

Spatiotemporally resolved black carbon concentration, schoolchildren's exposure and dose in Barcelona

Abstract At city level, personal monitoring is the best way to assess people's exposure. However, it is usually estimated from a few monitoring stations. Our aim was to determine the exposure to black carbon (BC) and BC dose for 45 schoolchildren with portable microaethalometers and to evaluate the relationship between personal monitoring and fixed stations at schools (indoor and outdoor) and in an urban background (UB) site. Personal BC concentrations were 20% higher than in fixed stations at schools. Linear mixed-effect models showed low R^2 between personal measurements and fixed stations at schools ($R^2 \leq 0.28$), increasing to $R^2 \geq 0.70$ if considering only periods when children were at schools. For the UB station, the respective R^2 were 0.18 and 0.45, indicating the importance of the distance to the monitoring station when assessing exposure. During the warm season, the fixed stations agreed better with personal measurements than during the cold one. Children spent 6% of their time on commuting but received 20% of their daily BC dose, due to co-occurrence with road traffic rush hours and the close proximity to the source. Children received 37% of their daily-integrated BC dose at school. Indoor environments (classroom and home) were responsible for the 56% BC dose.

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Practical Implications

This study provides valuable information on the BC dose that schoolchildren receive during weekdays. Owing to the time spent in indoor microenvironments (considering classroom and home, 82%), children receive around half of their BC dose (56%) there. School (classroom and playground) contributes to a third of schoolchildren's daily dose. However, the highest dose:time intensity (3.5:1) is found during commuting activities. Policies focusing on reducing traffic intensities around schools should be enhanced.

Introduction

Many epidemiological studies demonstrate that exposure to atmospheric pollutants, specially particulate matter (PM), has important and varied adverse effects on human health (Beelen et al., 2014; Raaschou-Nielsen et al., 2013; WHO, 2013).

Human exposure was defined by Ott (1982) as ‘the event when a person comes into contact with a pollutant of a certain concentration during a certain period of time’. Traditionally, urban population exposure to air pollutants has been assessed based on data from air quality monitoring sites, which usually provide data of a wide variety of pollutants, albeit for few points in a city (Steinle et al., 2013) that might not be representative for all the population. Although outdoor air pollution estimates have been associated with health, a more refined exposure assessment may be needed to reduce exposure misclassification and find stronger associations with health outcomes. Therefore, for an accurate personal exposure assessment the different places in which time is spent (Ashmore and Dimitroulopoulou, 2009) should be considered, as, in fact, people spend around 90% of their time indoors (Buonanno et al., 2012; US-EPA, 2008). Therefore, direct personal exposure measurements are the most representative of people’s exposure (Jantunen et al., 2002). However, these personal assessments raise new uncertainties such as how many personal measurements might be taken to be representative of a population or a conurbation, how should be the subjects distributed within city to control for spatial variability or which personal characteristics need to be controlled in order to characterize all existent time–activity patterns within a population. Finally, personal monitoring also puts a burden on people and is labor and resource intensive.

It is important to investigate the relationship between personal exposure and outdoor background concentrations from fixed monitored stations. Previous studies evaluating this relationship for PM_{2.5} came to different conclusions: some of them finding relatively low correlation between exposure from personal and fixed air quality monitoring station (Adgate et al., 2002; Borgini et al., 2011; Brown et al., 2008; Crist et al., 2008) and some others showing high correlations (Janssen et al., 1999; Montagne et al., 2014b). Although Montagne et al. (2014b) found generally high correlation between temporal variation of the outdoor concentration and personal monitoring for different PM_{2.5} components, Montagne et al. (2014a) found that the Land-Use Regression Models did not predict the spatial variation of the same components. Hence, using only the outdoor air component of exposure might not be enough to characterize human exposure to air pollutants (Steinle et al., 2013).

Adults have been the main target of personal monitoring, but less is known about children’s personal

exposure (Borgini et al., 2011; Buonanno et al., 2013; Crist et al., 2008; Van Roosbroeck et al., 2007). Children are more susceptible than young adults to air pollutants (Kulkarni and Grigg, 2008); therefore, their exposure is a major health concern (Mejía et al., 2011). Differences between adults and children can be summarized in the day-to-day activities which are in conjunction with being in different microenvironments (ME) depending on the activity, and the substantial difference in breathing heights that makes children closer to some pollution sources such as road traffic (HEI Panel on the Health Effects of Traffic-Related Air Pollution, 2010; Reche et al., 2011).

Black carbon (BC) personal measurement studies (Adams et al., 2002; Buonanno et al., 2013; Dons et al., 2011, 2012, 2013) are scarce in the literature, even though it is a good tracer of traffic emissions (especially from diesel engines), a source of major concern in urban environments. Although BC may not be toxic itself, it may have an indirect key role in toxicity as it is supposed to operate as a universal carrier of a wide variety of chemical components, such as semi-volatile organics and other compounds co-released in combustion processes (WHO, 2012). Therefore, a reduction in the exposure to BC should lead to a reduction in negative health outcomes derived from these toxic constituents.

In addition, Morawska et al. (2013) identified two important gaps in literature which are covered in the present work: the relationship between ambient concentration and personal monitoring, and the contribution of school exposure to a child’s daily exposure and dose with respect to other MEs.

Framed within the ERC-Advanced Grant (FP7) BREATHE Project, this work’s aim was to evaluate (i) the relationship between personal BC exposure of school children assessed by portable microaethalometers and (a) school BC concentrations, obtained by the same instrument at an indoor and outdoor fixed stations at the schools and (b) ambient BC concentration in a reference Urban Background (UB) site; and (ii) the daily-integrated exposure and dose of schoolchildren in Barcelona. In a previous study, we evaluated the relationship between spatial models and personal exposure (Nieuwenhuijsen et al., 2015), while the focus here is on in-depth analyses of the commuting data, and spatiotemporal relationships between ambient and personal BC and dose.

Materials and methods

Instrumentation and calibration

Both personal and fixed stations at schools (indoor and outdoor) were monitored using a MicroAeth AE51 (AethLabs, USA). It is a small (117 × 66 × 38 mm), light (0.28 kg), and battery-powered device which

can be easily carried in a belt bag, reducing wearer nuisance. A Multi Angle Absorption Photometer (MAAP Thermo ESM Andersen Instruments) was employed at a UB site which was monitoring BC during the whole sampling period.

The MicroAeth provides data on BC concentrations derived from absorption values. The factor applied to convert the absorption values into mass concentrations ($\mu\text{g}/\text{m}^3$) varies in every region, and therefore, the MicroAeths placed on the fixed stations at schools were cross-correlated with offline measurements of EC in gravimetric samples by thermal-optical transmission (Sunset Laboratory OCEC Analyser) collected *in situ* during the sampling campaigns. The site-specific calibration factor applied to convert the BC measured by the MicroAeths to equivalent black carbon (EBC) concentrations was 0.54 ($\text{EBC} = 0.54 \cdot \text{BC}$, $R^2 = 0.88$, Figure S1). Moreover, prior and after the two monitoring campaigns, all the MicroAeth were compared between them, and the corresponding correction factor with respect to the reference one was applied to the data of each individual instrument (see Supplementary Material for further information). BC concentrations measured with the MAAP in the background site were converted into EBC by an experimental Absorption/EC factor of 9.2 previously determined versus *in situ* thermo-optical EC filter data by Reche et al. (2011).

The effect of filter loading on BC measurements was kept to a minimum by replacing the filter strips every 24 h and setting a flowrate of 100 ml/min. The time-base was set to 5 minutes to minimize the noise in the measurements due to the sensitivity of MicroAeths to vibration, as well as to extend battery life.

EBC monitoring

Fifty-three children (7–10 years old) were initially involved in the personal measurements during 48 h

each, which took place from 19 March 2012 to 22 February 2013. Sampling was carried out only during weekdays. Data from 8 children were discarded because of measuring errors recorded by the MicroAeth and other operational problems, resulting in 45 children being finally included in the study (with at least 24 h of valid data). Children carried the instrument in a belt bag, with the inlet tube always exposed and placed in the breathing zone. To minimize any annoyance derived from wearing the instrument, pupils were allowed to leave the device on the table or hang it from their chair during teaching hours and to leave it on the night stand (and carry the batteries) during sleeping time. It was stressed that it was important to wear the instrument every time they changed location (even between school classrooms), went to the playground or commute.

The children were also instructed to fill in a time-activity diary reporting every time they changed location and activity. The personal measurements sampled by the MicroAeth were classified based on the location (or MEs) of the children as 'classroom' (if the child was inside the school building regardless if it was specifically a classroom), 'school playground', 'home', 'commuting', and 'other' (this category includes MEs of many kind such as public library, swimming pool, shop, ...) according to the information registered in the time-activity diaries. Some commuting time might be misclassified into the 'other' category, as some trips were not clearly specified in the diaries.

The 45 children were attending 25 different schools, which were part of the BREATHE Project. The schools were located in the city of Barcelona (16 000 inhabitants/ km^2 , IDESCAT, 2012), except two, which were in Sant Cugat del Vallès (1800 inhabitants/ km^2 , IDESCAT, 2012, Figure 1). At the same time as personal monitoring, online BC concentrations with the MicroAeth device were simultaneously monitored at

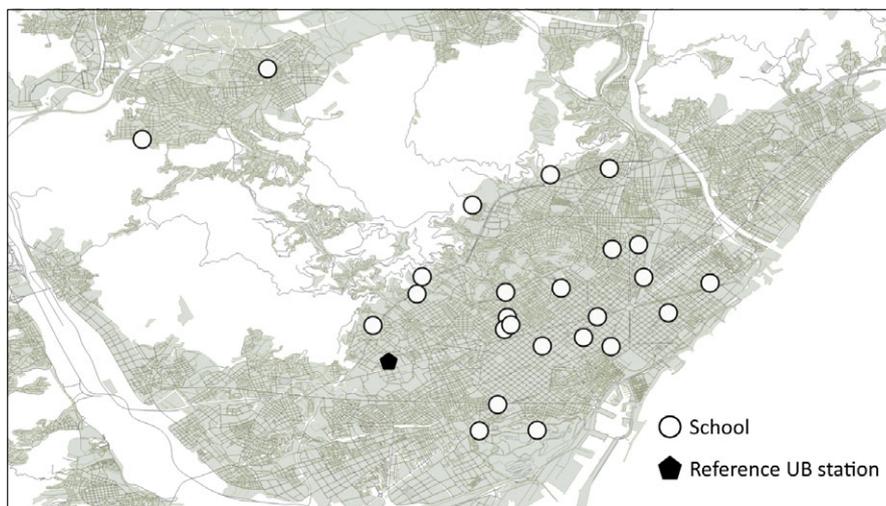


Fig. 1 Location of the reference urban background station and the schools that the children were attending to

schools, both indoors (in a classroom with pupils from 7–10 years old) and outdoors (in the playground or in a balcony) throughout 24 h a day at a height between 0.7 and 1.5 (children's breathing height). Moreover, BC concentrations were also monitored in the reference UB station of Palau Reial, located in the garden of the IDAEA-CSIC building (41°23'14" N, 02°06'56" E, 78 m.a.s.l). Therefore, we measured BC concentration across three different spatial units of analysis: personal, school, and city scale (Mejía et al., 2011; Morawska et al., 2013).

Further information about the data collection and air quality assessment at fixed stations at schools can be found elsewhere (Rivas et al., 2014).

Data analysis

Negative measurements were not removed from the analyses (McBean and Rovers, 1998), as the MicroAeth detects the change in optical absorption, and small shifts in the light beam or the filter ticket can cause a temporary decrease in measured absorption. The aethalometer computes the difference with the previous measurement, and therefore, negative measurements are considered offsets in the next observations. On the other hand, negative values in MicroAeth can also be due to sharp changes in relative humidity (i.e., when moving from the indoor to the outdoor environment), as demonstrated by Cai et al. (2014). Although negative values have been included into the analyses, the 1st percentile has been considered as the minimum when reporting the range in the results section as negative concentrations have no physical meaning.

When comparing data between the different monitoring stations, only data for which simultaneous measurements were available were used (casewise deletion). To assess the relationship between EBC personal measurements and EBC measurements at fixed stations, we carried out linear mixed-effects models (LMMs) with school and student as random effect to account for the repetitive measurement and assessed the proportion of variance that explains the fixed part of the model (R^2).

All the statistical analyses (data management, descriptive statistics, time series and LMMs) were performed with the 10 min averaged values with the R statistical software (v 3.1.0., R Core Team, 2014) and the packages *openair* (Carslaw and Ropkins, 2012) and *lme4* (Bates et al., 2014), among others.

Results and discussion

EBC concentrations

The EBC concentrations measured in the different monitoring sites as well as in personal measurements during the overall experimental campaign are shown in Figure 2 and Table S2. Similar EBC levels were

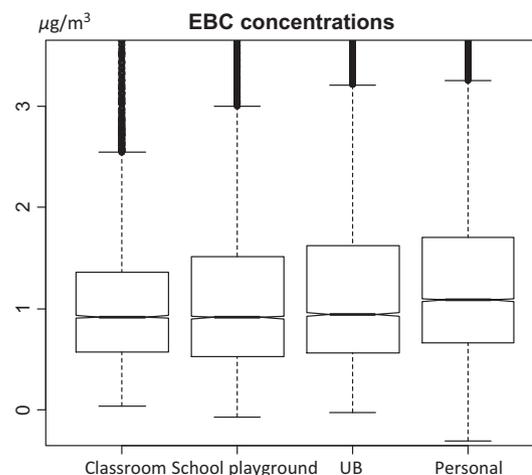


Fig. 2 Boxplot showing EBC concentrations ($\mu\text{g}/\text{m}^3$, 10-minute time resolution) measured at different monitoring stations (at indoor school classrooms, school playgrounds, urban background, and personal monitoring). Boxes represent the interquartile range (IQR, 25–75 percentile), the line shows the median of the data. The whiskers add and subtract 1.5 the IQR to 75 and 25 percentiles, respectively. The notch displays the confidence interval around the median

obtained in all the stations. The geometric mean (GM) EBC concentration in the fixed stations (schools playgrounds, classrooms, and UB) was $0.9 \mu\text{g}/\text{m}^3$. The similarities between indoor and outdoor environments could be explained by the relative location of the sampling sites within schools, whereby in some schools, the classroom was relatively closer to outdoor traffic than the outdoors and EBC easily infiltrates from the outdoor to the indoor environment (Rivas et al., 2015). Schools were distributed among high- and low-trafficked areas of the city, which results in mean concentrations similar to the UB site. The increasing variability from the classroom toward the UB site (Figure 2) was interpreted as resulting from the fact that the outdoor environments (school playground and UB) are influenced to a larger degree than the classroom by meteorological factors, which impact EBC levels by diluting or concentrating this pollutant as a function of atmospheric dispersion. The highest GM EBC concentration was obtained for personal measurements ($1.0 \mu\text{g}/\text{m}^3$; 20% higher than at the school fixed stations, 10% higher than at UB). The range of 10-min EBC concentrations from personal measurements was the widest one ($0.1\text{--}53.3 \mu\text{g}/\text{m}^3$) when compared to the fixed school and UB sites and, and the highest values were due to peak concentration events that took place mainly during commuting time and will be discussed later. There are only few publications reporting personal measurements of BC data on children. Buonanno et al. (2013) monitored 103 children during 48 h (8–11 years old) in Cassino (Italy) and obtained an average of $5.1 \mu\text{g}/\text{m}^3$ (range $0.1\text{--}521 \mu\text{g}/\text{m}^3$) which is much

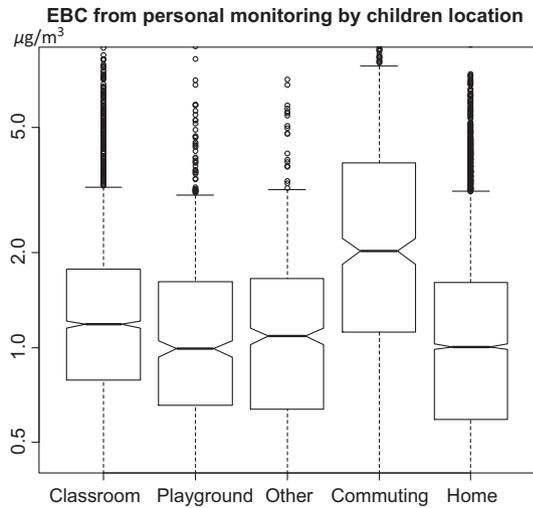


Fig. 3 EBC concentration ($\mu\text{g}/\text{m}^3$, logscale, 10-minute time resolution) ranges from personal monitoring by time periods corresponding to the ME where children reported to be in the diary. Boxes represent the IQR, the line shows the median of the data. The whiskers add and subtract 1.5 the IQR to 75 and 25 percentiles, respectively. The notch displays the confidence interval around the median

higher than ours (arithmetic mean, $\text{AM}=1.5 \mu\text{g}/\text{m}^3$ EBC, $2.7 \mu\text{g}/\text{m}^3$ of uncorrected BC; Table S2).

In Figure 3, we split the concentrations measured by personal monitoring by the ME where children reported to be in the diary each time period (the equivalent figures for the fixed stations are presented in Figure S2). The highest EBC levels from personal monitoring were measured during commuting times ($\text{GM}=2.0 \mu\text{g}/\text{m}^3$, which were significantly higher than in the rest of ME; Figure 3, Table S2), followed by the concentrations to what children were exposed when being in the classroom ($1.2 \mu\text{g}/\text{m}^3$) and in the school playground ($1.0 \mu\text{g}/\text{m}^3$). The lowest concentrations were obtained during periods when children were at home or at other ME ($0.9 \mu\text{g}/\text{m}^3$ in both ME). The previous results were expected, as children (and citizens in general) are very close to traffic when commuting, while the lowest EBC levels are expected during nights, when children are at home. For epidemiological studies with large populations, knowing the amount of time that the subjects spend in commuting (i.e., through questionnaires) may be used as a qualitative indicator of the degree by which personal exposure is higher than fixed monitor EBC concentrations.

The impact of each transport mode used by children for commuting was evaluated and the results are shown in Table 1. Children were asked to write down in the time-activity diary the mode of transport that they used for commuting, but in the 45% of the trips (103 of 229 of identified trips) the transport mode was unknown. For those cases in which we have information, the lowest concentrations were measured for the car mode ($\text{GM}=1.7 \mu\text{g}/\text{m}^3$, $\text{AM}=2.3 \mu\text{g}/\text{m}^3$, probably

Table 1 Mean EBC concentrations from the personal measurements for the different means of transports (reported in the time-activity diary)

Parameter	On foot	Bus	Metro	Car	Mixed	Unknown
N (trips)	75	12	9	21	9	103
Mean trip duration (min)	26	40	39	37	50	20
EBC, GM ($\mu\text{g}/\text{m}^3$)	1.9	3.9	3.8	1.7	3.6	1.8

GM, geometric mean.

influenced by air recirculation; Hudda et al., 2012; Knibbs et al., 2010) followed by the levels found when commuting on foot ($\text{GM}=1.9 \mu\text{g}/\text{m}^3$, $\text{AM}=2.4 \mu\text{g}/\text{m}^3$). Children commuting by metro were facing a GM EBC concentration of $3.8 \mu\text{g}/\text{m}^3$ ($\text{AM}=4.0 \mu\text{g}/\text{m}^3$) with the highest concentrations observed for the bus mode ($\text{GM}=3.9 \mu\text{g}/\text{m}^3$, $\text{AM}=5.0 \mu\text{g}/\text{m}^3$). These results agree to what Dons et al. (2012) observed for 62 adults in Belgium, where bus transportation was the one with the highest BC levels ($6.6 \mu\text{g}/\text{m}^3$ for bus passengers). However, in their study car was the second highest transportation mode with BC concentrations of $6.4 \mu\text{g}/\text{m}^3$ for car drivers and $5.6 \mu\text{g}/\text{m}^3$ for car passengers, which in our case showed the lowest mean concentrations (which may be due to differences in air recirculation settings, but this information is not available and cannot be assessed).

EBC Time series

The analysis of the EBC concentration time series allows the identification of peak concentration events and relation to specific activities (when identified in the diary). Moreover, we can evaluate how EBC concentrations monitored in different ME by the different fixed monitoring sites relate to concentrations measured by personal monitoring.

As an example, Figure 4 shows the time series of EBC concentration measured by personal monitoring of 4 children from 2 different schools. EBC concentrations measured by the fixed monitoring stations are also shown. Although the schools were located in different areas of Barcelona, with different traffic intensities (Rivas et al., 2014), most of them showed the morning and afternoon road traffic rush hour, which were not only identified in outdoor monitoring stations but also inside the schools owing to a high EBC infiltration (Rivas et al., 2015). The morning rush hour coincided with children commuting to school. In fact, most of the commuting periods were clearly evident in the personal measurements because of (extremely) high EBC peaks (Figure 4), which were an average of 2.9 times higher than mean concentrations measured at home. The ratio $\text{EBC}_{\text{commuting}}/\text{EBC}_{\text{home}}$ ranged between 0.8 and 26.7 (median = 2.5). The 26.7 was an extreme case of a child exposed to very high concentrations during commuting but having very low concentrations while being at home. In fact, the second maximum of this ratio drops

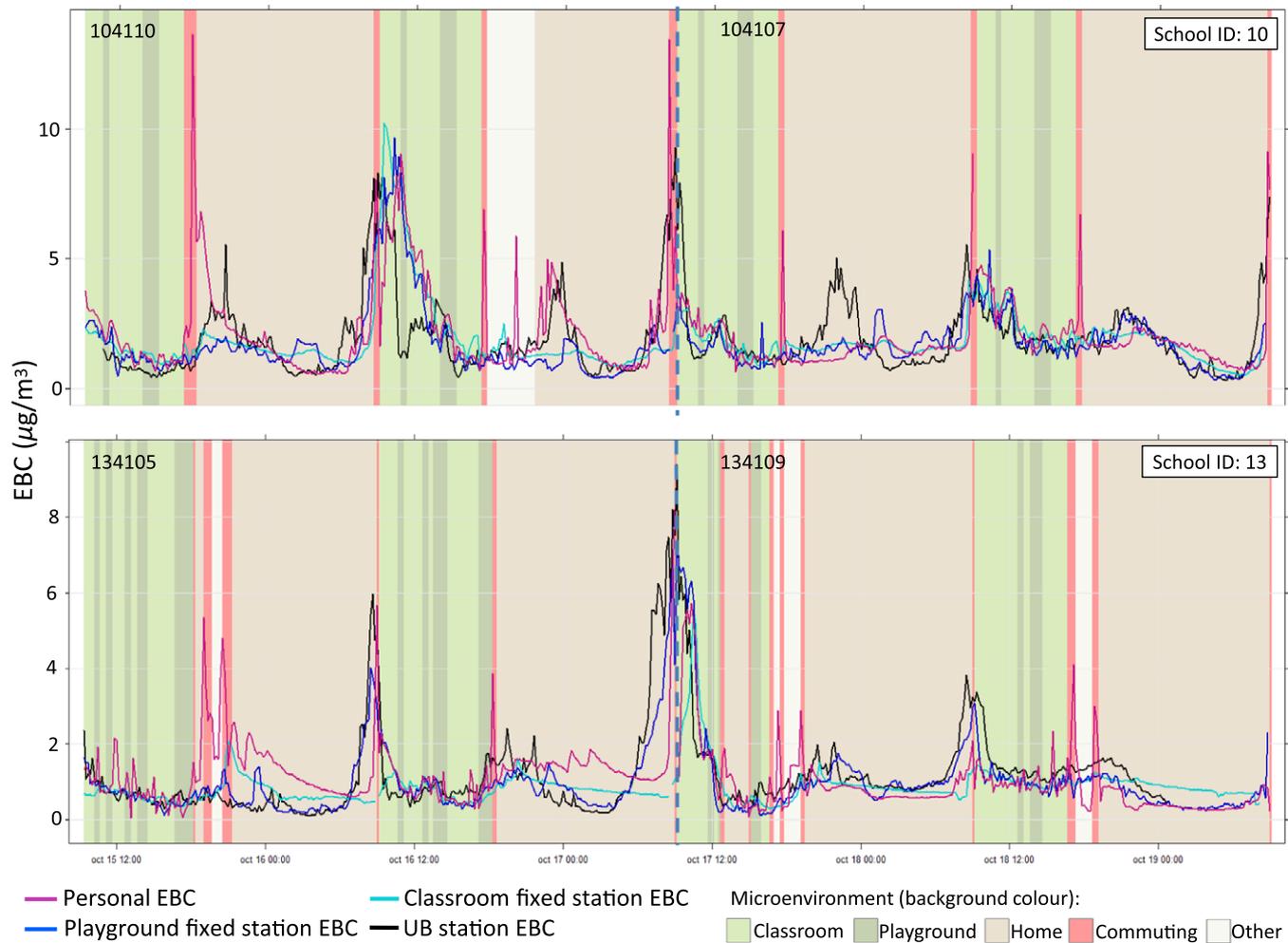


Fig. 4 Time series corresponding to 4 different children (from 2 schools). Lines indicating EBC concentrations ($\mu\text{g}/\text{m}^3$) measured in the personal monitor (pink), school classroom (light blue), school playground (blue), and in the urban background (black) are shown. Background shadow indicates in which microenvironment were children located at each time step

drastically to 5.2. The three cases with the highest ratios are presented in Figure S3.

Although monitored children were not always attending the same classroom where the air quality monitoring was carried out, during school hours, the majority of EBC concentration from personal measurements followed approximately the same levels and trends as the ones measured by indoor school monitoring stations. Generally, indoor and outdoor school levels also followed the UB trends, confirming the previously observed infiltration and the influence of traffic emissions. However, levels at school might be higher or lower than at UB, depending on traffic conditions in the specific streets surrounding the school. Therefore, this makes the estimation of the personal exposure based on a simple (or few) fixed stations difficult, as a more local-scale characterization is needed. In addition, sometimes a lag time between the morning EBC peak in the UB station and in schools can be seen, mainly because the UB station is located near one of the main access roads to the city. This lag time is usu-

ally longer when referring to indoor environments, since extra time should be added to account for EBC transport and infiltration from outdoor to indoor air. There is a great variability among each child's time series, probably associated to the living area location (school and home, principally), which might be influenced by different traffic intensities and, thus, different EBC concentrations. In accordance with what Dons et al. (2011) concluded, differences in EBC concentrations to what children are exposed are due to differences between their time-activity pattern and the corresponding location visited.

Agreement between personal measurements and different monitoring sites

Assuming personal monitoring as the most representative measure for exposure (Jantunen et al., 2002), we performed LMMs to test the agreement between personal measurements and those recorded by fixed stations in different locations. The regression coefficients

(RC, which is the equivalent to the slope in a simple linear regression), intercepts, and R^2 (defined here as the proportion of the variance explained by the fixed effect) are shown in Table 2 for all the data and separately by warm and cold season. Low R^2 between personal measurements and fixed stations at schools were found ($R^2 = 0.28$ and $R^2 = 0.26$, classroom and playground, respectively), being the R^2 much higher during the warm (mean temperature >20 °C) than the cold season (especially for the school fixed stations). The coefficients are also higher (closer to 1) for the warm season period, indicating a better prediction from the fixed station (especially the indoor stations at schools, with a RC = 1 and $R^2 = 0.52$) during this season. Studying the agreement between personal measurements and fixed stations in the different microenvironments allows us to have a deeper understanding about which are the microenvironments of which the fixed stations fail to be representative.

Focusing only in the periods when children were at the classroom microenvironment we can observe an important increase of the R^2 (being 0.79 for the classroom and 0.75 for the playground station) when compared to the whole day, indicating the importance of

the spatial unit of analysis when assessing human exposure. During the warm season, the coefficients are much closer to 1 than during the cold season, and this worse prediction from the outdoor fixed stations is due to closed windows during colder periods that partially hinder EBC infiltration into the indoor environment. On the other hand, it should be highlighted that during both seasons, the coefficients for the classroom station are close to 1 during classroom and home time (although lower R^2 are found when the children were at home), what indicates that these two indoor environments followed not only similar patterns but also similar levels. Considering the important amount of time spent in the indoor environments, these results suggest the necessity to characterize indoor school environments for an accurate assessment of exposure to EBC of schoolchildren.

The relationship between personal EBC concentrations when children were at school and concentrations at UB fixed station was also assessed by LMM, obtaining an $R^2 = 0.45$ in both classroom and playground times (R^2 between UB and schools are 0.37 for the indoor station and 0.39 for the outdoor one), which was much higher than the R^2 obtained when

Table 2 Regression coefficients (RC), intercept, and R^2 from the linear mixed-effects models performed for EBC concentration from personal measurements as the outcome and fixed stations (in different locations) as fixed-effect predictor. Models were performed for the complete day (including all microenvironments) and only considering the time spent in each of the microenvironment separately

Fixed-effect predictor ($\mu\text{g}/\text{m}^3$)	All seasons			Cold season			Warm season		
	RC	intercept ($\mu\text{g}/\text{m}^3$)	R^2	RC	intercept ($\mu\text{g}/\text{m}^3$)	R^2	RC	intercept ($\mu\text{g}/\text{m}^3$)	R^2
All day									
EBC classroom fixed	0.95*	0.4*	0.28	0.85*	0.5*	0.17	1.00*	0.3*	0.52
EBC playground fixed	0.65*	0.6*	0.26	0.53*	0.7*	0.15	0.72*	0.5*	0.50
EBC UB fixed									
Unadjusted	0.39*	0.9*	0.18	0.28*	0.9*	0.14	0.49*	0.9*	0.29
Adjusted ^a	0.39*	0.9*	0.18	0.28*	1.2*	0.14	0.49*	0.3	0.29
Classroom time									
EBC classroom fixed	0.94*	0.3*	0.79	0.81*	0.4*	0.68	0.99*	0.2	0.79
EBC playground fixed	0.73*	0.5*	0.72	0.46*	0.7*	0.57	0.80*	0.5*	0.73
EBC UB fixed	0.49*	0.9*	0.45	0.11*	1.1*	0.41	0.61*	1.0*	0.40
Playground time									
EBC classroom fixed	1.00*	0.1	0.73	0.87*	0.2*	0.49	0.99*	0.1	0.87
EBC playground fixed	1.02*	0.1	0.75	0.87*	0.2*	0.48	1.01*	0.1	0.89
EBC UB fixed	0.53*	0.8*	0.45	0.18*	0.9*	0.31	0.64*	0.9*	0.47
Home time									
EBC classroom fixed	1.00*	0.3*	0.48	0.92*	0.4*	0.46	1.09*	0.2	0.41
EBC playground fixed	0.48*	0.7*	0.47	0.50*	0.7*	0.46	0.49*	0.7*	0.40
EBC UB fixed	0.31*	0.9*	0.43	0.38*	0.7*	0.46	0.34*	0.9*	0.34
Commuting time									
EBC classroom fixed	0.93*	1.9*	0.30	0.63	2.3*	0.29	1.12*	1.0*	0.43
EBC playground fixed	0.76*	2.0*	0.32	0.43	2.5*	0.29	0.85*	1.3*	0.55
EBC UB fixed	0.53*	2.2*	0.30	0.18	2.7*	0.29	0.65*	1.5*	0.41
Other time									
EBC classroom fixed	-0.01	1.6*	0.37	-0.19	2.2*	0.38	0.13	1.1*	0.18
EBC playground fixed	0.00	1.5*	0.37	0.11	1.9*	0.37	0.14	1.1*	0.18
EBC UB fixed	0.10	1.4*	0.37	0.14	1.7*	0.39	0.15	1.1*	0.19

^aAdjusted by distance between school and UB and by traffic density at school.

*P-value <0.05.

considering the whole day ($R^2 = 0.18$, Table 2). This lower coefficient of determination when compared to the fixed sites in schools is due to specific characteristics of each ME where the children spend their time but also to the spatial variability among the city for this pollutant observed during BREATHE campaigns.

When children were commuting, the corresponding R^2 was around 0.30 in all stations with higher R^2 during the warm season. The high intercepts indicate that children receive a contribution of around $2 \mu\text{g}/\text{m}^3$ of EBC that is not accounted by the fixed stations. The low R^2 for commuting periods (≤ 0.32) can be explained by the proximity of children to the main source of EBC, traffic, and also because their breathing height is very close to exhaust pipes (Mott et al., 1997).

As shown in Table 2, the R^2 for the ‘other’ microenvironments is also low and the coefficients are not significant. This may be due to the fact that this single category includes very different microenvironments with different characteristics.

In addition to season, distance from school to the UB station and traffic density (traffic counts were performed during 15 min twice per week in each school) were also included in the model as possible predictors of personal EBC concentrations (adjusted model for UB in Table 2). These two variables did not contribute to improve significantly the model. Moreover, distance to school was further studied (Figure S4) and the correlation coefficient between personal EBC measurements and EBC concentrations at UB for each child showed no linear relationship with distance. Other possible influential variables (e.g., architectural features, wind speed, and direction) that were not assessed in this study may have an important role.

Moreover, correlations between five pairs of children (from three different schools) that were monitored simultaneously resulted in a coefficient of determination of 0.08 when correlating the EBC concentration between them. Again, the low relationship between the concentrations these children were exposed to seems to be explained by the distance to road traffic in each specific moment. The fact that the correlation between exposure measurements for different children is low illustrates the difficulties to obtain representative exposure data at individual level.

Children’s daily-integrated exposure and dose to EBC

Exposure is often confused with concentration (Morawska et al., 2013), the latter being the most frequently quantified. The concept of exposure incorporates the duration of the contact to a certain concentration by integrating over time (Duan, 1982; Ott, 1982). Determining the dose is a step further than the exposure, and it corresponds to the product of the exposure by a dosimetry factor (Morawska et al., 2013). In our case,

Table 3 Inhalation rates (m^3/h) for children (6–10 years) as a function of the activities usually carried out in each of the microenvironments considered

Microenvironment	Activity associated	Inhalation Rate ^a (m^3/h) (age group: 6–10 years)
School indoor	School/studying/eating	0.42
School outdoor	Playing outdoor	1.27
Commuting	Transportation	0.91 ^b
Home (non sleeping time)	Sedentary activities/eating	0.42
Home (sleeping time)	Sleeping and resting	0.31
Others	Entertainment indoor and outdoor	0.91

^aInhalation rates obtained from Buonanno et al. (2011).

^bThe original inhalation rate from Buonanno et al. (2011) was $0.58 \text{ m}^3/\text{h}$.

the dosimetry factor is the inhalation rate and the ones being employed are presented in Table 3 and were obtained from Buonanno et al. (2011), which were adapted from Adams (1993) and US-EPA (2009). The dosimetry factor for transportation was originally $0.58 \text{ m}^3/\text{h}$. However, for the present work, the inhalation rate for a non-sedentary job was considered as the most appropriate for commuting activities, as they might include active transportation that may involve some increment on inhalation rates. To accurately determine the dose, for this section the ‘home’ has been split into two, considering if the children were sleeping (the activity with the lowest inhalation rate) or doing other sedentary activities. In this study, only working days were considered. The exposure and dose received during weekends may vary considerably from weekdays.

The mean daily-integrated exposure to EBC for the 45 children was $34.6 \mu\text{g}/\text{m}^3/\text{h}/\text{day}$, and it showed a high variability among the children (standard deviation: $13.8 \mu\text{g}/\text{m}^3/\text{h}/\text{day}$, range: $12.8\text{--}72.9 \mu\text{g}/\text{m}^3/\text{h}/\text{day}$, Figure S5). For the daily-integrated dose, the mean accounted for $18.2 \mu\text{g}/\text{day}$ (standard deviation: $7.7 \mu\text{g}/\text{day}$, range: $6.5\text{--}40.8 \mu\text{g}/\text{day}$, Figure 5) and the variability observed within the exposure was maintained for the dose. This variability was a result of the different time–activity–geography patterns of each child, who can carry out very different activities in locations with different EBC concentrations. Exposure and dose could be significantly different even between children attending the same school, and this variability could not be taken into account only with the fixed stations. Mullen et al. (2011) also observed a high variability in ultrafine particle number concentration among 13 occupants of 4 apartments. This highlights the usefulness of personal monitoring for a precise estimation of the exposure/dose of each subject.

Home was the ME with the lowest EBC concentration. Notwithstanding, from the total daily-integrated exposure, children received the 50% while being at home (30% during sleeping time), where they spent around 58% of their daily time (Figure 6, Figure S6). Children received the highest exposure at

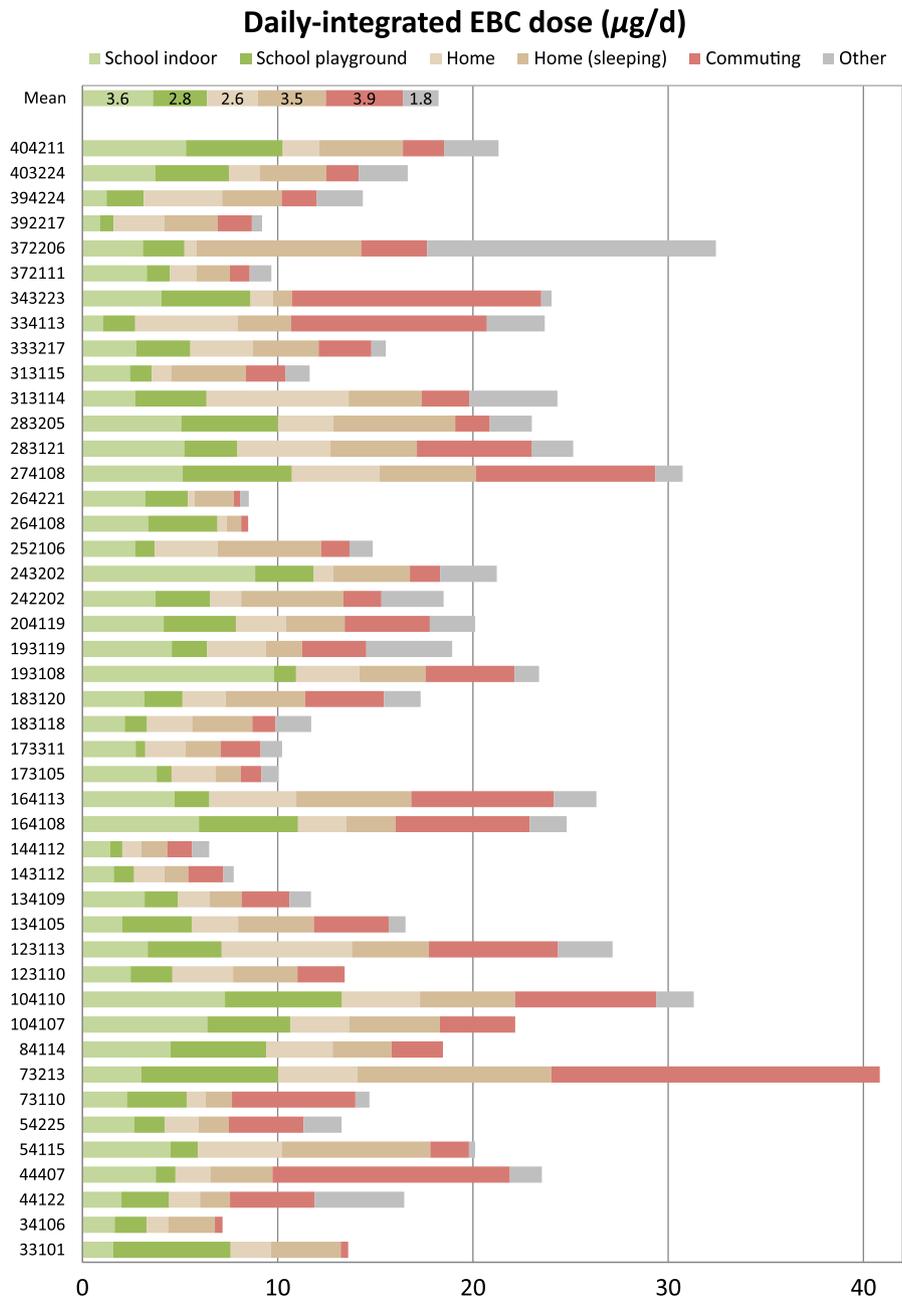


Fig. 5 Estimated daily integrated EBC dose ($\mu\text{g}/\text{m}^3$) for the 45 children and their mean (for %, refer to Figure S4). The integrated dose represents the product of the exposure in each of the microenvironments ($\text{ng}/\text{m}^3/\text{h}/\text{day}$) by the inhalation rate (m^3/h)

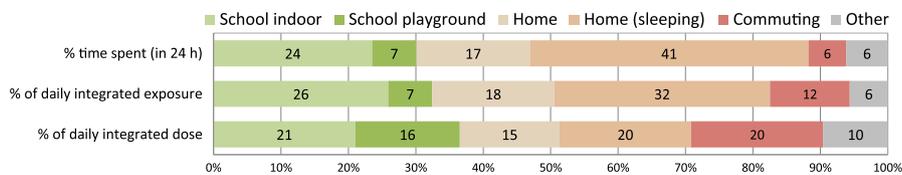


Fig. 6 Mean % of the daytime spent and percentage of daily integrated exposure and dose corresponding to each microenvironment for the 45 children

home because of the large time spent there, since it accounts for the night period. However, since the activities usually carried out at home during week-

days are not very active, the home contribution to the daily-integrated dose decreased to only 35% (20% corresponds to sleeping time). Actually, the

lowest ratio of exposure and dose with respect to the time spent was observed at home during sleeping time (ratio exposure:time = 0.77:1, dose:time = 0.47:1). Children spent 31% of their weekday at schools, where they received 33% of their daily-integrated exposure to EBC (26% in the classrooms and 7% at playgrounds) and 37% of the daily-integrated dose (21% and 16%, classroom and playground, respectively). Indoor environments (classroom + home) accounted for the 82% of the daily time of schoolchildren during weekdays. The corresponding daily-integrated exposure and dose received in the indoor environments was 76% and 56%, respectively. Therefore, children received more than half of the dose in the indoor environment. Although the dose received at home is higher, policies for the reduction of EBC emissions around schools would benefit a large number of children given that they spend a considerable portion of their weekdays in a shared location (school).

However, the highest ratio of exposure and dose with respect to the time spent was observed during commuting. It was responsible for 12% of the daily exposure and around 20% of the daily dose whilst it only accounted for the 6% of the time, so a relation 2.1:1 (3.5:1) of exposure:time (dose:time) is observed. The high exposure was explained by the high concentrations found during commuting, and the dose is a combination of the former and the moderate physical activity intensity usually involved in commuting. In fact, the inhalation rate factor employed for commuting may vary considerably according to the mode of transport, being considerably higher in the case of active travel (De Nazelle et al., 2012). However, as 35% of the commuting modes were not reported by children, the same inhalation rate has been used for this activity regardless of the transport mode. Buonanno et al. (2013) obtained a similar percentage of time spent in the different ME for 103 children in Cassino (64% at home, 24% at school, and 4% in transport versus 58%, 30%, and 6% in our study) and also a similar distribution of the exposure contribution (60% at home, 20% at school, and 11% in transport versus 50%, 32%, and 12% in our study), although with a much higher dose (39.2 $\mu\text{g}/\text{day}$ versus 18.2 $\mu\text{g}/\text{day}$ in our study). On the other hand, Dons et al. (2012) obtained a higher exposure:time and dose:time relationship in transport for 62 adults in Belgium (3.3:1 versus our 2.1:1 of exposure:time; 4.8:1 versus our 3.5:1 for dose:time), with people spending around 6% of their days commuting and receiving the 21% of their daily-integrated exposure and 30% of their dose. In the case of the exposure, it might be due to differences in activities schedule between children and adults (or between regions) and, in the case of the dose, it should also be considered that inhalation rates depend on the person age (increases with age).

Policies to reduce EBC levels should be enhanced throughout the urban area. As more than a third of the daily-integrated dose takes place at schools and commuting has the highest dose:time relationship, specific policies focused on reducing traffic intensities around schools should be implemented. These school targeted actions will favor the abatement of the exposure of a wide fraction of the population, which are also one of the most vulnerable to air pollutants threats.

Conclusions

The present work aimed to contribute to the current knowledge on spatial and temporal monitoring of EBC, specifically when assessing personal exposure of schoolchildren. To accomplish this objective, continuous personal monitoring of EBC was carried out with microaethalometers for 45 schoolchildren (aged 7–10), and fixed monitoring stations were located in indoor and outdoor school microenvironments and at an urban background site. The highest (geometric) mean EBC concentrations corresponded to personal monitoring (20% higher than at the school fixed stations, 10% higher than in the urban background) owing to peak concentration events during commuting times. This was due to two reasons: the co-occurrence of children commuting times and road traffic rush hours, and the closest proximity to the source (road traffic) while commuting. In fact, children spend only the 6% of their daily time in commuting, while based on our estimations, they received around 20% of their total daily EBC dose during this activity. This estimate will vary as a function of breathing rates, especially in transport microenvironments for which their variability is expected to be large. High R^2 from LMM correlations ($R^2 \geq 0.70$) were found between EBC from personal monitors and school fixed sites (both in classroom and playground) when considering only the time periods when children were in each of the microenvironments. On the other hand, the LMM relating personal measurements with the urban background station was weaker ($R^2 = 0.45$) for the same period, thus indicating the importance of the spatial unit of analysis when assessing human exposure. Due to opened windows that facilitate the entrance of outdoor pollutants to indoor environment, during the warm season, the outdoor fixed stations were more representative (higher R^2 and coefficients closer to 1) of the personal exposures than during the cold one. Children spent 82% of their time in indoor environments (classroom and home), where they received 76% and 56% of their daily-integrated exposure and dose, respectively. Considering the important amount of time spent in the indoor environments, it is important to characterize indoor environments for an accurate exposure assessment to EBC. The contribution from schools (including classroom and

playground) to the total daily-integrated EBC dose was the 37%, and the highest dose:time intensity was observed during commuting times. Reducing traffic intensities around schools should be enhanced to minimize the exposure of a wide fraction of the population who spend a large portion of their weekdays in a shared location.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Appendix S1. Instrumentation calibration and supplementary data.

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