticles: comparing in vitro measurements to in vivo pulmonary toxicity profiles. Toxicol Sci 97:163–180
- Semmler M, Seitz J, Erbe F, Mayer P, Heyder J, Oberdörster G, Kreyling WG (2004) Long-term clearance kinetics of inhaled ultrafine insoluble iridium particles from the rat lung, including transient translocation into secondary organs. Inhal Toxicol 16:453–459
- SMBS (2014) Report on National Economic and Social Development of Shanghai for 2013., http://www.stats-sh.gov.cn/sjfb/201402/ 267416.html (February, accessed on 07.26.14, (in Chinese))
- Sørensen M, Autrup H, Møller P, Hertelc O, Jensenc SS, Vinzentsa P, Knudsena LE, Loft S (2003) Linking exposure to environmental pollutants with biological effects. Mutat Res Rev Mutat Res 544: 255–271
- Spira-Cohen A, Chen LC, Kendall M, Lall R, Thurston GD (2011) Personal exposures to traffic-related air pollution and acute respiratory health among Bronx schoolchildren with asthma. Environ Health Perspect 119:559–565
- Stabile L, Fuoco FC, Buonanno G (2012) Characteristics of particles and black carbon emitted by combustion of incenses, candles and antimosquito products. Build Environ 56:184–191
- U.S. EPA (2006) Air quality criteria for particulate matter. National Center for Environmental Assessment, US Environmental Protection Agency, Research Triangle Park, North Carolina
- Vanderstraeten P, Forton M, Brasseur O, Offer ZY (2011) Black carbon instead of particle mass concentration as an indicator for the traffic related particles in the Brussels Capital Region. J Environ Prot 2: 525–532
- Wang ZS, Wu T, Duan XL, Wang S, Zhang WJ, Wu XF, Yu YJ (2009) Research on inhalation rate exposure factors of Chinese residents in environmental health risk assessment. Res Environ Sci 22:1171– 1175 (in Chinese)
- Wang L, Morawska L, Jayaratne ER, Mengersen K, Heuff D (2011) Characteristics of airborne particles and the factors affecting them at bus stations. Atmos Environ 45:611–620
- Weichenthal S (2012) Selected physiological effects of ultrafine particles in acute cardiovascular morbidity. Environ Res 115:26–36
- Weingartner E, Saathoff H, Schnaiter M, Streit N, Bitnar B, Baltensperger U (2003) Absorption of light by soot particles: determination of the absorption coefficient by means of aethalometers. J Aerosol Sci 34: 1445–1463
- Westerdahl D, Wang X, Pan X, Zhang KM (2009) Characterization of onroad vehicle emission factors and microenvironmental air quality in Beijing, China. Atmos Environ 43:697–705
- Yang W, Lee K, Yoon C, Yu S, Park K, Choi W (2011) Determinants of residential indoor and transportation activity times in Korea. J Expo Sci Environ Epidemiol 21:310–316