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$PM_{2.5}$ exposure in highly polluted cities: A case study from New Delhi, India

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ABSTRACT

Personal exposure (PE) to air pollutants is driven by a combination of pollutant concentrations in indoor and outdoor environments, and time-activity pattern of individuals. The objectives of this study were to estimate personal exposure to $PM_{2.5}$ and black carbon (BC), and assess the representability of ambient air quality monitoring stations to serve as surrogates for PE in New Delhi. Personal exposure to air pollutants ($PM_{2.5\text{-}PE}$ and BC_{PE}) was measured using portable, battery-operated instruments ($PM_{2.5}$- pDR1500 and BC- microAethalometer AE51) in a small cohort of healthy adults (n = 12 in summer, n = 6 in winter) with no occupational exposure. Average $PM_{2.5\text{-}PE}$ and BC_{PE} ($\mu g/m^3$) were 53.9 ± 136 and 3.71 ± 4.29 respectively, in summer and 489.2 ± 209.2 and 23.3 ± 14.9 respectively, in winter. Activities associated with highest exposure levels were cooking and indoor cleaning for $PM_{2.5}$, and commuting for BC. Within transport microenvironments, autorickshaws were found to be the most polluted, and lowest BC exposure was registered in public buses. Comparison of fixed-site ambient monitoring data showed a higher correlation with personal exposure dataset in winter compared to summer (r^2 of 0.51 (winter) and 0.21 (summer); 51% (winter) and 20% (summer)). This study highlights the need for detailed assessment of PE to air pollutants in Indian cities, and calls for a denser network of monitoring stations for better exposure assessment.

1. Introduction

Exposure to air pollutants in outdoor and indoor environments has been identified as a risk factor for morbidity and mortality, particularly in the developing countries (Lim et al., 2012; Brauer et al., 2015). Within the same microenvironment (defined as '*a chunk of air space with homogenous pollutant concentration*', (Duan, 1982), referred to as ME hereafter), personal exposure (PE) to particulate matter (PM) depends on several factors include age, gender, time spent in different micro-environments, and susceptibility to air pollutants. Lifestyle and activity patterns as well as occupation and the socio-economic status of an individual can significantly influence exposure levels (Dons et al., 2011; Hajat et al., 2015; Steinle et al., 2015). Thus, a good understanding of exposure patterns (e.g. acute exposure during cooking) can improve modelling analyses, and reduce misclassification errors (Delgado-Saborit, 2012).

Pollution levels are almost universally higher in India compared to levels reported in Europe and North America, but paradoxically, there are a limited number of studies on PM in India (Guttikunda et al., 2014; Gurjar et al., 2016). Furthermore, these studies have typically been conducted at one or two locations, which does not account for heterogeneity in ambient conditions, much less the exposure for individuals. This becomes very relevant in India because under similar air quality conditions, individual exposure can vary significantly due to differences in activity patterns (type of job, location of work, time spent indoor vs. outdoor), rendering it difficult to generalize exposure concentrations for a large population using a limited number of monitoring stations. A recent review article highlighted the paucity of data on PE to air pollution in urban India (Pant et al., 2016b).

New Delhi is one of the most polluted cities in the world, and a recent modelling study estimated that exposure to air pollution can reduce the life expectancy of Delhi's citizens by six years (Ghude et al., 2016). Other analyses on health effects have also reported an increasing number of cases of respiratory diseases, as well as pollution-related morbidity and mortality in the city (Guttikunda and Goel, 2013; Nagpure et al., 2014). Air pollution exposure assessment is vital especially in the context of mixed-land use, and high population density in Indian cities, which often leads to high pollution exposures for a significant proportion of the population. To date, work published in the literature has either focused on characterization of ambient PM

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(Siddique et al., 2010; Khillare and Sarkar, 2012; Tiwari et al., 2014, 2009; Pant et al., 2015, 2016a), assessment of microenvironment-specific air quality (Apte et al., 2011; Firdaus and Ahmad, 2011; Goel et al., 2015a; Kumar and Gupta, 2016) or modelling of air pollution related health impacts for Delhi (Foster and Kumar, 2011; Guttikunda and Goel, 2013). However, none of these studies have provided an overarching assessment of human exposure in an urban context that can integrate a variety of typical exposures across a typical day, and be useful in comparing different types of exposures throughout a person's daily activity. A limited number of studies have characterized personal exposure in urban areas, but focused on student population (Jai Devi et al., 2009; Devi et al., 2013; Ashok et al., 2014). While such analyses have reported exposure profiles, it is important to consider that the time-activity patterns for the student population would be different from the general population. In addition, there is a significant gap in our understanding on personal exposure in urban areas in India, as was highlighted in a recent review (Pant et al., 2016b). Quantification of exposures across different microenvironments and activity types can lead to better health impact assessment, and inform policy initiatives such as the Government of India Smart Cities Mission to include clean air as part of the program.

Thus, the objectives of this study were- (i) to estimate personal exposure to air pollutants ($PM_{2.5}$ [PM with aerodynamic diameter less than 2.5 μm] and black carbon [BC]) in New Delhi, India in the general population (i.e. people with no significant occupational exposure) and, (ii) to assess the representability of ambient air quality monitoring stations to serve as surrogates for PE in New Delhi.

2. Materials and methods

2.1. Ambient measurements

Sampling was conducted in New Delhi (S1, SI), India in three phases between December 2014 and May 2016, and the sampling campaigns are summarized in Table 1. Ambient PM and BC samples were collected at two locations- Indian Institute of Technology (IIT), an urban site and Laxmi Bai Nagar (LBN), a residential site in summer 2015 while in winter 2014–15 and summer 2016, samples were only collected at the residential site. IIT is a university campus spread over 325 acres in South Delhi, and includes academic, residential and recreational buildings. There are no known major sources of air pollution in the area, except for common urban Indian sources such as vehicles and diesel backup generators. Sampling was conducted on top of a building (~10 m from ground level). LBN is a residential area, and sources include a nearby collector roads, and government and private housing. The two sites were considered for ambient sampling in order to get an estimate for average ambient pollution in residential areas. Real-time data from a central monitoring station for Delhi- the US Consulate monitoring station in Chanakyapuri (CNP) was used for comparison against the measurements. This site is at a distance of ~3.7 kms from LBN and 9.5 kms from IIT. There are no known major PM sources in the area except for local traffic, and this site was chosen as a reference urban site.

Table 1
Summary of sampling campaigns in Delhi, India (n = number of days of sampling).

Site	Sample type		Phase		
	Nephelometer and microAethalometer, real-time sampling	Integrated filters for PM analysis	Winter 2014-15	Summer 2015	Summer 2016
Ambient Air Sampling					
IIT	✓	✓	–	19	
LBN	✓	✓	6	9	~12
Personal Exposure (PE) Sampling					
Various	✓	✓	12	24	
Microenvironment (ME) Sampling					
Transport	✓	✓		✓	✓
Kitchen	✓		1.5	2	
Office	✓			3	
Home	✓		6		

2.2. Personal exposure measurements

Healthy volunteers between the ages 18–60 were recruited for 48 h PE monitoring (n = 12 in summer, n = 6 in winter). Each subject was sampled for a period of 48 h, although in some cases, the sampling duration was shorter due to instrument malfunction or other logistical issues. Demographic and health data was collected from each volunteer during both phases; volunteers with cardiovascular diseases and other health conditions were ineligible for the study. Seven female and five male subjects participated in the study during summer, and three female and three male subjects participated in the study during winter. With the exception of one subject during the summer campaign, all subjects were non-smokers, and all subjects used LPG gas as a cooking fuel at home. Most of the study subjects spent between 40 and 90 min in their daily commute, and between 8 and 10 h at work. Characteristics of the study group are summarized in Table S1. Most participants worked in research or commercial institutions, and spent most of the work hours in an office.

The study design was reviewed and approved by the Institutional Review Board of Human Subjects Research at University of Massachusetts, Amherst (2014–2309), and written consent was obtained from all volunteers before sampling began. During sampling, volunteers were requested to follow their usual activity patterns, and keep instrument inlets at breathing heights and away from obstruction.

2.3. Microenvironment sampling

For transport ME sampling, three main transport modes were assessed during this study including bus, three-wheeler and car (personal vehicle, diesel). A limited set of samples was also collected for two-wheelers. The sampling route (22 km in each direction) included a section of Mathura Road- National Highway 2 (NH 2) which is one of the busiest roads in Delhi, and includes intra- and inter-state goods vehicles in addition to passenger vehicles and public transport, as well as Prithviraj Road which has a low percentage of heavy duty vehicle traffic usage. Mathura Road is one of the major arterial roads in Delhi with an average traffic flow of 170,000 vehicles per day (Pant et al., 2015). Sampling was conducted along the route during peak (8–10 AM and 6–8 PM) and off-peak (12-2 PM) hours. Samples were also collected in indoor MEs including kitchen, office and a bedroom. None of the sampling locations had mechanical ventilation, and sampling equipment was placed at a height of ~1.5–2 m in all cases. Kitchen and bedroom samples were collected in a two-bedroom house in South Delhi where the kitchen is away from the bedroom.

2.4. Instrumentation and chemical analysis

To measure $PM_{2.5}$, a portable scattering nephelometer (pDR-1500, Thermo Fisher, Franklin, MA, USA) equipped with a $PM_{2.5}$ cyclone was used at a flow rate of 1.5 L/min and a time base of 5 min. Zero check was performed on the instrument using a clean HEPA filter before every run, and calibrations were checked before field deployment. In addition, this instrument was used to collect 24-h integrated measurements of $PM_{2.5}$ on PFTE filters (Zefon International, 37 mm, 2.0 μm). BC was measured using a microAethalometer AE51 (AethLabs, San Francisco, CA, USA) operated at a flow rate of 50 mL/min and a 5-min time base. For transport ME sampling in summer 2015, the same time base was used, but in summer 2016, 1- minute time base was used in order to

account for some of the variability in on-road emissions. A condensation particle counter (CPC 3007, TSI Inc., Shoreview, MN) was used to for particle number concentration (PNC) measurements (0.01–1 μm). At concentrations higher than 10^5 cm^{-3}, the CPC can under-report true concentrations, and all values higher than this threshold were adjusted using a correction factor (Westerdahl et al., 2005). A GPS module (DG-200, USGlobalSat Inc., CA, USA) was used to collect location and speed during sampling.

All instruments were battery operated and portable, and allowed for simultaneous measurement of $PM_{2.5}$ mass and BC concentrations and geolocation information. For PE measurements, each volunteer was provided with a handbag containing the instruments and battery pack and request to keep the instrument inlet at breathing height. For transport ME sampling, the instruments were hand-carried in a cardboard box with insulation, while for other ME sampling (i.e. home, office and kitchen), the instruments were placed at a stationary location. Integrated 24 h filter samples were weighed twice using a microbalance (MT XP6, Mettler) before and after sampling. Before gravimetric measurements, filters were equilibrated under controlled temperature (20–30^{0}C) and RH (40–50%). Subsequently, the samples were analyzed for metals using energy dispersive X-ray fluorescence (ED-XRF, Quant'x filter XRF) following standard procedures. XRF calibration was performed using thin film (nanoXRF, Fort Worth, TX) standards. Method detection limits (MDLs) were determined using blank Teflon filters (3*standard deviation).

2.5. Data processing and analysis

Raw data was checked for missing values, anomalies (e.g. non-physical data such as negative concentrations, impossibly high concentrations, or significant variation from the specified flow rates) and any other errors (e.g. missing data due to instrument malfunction). Data was removed in a manner consistent with our laboratory standard operating procedures, and thus, excluded from the analysis. Subsequently, summary data was generated, and all datasets were tested for normality using quantile-quantile plots and Shapiro-Wilk Test. Descriptive statistics were used to characterize ambient, micro-environment and PE pollutant data. Spearman's Rank correlation analysis (r_s) was used for understanding correlations, and linear regression analysis was used to examine relationships between different variables. For metals, data below MDL was replaced with 0.5*DL and Pearson correlation (r_p) was used to understand the relationship between metals. Data analysis was conducted using Microsoft Excel 2010 and R (version 3.2.4, R-project.org).

3. Results and discussion

3.1. Ambient $PM_{2.5}$ and BC using stationary sampling

Summary statistics of ambient $PM_{2.5}$ and BC measurements across the three sites are presented in Table 2. $PM_{2.5}$ concentrations were found to vary across seasons, with winter concentrations being significantly higher than the summer. Geometric mean [GM] $PM_{2.5}$ concentrations in winter were estimated as 488(1.63) μg/m^3 and 238(1.54) μg/m^3 at LBN and CNP (Table S2) respectively. Concentrations were much lower in summer 2015 with average GM hourly concentrations of $PM_{2.5}$ ranging between 39.2–45.9 μg/m^3 at the three sites. Annual average $PM_{2.5}$ concentration in New Delhi was reported to be 130 ± 103 μg/m^3 based on measurements at an urban background location (Tiwari et al., 2014) while Pant et al. (2015) reported average 12 h $PM_{2.5}$ concentrations of 58.2 ± 35.0 μg/m^3 and 276.9 ± 99.9 μg/m^3 for summer and winter respectively at an air pollution hotspot. For BC, average concentration in winter was more than five time higher than in the summer at LBN [GM = 17.0(1.82) and 3.08(2.25) μg/ m^3 for winter and summer respectively].

Correlation analysis was performed using 5-min $PM_{2.5}$ and BC concentration data. In winter 2014 and summer 2015, ambient $PM_{2.5}$ at two sites (LBN and CNP) were found to be moderately correlated ($r_s = 0.65$, $p < 0.05$, $n = 133$ and $r_s = 0.56$, $p < 0.05$, $n = 227$) while in summer 2016, a strong correlation was observed ($r_s = 0.82$, $p < 0.05$, $n = 287$). Additionally, a strong correlation was observed between measurements at CNP and IIT ($r_s = 0.76$, $p < 0.05$, $n = 460$) in summer 2015.

Integrated $PM_{2.5}$ filter samples were also analyzed for elemental concentrations, and the results are discussed in Section S2 of the supplementary information. This data was analyzed in order to provide a frame of reference for the PE observations.

3.2. Personal exposure to PM and BC using mobile sampling

Different individuals have different exposures, and associated health effects; with factors such as age, gender, occupation and socio-economic status significantly affecting PE to air pollutants. PE patterns across a section of the population can help pinpoint the key variables driving exposure to air pollution, and inform strategies for exposure reduction, as well as health assessment studies. In Delhi, mean hourly $PM_{2.5\text{-}PE}$ (i.e. personal exposure $PM_{2.5}$) and BC_{PE} (i.e. personal exposure BC) were 32.5(2.36) μg/m^3 and 2.41(2.65) μg/m^3 and 432(1.66) μg/m^3 and 18(2.02) μg/m^3 in summer and winter respectively. A summary is presented in Table 3. In comparison, mean $PM_{2.5\text{-}PE}$ was reported to be 102.5 μg/m^3 in Beijing (China) (Du et al., 2010); in Australia, William and Knibbs (2016) reported average 24 h BC_{PE} of 0.60 μg/m^3 and in Birmingham (UK), Delgado-Saborit (2012) reported BC_{PE} of 1.3(± 2.2) μg/m^3.

In winter 2014, $PM_{2.5\text{-}PE}$ and BC_{PE} were found to be strongly correlated ($r_s = 0.90$, $p < 0.05$, $n = 190$) taking hourly measurements from the entire cohort into consideration while individual correlations (r_s, $p < 0.05$) ranged between 0.32 and 0.88. In summer 2015, individual personal $PM_{2.5}$-BC correlation (r_s, $p < 0.05$) ranged between −0.27 and 0.86, while the overall correlation was moderate ($r_s = 0.50$, $p < 0.05$). In general, the correlations improved when daily data was used.

Time spent in different MEs was split into six categories- commuting, indoor-home, indoor-office, indoor-kitchen, outdoor and indoor-other. Overall, $PM_{2.5\text{-}PE}$ varied from ~3–3000 μg/m^3, and short-term peaks were associated with commuting, cooking and cleaning.

Table 2
Summary statistics for ambient $PM_{2.5}$ and BC (μg/m^3) (N refers to # of 5 min observations, AM- arithmetic mean, SD- standard deviation, GM- geometric mean, GSD- geometric standard deviation, Min- minimum, Max- maximum).

Site	Study Period	Pollutant	N (# of days)	AM (SD)	GM (GSD)	Min-Max
LBN	Winter 2014-15	$PM_{2.5}$	1741 (~6)	545 (257)	488 (1.63)	85.4- 2500
	Winter 2014-15	BC	376 (~1.3)	19.3 (8.22)	17.0 (1.82)	0.87- 54.7
	Summer 2015	$PM_{2.5}$	2664 (~9)	45.0 (27.9)	38.7 (1.74)	3.43–498
	Summer 2015	BC	1333 (~4.6)	3.99 (2.66)	3.08 (2.25)	0.04–15.1
	Summer 2016	$PM_{2.5}$	3443 (~12)	97.6 (98.4)	68.8 (2.26)	6.68–668.2
IIT	Summer 2015	$PM_{2.5}$	5561 (~19)	60.9 (59.2)	45.4 (2.04)	5.7–437.6
	Summer 2015	BC	828 (~3)	3.89 (3.34)	3.07 (1.94)	0.22–30.1

Table 3
Summary statistics for personal exposure data ($PM_{2.5}$ in μg/m^3 and BC in μg/m^3, N refers to # of 5 min observations; each volunteer was sampled for 48 h).

PE campaign	Pollutant	AM	SD	GM	GSD	Min-Max
Winter 2014–15	$PM_{2.5}$	484	230	432	1.66	18.8–2246
	BC	22.6	14.9	18.0	2.02	0.84–71.2
Summer 2015	$PM_{2.5}$	53.9	136	32.5	2.36	2.64–3020
	BC	3.71	4.29	2.41	2.65	0.008–71.6

Cooking/ time spent in kitchen, cleaning activities and commuting were found to correspond to highest PM peaks (peak exposure levels) while BC peaks were typically highest during commuting or when the subjects were outdoors. Table 4 summarized the concentrations across all microenvironments. Overall, concentrations were lower in indoor office MEs, and this is likely due to the nature of offices where the samples were collected. In most cases, subjects from this cohort worked in air-conditioned offices, with closed windows and mechanical ventilation.

An example exposure profile is illustrated in Fig. 1. In this case, highest concentrations of $PM_{2.5}$ and BC were observed during cooking and commuting, and the major peaks on June 23rd and 24th (summer) correspond to cooking and commuting events. Indoor (home) ME contributed 62% and 52.5% of the total $PM_{2.5}$ and BC exposure for this individual, while indoor (kitchen) ME contributed 17.1% and 13% of $PM_{2.5}$ and BC respectively. Commuting contributed more than 15% of the total BC exposure, while office contributed 12% and 19% of the total $PM_{2.5}$ and BC exposure. Fig. 1b represents an example profile for the winter season. In this case, the highest concentrations were associated with cooking, incense burning and commuting activities.

Differences were also observed in the exposure profiles of study subjects in terms of pollutant exposure (Fig. 2). For study subjects 1 and 7 (both females), kitchen ME was an important contributor to both $PM_{2.5}$ and BC exposure in the context of peak exposures, while kitchen was not relevant for subjects 8 and 10 (males). This is an important observation since women bear a significant responsibility in terms of cooking in India, and often spend a significant amount of time in the kitchen, especially in poor households (Dutta and Banerjee, 2014). Thus, for women that are the primary cook(s) in the household cooking, PM exposures can be very high, albeit for short periods of time. Highest peak PM exposure level was reported for subject 7, who conducted cooking as well as cleaning on both days of sampling, and corresponding peaks were seen in the exposure profile (Fig. 1). All the participants in this study used LPG as a cooking fuel, and thus, we do not expect a substantial contribution from the choice of cooking fuel. However, Asian-style cooking has been reported to generate more PM compared to Western cooking (Abdullahi et al., 2013), and in northern India, where this study was conducted, the culinary style involves pan-frying as well as deep-frying. In case of one of the volunteers, concentrations as high as 870 μg/m^3 were observed, and the highest concentrations corresponded with time spent in the kitchen while in another case, the highest 5-min concentration was 1283 μg/m^3. However, not all women reported cooking in the kitchen, and the exposure due to cooking is expected to vary from person to person, and across the socio-economic spectrum, due to fuel choices as well as time spent in the kitchen.

On average, subjects spent between 5–10% of their time in commuting, which is in agreement with the reported data in the literature (Goel et al., 2015b; Pant et al., 2016b). On average, BC exposure during commuting varied between 10% and 25% of the total BC exposure across all microenvironments, with the exception of two subjects where the contribution of commute to BC levels was much lower (1–5% of total BC). Fig. 2 compares PE for four individuals across all microenvironments; subjects 1 and 7 are females and subjects 8 and 10 are males. All four subjects spent at least 85% of their time in indoor MEs, and commute time varied between 6–14%. Subject 8, used public buses for commuting, and had the lowest BC exposure, while for subjects 1, 7 and 10, commute was a significant contributor to their total BC exposure, and all three used cars for commuting (two of them used personal vehicles, while the third used a shared taxi). Further, all three subjects travelled along routes with variable traffic volumes. Lower BC exposure in bus is discussed further in Section 3.3.1. For Subject 1, the total time spent in commuting was 10.9% (over a 48 h period), but the contribution of the commute to the total $PM_{2.5}$ and BC exposure was 17.6% and 25.4% respectively. In Australia, commuting was estimated to contribute to ~32% of the total BC_{PE} while in Belgium, commuting was found to contribute to 21% of the total BC_{PE} (Dons et al., 2012; Williams and Knibbs, 2016). On the other hand, Subject 7 had substantial exposure from kitchen/cooking emissions, as well as indoor cleaning activities. In this context, it is also important to remember that several variables including type of commute and housing location and type can depend on the socio-economic status of the individual, and previous studies have shown that individuals with the same socio-economic status, as well as lifestyle can have different exposures, due to differences in time-activity patterns (e.g. time and duration of commute) (Dons et al., 2011). In this study, we did not explore the role of socio-economic status on personal exposure levels, but future studies should include such an assessment.

Similar to ambient air (see SI), Si and Fe were the most abundant crustal elements in PE samples, followed by Zn, Cu and Pb (Table 5). In both seasons, crustal elements- Ca, Fe and Si were found to be positively and strongly correlated ($r_p = 0.74–0.89$, $p < 0.05$). However, surprisingly, Al was not significantly correlated with the other elements while Ti had a positive and significant correlation with Ca, Fe and Si ($r_p = 0.59–0.78$, $p < 0.05$). Pb, Zn and Ni were also found to be strongly correlated ($r_p = 0.77–0.80$, $p < 0.05$). In summer, Pb was also found to be weakly correlated with Cu and Ni ($r_p = 0.47–0.48$, $p < 0.05$).

3.3. Pollutant concentrations in microenvironments

3.3.1. Transport microenvironments

On average, between 11 and 25 h of measurements were conducted in each transport microenvironment. Average $PM_{2.5}$ concentrations (1-min average) for autorickshaw and bus were 59.4(2.37) and 53.9(1.98) μg/m^3 while the BC concentrations for the two modes were 23.4(1.95) and 14.1(1.93) μg/m^3, respectively. Results are summarized in Table 6 (and Table S4, SI). Broadly similar concentrations were observed during a more limited sampling campaign in summer 2015 (Table S4) where average $PM_{2.5}$ and BC concentrations (5-min average) across three transport modes- autorickshaw, bus and two-wheeler were 78.0 ± 39.7, 51.0 ± 15.7, and 55.0 ± 28.8 μg/m^3 and 31.7 ± 34.7, 8.34 ± 8.51, and 23.8 ± 11.7 μg/m^3 respectively. In-vehicle measurements were

Table 4
Average $PM_{2.5}$ and BC concentrations (μg/m^3) across the different microenvironments.

Pollutant	Home	Kitchen	Outdoor	Commute	Office	Other indoor	Unspecified
PM	57.7 (± 40.8)	310.0 (± 456.2)	113.9 (± 71.6)	49.3 (± 21.8)	25.8 (± 12.4)	40.5 (± 33.0)	18.1
BC	2.78 (± 1.35)	6.76 (± 7.24)	4.75 (± 2.42)	4.31 (± 2.28)	2.38 (± 1.50)	2.38 (± 2.19)	2.99

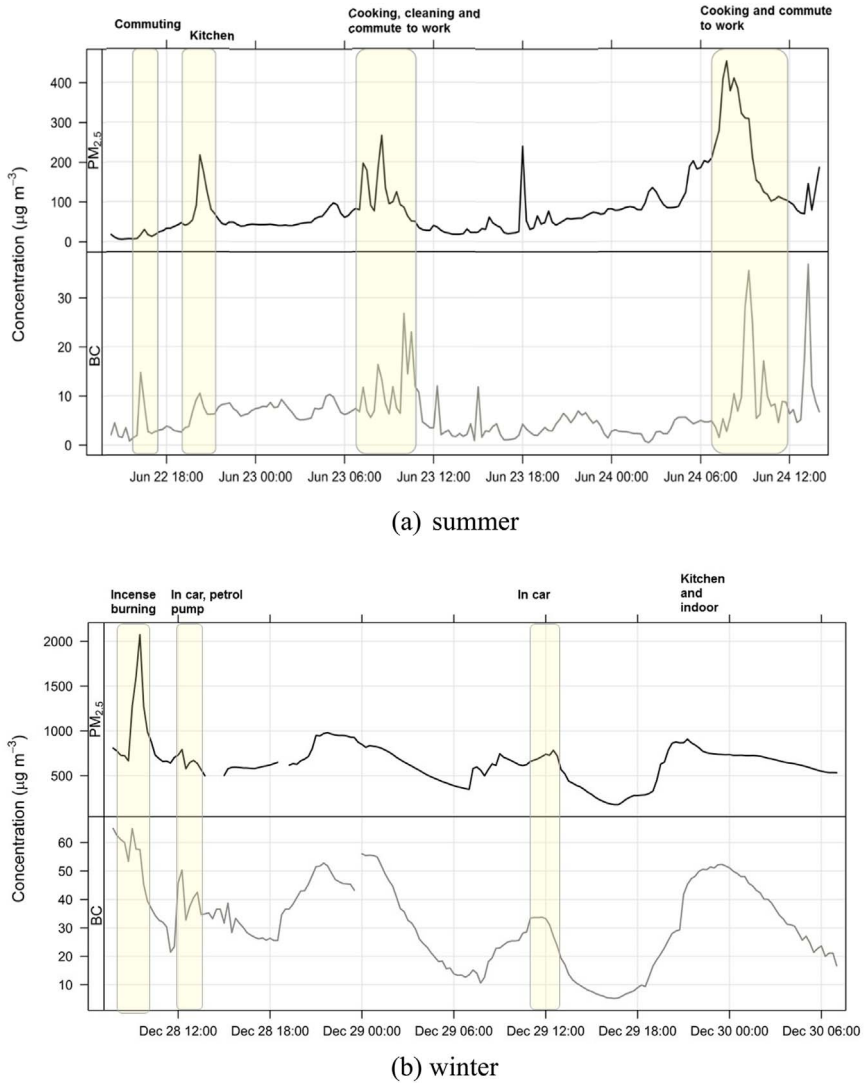

Fig. 1. Exposure profile for one of the study subjects (a- summer, b- winter).

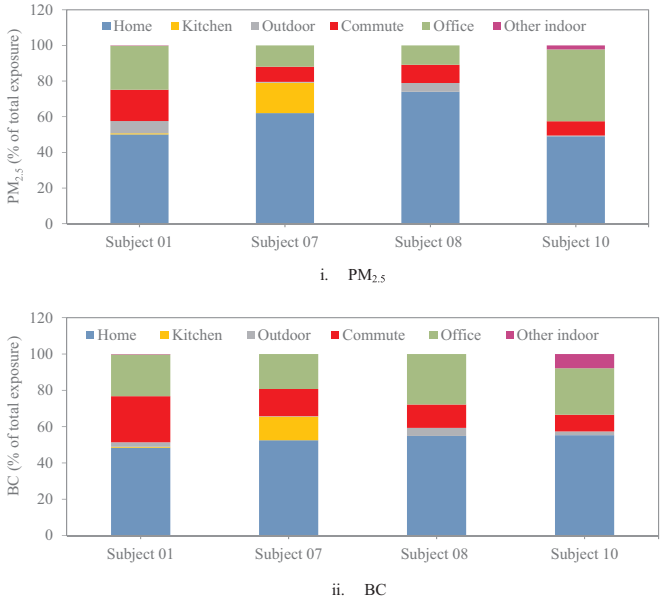

Fig. 2. Comparison of average personal exposure in different MEs for (i) $PM_{2.5}$ and (ii) BC for select subjects.

Table 5
Summary of PE $PM_{2.5}$ element concentrations (*in µg/m^{3}and other elements in ng/m^3).

Element	Summer 2015		Winter 2014-15	
	AM (SD)	Min –Max	AM (SD)	Min- Max
Al*	0.78 (0.54)	0.02–1.92	0.37 (0.49)	0.01–1.58
Si*	1.05 (0.52)	0–2.14	0.78 (0.59)	0–2.1
Ca*	0.63 (0.29)	0.07–1.19	0.55 (0.55)	0.2–1.98
Fe*	0.61 (0.23)	0.1–1.02	0.62 (0.21)	0.36–0.99
Ti	50.8 (72)	2.1–294.9	45.5 (44.4)	6.14–166.8
Mn	54.5 (41.2)	3.1–139.3	63.8 (45)	8.76–127.5
Ni	47 (28.1)	3.3–120.8	63.3 (22.4)	19.9–92.9
Cu	185.8 (142.4)	78.7–707.1	189.8 (43.2)	134.8–255.7
Zn	296.7 (214.6)	57.9–966.1	563.1 (149.1)	273.4–823.9
Pb	97.6 (83)	2.5–281.5	270.1 (156.7)	61.3–569.9

also conducted for cars, but measured BC concentrations were found to exceed $PM_{2.5}$ concentrations. Since this is a highly unusual observation, and we were unable to explain this, or replicate the measurements, the data has not been included in the discussion.

Both BC ($r_s = 0.67$, $p < 0.05$) and PNC ($r_s = 0.56$, $p < 0.05$) were found to be moderately correlated with $PM_{2.5}$ in autorickshaw, while only BC was found to be correlated to $PM_{2.5}$ ($r_s = 0.74$, $p < 0.05$) in bus. Vehicle speed was not found to be correlated with pollutant concentra-

Table 6
Summary of pollutant concentrations in transport microenvironments (BC in μg/m^3, $PM_{2.5}$ in μg/m^3, PNC in pt/cc).

Mode	Average Speed	Pollutant	N	AM	SD	GM	GSD	Min-Max
Summer 2016 (N # 1 min observations)								
Car	20.4 ± 18.2	PNC	673	25573	15333	21720	1.76	4900–77500
Bus	13.1 ± 14.8	BC	1094	17.1	10.6	14.1	1.93	0.35–89.6
		$PM_{2.5}$	1052	69.1	55.8	53.9	1.98	8.27–310
		PNC	724	46636	33553	36720	2.04	340–200000
Autorickshaw	18.2 ± 15.4	BC	1345	29.0	21.7	23.4	1.95	0.25–247
		$PM_{2.5}$	1447	88.7	89.4	59.4	2.37	10.4–447
		PNC	1079	68551	45867	55260	1.97	5900–300500

tions in any of transport modes in 2016, but for the 2015, a moderately weak but positive relationship ($r_s = 0.44$) was observed for bus. For two-wheelers, BC and speed were also found to be weakly but positively correlated ($r_s = 0.36$).

Since this was a small dataset, and the data were positively skewed, a Kruskal-Wallis test ($p < 0.05$) was used to assess differences between the three modes. For summer 2016, the Kruskal-Wallis test showed a statistically significant difference between autorickshaw and bus for BC and PNC, while for $PM_{2.5}$, the difference was not statistically significant.

For each transport mode, a ratio of BC/$PM_{2.5}$ was calculated in order to understand the contribution of BC to total in-vehicle $PM_{2.5}$ concentration. This ratio was found to be the lower for buses (0.29 ± 0.15), compared to autorickshaw (0.46 ± 0.40), indicating that the relative contribution of BC to $PM_{2.5}$ is lower for buses. This is not surprising given that public buses in Delhi run on compressed natural gas (CNG) which is not likely to produce much BC. Additionally, the ratio between $PM_{2.5}$ in transport mode and ambient $PM_{2.5}$ was also calculated, and was observed to be the highest for three-wheelers (0.67 ± 0.46) compared to buses (0.59 ± 0.43).

In terms of $PM_{2.5}$ chemical composition, highest concentrations of elements associated with crustal dust [silicon (Si), iron (Fe) and calcium (Ca)] were observed in buses, while the concentrations of these elements were lowest in cars. Highest in-vehicle concentrations for copper (Cu) and zinc (Zn) were observed for car and bus respectively, while lead (Pb) was the highest in the case of autorickshaw.

3.3.2. Indoor microenvironments

In the kitchen, average $PM_{2.5}$ and BC concentrations were 44.3(2.38) μg/m^3 and 3.57(2.19) μg/m^3 respectively while average PNC was 71900 (2.06) particles/cm^3 (Table 7). $PM_{2.5}$ was strongly correlated with BC ($r_s = 0.61$, $p < 0.05$) as well as PNC ($r_s = 0.74$, $p < 0.05$), although BC and PNC were not found to be correlated.

Table 7
Summary of pollutant concentrations in indoor microenvironments (BC and $PM_{2.5}$ in μg/m^3, PNC in pt/cc; N refers to 5-min observations).

ME	Pollutant	N (# of days)	AM	SD	GM	GSD	Min-Max
Kitchen[a]	$PM_{2.5}$	921 (~3)	77.2	179	44.3	2.38	12.9–3497
	BC	917 (~3)	4.86	4.13	3.57	2.19	0.44–35.9
	PNC	38	92824	68002	71900	2.06	24630–234800
Office[b]	$PM_{2.5}$	854 (~3)	30.5	15.5	26.8	1.70	8.33–82.5
Home[c]	$PM_{2.5}$	1823 (~6)	405	4.93	336	1.84	88–1874
	BC	390 (~1.5)	16.6	264	15.8	1.37	5.7–41

[a] Average of summer and winter.
[b] Summer.
[c] Winter.

Concentrations of $PM_{2.5}$ in the kitchen ME were observed to be broadly similar to a previous studies from Mysore (Karnataka) and Nagpur (Maharashtra) (Andresen et al., 2005; Mönkkönen et al., 2005). Significant peaks were seen for $PM_{2.5}$ concentrations, especially during cooking events, and this can be attributed to particle generation during the cooking process.

In winter 2014-15, $PM_{2.5}$ and BC were monitored inside a two-bedroom apartment at LBN over a two day period. Electrical space heating was used during evening, and cooking fuel was LPG. There were no smokers in the house, and ventilation was passive, with windows manually opened and closed during the day as needed. Average $PM_{2.5}$ was recorded as 336(1.84) μg/m^3, while average BC was 15.8(1.37) μg/m^3. Based on this limited set of observations (Fig. 3), hourly indoor and ambient BC were found to be strongly correlated ($r_s = 0.90$, $p < 0.05$) and indoor and ambient $PM_{2.5}$ were found to be strongly correlated ($r_s = 0.88$, $p < 0.05$) indicating the influence of ambient pollutant concentrations on indoor levels, for both air pollutants. However, poor correlation was observed between indoor BC and $PM_{2.5}$ ($r_s = 0.20$, $p < 0.05$), and is indicative of different sources for the two pollutants.

Offices are another microenvironment that can contribute to the daily exposure. In Delhi, average $PM_{2.5}$ in the office microenvironment (natural ventilation) was observed to be 30.5 ± 15.5 μg/m^3 during summer season. In comparison, Goyal and Kumar (2013) reported $PM_{2.5}$ concentrations ranging between 16.9 and 102.6 μg/m^3 in a mixed-use commercial building and Goyal and Khare (2009) reported average concentrations of 71 and 359.9 μg/m^3 in a naturally-ventilated school building for non-winter and winter periods respectively, both in Delhi.

3.4. Relationship between ambient and personal exposure pollutant concentrations

The relationship between personal and ambient $PM_{2.5}$ concentrations was assessed using the following approach: first, Spearman correlation coefficients were used (r_s) to assess correlation between the variables, and following that, linear regression analysis was used to understand the relationship between ambient and personal exposure PM.

In winter, correlations between hourly personal exposure $PM_{2.5}$ ($PM_{2.5\text{-}PE}$) and ambient $PM_{2.5}$ ($PM_{2.5-A}$) and BC_{PE} and $PM_{2.5-A}$ were strong ($r_s = 0.78$, $n = 173$ and $r_s = 0.72$, $n = 204$ respectively, $p < 0.05$) [Fig. S1a]. In summer 2015, however, the correlations were found to be weaker [Fig. S1b]. Correlation between $PM_{2.5\text{-}PE}$ and $PM_{2.5-A}$ was moderate ($r_s = 0.48$, $p < 0.05$), but weak for BC_{PE} and $PM_{2.5-A}$ ($r_s = 0.28$, $p < 0.05$). In comparison, a moderately strong correlation was reported between ambient and PE $PM_{2.5}$ in Toronto (Kim et al., 2005) while in Gothenburg (Sweden) researchers reported a weak correlation between $PM_{2.5}$ and BS (black smoke, a surrogate for BC) (Johannesson et al., 2007). Similarly, in USA, a weak correlation was reported between personal and ambient $PM_{2.5}$ (Hsu et al., 2012). Several factors including differences in air exchange rates (summer vs

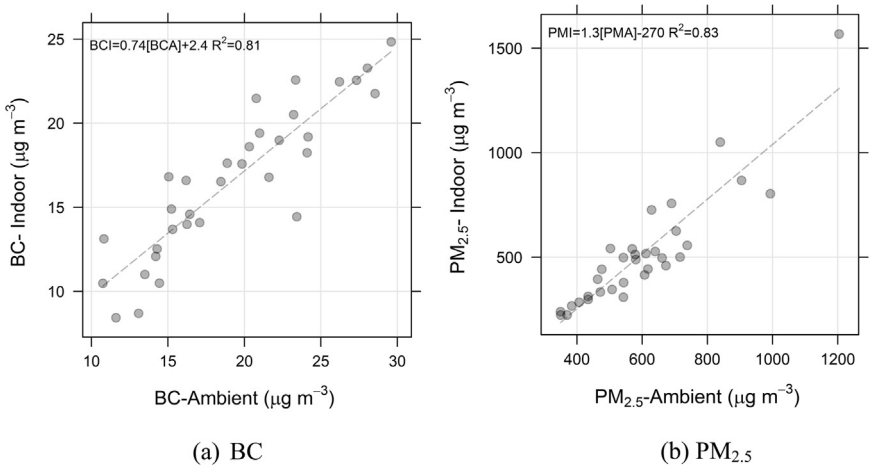

Fig. 3. Scatter plot for hourly indoor and outdoor (a) BC and (b) $PM_{2.5}$ concentrations in winter 2014.

winter), distance from the ambient monitoring station and time spent in different microenvironments (indoor vs outdoor) can influence the correlation patterns between ambient and personal exposure data.

$PM_{2.5\text{-}PE}$ and BC_{PE} were also regressed against $PM_{2.5-A}$ using log-transformed data (owing to a non-normal distribution of data) and results are summarized in Table 8. For the pooled winter hourly dataset, 51% of the variance in $PM_{2.5\text{-}PE}$ can be explained using $PM_{2.5-A}$, while for summer, only 20% of the variance is explained by ambient PM. For BC_{PE}, 65% of the variance can be explained by ambient PM in winter while in summer, a mere 7% variance in concentrations is explained by $PM_{2.5-A}$. The gradient for $PM_{2.5\text{-}PE}$ in summer was almost half of that in winter, but the intercepts were very similar in both seasons.

4. Conclusions

This pilot study presents data on personal exposure to $PM_{2.5}$ and BC in New Delhi, and presents a novel dataset to understand the PE concentrations compared to ambient air pollution in a highly polluted city. The analysis elucidates personal exposure levels for Delhi's residents based on a small, non-occupational, general population cohort, and average exposure levels were found to be higher than concentrations reported for similar cohorts in Europe and N. America. Commuting, indoor cleaning activities and cooking were identified as activities contributing to high levels of pollutant exposures, and indoor MEs were found to make the largest overall contribution. Commuting has been identified as a high-exposure activity in studies elsewhere (Dons et al., 2012; Karanasiou et al., 2014), and in Delhi, traffic ME was one of the major contributors to BC exposure. Further, similar to ambient air pollution trends, PE concentrations were higher in winter compared to summer, and choice of commute was seen to influence the total daily exposure. Autorickshaws were found to be associated with the highest exposure levels among the commute types monitored in this study. This is very relevant in the policy context, since in areas with poor or no public transportation, autorickshaws are often the most common mode of commute, both in Delhi and in other cities and towns. It was also interesting to note that the lowest levels of BC were observed in buses, and this issue warrants further investigation. Future studies should look at in-vehicle exposure with respect to land use and traffic characteristics, and personal exposure data should be used as an input for land use zoning and urban transportation planning.

Research elsewhere has reported that representative fixed site monitors can often serve as surrogates for personal exposure PM (Kim et al., 2005; Hsu et al., 2012), particularly in cases where regional pollution is an important source (e.g. Delhi in winter season). The same pattern seems to be evident in the current dataset, but needs more detailed investigation. In terms of the representability of ambient PM measurements as a surrogate for personal exposure, higher correlation was seen in the dataset for winter compared to summer, indicative of a higher contribution of ambient air pollution to PE in winter. Several studies focused on analysis of ambient air have reported significant differences between pollutant concentrations in summer and winter seasons, and this is attributed to a mix of meteorological factors, and an increase in local (space heating) and regional (crop residue burning) combustion sources.

One of the limitations of this study is that a single reference monitor was used for ambient $PM_{2.5}$ for the entire cohort, and this is not representative for all subjects. Although there is limited data on spatial heterogeneity of air pollutants in New Delhi, it is reasonable to assume that pollutant concentrations vary across the city. It is also important to acknowledge that the dataset analyzed in this pilot study was limited, and cannot be directly extrapolated for larger scale analyses. The sample population was comprised of middle class individuals, and the observed concentrations were not likely representative of all individuals in the city. Further, previous research has indicated confounding effect of exposure to gaseous pollutants (Kim et al., 2005), and this was not explored in the current study. Future studies should include an assessment of both particulate and gaseous pollutants.

Table 8
Summary of linear regression analysis using log-transformed data.

Season	Metric		Explanatory Variable		Model		
			Gradient	Intercept	R^2	F	p-Value
$PM_{2.5\text{-}PE}$	Winter	$PM_{2.5-A}$	1.04 ± 0.12	0.68 ± 0.05	0.51	177.1	< 2.2e−16
	Summer		0.45 ± 0.09	0.66 ± 0.06	0.21	135.3	< 2.2e−16
BC_{PE}	Winter		- 1.31 ± 0.14	1.10 ± 0.06	0.65	41.4	2.86e−10
	Summer		- 0.28 ± 0.11	0.43 ± 0.07	0.07	376.5	< 2.2e−16

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.envres.2017.03.024.

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