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Variations in individuals' exposure to black carbon particles during their daily activities: a screening study in Brazil

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Abstract
Black carbon (BC) is a fraction of airborne $PM_{2.5}$ emitted by combustion, causing deleterious effects on human health. Due to its abundance in cities, assessing personal exposure to BC is of utmost importance. Personal exposure and dose of six couples with different working routines were determined for 48 h based on 1-min mobile BC measurements and on ambient concentrations monitored simultaneously at home (outdoor) and at a suburban site. Although couples spent on average ~ 10 h together at home, the routine of each individual in other microenvironments led to 3–55% discrepancies in exposure between partners. The location of the residences and background concentrations accounted for the differences in inter-couple exposure. The overall average exposure and dose by gender were not statistically different. The personal exposure and dose calculated with datasets from fixed sites were lower than the calculations using data from mobile measurements, with the largest divergences (between four and nine times) in the transport category. Even though the individuals spent only 7% of the time commuting, this activity contributed to between 17 and 20% of the integrated exposure and inhaled dose, respectively. On average, exposure was highest on bus trips, while pedestrians and bus passengers had lower doses. Open windows elevated the in-car exposure and dose four times compared to settings with closed windows.

Keywords Air pollution · Personal exposure · Mobile monitoring · Transport modes · Spatial analysis · Short-lived climate pollutant

Introduction

Over the last couple of years, air quality and population exposure to air pollutants have received special attention due to scientific evidence of causal relationship between contact with air pollutants and negative health outcomes, particularly for individuals with cardiovascular or respiratory diseases (Brook et al. 2010; Gan et al. 2011). Although some studies have shown the effects of multiple air pollutants on human health, such as surface ozone (e.g., Goldberg et al. 2001) and dioxins (e.g., Kogevinas 2001), a vast number of epidemiological studies have focused on particulate matter (PM) due to its abundance in urban environments and the greater effect caused by the complex mixture of organic and inorganic species within the PM. More specifically, the inhalation of fine ambient PM (with diameter smaller than 2.5 μm, $PM_{2.5}$) accounts for three million annual deaths worldwide (WHO 2016).

Black carbon (BC) is a fraction of $PM_{2.5}$ mainly emitted by the combustion of fossil fuels and biomass, ubiquitous in the atmosphere of urbanized areas and downwind of biomass burning sources. The lifespan of BC in the lower atmosphere ranges from days to weeks, which is long enough to alter the Earth's radiation balance through the absorption of incoming solar radiation (Jacobson 2001). Because of its nanometer size, BC may be deposited onto the walls of the respiratory system, causing cardio-respiratory diseases (Mauderly and Chow 2008; Gan et al. 2011). BC is considered a robust indicator to assess the adverse effects of particulates on human health where combustion processes prevail, showing clearer associations with morbidity, daily mortality, and life expectancy than solely $PM_{2.5}$. For example, Janssen et al. (2011) used the average and 95% confidence interval (CI) of concurrent roadside BC and $PM_{2.5}$ street increments (calculated as the

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difference between traffic and background concentrations) to estimate the effects on health of the increase in concentrations. They concluded that a 1 $\mu g \, m^{-3}$ increase in $PM_{2.5}$ was associated with a 0.19% increase (95% CI 0.03, 0.35%) in all-cause mortality and a 0.29% increase (95% CI 0.07, 0.50%) in cardiovascular mortality. Comparatively, the effect estimates for the proportional increase in BC concentrations were seven to eight times larger than for $PM_{2.5}$. Moreover, a 1 $\mu g \, m^{-3}$ decrease in $PM_{2.5}$ would lead to an increase in life expectancy of 21 days per person, while a proportional decrease in BC concentration yields an increase between 3.1 and 4.5 months per person.

Assessing personal exposure to ultrafine particles (with diameter smaller than 100 nm, UFP) in terms of number concentration is also relevant due to the variety of emission sources and secondary particle formation processes in urban environments (Kumar et al. 2013, 2014). Just like BC particles, UFP can potentially penetrate the respiratory system and cause deleterious effects on human health (e.g., Shah et al. 2008). However, hand-held particle counters available on the market are bulkier and heavier that BC monitors, and most models use some type of alcohol to make particles grow into detectable sizes. These aspects can make mobile measurements challenging, particularly when it involves volunteers that are not familiar with monitoring and caution has to be taken to avoid tilting the particle counter due to the risk of flooding the optical chamber.

Traditionally, personal exposure to air pollutants is assessed with data from fixed monitoring sites (Steinle et al. 2013). However, because of the large spatial variability of airborne particles in a range of a few meters (Targino et al. 2016, 2018; Hankey and Marshall 2015), ambient concentrations measured at a fixed station can significantly weaken associations between health outcomes and exposure, especially in transport microenvironments or in certain urban settings, such as within urban canyons or close to highly trafficked roads. Personal exposure data collected by portable monitors show that most of the population is exposed to much higher levels of air pollutants than the average estimated by data from fixed monitoring sites (Jerrett et al. 2005). Placing air pollution samplers closer to individuals' homes may reduce the errors of exposure estimates; however, this strategy may not circumvent the under-representation of concentrations of traffic-related particles. Although most urban dwellers spend only between 6 and 10% of their daily time commuting (e.g., Dons et al. 2012 in a study conducted in Belgium; Fondelli et al. 2008 sampling in Florence, Italy), the high pollution levels within transport microenvironments may contribute to between 20 and 36% of the daily exposure to BC particles, depending on the transport mode (Williams and Knibbs 2016 in a exposure study in Brisbaine, Australia; Dons et al. 2012).

Thus, this paper assesses the personal exposure to BC of couples with different daily routines by using high-frequency, geo-referenced, continuous samples collected by personal monitoring (mobile), fixed at home (outdoor) and at a fixed station in a mid-sized city in Southern Brazil. In a literature search, we found that studies investigating differences in air pollution exposure by gender are scarce. Studies on personal exposure to BC using portable monitors have been conducted in several countries (e.g., Fruin et al. 2004; Dons et al. 2011; Hankey and Marshall 2015; Li et al. 2015; Pattinson et al. 2018), but, to the best of our knowledge, this screening study focusing on gender and using a combination of mobile and stationary measurements is pioneering in Brazil.

In relation to BC emission sources, Brazil presents a unique combination of characteristics, such as frequent events of long-range transported pollution during the biomass burning season (Oliveira et al. 2016), large emissions from open burning of domestic waste in urban areas (Wiedinmyer et al. 2014), and a fleet composed of quite old heavy-duty diesel vehicles (10-year average; SINDIPEÇAS 2017) running without diesel particle filters. Diesel passenger cars are not allowed in Brazil.

Specifically, we sought (i) to determine which microenvironments yielded the highest BC concentration, exposure, and dose; (ii) to assess the inter- and intra-couple's BC exposure variability, and to compare differences in exposure between genders with well-controlled socioeconomic factors; (iii) to quantify the discrepancy in exposure and dose between calculations carried out with data from personal and fixed monitoring, and whether home outdoor and fixed-site concentrations can be used as proxies for calculations of personal exposure; (iv) to pinpoint the commute mode that most affects the exposure and dose, and (v) to determine the spatial distribution of BC concentrations in the city by using the mobile measurements carried out by the volunteers while commuting.

Methods

Study area

The monitoring was performed in the city of Londrina, with a population of 554,000 inhabitants located in Southern Brazil (latitude 23° 22′ S; longitude 51° 10′ W; average altitude 585 m). Its climate is humid subtropical (Cfa in the Köppen-Geiger classification) with an annual mean temperature of 21.0 °C, annual mean precipitation of 1630 mm, abundant rainfall in summertime (December to February), and plenty of sunshine throughout the year (average of 2600 h) (Krecl et al. 2016).

Londrina has a fleet of 370,000 vehicles (DETRANPR 2016), of which 63% are passenger cars, 22% motorcycles, 11% light commercial vehicles, 3% trucks, and 1% buses. According to the local emissions inventory, 67% of the PM

is emitted by the industry and 33% by the road transportation sector (IAP 2013). Furthermore, the air quality in the city is affected by illegal burning of household waste, especially in the suburbs (Targino and Krecl 2016), and by the large-scale transport of pollutants emitted by biomass burnings (e.g., burning of *Cerrado* in central Brazil) (Krecl et al. 2016; Targino and Krecl 2016).

Instrumentation and data collection

The study comprised both mobile and fixed-site BC measurements carried out with hand-held and benchtop aethalometers (models AE51, AethLabs, USA and AE42, Magee Scientific, USA, respectively). Both the AE51 and the AE42 are optical instruments whose operating principle is based on the absorption of electromagnetic radiation by particles, according to the Beer-Lambert law. Particulates suspended in the air are carried through the sampling tube with a constant volumetric flow rate (Q) and deposited onto a filter spot of area A. A beam of electromagnetic radiation is transmitted through the filter and collected by a photodetector. The AE51 operates at only one wavelength ($\lambda = 880$ nm) while the AE42 operates at seven wavelengths ($\lambda = 370, 470, 525, 590, 660, 880$, and 950 nm).

The particles deposited on the filter absorb and attenuate the impinging radiation and the attenuation (ATN) is calculated by monitoring the intensity of the radiation transmitted through a blank filter spot (I_0) and through the filter spot onto which the particles accumulate (I):

$$\text{ATN} = \ln \frac{I_0(\lambda)}{I(\lambda)} \tag{1}$$

By monitoring the variation of the attenuation (ΔATN) in a time interval (Δt), the absorption coefficient (b_{abs}) can be calculated as:

$$b_{abs}(\lambda) = \frac{A}{Q} \frac{\Delta \text{ATN}(\lambda)}{\Delta t} \tag{2}$$

The method assumes that the relation between b_{abs} and the BC concentration is linear and that BC is the only absorbing material in the sample. The BC mass concentration (μg m^{-3}) is related to the absorption coefficient by the wavelength-dependent cross-sectional absorption coefficient σ_λ (m^2 g^{-1}) via the equation:

$$\text{BC}(\lambda) = \frac{b_{abs}}{\sigma_\lambda}, \tag{3}$$

and we used the σ_λ values provided by the manufacturers (12.5 m^2 g^{-1} for the AE51 at $\lambda = 880$ nm and 16.6 m^2 g^{-1} for the AE42 at $\lambda = 880$ nm). All BC concentrations reported in this study correspond to $\lambda = 880$ nm.

Study design

The mobile data collection was performed with six non-smoking couples, with ages between 20 and 50 years, living in different areas of the city and not working in the transport sector. As a criterion, each couple should reside in the same home, but each member should work in different places. This strategy allowed us to control for housing characteristics and, thus, to evaluate the effect of differences in activity patterns on personal exposures when studying intra-couple variations. Participants were recruited based on personal contacts and no financial compensation was offered. Table 1 shows some of the couples' personal data (gender, age, and occupation), housing features, and sampling periods (selected days in the period August 09–December 09, 2015). Fixed measurements were conducted at the campus of the Federal University of Technology (UTF) and at the volunteers' homes sampling outdoor air (Fig. 1).

The mobile monitoring was performed for 48 h during which each volunteer continuously carried an AE51 and a GPS logger (D-100, Globalsat, Taiwan) to determine their geographical position across the city. Participants were asked to wear the sampler with them at all times with the inlet positioned between the shoulder and waist. Even though the AE51 monitors are quite silent, we instructed volunteers to leave them in the living room while at home to avoid disturbances during their sleep. According to the household layouts, any combustion in the kitchen would affect the bedroom and living room in a similar fashion, why we consider that the nocturnal measurements are representative of the bedroom.

Since both personal samplers were placed next to each other in the living room at night, the data were used for inter-comparison checks and to help validate the experimental data.

The participants made notes of their time-activity with paper and pencil throughout the sampling period, detailing their activities (such as commuting, cooking, and at work) and peculiar events (e.g., close proximity to any smokers, and presence of heavy traffic) and for how long. This information was used for subsequent linkage with the BC time series. Couples were monitored during 48 h to reduce interferences in their daily routine, since some studies conducted during longer sampling periods reported dissatisfaction of some volunteers (e.g., Dons et al. 2012), which affected the quality of the data. For each couple, another AE51 unit was installed at the home and operated during the same period. The unit was housed indoors with a 0.5-m conductive sampling tube going to the outdoors through a window opening. The only source of indoor combustion in the homes was gas stoves.

The AE51 units were configured to operate with a flow rate of 100 ml min^{-1} and time resolution of 1 min (mobile monitoring) and a flow rate of 50 ml min^{-1} and time resolution of 1 min (fixed monitoring). Before each data collection session with the couples, time synchronization and flow calibration

Table 1 Experimental details related to the volunteers. Traffic rate is expressed as total number of vehicles, and the rate of heavy-duty diesel vehicles are shown in square brackets

Couple	Gender	Age	Occupation	Home floor	Traffic rate[a] (vehicle h^{-1})	Sampling period
1	Female	26	Graduate student	6	302[b]	August 09 to 11, 2015
	Male	27	Office clerk		[2]	
2	Female	23	Laboratory technician	9	2757[b]	August 30 to September 01, 2015
	Male	22	Undergraduate student		[77]	
3	Female	23	Business consultant	1	1213[b]	September 16 to 18, 2015
	Male	29	Teacher		[36]	
4	Female	48	Teacher	House	13[c]	September 28 to 30, 2015
	Male	50	Engineer		[2]	
5	Female	27	Nurse	9	2402[b]	November 04 to 06, 2015
	Male	30	HR clerk		[30]	
6	Female	47	University lecturer	5	981[b]	December 07 to 09, 2015
	Male	41	University lecturer		[11]	

[a] Mean traffic rate in front of the participants' home at rush hours (08:00–09:00; 17:00–18:00)

[b] AE51 installed at home facing the street

[c] AE51 installed at home away from traffic

were carried out, and a new filter strip was installed at the end of the first day of sampling to minimize the effect of filter loading (Virkkula et al. 2007), which could affect the linearity of Eq. 3. Download of GPS data and battery changes were carried out daily by the researchers when the participants were at home.

The benchtop AE42 was operated at the UTF campus with a 2.5-μm cyclone fitted to the inlet, a flow rate of 5 L min^{-1}, and time resolution of 2 min. The campus is located 5 km away from the city center in a sparsely populated suburban area, with the closest neighborhood about 400 m away. The road that gives access to the campus and to a residential cluster development has a reduced traffic rate (2700 vehicles day^{-1}, of which 8.4% are heavy-duty vehicles) (Targino and Krecl 2016). All BC monitors were intercompared for 18 h in

ambient air with the same flow configuration and sampling frequency used in the fieldwork, showing excellent reproducibility and a high linear correlation (coefficient of determination $R^2 = 0.99$).

To assess the influence of the positioning of the sampling tube on the BC concentrations, we performed a test with a volunteer who carried two AE51 simultaneously during 24 h. One inlet was placed within the breathing zone (that is, 30 cm from the nose and the mouth) and the other inlet in the region between the shoulder and the waist, depending on the activity and convenience.

Local traffic emissions can have a large influence on BC concentrations measured close to busy urban roads, with concentrations particularly high on roads with a large fraction of diesel-powered vehicles (e.g., Krecl et al. 2014, 2016; Targino

Fig. 1 Location of the monitoring sites: couples' residences and UTF site
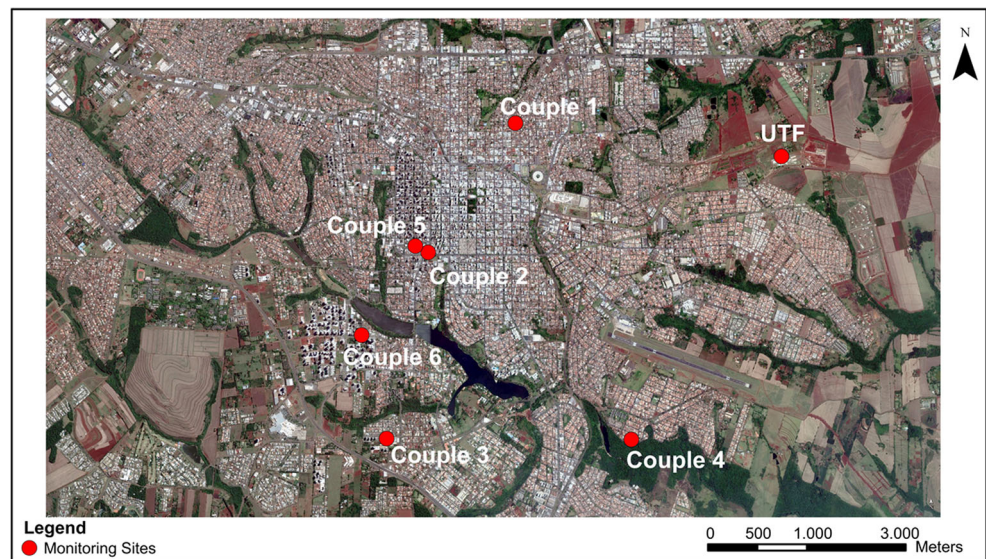
et al. 2016). Due to the lack of official information on traffic rates, we counted vehicles at rush hours (between 08:00 and 09:00, and between 17:00 and 18:00) on weekdays using the method described in Targino et al. (2016). In short, vehicles were manually counted and split into four categories (cars, motorcycles, buses, and trucks) during two 15-min intervals with alternating 15-min pauses. Hourly traffic rates were computed as twice the counts observed in the two 15-min intervals. Due to limited manpower for manual counting in this study, traffic rates were only reported at points in front of the participants' homes.

Data analysis

To calculate the personal exposure and dose, the volunteers' activities were divided into four categories that represented different microenvironments: in transport, at work, at home, and others (e.g., shopping, gym). Following Monn (2001), we calculated the time-averaged personal exposure for each microenvironment j ($\overline{E}_j$) as the mean value of the BC concentrations measured with the AE51 monitor when the volunteer was in microenvironment j. The average inhaled dose for each microenvironment j ($\overline{D}_j$) was evaluated as:

$$\overline{D}_j \left[\mu g \ min^{-1} \right] = \overline{E}_j . V_{j,k}, \tag{4}$$

where $V_{k,j}$ is the inhalation rate for the activity k in the microenvironment j (Monn 2001). The inhalation rates depend on the age of the exposed person, gender, activity, and specific exertion level within each activity and were taken from the work by Allan and Richardson (1998).

The spatial analysis of BC concentrations for the mobile data was performed with tools of geographic information system (GIS) and only considered measurements in the transport category. We defined 100-m polygons along the sampled streets and all data points that fell into each individual polygon were used to calculate an aggregated median BC concentration. This method has been used by several authors (Targino et al. 2016, 2018; Brantley et al. 2015) as a means of reducing the influence of sporadic extreme concentrations caused by emissions from vehicle exhaust.

Since the BC monitoring was not performed simultaneously for the six couples (12 days sampled between August and December 2015), some measurements may have been influenced by local and/or regional air pollution events (such as urban fires and long-range transport of biomass burning). To account for changing background conditions and to highlight the spatial variability due to traffic contributions, a temporal adjustment was applied to the mobile measurements for the transport category. We applied additive or multiplicative corrections (Dons et al. 2012; Krecl et al. 2014), according to the relation between the BC data collected with mobile samplers

during transit and those sampled at a fixed reference location (in our case, the UTF site). The additive correction (Krecl et al. 2014) was used when the mobile concentration on a particular day and time was higher than concurrent concentrations at the UTF site, which means that local emission sources were strong and influenced the in-transport measurements. When the mobile concentrations on a particular day and time were lower than concurrent concentrations at the UTF site, we used a multiplicative correction (Krecl et al. 2014) to prevent the temporally adjusted BC concentrations to become negative. In this case, we assumed that there were no local sources of BC affecting the in-transport measurements, and the mobile measurement was corrected for the variation observed at the reference site.

Results and discussion

Breathing zone vs. waist

Table 2 summarizes the descriptive statistics of the BC data collected for 24 h simultaneously with the inlets in the waist and in the breathing zone. The largest BC concentration was measured by the AE51 unit whose inlet was placed close to the waist (26.68 μg m^{-3}), while the one placed within the breathing zone recorded a maximum concentration of 20.61 μg m^{-3}. The other statistical indicators, such as mean, median, and even the 95th percentile, showed very similar values between the datasets. This indicates that the instruments were likely to report discrepant values for extreme sporadic concentrations larger than the 95th percentile. The Mann-Whitney test indicated that there were no statistically significant differences (95% confidence level) between the two time series and, thus, placing the inlet within the breathing zone or close to the waist will not cause substantial differences in the results of personal exposure.

Concentration, personal exposure, and dose

In our study, the volunteers spent on average 67% of their time at home, 23% at work, 7% in transport, and 3% conducting other activities (shopping, gym, visiting friends, and family, etc.). Boxplots of BC concentrations measured with personal and fixed-site monitoring are shown in Fig. 2. The concentrations varied substantially per microenvironment, with highest levels and variability in transport (mean of 4.01 μg m^{-3} and STD of 5.35 μg m^{-3}). Conversely, mean concentrations at work were lower and showed less variability (mean of 1.42 μg m^{-3} and STD of 1.45 μg m^{-3}). On average, outdoor concentrations sampled at home were higher (mean of 1.43 μg m^{-3}) than those observed at the UTF site (mean of 1.16 μg m^{-3}). This feature can be attributed to the fact that most of the couples' homes were located in the vicinity of

Table 2 Statistical summary of BC concentrations measured with two AE51 units

BC conc. ($\mu g\ m^{-3}$)	Breathing zone	Waist
Minimum	0.01	0.01
Maximum	20.61	26.68
Mean	1.19	1.22
Median	0.83	0.85
Standard deviation	1.71	1.92
5th percentile	0.10	0.21
25th percentile	0.46	0.58
75th percentile	1.31	1.21
95th percentile	3.18	3.56

busy avenues and streets (Table 1, Fig. 1), while the UTF was located in a less trafficked area. Mean BC concentrations were very similar indoor (at home) and outdoor (home out) (Fig. 2), but note that indoor sampling only occurred when the volunteers were at home, generally in the evening/night. We calculated infiltration ratios for these coincident periods and found values between 0.82 and 0.93 (average of 0.87). These high infiltration ratios were caused by the windows frequently left open even at night due to the warm climate. Thus, in trafficked areas, the contribution of outdoor sources to BC indoor concentrations can be substantial.

We observed a great inter- and intra-couple variability in BC exposure and dose (Fig. 3). The average personal exposure between partners varied up to 55% (couple 4) and the difference was up to 3.4-fold for individuals not living in the same residence (1.17 vs. 3.94 $\mu g\ m^{-3}$). These differences are related to the type of activities developed during the sampling period and the time spent within each microenvironment, showing that even individuals living in the same home may experience different dose and exposure values. For all cases, the transport category contributed the most to the exposure (average of 4.73 $\mu g\ m^{-3}$) and dose

(average of 0.05 $\mu g\ min^{-1}$) and displayed a great heterogeneity among the sampled individuals.

Couple 2 was the most exposed and with the highest inhaled dose, while couple 5 was the least exposed and with the lowest inhaled dose. In the home category, couple 2 also stood out with average exposures of 2.74 to 2.46 $\mu g\ m^{-3}$ for the man and woman, respectively. This couple lives in Londrina's city center, next to roads with high vehicular flow (Table 1). Despite living in the ninth floor, the windows in the apartment were usually kept open, facilitating the infiltration of outside air.

The volunteers that experienced the largest BC concentrations in the workplace were the woman of couple 6 and the man of couple 2. Both volunteers worked at UTF campus, and during the sampling period fire foci were reported next to the campus, which contributed to the enhancement of BC concentrations. The average exposure for the man in this category was 1.38 $\mu g\ m^{-3}$ (10% of his total average exposure) and 0.99 $\mu g\ m^{-3}$ for the woman (16% of her average total exposure).

In the data sampled within "other" environments, the woman of couple 1 presented the highest fraction to the total daily exposure (28%), which was greatly influenced by a trip to the supermarket. Dons et al. (2014) also showed that the average exposure concentrations were relatively high for social and leisure activities (2.45 $\mu g\ m^{-3}$), exceeding the average of the concentrations found in our study (2.13 $\mu g\ m^{-3}$).

The highest exposure and dose in the transport category occurred with the woman of couple 6 (5.61 $\mu g\ m^{-3}$ and 0.07 $\mu g\ min^{-1}$, respectively), which corresponds to 66% of her exposure and 68% of her dose. She made an intercity trip and spent about 5 hours of her day in motorized transport. The time spent in the transport microenvironment accounted for 42% of the man's exposure (2.40 $\mu g\ m^{-3}$) and 27% of his dose (0.03 $\mu g\ min^{-1}$). A similar study conducted by Dons et al. (2011) in Belgium found that the time spent in or near transport may result in large dissimilarity in personal exposure

Fig. 2 Boxplot of BC concentrations for personal (all volunteers) and fixed-site monitoring. Represented are the 5th, 25th, 50th, 75th, and 95th percentiles, and mean (x) values. Note that "At home" corresponds to indoor sampling at night, and "Home (outdoor)" to external air sampling during 48 h
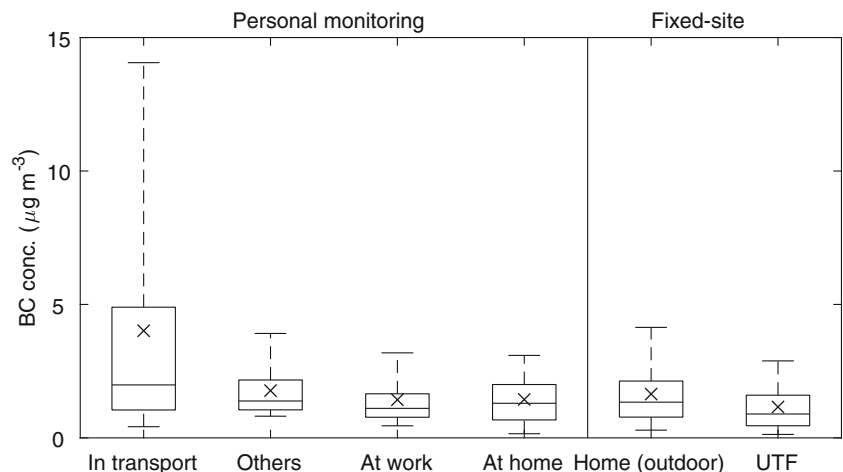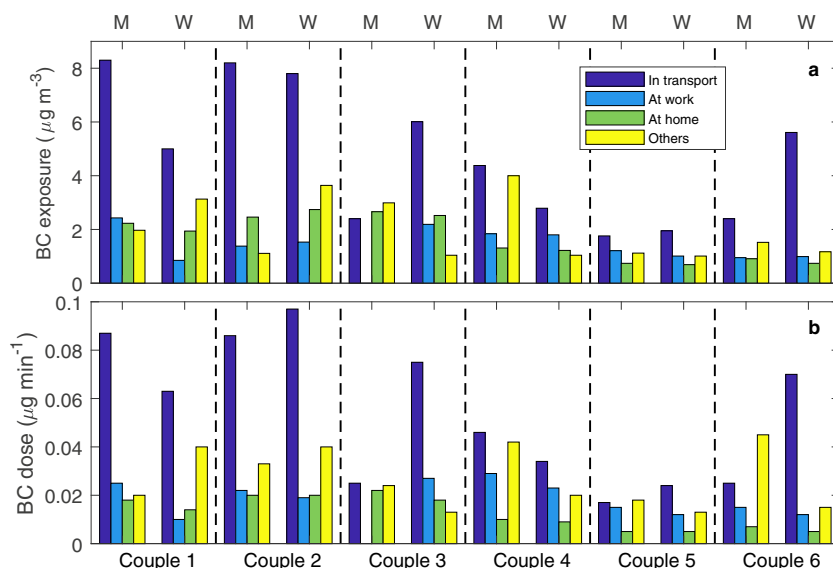
Fig. 3 BC exposure and dose per volunteer and microenvironment. *M* man, *W* woman

between two people who live in the same home, reaching a difference of up to 30%.

The overall average exposures for men and women were 2.54 and 2.43 µg m^{-3}, respectively, but this difference was not statistically significant (Mann-Whitney test at 95% confidence level), not even when considering the average exposure for each microenvironment. In relation to the inhalation dose, the same average value was observed for men and women when considering all microenvironments together (0.028 µg min^{-1}) and the differences were not significant when comparing mean and women average doses for each microenvironment.

Comparison between fixed and mobile monitoring

To illustrate the discrepancy in average personal exposure when using concentrations gathered with portable monitors and measured at fixed sites, we selected the time series recorded by couple 2 (mobile and home outdoor) and at the UTF site (Fig. 4). Couple 2 lives in the vicinity of highly trafficked roads and commute by bus to reach their workplaces. The time series for the portable samplers show a large temporal variability, responding to the transit across the different microenvironments. For both the man and the woman, the spikes in BC concentrations appeared when they were commuting. The concentrations increased rapidly and were persistent for the duration of the commutes, reaching values as high as 76.7 µg m^{-3} at about 07:00 h on September 01, when the man was at the bus terminal. This is a busy commuter terminal served by 62 diesel bus lines, corresponding to about 2260 buses day^{-1} on weekdays. Targino et al. (2018) surveyed the BC concentrations during bus commutes in Londrina and also recorded the largest concentration at this terminal.

We observed that the concentrations at the fixed monitoring stations varied very little throughout the day (interquartile ranges 1.60 µg m^{-3} for the home outdoor and 1.06 µg m^{-3}

for the UTF), unlike the mobile concentrations measured by the couple. The concentrations at the UTF campus were especially low (95th percentile of 3.53 µg m^{-3}). As mentioned before, the campus is located in a sparsely populated area with low traffic rate. This reinforces that data from fixed sites cannot capture the fine variability of BC concentrations, especially within transport microenvironments or caused by sporadic events and are not, therefore, accurate for assessing personal exposure.

The average concentrations of couple 2 at work and during commutes were 1.53 and 7.82 µg m^{-3} for the man and 1.37 and 8.25 µg m^{-3} for the woman, respectively. The values found during commuting were higher than those reported by Dons et al. (2012), who found mean concentration of 1.07 µg m^{-3} at work and 5.13 µg m^{-3} in transit modes in Flanders (Belgium), but agree with the value observed on urban buses in Shanghai (China), with mean BC concentration of 7.28 µg m^{-3} (Li et al. 2015). However, de Nazelle et al. (2017) argued that comparing exposure values during passive commutes is not straightforward due to particularities in the characteristics of the fleet (such as the type of fuel, fleet technology, and ventilation settings) which causes discrepancies across different studies.

We calculated R^2 between BC concentrations from the mobile and fixed measurements, using 10-min and hourly averaged data, for the 48-h sampling period and the nighttime hours (20:00–06:00 when both volunteers were at home). As expected, 1-h correlations were higher than 10-min correlations since short-averaging times are connected to large variability in local emissions and/or local meteorological conditions. Regardless of the chosen averaging time (10 min or 1 h) and datasets length (48 h or nighttime), no correlation was observed between (i) the two fixed sites (home outdoor and UTF), and (ii) BC personal data and concentrations at UTF. Rivas et al. (2015) reported low correlation ($R^2 = 0.28$)

Fig. 4 BC time series for personal monitoring of couple 2 (man and woman, 1 min), home outdoor (1 min), and UTF site (2 min). Dashed lines separate periods when volunteers are at home (night)
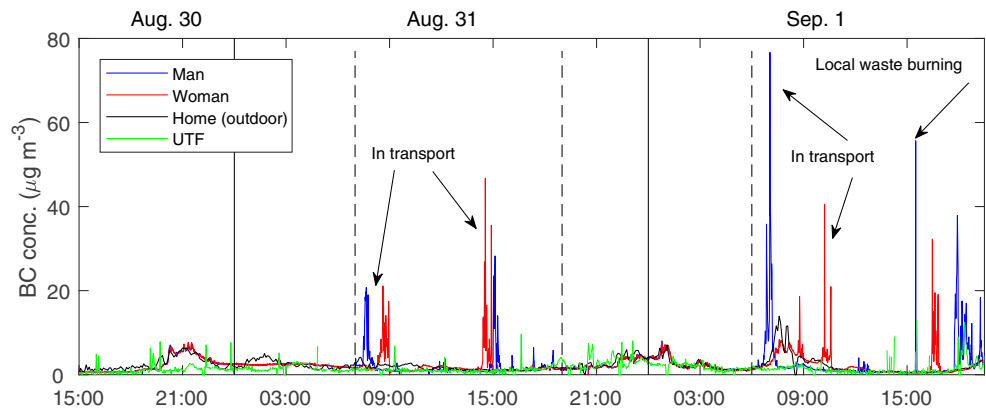
between indoor and outdoor BC data when the individuals were near a monitoring station in Barcelona and an even lower correlation ($R^2 = 0.18$) when the distance increased. In our study, the correlation increased slightly ($R^2 = 0.30$) when comparing woman and home outdoor datasets over the 48-h experiment and was higher than the correlation man vs. home outdoor, since he spent a shorter time at home.

Comparison between the couple's BC home indoor concentrations and the home outdoor concentrations collected at night presented moderate correlations ($R^2 = 0.71$–0.72 for 10-min data, $R^2 = 0.77$ for 1-h data). This increase in correlation at night might be explained by the fact that all AE51 instruments were collocated at home, with indoor instruments partially sampling outside air in certain time periods when the windows remained open due to the warm weather. In a study performed in Boston (USA) by Brown et al. (2008), the authors assessed whether data of outdoor and indoor environments can be used to better estimate the personal exposure. For elemental carbon in summertime, they found a low correlation between personal concentrations and home outdoor ($R^2 = 0.22$) and no correlation with a fixed station located in the city center.

In this study, the linear correlation was highest between the two A51 units placed side by side in the living room at night (Table 3), indicating an excellent agreement between them and, thus, validating our measurements. This finding contrasts with the poor correlation found between BC concentrations measured by the woman and the man of couple 2 during the entire sampling period ($R^2 < 0.15$), hence indicating the large influence of the time-active patterns on the measurements.

The personal exposure and average dose for couple 2 were calculated using data of the mobile monitoring, the home outdoor, and UTF concentrations. The calculations using home outdoor and UTF data underestimated the exposure for all microenvironment categories. However, the largest divergences (between four- and nine-fold) were found in the transport category (Table 4). These results agree with those of other studies (Avery et al. 2010; Briggs et al. 2000) who attributed the exposure in transport microenvironments to be the most under-represented when using data from fixed monitors, since

people travel across places with high concentrations that are seldon detected by fixed equipment.

Spatial analysis in transport

For the spatial analysis during transportation, we considered BC concentrations measured by the volunteers during motorized commutes within the perimeter of the municipality of Londrina only, and excluded data from intercity trips. Following Dons et al. (2012), we corrected 1964 BC values, 88% using the additive method and 12% with the multiplicative approach. The Mann-Whitney test applied to the raw and corrected BC data (95% confidence level) showed that there were statistically significant differences between the time series (median of 2.18 and 1.98 $\mu g\ m^{-3}$, respectively). Therefore, the analysis was conducted with the corrected BC dataset.

The largest concentrations were found in the busiest areas of the city: Juscelino Kubitschek (JK) avenue, Celso Garcia Cid (CGC) avenue, Higienópolis (Hig.) avenue, Sergipe

Table 3 Coefficient of determination between fixed and mobile measurements for couple 2 considering 48 h and only nighttime data (20:00–06:00), calculated using 10-min and 1-h mean BC concentrations

Datasets	Period	R^2 (10 min)	R^2 (1 h)
Woman and UTF	48 h	0.01	0.03
	Night	< 0.01	0.02
Man and UTF	48 h	0.08	0.16
	Night	< 0.01	0.02
UTF and home outdoor	48 h	0.05	0.12
	Night	0.01	0.06
Woman and home outdoor	48 h	0.30	0.45
	Night	0.71	0.77
Man and home outdoor	48 h	0.08	0.41
	Night	0.72	0.77
Man and woman	48 h	0.02	0.14
	Night	1.00	1.00

Table 4 Average personal exposure and inhaled dose per category for couple 2 calculated using different datasets

		Woman			Man		
	Category	Mobile	Home outdoor	UTF	Mobile	Home outdoor	UTF
Exposure ($\mu g\ m^{-3}$)	House	2.74	2.41	1.59	2.45	2.42	1.68
	Work	1.53	1.35	0.92	1.38	1.83	1.02
	Transport	7.82	1.55	0.85	8.25	2.27	1.24
	Others	4.54	3.55	1.67	1.11	1.19	0.81
Dose ($\mu g\ min^{-1}$)	House	0.020	0.018	0.012	0.020	0.020	0.013
	Work	0.019	0.017	0.012	0.022	0.029	0.012
	Transport	0.097	0.019	0.010	0.086	0.023	0.013
	Others	0.059	0.046	0.021	0.033	0.035	0.024

street, and the commuter's terminal (CT) (Fig. 5). These roads are hotspots of air pollutants and were also identified in the study by Targino et al. (2016). They analyzed the spatio-temporal variability of BC concentrations in Londrina's city center and reported high BC concentrations on Higienópolis and JK avenues and at the commuter's terminal, which were strongly correlated with the number of heavy-duty diesel vehicles. For example, Higienópolis Ave. has a traffic rate of 25,000 vehicles day^{-1} on weekdays, of which 2.4% are heavy-duty vehicles (Targino and Krecl 2016). The mean BC concentrations measured at the terminal were 10.24 and 20.14 $\mu g\ m^{-3}$, in the morning and in the afternoon, respectively, which are similar to BC concentrations reported by Targino et al. (2016) (6.53 and 21.50 $\mu g\ m^{-3}$) at the same terminal. The large number of diesel-fueled buses serving Londrina commuter's terminal combined with poor ventilation settings contributed to increase the air pollution levels.

Analysis according to the transport mode

In our study, the couples spent around 7% of their time commuting, with the following share per transport mode: seven volunteers used cars, two used buses, one travelled by bus and car, one walked, and one combined bus, car, and on foot. Figure 6 displays the boxplots of BC concentrations for trips by car, bus, and on foot for all volunteers together, with mean values of 3.58, 5.80, and 5.34 $\mu g\ m^{-3}$, respectively. The highest BC concentrations were found for bus trips (95th percentile of 17.52 and maximum of 54.16 $\mu g\ m^{-3}$) matching the results reported by Targino et al. (2018) in a previous study comparing personal exposure on passive and active transport modes in Londrina.

Bus passengers and pedestrians presented higher average inhaled doses (0.080 and 0.075 $\mu g\ min^{-1}$, respectively) than car commuters (0.041 $\mu g\ min^{-1}$). Note that pedestrians can have their personal dose aggravated due to the higher inhalation rates when compared to passive commuters, as reported by Targino et al. (2018), Moreno et al. (2015), and Li et al. (2015).

The woman of couple 6 was the only one who used the three transport modes, with the highest exposure when she commuted on foot (5.53 $\mu g\ m^{-3}$). She also reported the highest total inhaled dose (0.13 $\mu g\ min^{-1}$). Proximity to the traffic and high concentrations may have significantly contributed to her exposure and dose when walking in the city center, noting that the walking time was shorter than the time she traveled by bus (16 vs. 323 min).

Couple 5 presented average exposures of 1.95 $\mu g\ m^{-3}$ for the woman (who only commuted by car) and 1.76 $\mu g\ m^{-3}$ for the man (who only commuted on foot). Conversely, the average dose was lower for the woman (0.01 $\mu g\ min^{-1}$) than for the man (0.02 $\mu g\ min^{-1}$). These results compare favorably with the findings by Dons et al. (2012) in Belgium, where exposure in motorized transportation was higher than that on foot (6.3 and 3.3 $\mu g\ m^{-3}$, respectively) and the highest dose corresponded to pedestrians.

Besides transport mode and travel time, the route characteristics (total traffic rate, share of heavy-duty diesel vehicles, and street configuration) can largely influence the personal exposure in transportation. For example, less trafficked routes can significantly reduce BC concentrations as shown by Hankey and Marshall (2015) in a study in Minneapolis (USA), with a 20% average decrease by moving one block away from major roads to adjacent local roads. Figure 5 shows clearly that the highest concentrations were found in heavy-trafficked streets in the city center, resulting in higher personal exposures than if volunteers had opted for routes with lower vehicle rates.

In the case of passive modes, we can also add other factors affecting BC concentrations inside the cabin, such as vehicle technology, fuel quality, and ventilation rates. Personal exposure of bus passengers to PM depends on the bus characteristics and may vary significantly from country to country. For example, couple 2 presented relatively high average exposure (8.25 $\mu g\ m^{-3}$ for the man and 7.82 $\mu g\ m^{-3}$ for the woman) when travelling by bus in Londrina, where the bus fleet runs on diesel, is largely dominated by EURO-III equivalent vehicles (72%), and circulates with open windows most of the

Fig. 5 Spatial distribution of adjusted BC concentrations during motorized commute for all volunteers together
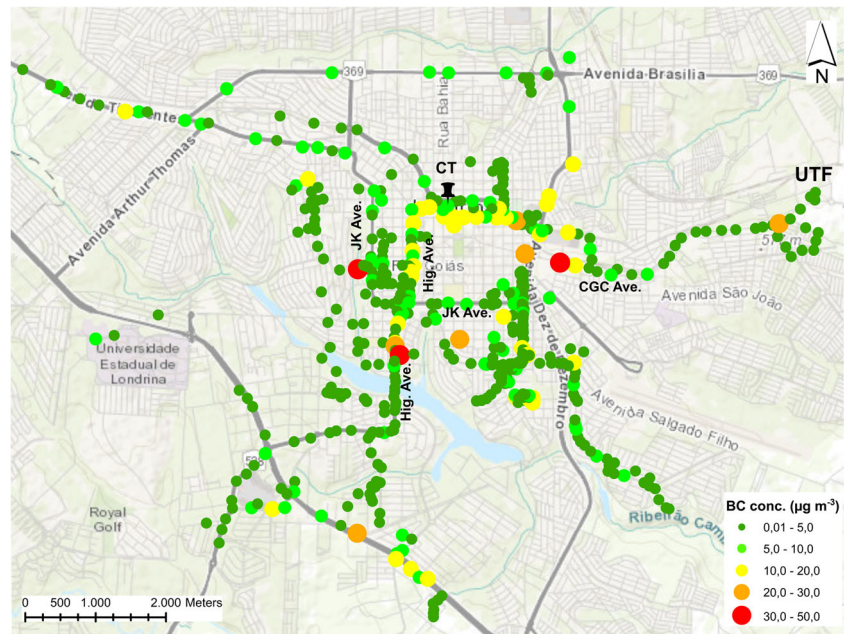
time. In Dublin (Ireland), public buses also run on diesel, and personal exposure to $PM_{2.5}$ was reported highest on buses when compared to on foot, car, and bicycle commutes along two fixed routes (McNabola et al. 2008). On the other hand, a study conducted in Barcelona (Spain)—where the bus fleet consists of hybrid buses that use natural gas and electric power—reported inhaled BC doses 50% higher for car commuters compared to bus passengers when travelling on the same prescribed routes (de Nazelle et al. 2012).

A high ventilation rate allows external pollutants to enter the cabin (Zuurbier et al. 2010) and, thus, increases personal exposure. In our study, only couple 6 made notes about the car's windows being open or closed. The woman had average exposures of 5.78 μg m^{-3} (window open) and 2.67 μg m^{-3} (window closed), whereas the man was exposed to lower levels (3.91 μg m^{-3} open and 0.90 μg m^{-3} closed). The dose

was up to four-fold higher when the vehicle windows were open, collecting outside air. Li et al. (2015), in a study conducted in Shanghai (China), proved that BC concentrations were higher when bus windows were open, verifying BC concentrations nearly three-fold higher than the level measured on the streets. Williams and Knibbs (2016) carried out a study in Brisbane (Australia), reporting that in-cabin BC concentrations were 2.6-fold higher when the windows were open than when they were closed. Another work by Quiros et al. (2013) verified that exposure to ultrafine particles was ~2-fold higher while repeatedly driving a car with open windows on an urban residential street in Santa Monica (USA).

Conclusions

The results showed a great variability in BC concentrations, exposure, and dose among the 12 volunteers. Personal monitoring indicated that exposure may be different (up to 55%) for couples living in the same home, but working in different locations. This variability is related to the type of activities performed and the time spent within each microenvironment, being transport the category that contributed the most to the exposure and dose. We did not observe any statistically significant differences in average exposure and dose between genders.

Ambient BC concentrations measured at home and the UTF campus disagreed with the values simultaneously monitored with portable equipment. The calculated exposure and dose were higher when using personal monitoring data
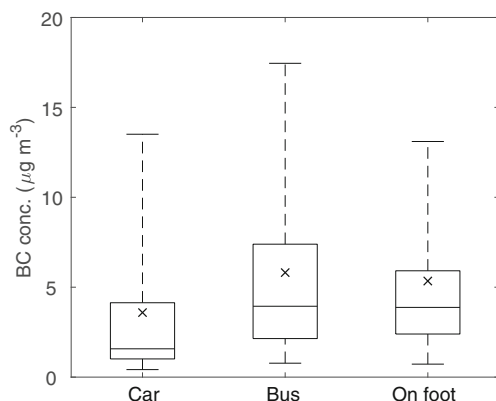

Fig. 6 Boxplot of BC concentrations per commuter mode. Represented are the 5th, 25th, 50th, 75th, and 95th percentiles, and mean (x) values

compared to (proxy) fixed-site data (either home outdoor or UTF datasets). Moreover, concentrations measured at fixed sites were not correlated with the personal monitoring during the day, since BC featured a large spatial variability. Therefore, the assessment of personal exposure by proxy data is not recommended, particularly during commuting.

In the transport category, we observed that the average exposure was higher when the volunteers commuted by bus and lower when travelling by car. In the on foot category, proximity to traffic contributed to relatively high values of exposure and the dose was highest because of the increased breathing rate for this active mode. Exposure during transit strongly depended on the route taken and the travel time. In addition to these factors, we verified that concentrations inside the car were higher when the windows were open, making the average exposure to be up to three times higher and the dose up to four times higher, when compared with the windows closed.

This screening study highlights the importance of assessing personal exposure to BC with mobile monitoring in a medium-sized city (100,000–550,000 inhabitants), particularly during transport. In Brazil, cities this size are abundant (267 cities) and account for 27% of the total population (~55.5 million of inhabitants). Hence, our findings could be valid in this large group of cities because of similar activity patterns, vehicle fleet share, fuel composition, and urban design. However, this pioneering study should encourage the undertaking of urban measurements considering larger group of volunteers to accurately calculate the personal exposure to BC.

The methodology could be improved by continuously monitoring outdoor and indoor concentrations at home, including couples living in suburban and rural areas, and volunteers that use bicycle as transport mode. In large study areas, only one station could not be representative of background conditions to temporally adjust the BC concentrations during transport and more fixed stations should be required. Finally, personal monitoring for longer time periods (over 1 week) should be considered when possible or, alternatively, conducting a second sampling campaign in a different season to have a better representativeness of variations in meteorological conditions and in emission sources along the year.

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