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Personal exposure to equivalent black carbon in children in Milan, Italy: Time-activity patterns and predictors by season☆

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ABSTRACT

Air pollution is a global threat to public health, especially when considering susceptible populations, such as children. A better understanding of determinants of exposure could help epidemiologists in refining exposure assessment methods, and policy makers in identifying effective mitigation interventions. Through a participatory approach, 73 and 89 schoolchildren were involved in a two-season personal exposure monitoring campaign of equivalent black carbon (EBC) in Milan, Italy. GPS devices, time-activity diaries and a questionnaire were used to collect personal information. Exposure to EBC was 1.3 ± 1.5 μg/m^3 and 3.9 ± 3.3 μg/m^3 (mean ± sd) during the warm and the cold season, respectively. The highest peaks of exposure were detected during the home-to-school commute. Children received most of their daily dose at school and home (82%), but the highest dose/time intensity was related to transportation and outdoor environments. Linear mixed-effect models showed that meteorological variables were the most influencing predictors of personal exposure and inhaled dose, especially in the cold season. The total time spent in a car, duration of the home-to-school commute, and smoking habits of parents were important predictors as well. Our findings suggest that seasonality, time-activity and mobility patterns play an important role in explaining exposure patterns. Furthermore, by highlighting the contribution of traffic rush hours, transport-related microenvironments and traffic-related predictors, our study suggests that acting on a local scale could be an effective way of lowering personal exposure to EBC and inhaled dose of children in the city of Milan.

1. Introduction

Ambient and household air pollution are currently recognized as one of the main public health concerns worldwide (WHO 2018a, b). It does not affect the whole population in the same way; a larger impact is reported in the most susceptible subgroups (Makri and Stilianakis 2008). In particular, children are among the most affected as they breathe closer to the source (i.e. exhaust pipes of vehicles), they are generally more active than adults, present higher respiratory and metabolic rates, and their immune system is not completely developed (Bateson and Schwartz 2008). In the last decade, numerous studies suggested significant associations between exposure to ambient air pollution and a broad spectrum of adverse health effects in children (Perera, 2017; Rojas Rueda et al., 2019). At the 2018 Global Conference on Air Pollution and Health, the World Health Organization (WHO) has called on policy makers, researchers and health practitioners to strengthen the actions to protect children and enhance education on air pollution as a key factor for improving health and quality of life (WHO 2018a, b).

This applies to Italy where more than 98% of children are exposed to PM2.5 concentrations above the annual average guideline value of 10 μg/m^3 (WHO 2018a, b), and more specifically to the Po Valley region, which is among the most densely populated, motorized, industrialized, and polluted areas in Europe (EUROSTAT 2019; EEA, 2018). In this region, Greater Milan is the biggest urban area, with approximately 3.2 million inhabitants.

Among the airborne contaminants black carbon (BC) represents

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primary fine and ultrafine particles strictly linked with fossil fuel and biomass combustion (U.S.EPA 2012). It can overcome human membranes and reach the circulatory system, brain, and even the fetal side of the placenta (Brockmeyer and D'Angiulli 2016; Bové et al., 2019), operating as a carrier of harmful chemical components (Dachs and Eisenreich, 2000). Since it is well known as a suitable marker to study the impact of air pollution on health (Janssen et al., 2011), increasing the knowledge on personal exposure to BC has become a matter of interest to the public health field. For instance, through a better understanding of time-activity patterns, temporal variability, and determinants of exposure, policy makers can more easily identify effective mitigation interventions, while epidemiologists could produce more robust associations with health outcomes (Caplin et al., 2019, Dons et al., 2011, Klepeis et al., 2001). Khreis and Nieuwenhuijsen (2017) recently stressed the need for more refined exposure assessment methods to study the impact of air pollution on children's health by incorporating time-activity and mobility patterns.

In recent years, several scientific contributions investigated personal exposure of children to EBC, focusing especially on microenvironments (MEs) and activities (Buonanno et al., 2013, Jeong and Park 2017; Rivas et al., 2016; Panella et al., 2016; Paunescu et al., 2016), however, without comprehensively studying temporal patterns and without analysing predictors of personal exposure. This contribution reports the findings from a two-season intensive personal monitoring campaign carried out in the city of Milan involving 73 and 89 schoolchildren during May/June 2018 and January/February 2019, respectively. Each weekday, from 2 to 5 children were simultaneously monitored allowing the investigation of within- and between-day variability. Seasonal linear mixed-effect models were developed highlighting the most important predictors of the mean and peak (99th percentile) personal concentrations and estimated inhaled dose of BC, here presented as equivalent BC (EBC; Petzold et al., 2013). With this study, we aim to extend knowledge on personal exposure and inhaled dose of schoolchildren to air pollution, and particularly BC, by focusing on temporal patterns (both seasonal and daily), microenvironments and possible predictors.

2. Materials and methods

2.1. MAPS MI project: study design

This work is part of the Mapping Air Pollution in a School catchment area of Milan (MAPS MI) project. This was a research project consisting of three different stages: 1) the analysis and modelling of the spatial distribution of EBC in the school catchment area, conducted with a participatory approach (Boniardi et al., 2019a); 2) an environmental education intervention to raise awareness and involve schoolchildren, their teachers and parents in the research process; 3) a two-season personal monitoring campaign to study the exposure of schoolchildren and validating previously developed Land Use Regression (LUR) models (Boniardi et al., 2019b).

The proposal was approved by the ethical committee of the University of Milan and the elementary school board. We informed teachers, children, and their parents about the foreseen activities. In particular, as required by the new General Data Protection Regulation of the European Union (EU, 2016), children and their parents were asked to sign three forms: 1) an informed consent sheet about the project specifically prepared to be understood by children; 2) an informed consent sheet about the project for parents or legal guardians; 3) a consent sheet to process personal data.

The entire project took place in Milan, Italy. Measurements were performed involving 8 to 9-year-old children who attended the same school located in the north-western part of the city (Fig. 1). The study area is characterized by a mid to high socioeconomic background (Petsimeris & Rimoldi, 2015) and is affected by a major access road into the city (A8 highway), with intense motorized traffic especially during the morning rush hour (>50,000 veh/day). In general, in the Metropolitan area of Milan, air pollution is mainly affected by traffic and heating system sources (ARPA, 2018). In particular, especially with regards to EBC, biomass heating systems are important sources of air pollution, especially during winter (Boniardi et al., 2019a; Hakimzadeh et al., 2020).

2.2. A participatory approach to involve teachers, schoolchildren, and their parents in the research process

A two-module environmental education intervention was conducted in the selected elementary school. The design of the intervention was first discussed with the Teaching Committee of the school and planned accordingly. The two modules involved the third-grade classes of the school during March–April and October–November 2018 and consisted of playful and experience-based laboratories following the Investigation, Vision, Action, Change (IVAC) methodology (Jensen and Schnack 1997). In particular, the first module (Investigation, Vision) aimed at making children familiar with the topic, and to investigate EBC in the neighbourhood, while the second module (Action for Change) was more focused on how our society can address air pollution issues. The modules were deliberately performed just before the two personal monitoring campaigns to increase the level of engagement and the chance to recruit active and aware volunteers. Given this, the participation in the personal monitoring campaigns was designed to be a fully-fledged part of the intervention by asking schoolchildren to become active researchers.

2.3. The design of the personal monitoring campaign

The two-season personal monitoring campaign was carried out during spring 2018 (April, May and June) and winter 2019 (January and February). Involved children were asked: 1) to wear a GPS, model i-gotU GT600 (Mobile Action, Taiwan), 2) to complete a time-activity diary (supplementary materials, S15) to collect information on their activities and locations, and 3) to wear a shoulder bag equipped with a micro-aethalometer AE51 (Aethlabs, San Francisco, CA) to measure personal exposure to EBC (supplementary materials, S16). To identify possible confounders and other influencing variables, parents were asked to fill in a questionnaire. All the operations within the school were carried out in the former infirmary room of the school. Starting on Monday afternoon and up to Thursday afternoon, each day, at 4 p.m., a group of 2–5 children was enrolled in the study, wearing the equipped shoulder bag; then, after the home-to-school commute of the very next day, between 8 and 9 a.m., children returned the shoulder bag. During the afternoon meeting, children were trained on how to handle the equipment during their personal monitoring: for instance, they were asked to behave as on a normal day, wearing the shoulder bag as long as possible, and always to leave it in their proximity; e.g. when a child went swimming or performed other indoor sports, he/she was instructed to leave the shoulder bag in the dressing room. When involved in outdoor activities, children were instructed to leave the equipment as close as possible in the outdoor environment. Moreover, children were warned on the importance of leaving the equipment in an open environment (i.e. not locked in a wardrobe) and with the inlet tube free from obstruction. In all cases, the same information was also given to the parents. In addition, a micro-aethalometer MA200 (Aethlabs, San Francisco,

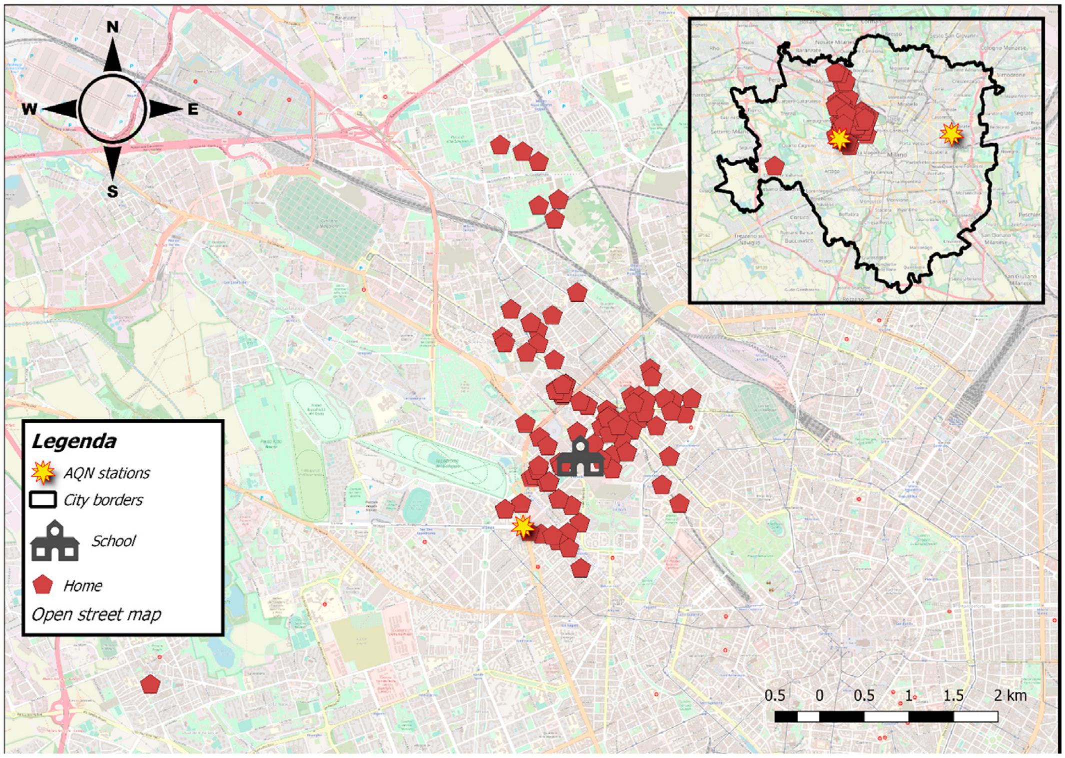

Fig. 1. *Study area with an approximation of the home locations of the participants (dark red polygons) and the School (black building icon). In the small inset the study area within the city of Milan is shown. For privacy, home locations were slightly moved from their real addresses.* (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

CA) was placed inside the former infirmary room of the school measuring EBC continuously.

2.4. 24-Hour exposure attribution

Each 24 h EBC personal monitoring period was reconstructed beginning from midday of the first day until midday of the second day, as follows:

- From 12 p.m. to 4 p.m. of the first day, the personal EBC concentration was estimated using the EBC concentration measured by the MA200 device placed inside the former infirmary room of the school (4 h);
- From 4 p.m. of the first day to 8.40 a.m. of the second day, the personal EBC concentration was measured by the worn AE51 aethalometer (~16 h);
- From the 8.40 a.m. to 12 p.m. of the second day, personal EBC concentration was estimated using the EBC concentration measured by the MA200 (~4 h);

Due to technical issues, during the warm season, the site inside the school was not monitored for about two weeks. To estimate the missing EBC concentrations, we extrapolated hourly EBC concentrations from the urban background station operated by the air quality network (AQN) of Milan (ID station 10283, via Ponzio 34/6 – Pascal Città Studi, ~6 km from the school)(Fig. 1). Hourly correction factors ranged from 0.75 to 1.25. The latter were similar to the regression slopes between the MA200 and the AQN site computed for the available days. The same procedure was applied to the cold season monitoring campaign as well, even if extrapolation was necessary for only a few hours. Scatter plots of regression models are reported in the supplementary materials (S1 and S2).

2.5. Micro-aethalometers and GPSs

Aethalometers are optical devices that estimate black carbon concentrations by measuring the rate of attenuation (ATN) of a beam of light, usually at a wavelength equal to 880 nm, through a filter over which the sampled air is drawn. Concentrations are finally derived by applying an assumed mass absorption cross-section (MAC) (Cheng and Lin, 2013). For this reason, BC estimated by the micro-aethalometer is referred to as equivalent black carbon (EBC) (Petzold et al., 2013). The micro-aethalometer series are commonly used in EBC personal monitoring assessments because of their high portability, and adaptability according to different scenarios. During the two monitoring campaigns, five AE51s and one MA200 were used with the pump flowrate set to 100 mL/s on a 60-s time resolution to deal with both durability and high time resolution needs. Polyurethane sampling tubes, approximately 15 cm long, designed to dissipate electrostatic discharge from air passing through the tube were used.

The GPS devices used in this study were all i-gotU GT600 (Mobile Action, Taiwan), a highly portable device equipped with a SIRF III GPS chipset, a built-in patch antenna, 20 channels, and a sensitivity of −159 dBm. In this study GPS data (300-s and 60-s time resolution, respectively for warm and cold season) were used to check and improve the quality of the reported time-activity diaries (e.g. check timing of a car trip).

2.6. Data handling

EBC raw data were post-processed by removing all the reported errors, by smoothing with the Optimized Noise-reduction Algorithm (ONA) (Hagler et al., 2011), by applying correction factors to account for differences between devices, and by correcting for the loading effect of the particulate matter collected on the filter (Virkkula et al., 2007). Besides, to keep the loading effect-related bias as low as possible, filters were replaced every day, so that each child corresponded with one individual filter. After the application of the ONA algorithm, no negative values were found. After that, the algorithm proposed by Virkkula et al. (2007) was applied, only to the AE51s data. The compensation factor $k_{personal} = 0.007$ was used following Good et al. (2017). Considering all of this, the following algorithm for the calculation of $BC_{corrected}$ starting from the raw AE51s data (BC_{raw}) was applied:

$$BC_{corrected} = BC_{raw} \times (1 + ATN \times 0.007) \times F_{IC}$$

where F_{IC} represents the regression slopes between MA200 and AE51s EBC concentrations measured during the four intercomparison exercises conducted pre and post both seasonal monitoring campaigns. These were performed by running all devices simultaneously and next to each other for about 24 h in a fixed urban background position. The MA200 was the device most recently calibrated, so it was considered as the gold standard. Again, only AE51s data were post-processed by applying ONA and Virkkula's algorithm applying compensation factors for fixed sites. We applied compensation factors $k_{warm\text{-}fixed} = 0.0007$ and $k_{cold\text{-}fixed} = 0.0054$ respectively for the warm and the cold season, following Virkkula et al. (2007). The final F_{IC} ranged from 0.78 to 1 (supplementary materials, S3 and S4) and were calculated as the mean value between the pre- and post-monitoring intercomparisons.

2.7. Contribution of microenvironments to time, daily integrated personal exposure, and dose

The information reported in the time-activity diary was manually cross checked with the GPS data to minimize possible misclassification. Then, each 1-min EBC concentration was associated to different locations and microenvironments attended by the children as reported in the diary. The following microenvironments (MEs) were assigned: a) Home (sleeping), i.e. the time spent at home during sleeping time; b) Home (other activities), i.e. the time spent at home, different from sleeping time; c) School; d) Indoor (other), i.e. the indoor MEs different from School and Home; e) Transportation, i.e. the total time spent in transport microenvironments not distinguishing between modes of transport and including home-to-school commuting; f) Outdoor (other), i.e. all those outdoor MEs different from transportation. For each child, the time associated with each ME ($Time_{ME_i}, min$), was used to calculate the fraction of daily time (1440 min) spent in each ME:

$$\%Time_{ME_i} = \frac{Time_{ME_i}}{1440\ min} \times 100$$

Afterwards, the exposure to EBC associated to each ME in which the child spent time from t_1 to t_2, and computed as $\int_{t_1}^{t_2} BC_{ME_i} \Delta t \left(\frac{\mu g}{m^3} \times min\right)$, was used to calculate the contribution of each ME ($\%BC_{ME_i}$) to the daily integrated personal exposure ($\int_{t_0}^{1440} BC\Delta t$) by applying the following formula:

$$\%BC_{ME_i} = \frac{\int_{t_1}^{t_2} eBC_{ME_i} \Delta t}{\int_{t_0}^{1440} eBC \Delta t} \times 100$$

A similar approach was also used to estimate the daily integrated dose. First, the ME-related dose ($Dose_{ME_i}$) ($\mu g \times min$), was estimated by multiplying each 1-min EBC concentration by an activity-related ventilation rate (VR_j) value (U.S.EPA, 2009)(supplementary materials, S6), with j related to each activity performed in the ME_i, and in particular:

$$Dose_{ME_i} = \int_{t_1}^{t_2} eBC_{ME_i} \times VR_j \Delta t$$

Afterwards, each $Dose_{ME_i}$ was used to calculate the contribution of each ME ($\%Dose_{ME_i}$) when compared to the daily dose ($Dose_{1440}$) by applying the following formula:

$$\%Dose_{ME_i} = \frac{Dose_{ME_i}}{Dose_{1440}} \times 100$$

The information collected from the time-activity diary was supplemented with a series of assumptions. Children started to provide information once they left the school (4.30 p.m.), so no information was available about activities at school. To deal with this lack of information, we profiled a typical school day by asking information from the teachers (supplementary materials, S5), and then we applied this profile to all the children. Also, the sleeping period, defined here as Home (sleeping), was assumed to be the same for all children according to a previous study conducted in Italy (Russo et al., 2007). Finally, the non-dimensional dose intensity related to each microenvironment (I_{ME_i}), as already defined by Buonanno et al. (2012) was computed by using the following formula:

$$I_{ME_i} = \frac{\%Dose_{ME_i}}{\%Time_{ME_i}}$$

where $\%Dose_{ME_i}$ is the fraction of dose associated with the ME_i, while $\%Time_{ME_i}$ is the fraction of daily time spent in the ME_i.

2.8. Meteorological variables

Wind speed data were collected from the nearest AQN station (ID station 19005, piazza Zavattari, ~1 km from the school reference site) (Fig. 1). The hourly atmospheric mixing layer height (hmixdia in this paper) was extracted by the Emilia-Romagna Regional Environment Protection Agency (ARPAE) from the Limited Area Model Analysis (LAMA) data set. This is an initialized local analysis obtained with the limited area model of the Consortium for Small-scale Modelling (COSMO) combining the operational analysis produced by the European Centre for Medium Range Forecasts (ECMWF), as a first guess, and observational Global Telecommunication System (GTS) data over the area. The mixing layer height seasonal hourly boxplots and wind roses are given in the supplementary materials (S9, S14).

2.9. Statistical analysis

R (R Core Team 2013) was used to manipulate and clean data, as well as to perform the statistical analysis (Wickham et al., 2019; Wickham, 2009; Bates et al., 2015; Barton, 2009). The nonparametrical Mann-Whitney test was used to check differences between seasons. Since our data were clustered by days, the different components of the variance (between and within days) were studied by developing an empty linear mixed model with only the random effect intercept for each day. The Intra-Class Correlation coefficient (ICC) was computed as the ratio between the random effect variance and the total variance, i.e. the sum of the random effect variance and the residual variance. The result can be interpreted as the proportion of the total variability explained by between-days variability. Moreover, random-effect intercept models were developed to investigate the influence of possible explanatory variables on mean and 99th percentile personal exposure, and inhaled dose. Response variables were log transformed to comply with linear modelling assumptions. All the tested predictors are reported in the supplementary materials (S7). The variables were collected from different sources and in particular: a) general information variables (i.e. smoking habits of parents etc.) and time-activity variables (i.e. transport mode during home to school commuting) were extracted from questionnaires and time-activity diaries; b) spatial variables were calculated using Quantum GIS (QGIS Development Team, 2016) based on schoolchildren's home addresses.

A forward stepwise procedure was used to select the predictors and to build the final models. Firstly, all the collected variables were checked in a single-variable model and only those with p-value<0.1 and having an expected direction of effect were selected. Secondly, we introduced the variables one by one in an empty model, starting from the predictors with the highest effect on the Akaike Information Criterion (AIC) estimate. For each iteration, the new variable was retained if the AIC of the new model significantly decreased. The significance ($p < .01$) was tested by comparing the previous-step model AIC with the new model AIC, by means of Analysis of Variance (ANOVA) test. Marginal and conditional coefficients of determination (R^2_m and R^2_c) were computed following Nakagawa and Schielzeth (2013). The leave one out cross validation (LOOcV) technique was used to test the performance of the models focusing on Root Mean Square Error (RMSE) and R^2. Multicollinearity was checked with the variance inflation factor (VIF>5). Influential observations were checked by means of the Cook's distance estimate with a threshold equal to $Q3+3 \times IQR$ where Q3 is the third quartile of the estimated values and IQR their interquartile range (Loy and Hofmann, 2014). When necessary, the observations were discarded, models were checked, and predictors removed if their p value was >0.1.

In one case, an observation of potential influence close to the Cook's D threshold was maintained as it was evaluated to be in the range of expected values and representing important information for the explanatory capacity of the model. Finally, residuals were investigated to check for homoscedasticity and normality.

3. Results

3.1. General characteristics of the sample

Altogether, 128 children from 6 third-grade classes (age 8–9 yrs), as well as their parents and teachers were involved in the environmental education intervention. Out of these children, 85 (66%) and 109 (85%) expressed their interest to be involved in the warm and cold season personal monitoring campaigns, respectively. In the supplementary materials, figure S8 shows the sample flow chart with exclusion criteria. In particular, a few children did not confirm their participation due to illness or without explicit reason (n = 3, n = 11 in the warm and cold season, respectively), others were excluded from the analysis due to instrumental issues (n = 6, n = 1) or lack of information on MEs (n = 3, n = 8). The analysis finally involved 73 and 89 children, and among them, 65 children participated in both monitoring campaigns. Either way, females were the most represented; children with parents who smoke were close to a third of the sample; gas cookstoves represented the most prevalent type of facilities for cooking; and children mostly lived less than 1 km away from the school (Table 1). Moreover, the time-activity pattern showed that children spent most of their time at home, while only a small fraction of them frequented other outdoor environments (i.e. public parks, sport facilities etc.). The average time spent in transport was similar for both seasons (~47 min), and a significant fraction of time was spent in cars (about 20 min) (S7). Cooking time was on average about 25 min. Regardless of the season, children mostly went to school on foot (~50%), but a significant number of them were transported by cars (~30%). Other modes of transport, mostly equally represented, were bus, bicycle, motorcycle, and subway. Finally, on average children were living on the 3rd floor, they were located near streets with more than 20,000 vehicles per day and with a Sky View Factor (SVF, portion of visible sky computed in a circular buffer of radius equal to 25 m around children's home) of 0.55, indicating a dense urban environment with street canyons (Middel et al., 2018).

3.2. Personal exposure to EBC and estimated inhaled dose

In Table 2, an overview of the descriptive statistics of personal exposure to EBC is given (Table 2). The mean ± SD personal EBC concentrations in the cold season were 3.9 ± 3.3 $\mu g/m^3$, a 3-fold higher than in the warm season (1.3 ± 1.5 $\mu g/m^3$), while it was 2.8 $\mu g/m^3$ (range: 0.2–11.9 $\mu g/m^3$) when considering overall data. The total variability of the mean personal EBC concentration was mostly explained by the between-day component of variance for both the warm (80%, 95% CI: 65–90%) and the cold season (93%, 95% CI: 88–97%). When considering the 99th percentile (representative of peak exposure), the between-day variability explained respectively 33% (95% CI: 30–60%) and 86% (95% CI: 76–93%) of the total variability. Regardless of seasonality, transportation was the ME

Table 1
Overall characteristics of the participating children.

Variables *(abbreviation)*	Unit, statistics	Warm	Cold
Number of participants	n	73	89
Gender	n (%), Female	42 (58)	46 (52)
Age	years, mean ± SD	8.5 ± 0.7	9 ± 0.5
Parental education	n (%), University degree	55 (75)	69 (78)
Gas cookstoves	n (%)	68 (93)	72 (81)
Biomass heating system	n (%)	1 (1)	1 (1)
Number of children going to school on foot	n (%)	38 (52)	47 (53)
Distance between home and school	meters (m), mean ± SD	855 ± 700	892 ± 613

Table 2
Statistics of the seasonal and overall personal EBC concentrations (μg/m^3) according to the different microenvironments (MEs). Personal EBC concentrations are presented as median, range, arithmetic mean (AM), standard deviation (SD), and geometric mean (GM).

MEs	Warm					Cold					Overall				
	Personal EBC (μg/m^3)					Personal EBC (μg/m^3)					Personal EBC (μg/m^3)				
	n[c]	Median	Range[b]	AM (SD)	GM	n	Median	Range[b]	AM (SD)	GM	n[c]	Median	Range[b]	AM (SD)	GM
Home (other activities)	73	1.2[a]	0.4–4.4	1.4 (2.0)	1.2	89	2.7	0.2–11.5	3.2 (3.3)	2.4	162	1.7	0.3–10.5	2.4 (2.9)	1.8
Home (sleeping)	73	0.9[a]	0.3–3.3	1.1 (0.6)	0.9	89	3.6	0.2–12.8	4.3 (3.2)	3.0	162	1.5	0.3–11.7	2.8	1.8
School	73	1.2[a]	0.4–3.7	1.4 (0.8)	1.2	89	3.3	0.2–12.7	3.9 (2.9)	2.9	162	1.9	0.2–11.8	2.8	1.9
Indoor (other)	34	1.2[a]	0.4–3.8	1.3 (0.9)	1.2	52	2.0	0.1–6.4	2.3 (1.8)	1.6	86	1.6	0.1–6.0	1.9	1.4
Transportation	73	2.2[a]	0.1–22.5	3.5 (5.1)	2.2	89	4.1	0.2–22.9	5.7 (7.2)	3.8	162	3.2	0.1–22.7	4.7	3.0
Outdoor (other)	17	0.9[a]	<0.1–3.4	1.1 (0.9)	0.8	15	3.0	0.1–11.4	3.3 (2.3)	2.5	32	1.2	<0.1–8.8	1.8	1.9
24h exposure	73	1.1[a]	0.3–4.6	1.3 (1.5)	1.2	89	3.2	0.2–12.9	3.9 (3.3)	2.8	162	1.7	0.2–11.91	2.8 (3.0)	1.8

[a] tested differences between warm and cold personal EBC concentrations: p-value always <.001.
[b] computed as 1 st and 99th percentile.
[c] Number of children.

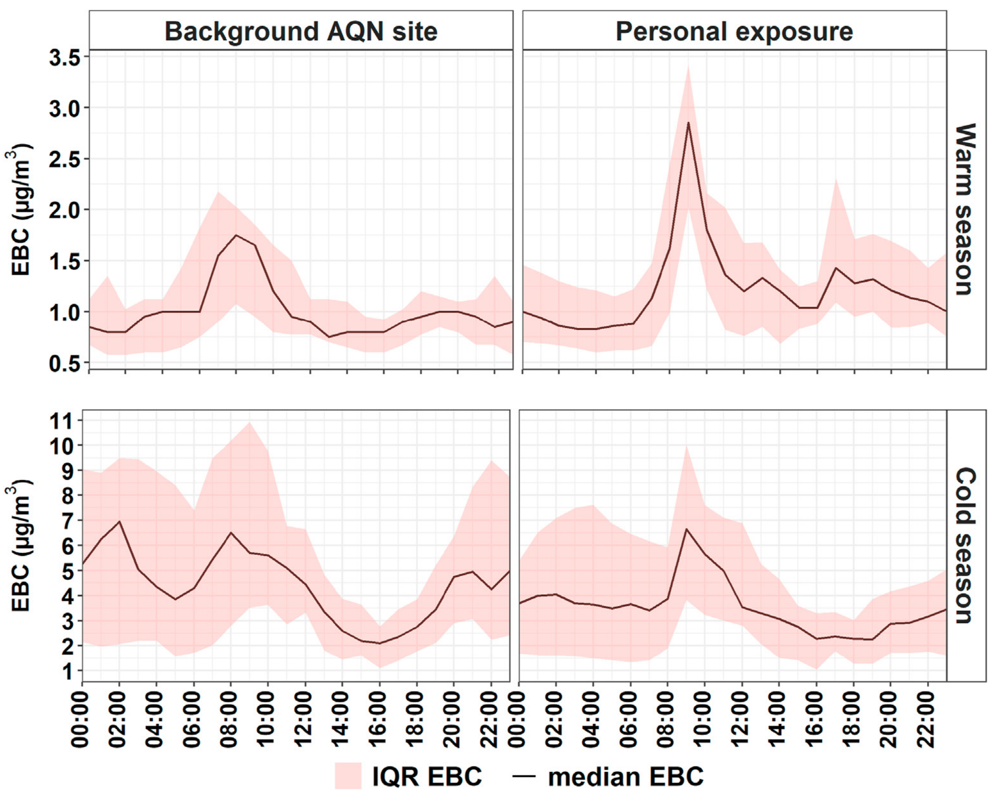

Fig. 2. Time trend of daily personal and AQN background site (ID station 10283, via Ponzio 34/6 – Pascal Città Studi, ~6 km from the school) EBC concentrations. Data are presented as warm and cold season median values (black line) and Inter Quartile Range (IQR) (red-shadowed area). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

linked with the highest mean and maximum personal EBC concentrations, while the lowest were related to Outdoor (other) and Indoor (other) MEs during the warm and the cold season, respectively.

In Fig. 2, the time trends by season of EBC concentrations measured by the AQN background site (ID station 10283) and by children are shown. A strong within-day variability was found with Morning Rush Hour represented the most critical daily time-window for EBC concentrations. Lowest values were generally detected during the afternoon and the night. However, during the cold season, an evening and night-time increment for both personal exposure and AQN site data was observed occurring together with the lowest detected mixing height (supplementary materials, S9). Trends were generally similar between personal exposure and AQN background site data, except for the steepness of peaks during the home-to-school (~8 a.m.) and school-to-home (~4 p.m.) commuting, especially during the warm season, and the presence of an evening peak of concentrations during the cold season that was detected only by the AQN site (Fig. 2).

The same seasonal pattern was found for the inhaled dose, with average values of 14.0 μg/day (range: 6.5–23.7 μg/day) and 39.2 μg/day (range: 5.6–83.9 μg/day) during the warm and the cold season, respectively. The overall mean inhaled dose was 27.8 μg/day (range: 5.6–79.7 μg/day). Finally, the between-day variability explained 78% (95% CI: 62–89%) and 93% (95% CI: 87–97%) of the total variability of the inhaled dose for the warm and cold seasons, respectively.

Considering the correlation between mean personal EBC

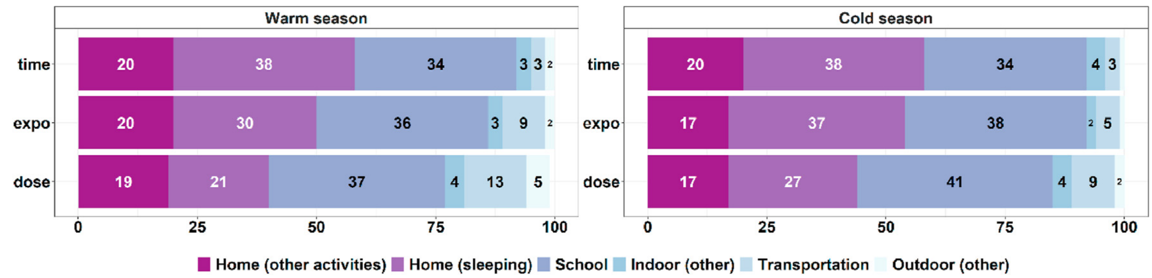

Fig. 3. Bar plots of the seasonal mean contribution (%) of different MEs to the daily time (time), daily integrated personal exposure (expo), and inhaled dose (dose). The numbers inside the boxes represent the relative contribution of MEs as percentage. Contributions <2% are not reported. Summary statistics of the relative contributions are reported in the supplement (S7).

concentration and inhaled dose, Pearson's r was 0.95 for the warm season and 0.98 for the cold season.

3.3. Contribution of MEs to time, daily integrated personal exposure, inhaled dose, and dose intensity

Fig. 3 shows the mean relative contribution of different MEs on daily time, daily integrated personal exposure, and inhaled dose. Summary statistics of both absolute values and relative contributions, as well as the results of the Mann-Whitney test for seasonal differences are reported in the supplementary materials (S10–S11). In particular, Home and School were always the main contributors, together accounting for 92%-86%–77% of the total daily time, personal exposure and inhaled dose in the warm season, and 92%-92%–85% in the cold season. Seasonal differences in time-related pattern were found only for the total time spent in Outdoor (other) and Indoor (other) MEs that were respectively the lowest and the highest during the cold season. The relative contribution of Home (sleeping) on the personal exposure was significantly higher in the cold season, while it was the opposite for Home (other activities), Outdoor (other) and Transportation. The relative contribution on total inhaled dose was significantly higher during the cold season when considering Home (sleeping) and School, while it was the opposite for Home (other activities), Outdoor (other) and Transportation. Finally, regardless of seasonality, Transportation and Outdoor (other) MEs strongly increased their influence when passing from time to personal exposure and inhaled dose, showing the highest dose/time intensity (supplementary materials, S12–S13).

3.4. Seasonal predictors of personal exposure to EBC and inhaled dose

Table 3 shows a summary of selected parameters for the six seasonal linear random intercept models. All potential predictors are reported in Table S7, together with their description and basic statistics. Altogether, the models explained from 71% up to 94% of the total variability (R^2_c) of personal exposure to EBC and inhaled dose. When focusing on the marginal effects of the explanatory variables (R^2_m), cold-season models were always the most explicative, except for the p99 warm-season model. In particular, when focusing on mean personal EBC concentrations (models I and II), the most predictive covariates (%R^2_m) were wind speed (Wind_speed, 41.7%), the time spent in a car (Time_car, 2.9%), the presence of parents who smoke (Smoke_parents, 1.7%), and the height of the home (Home_height, 0.6%) for the warm season, while the atmospheric mixing layer height (Hmixdia, 61.8%), the presence of parents who smoke (0.5%), and the distance of the house to the nearest road with traffic (Home_distance, 0.5%) were most predictive for the cold season. With regards to the 99th percentile personal EBC concentrations (models III and IV), the most predictive variables were the time spent in a car (23%), the time spent in home to school commuting (Time_homeschool, 5.5%), and the presence of parents who smoke (4.8%) for the warm season, while again the mixing layer height (24.3%), the time spent in home to school commuting (3.2%), and the presence of parents who smoke (0.9%) were most predictive for the cold season. Finally, regarding EBC inhaled dose (models V and VI), the most explicative covariates were wind speed (33.3%), time spent in a car (4.1%), and time spent at home (Time_home, 1.4%) for the warm season, while the mixing layer height (52.9%), the time spent in outdoor environment (Time_outdoor, 1.6%), time spent at home (0.5%), the presence of parents who smoke (0.4%), and the height of the home (0.2%) were most explicative for the cold season. Residuals complied with the normality assumption (Shapiro-Wilk test always >0.05), while a visual inspection of the studentized residuals versus the fitted values did not reveal homoscedasticity-related issues. The RMSE of the models were 2–5 times lower than the log-transformed standard deviations of the original data. The cross validation showed an R^2 ranging from 0.25 to 0.63, and higher RMSE if compared to the models, but lower than the standard deviations of the log-transformed original data.

4. Discussion

4.1. Participation, personal exposure to EBC, and dose

The participatory approach helped us to reach a high level of engagement, with a higher number of participants (+28%) in the second monitoring campaign compared to the first one. This higher response rate was probably due to: 1) the increasing level of engagement following the playful laboratories that were specifically planned right before the two monitoring periods, and 2) the positive experience of the first monitoring campaign and the following word of mouth among children and parents.

The overall personal EBC concentration (median = 1.8 $\mu g/m^3$) was comparable to the one reported by Donaire-Gonzalez et al. (2018) for 42 schoolchildren living in the industrial city of Sabadell, Spain (median = 1.7 $\mu g/m^3$), but it was higher than the one measured by Rivas et al. (2016) for 53 children in Barcelona (median = 1.1 $\mu g/m^3$).

In our study, we observed a strong seasonal contrast that was not found in literature: for instance, Paunescu et al. (2016) involved 98 schoolchildren in Paris, and found a geometric mean of EBC equal to 1.16 $\mu g/m^3$ and 1.64 $\mu g/m^3$ during warm and cold seasons, respectively. This is probably associated to the peculiar meteorology of the Po valley region. Indeed, during the cold season, the whole area experiences frequent stagnation events with thermal inversions, low atmospheric mixing layer heights, and consequent increases of air pollution concentrations (Caserini et al., 2017; EEA,

Table 3
Summary of parameters of the personal EBC mean concentrations, p99, and inhaled dose mixed-effect regression models. Only parameters of the selected covariates are shown.

	Personal EBC mean		Personal EBC p99		Potential inhaled dose	
	Warm (I)	Cold (II)	Warm (III)	Cold (IV)	Warm (V)	Cold (VI)
R^2_c [a]	0.82	0.94	0.71	0.90	0.82	0.92
R^2_m [a]	0.47	0.63	0.33	0.28	0.39	0.56
R^2_{LOOCV} [a]	0.38	0.61	0.28	0.37	0.25	0.60
RMSE[a]	0.10	0.13	0.21	0.14	0.10	0.12
$RMSE_{LOOCV}$ [a]	0.20	0.39	0.38	0.39	0.22	0.32
VIF[a]	1.06	1.03	1.14	1.01	1.07	1.02
Tot observations	66	85	65	87	66	76
Tot groups (days)	18	21	18	22	18	19
	estimate (95% CI)	**estimate (95% CI)**	**estimate (95% CI)**	**estimate (95% CI)**	**estimate (95% CI)**	**estimate (95% CI)**
Intercept	0.56 ** (0.45; 1.37)	13.26 *** (9.57; 16.95)	0.91 *** (0.69; 1.14)	7.81 *** (3.96; 11.66)	3.14*** (2.70; 3.49)	13.82 *** (10.02; 17.59)
Hmixdia		−6.38 *** (−2.94;-1.56)		−2.52 ** (−4.17;-0.86)		−1.85 *** (−2.56;-1.14)
Wind_speed	−0.76 ** (−1.11;-0.42)				−0.68** (−1.06;-0.027)	
Time_home					-3.35×10^{-4} (-8.14×10^{-4};-5.57×10^{-5})	-5.61×10^{-4} (−0.0011;-1.55×10^{-6})
Time_outdoor						0.16 ** (0.058; 0.26)
Time_car	0.0017 ** (1.77×10^{-4};0.0031)		0.0096 *** (0.0058; 0.013)		0.0018* (7.97×10^{-4};0.0039)	
Time_homeschool			0.016 * (0.0011; 0.032)	0.012 *** (0.0062; 0.018)		
Smoke_parents	0.080 * (0.010; 0.15)	0.088 * (0.0098; 0.17)	0.21 * (0.054; 0.36)	0.11 ** (0.030; 0.19)		0.077 (0.0023; 0.15)
Home_height	−0.0044 (−0.0087;-1.59×10^{-4})					
Home_road_distance		−0.0024 * (−0.0045;-3.08×10^{-4})				

*p < .05, **p < .01, ***p < .001.
[a] R^2_c = conditional R^2, R^2_f = marginal R^2, R^2_{LOOCV} = R^2 of the leave one out cross validation, VIF = highest Variance Inflation Factor, RMSE = Root Mean Square Error.

2018) (supplementary materials, S9 and S14). This finding is confirmed by the high winter personal EBC concentration (median = 3.8 μg/m^3) found by Buonanno et al. (2013) in 103 schoolchildren living in a similar environment (Liri valley) (ARPA Lazio, 2009). Given this picture, it is likely that meteorological variables are the main cause of the overwhelming contribution of the between-day component of variance on the total measured variability of personal EBC concentration.

Moreover, the correlation between average personal exposure and dose was very high (Pearson's R = 0.95 and 0.98). This result was expected since only a very small percentage of time was spent for those activities (transportation, sport etc) that could significantly increase the intake of air pollution. However, this could also be a consequence of the simplification we made by using standard ventilation rates and a small set of activities.

4.2. MEs contribution and dose intensity

In our study, the relative contribution of School and Home MEs to the daily integrated personal exposure to EBC (Fig. 3) is higher compared to previous studies. We found that school and home MEs contributed 58% and 34% to the personal exposure, while this was 50% and 32% in Barcelona (Rivas et al., 2016), and 50% and 22% in Seoul (Jeong and Park 2017). This is probably linked to the greater portion of time that our children spent in these environments. Additionally, a night-time peak was observed in Milan in the cold season. This phenomenon was likely linked to the combined action of traffic and heating system sources, as well as to the strong nocturnal thermal inversion that often occurs in the area (Vecchi et al., 2007), favouring the accumulation of pollutants in the lower layers of the atmosphere. This trend appears stronger for the AQN background site than the personal exposure data (Fig. 2) suggesting that outdoor EBC can only partially filter into indoor environments. On the contrary, the transportation related contribution was very small if considering time (3%), but much higher when focusing on exposure (7%) and dose (11%). The significant role played by transportation, especially focusing on peaks of exposure and dose (Fig. 3, supplementary materials S12–S13), is due to the proximity to the traffic source and the high ventilation rate associated with active modes of commuting (Zuurbier et al., 2009; Int Panis et al., 2010). These findings stress the urgency of mitigation actions to reduce traffic-related air pollution in the city of Milan. Finally, the lowest contributions and EBC concentrations were generally linked to indoor (other) and outdoor (other) MEs. This can be explained by the very restricted amount of time that children spent in these MEs, and by the time span of attendance that corresponded to the least critical daily time-window for EBC concentrations (late afternoon, Fig. 2).

4.3. Predictors of personal exposure to EBC and inhaled dose

Our most performant models were those focusing on mean exposure and dose for the cold season. In these cases, the great majority of the measured (or estimated) variability was explained by meteorological variables, and especially by the mixing layer height (hmixdia). This variable was reported to be one of the most important determinants of ambient concentrations of particulate matter (Miao et al., 2019), especially in the Po valley region (Bigi and Ghermandi 2016). Previously, Weichenthal et al. (2008), analysing personal exposure to ultrafine particles in transport microenvironments, found a strong collinearity between mixing layer height and wind speed, finally taking the latter in their regression analysis. Similarly, in our work the mixing layer height and wind speed were collinear, but only during the cold season ($R > 0.8$). However, since in this case the first was the most predictive, wind speed was not retained.

According to our findings, dose-related models were similar to the mean exposure models, except for the significant role played by the time spent at home (Time_home) and the time spent outdoor (Time_outdoor). They both suggest that the longer children stay outdoors, the more they increase their daily EBC dose. This is reasonable if we consider that children in outdoor MEs were involved in activities which imply high ventilation rates. Other important predictors that explained mean exposure and inhaled dose were the height of the home (Home_height) and its distance to the nearest road (Home_distance). This is probably linked to the peculiar atmospheric conditions that do not allow the dispersion of pollutants and thus increase the impact of local sources, such as traffic in proximity of the buildings.

Furthermore, in our study transportation-related variables were important predictors too, explaining the 99th percentile of personal EBC concentration. In this regard, it is important to highlight that peaks of exposure are particularly interesting when considering the trigger role that they can play on short-term health effects (Zhou et al., 2017). In particular, the important role played by the total time spent in a car (Time_car) was expected, since it is known that transportation MEs are more likely to cause peaks in personal exposure (Dons et al., 2019) and car trips may have a higher impact on personal EBC concentrations (De Nazelle et al., 2017; Dons et al., 2012). Additionally, the role of the time spent for home-to-school commuting (Time_homeschool) appears important as well, also pointing out the critical impact of traffic rush hours on children personal exposure to EBC. Finally, it is important to highlight that although we tested different transport-related variables, including time spent walking and other modes of home-to-school commuting, only the time spent in a car entered the models and was significantly linked to an increase of personal EBC concentration, especially when considering the warm season. These findings indicate that mitigation actions to reduce children's exposure to air pollution could focus on designing low-traffic home-to-school routes, and on promoting the use of non-motorized modes of transport. Moreover, our models confirm that environmental tobacco smoke (ETS) still is a matter of concern with regards to the health of children (Winickoff et al., 2009; Roberts et al., 2017).

In general, since in our dataset the between-day component of variance was much higher than the within-day variance, the overwhelming impact of meteorological variables was expected. These findings suggest that air pollution contributions from long-range transport and the urban background play a fundamental role in determining personal exposure to EBC in Milan. Furthermore, since temperature inversions and stagnation events in the Po valley are projected to increase due to climate change (Caserini et al., 2017), a drastic reduction of emissions is needed, and this calls for ambitious policies toward environmental protection in Northern Italy. Besides, to better understand the impact of local scale predictors, researchers should focus on how to maximize variability between participants and at the same time lowering the impact of urban and regional air pollution background, and time-related variability inside the sample.

4.4. Strengths, limitations, and further research

In our study a wide range of covariates was tested to explain schoolchildren's personal exposure to EBC, including meteorological conditions, traffic, personal behaviour, home characteristics, and family habits. Moreover, a special focus on seasonal patterns was given. This was achieved by recruiting schoolchildren attending the same school and living in the same area in Milan, in two seasonal monitoring campaigns lasting 5 weeks per season. It is possible that this intensive study design helped us in lowering the bias linked to the temporal and spatial variability that likely occur in similar experiences.

Our study has some limitations. For instance, we monitored only one working day for each season per child, and this may not be fully representative of the personal exposure profile. The study focused on a single pollutant, EBC, especially linked with traffic and biomass combustion; this confines the representativeness of the study to personal exposure to combustion-related air pollution. To deal with the lack of School EBC concentration data during the warm season, a significant number of EBC extrapolations have been made. By estimating about 14% of overall data, it is likely that an error was introduced, although the procedure we used to estimate missing data should have limited its magnitude. The monitor used to attribute EBC for each child while at school was placed in a room at ground level, while classrooms were located at the first floor. However, in all cases, windows overlooked the same courtyard reducing possible biases linked to the proximity of traffic. Moreover, some assumptions to complete the 24-h monitoring periods were made; in particular, time spent at the playground during school time was not considered, and an equal period of sleep was attributed to all the children due to lack of information in the time-activity diary. Especially focusing on the lack of data about the time spent in the playground, it is likely that in our study we underestimated personal EBC concentrations (and inhaled dose) during school-time since higher concentrations usually occurred outdoors, especially during the cold season, and children likely have higher ventilation rates there compared to activities inside school. Finally, most selection bias was avoided, as the recruitment of volunteers after the educational intervention reached an overall response rate of 77%, so it is likely that the sample was representative of the study area.

A possible next step for this kind of research should be diversifying the engaged schoolchildren and the study areas trying to be more representative of different socio-economic backgrounds. Besides, it would be interesting to investigate personal exposure of children using a sociological approach, by involving more closely children's parents and studying how their perceptions, knowledge and daily routines affect their children's exposure (Da Schio, 2020). Moreover, to study more in depth the determinants of exposure variability on a local scale, more efforts should be made in the attempt to disentangle the contribution of possible predictors from the overwhelming effect of meteorology by maximizing the within-observations variability and lowering the influence of regional and urban air pollution background. These next steps could help identifying new pathways toward more effective mitigation interventions.

5. Conclusion

In summary, our study aimed at extending knowledge on personal exposure and inhaled dose of schoolchildren to air pollution, and particularly EBC by focusing on seasonality, MEs and possible predictors. Our findings suggest that seasonality is linked to different patterns of personal exposure and dose, and especially to a different contribution of microenvironments. Mixed-effect models indicate that predictors vary depending on season and type of response variable (i.e. mean or peak exposure, or inhaled dose). In general, meteorological variables played a fundamental role in explaining variability, probably masking the real impact of other (local) predictors. Nevertheless, home location, transport and ETS-related predictors entered the model as well, suggesting that these variables could be important to refine exposure assessment methods in epidemiology. Besides, our findings suggest that maximizing variability between participants, lowering time-related variability inside the sample, and disentangling local and background air pollution, could help to better inform the effect of local mitigation actions. However, by highlighting the role of traffic rush hours and transport-related microenvironments on personal exposure, as well as by pointing out the role of home-to-school commuting in explaining peak exposure, our study suggests that reinforcing traffic-mitigation policies on a local scale could be an effective strategy to lower personal exposure to EBC and inhaled dose of children in the city of Milan.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.116530.

Main findings

Seasonal patterns in personal exposure to equivalent black carbon in elementary schoolchildren in the city of Milan were predicted by meteorological variables, traffic, and parental smoking.

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