Received: 18 July 2016 | Accepted: 17 November 2016
DOI: 10.1111/ina.12358

ORIGINAL ARTICLE

WILEY

Personal measurement of exposure to black carbon and ultrafine particles in schoolchildren from PARIS cohort (Paris, France)

A.-C. Paunescu[1] | M. Attoui[2] | S. Bouallala[3] | J. Sunyer[4] | I. Momas[1]

[1]Faculté de Pharmacie de Paris - Produits de Santé et Santé Publique, Université Paris Descartes, Paris, France

[2]Université Paris-Est Créteil Val de Marne Créteil, Île-de-France, France

[3]Service Evaluation de la Qualité de l'Air, ADEME Centre de Paris, Paris, Île-de-France, France

[4]Instituto de Salud Global Barcelona, Universitat Pompeu Fabra, Centro de Investigacion Biomedica en Red de Epidemiologia y Salud Publica, Barcelona, Catalonia, Spain

Correspondence
Isabelle Momas, Faculté de Pharmacie de Paris - Produits de Santé et Santé Publique, Université Paris Descartes, Paris, France.
Email: Isabelle.Momas@parisdescartes.fr

Funding information
Agence de l'Environnement et de la Maîtrise de l'Energie (ADEME), Grant/Award Number: Contract n° 1262C0010; French Agency for Food, Environmental and Occupational Health & Safety (ANSES), Research call Programme National de Recherche Santé Environnement Travail.

Abstract

This study aimed to measure in French children personal exposure concentrations of black carbon (BC) and ultrafine particles (UFP) and to quantify the contribution of different microenvironments (home, school, places of extracurricular activities, transport) to their total exposure. It was conducted on 96 9-year-old children from the PARIS birth cohort. BC and UFP were continuously measured by portable devices (microAeth® AE51 and DiSCmini®) for a minimum of 24 hours, while participating families simultaneously filled in a space-time-activities-budget questionnaire. BC exposure concentration was higher during trips (principally metro/train and bus), while UFP exposure concentration was higher during indoor activities (mainly eating at restaurants) and in trips. The most important UFP peaks were measured at home, especially during cooking. Home and school together accounted for much of the total exposure, 83.8% for BC and 85.3% for UFP. The contribution of transport to total exposure was 12.4% for BC and 9.7% for UFP, while extracurricular activities were responsible for 3.8% and 5% of the total exposure to BC and UFP, respectively.

KEYWORDS
black, carbon, exposure, particles, schoolchildren, ultrafine

1 | INTRODUCTION

Air pollution, especially traffic-related, plays a role in respiratory health and allergies, particularly in the development of symptoms and functional disturbances. Particles are one of the pollutants involved. The ultrafine particle fraction (UFP—particle size less than 100 nanometers) is a subject of recent interest, since these UFP can induce inflammatory effects[1] and oxidative stress[2] and contribute to airway remodeling in asthmatic patients.[3] Due to their concentration in terms of number per cm^3 of air, small diameter, and high surface area, UFP are not only able to transport other contaminants [organic compounds, such as polycyclic aromatic hydrocarbons (PAHs) and nitro-PAHs, small amounts of sulfates, nitrates, metals, and other elements in trace form[3]], but also to contribute with a fairly high efficiency to their deposition in alveoli.[4] The large contact area of the UFP is an important determinant of the reactivity of these particles. Their lung deposition surface area (LDSA) has been associated with acute lung inflammatory responses.[5] Negative associations were found between changes in pulmonary function tests and estimate of the individual daily dose deposited on alveolar surface area in children with allergic rhinitis, asthmatic, or sensitive to allergens.[6] Because of their characteristics, UFP are suspected to be the most damaging component of traffic-related air pollution for human health.[7] In addition, there are several domestic sources of UFP, such as cooking, heating and smoking.[3,8,9]

Black carbon (BC), constituent of $PM_{2.5}$ fine particulate matter (particles with an aerodynamic diameter less than 2.5 μm) resulting from the incomplete combustion of biomass and oil/diesel, is also suspected to be responsible for particle toxicity. However, toxicological studies *in vivo* and *in vitro*, although few, suggest that BC could not exercise its toxicity directly but through pollutants that it carries.[10] In any case, Janssen et al.[11] encouraged the use of this indicator in the study of adverse health effects caused by traffic-related air pollution.

Personal exposure to UFP and BC is poorly documented. Not long ago, UFP and BC concentrations could only be measured by stationary monitors measuring the outdoor/indoor air of homes or schools, which were not able to record the true subjects' exposure in all the microenvironments they visited during the day. These stationary measurements could lead to misclassification of the individual exposure. Recently, small portable devices that are easy to handle have been developed. The great advantage of these new devices is that measurements are taken closer to the individual's airway, and they can track human movements thanks to the autonomy provided by internal batteries. Daily personal measurements of BC and UFP exposure concentrations can thus be successfully made in all microenvironments, specifically on children.[6,12–17]

The work reported in this paper was carried out as part of an epidemiological study on respiratory health in 9-year-old children from the PARIS (Pollution and Asthma Risk: an Infant Study) cohort. This exposure study aimed to (i) measure personal individual BC and UFP exposure concentrations in a sample of 9-year-old children from the PARIS birth cohort; (ii) identify levels of BC and UFP measured in the main living environments (home and school); and (iii) quantify the contribution of different life microenvironments (home, school, places of extracurricular activities, transport) to total exposure to BC and UFP in children.

Practical Implications

- Thanks to portable devices, this study provides information on personal daily exposure concentrations of black carbon (BC) and ultrafine particles (UFP) in schoolchildren, especially in four microenvironments: home, school, place of extracurricular activities and transport. Exposure levels varied between microenvironments, transport being responsible for the greatest measured concentration. The most important UFP peaks were measured at home, due to household activities, mainly cooking. This study demonstrates the usefulness of BC and UFP indicators: the BC indicator measuring combustion sources—especially traffic-related, being more relevant outside, and the UFP indicator proving to be relevant in indoor environments.

2 | MATERIALS AND METHODS

2.1 | Study population

The study was conducted in a sample of 9-year-old children issued from a population-based birth cohort, the PARIS cohort, living in inner Paris (department 75) or in the suburbs (departments 77-78 and 91-94),[18] and invited to participate in a health examination between June 2014 and December 2015. Among the 141 solicited children, 96 agreed to participate in the BC-UFP-PARIS Study. The parents gave their written consent before measuring devices were provided, after being informed of the terms of the study by phone and an explanatory booklet.

2.2 | Study design

Portable devices continuously measured BC and UFP for a minimum of 24 hours prior to the health examination. The measurements were conducted during the weekdays and covered the child's time at home, in school, extracurricular activities, and trips.

In our study, exposure to BC and UFP is an estimate from concentrations measured by portable devices placed near the respiratory airways of participants (if they are moving with the backpack) or placed in the same room (at home and school) and is defined as quantity (mass, μg) for BC and particle number, for UFP per volume unit of air (m^3 or cm^3, for BC and UFP, respectively), expressed per unit of time (1 minute). While measurements were being carried out, participating families received an ongoing questionnaire containing a space-time-budget, and questions concerning the activities of their children and some of their habits at home (cooking, cleaning, smoking, etc.). The day of the health examination, the completed space-time-activities-budget questionnaire, and the measurement equipment were recovered. A week after, each participating family received a compilation of their child's BC and UFP measurements. All home addresses were geocoded and mapped. Google Maps estimates, made by plotting the walking distance between home and school address, and between home and the hospital where the health examination was held, were performed for each participant.

2.3 | BC and UFP measurements

2.3.1 | Measurement devices

To measure BC and UFP exposure concentrations, two portable devices were installed in an easy to handle and carry backpack. Three sets of instruments have been used alternately all along the study period. The **microAeth® AE51** (AethLabs, San Francisco, USA; the notation "MA" refers in this paper to the microAeth monitor) device measures the concentration of BC by the conventional method of absorption of infrared radiation. Particulate sampling and analysis are done in real time. A measurement per minute at a sampling flow rate of 100 mL/min was selected. The instrument measures 11.7×6.6×3.8 cm, weighs 250 g, and has an internal rechargeable battery with autonomy >24 hours for the selected flow rate. The measured concentration is given in nanograms BC per cubic meter (ng/m^3) of air, the measuring range being between 0 and 1 mg/m^3. According to the manufacturer, measurement accuracy is ±0.1 μg BC/m^3 at 1 minute average sampling and 150 mL/min flow rate[19]. The BC concentrations measured in ng/m^3 were expressed in this paper in $μg/m^3$.

The **DiSCmini®** (Diffusion Size Classifier miniature conceptualized by Matter Aerosol, Wohlen, Switzerland, and commercialized by Testo SE & Co. KGaA, Titisee-Neustadt, Germany; notation "DM" in this paper) is a battery-powered instrument that simultaneously measures the number of particles per cm^3 of air [from 1000 to 1 000 000 particles (pt)/cm^3], the mean (modal) diameter of particles [from 20 to 300 nanometers (nm)], and the surface coming into contact with the pulmonary mucosa [lung deposition surface area—LDSA ($\mu m^2/cm^3$)]. The DM is a diffusion battery based on unipolar charging of the aerosol, followed by detection in two electrometer stages. In the first diffusion stage, particles are deposited by diffusion. The resulting electrical current is measured with a sensitive electrometer. The remaining particles are collected in a second filter stage, and again, the current is measured. Small particles undergo larger Brownian motion, have a larger diffusion coefficient, and therefore are more likely to be collected in the diffusion stage, while larger particles are more likely to end up in the filter stage. From the calibration curve stored on the instrument, the particle size is determined, and then the particle number can easily be calculated[20,21]. The DM measures 18×9×4 cm, weighs less than 700 g, and has a rechargeable battery with autonomy of 8 hours.[21] To have the assurance that DM can operate about 24 hours, parents and teachers were asked to charge the device at 220V for the duration of the measurements at home or in the school. Participants used the internal battery only while commuting or engaging in activities. The three parameters are recorded continuously every second. For the purposes of our study, an average per minute was calculated. According to the manufacturer, the accuracy of the instrument is around 30%.[21]

Both MA and DM devices are equipped with memory cards, allowing the recovery of records and processing of data by computer, and both instruments were synchronized. Before each use in the field, the instruments were checked to ensure that they worked properly as indicated by the manufacturers (for MA, we verified the operating parameters by computer, and for DM, we verified if the current on each stage was "reasonably stable" and <2 fA and that the pump functioned properly). Concerning device maintenance, MA quartz filters were changed after each use to avoid saturation and subsequent measurement bias. DM impactors were cleaned after each use in an ultrasonic bath (Branson Model 1800, Fisher Scientific Co. L.L.C., Pittsburgh, PA, USA), in water filtered through a reverse-osmosis membrane for a period of one hour. The Tygon tubes were cleaned with 70% ethanol and dried in pure air, using a compressor (JUN-AIR 2000-25M; JUN-AIR Gast Manufacturing, Inc., Benton Harbor, Michigan, USA). After each month of experimental measurements, the instruments were reset, running them for one hour in a pure generated atmosphere.

2.3.2 | Laboratory validation experiments

Before planning BC and UFP measurements using MA and DM devices in our exposure study, we carried out laboratory validation experiments. Each device (MA and DM) and a reference instrument, a condensation particle counter (TSI Inc., Shoreview, MN; notation CPC), were exposed to the same atmosphere of monodispersed aerosol issued from a generator of poly-dispersed aerosol (standard diameters and concentrations) coupled downstream to a differential mobility analyzer (TSI SMPS 3081, notation DMA) used as classifier. Briefly for generated atmospheres of BC particles with four diameters (average of 30, 50, 70, and 100 nm), BC concentrations measured by MA were highly correlated with those calculated from the number of particles counted by the CPC (Pearson correlation coefficient r=.996, Spearman correlation coefficient r=.98, P<.0001); their precision (mean relative difference from the reference instrument) was 19.4%. The computed precision of the measured values by DM for aerosols containing a different number of particles per cm^3 of air with eight particle diameters varying between 30 and 100 nm was 24.8%. The DM has the ability to "respond" almost instantly to a sudden change (i) in number of particles per cm^3 of air and (ii) in mean diameter of the particles contained in the exposure aerosol. In both cases, only a few seconds (less than 10 seconds) was sufficient so that the DM could detect and record the new values. Details about method and main results are provided in Appendix S1.

2.3.3 | Field application of the study design to adult volunteers

Following the laboratory validation, we conducted a pilot study applying the design of the study planned for children to a group of 15 adult volunteers from the Paris area in order to ensure the correct procedures. Volunteer measurements were performed with the same devices for a longer period than that proposed for children (about 30 hours), in four microenvironments: home, work–office, places of different activities, and transport. Details about this field application and the main results are provided in Appendix S2. The proposed procedures were feasible in the field and applicable to the general population.

2.3.4 | Statistical analysis

The normality of BC and UFP distributions was tested and log-transformed, if necessary. Descriptive statistics of BC and UFP were provided [arithmetic mean (AM), standard deviation (SD), geometric mean (GM), median and range] for both total duration of measurements (about 24 hour) and time spent in each microenvironment (school, home, places of extracurricular activities, and transport).

We analyzed the details of each microenvironment. As the period when the child was at school ("day 1" and/or "day 2") and the duration of his or her presence varied from one child to another, a stricter temporary choice was proposed ("day 1" 8:30 to 11:30 a.m. and 2:00 to 4:30 p.m. and "day 2" 8:30 to 11:30 a.m.). Exposure concentrations at home were investigated during four specific time periods between 6:30 p.m. ("day 1") and 8:00 a.m. ("day 2") when children were constantly present at home ("day 1" 6:30 to 9:30 p.m. and 9:31 to 0:00 p.m.; "day 2" 0:01 to 6:30 a.m. and 6:31 to 8:00 a.m.). Exposure concentration during extracurricular activities was investigated by type of activity: "indoor" *versus* "outdoor." Exposure concentration during journeys was studied according to the purpose and destination of travel [(i) commuting school–home; (ii) journey to the hospital, and (iii) journey to another destination], and by transport modes [seven types have been described: (i) walking, (ii) bike or scooter, (iii) car or taxi, (iv) bus, (v) tramway, (vi) subway or train, and (vii) combined (several modes)]. Pearson and Spearman correlation coefficients were calculated between quantitative variables, especially concentrations of BC and UFP.

In order to compare average exposure concentrations between indoor and outdoor extracurricular activities, the Student's *t*-test was used while the comparison between average values at two time slots at school was tested by paired Student's *t*-test. Comparisons of average concentrations by trip destinations and transport modes were realized by variance analysis and between the four time periods spent at home, by variance analysis with repeated measurements. In the case of a significant time effect at home, and a significant trip destination effect, comparisons between groups 2 by 2 using Bonferroni correction were made. In case of a significant transport mode effect, comparisons between walking considered as the reference and each mode were performed using Dunnett's test.

The contribution (%) of each microenvironment (school, home, activities, transport) to the total exposure to BC and UFP was assessed. Comparisons of mean contributions by season ("warm" spring–summer and "cold" autumn–winter), geographical location (inner Paris and suburbs), or scholar calendar (school and vacation periods) were realized by Student's *t*-test. A *P*-value <.05 in bilateral situation was considered statistically significant. The statistical software was Stata 11 (StataCorp LP, College Station, Texas, USA).

3 | RESULTS

3.1 | Characteristics of participants

The study was conducted in 96 children for 63 days (43 days during the school period and 20 days during holidays) over 50 weeks, with 1, 2, or 3 children seen per week (29 in spring, 18 in summer, 31 in autumn, and 18 in winter). The average age of the 96 participants was 9.0 years (SD 0.2 years; median 9 years). Other characteristics of the participants are given in Table 1. The map distribution of participants' home addresses is given in Figure 1.

3.2 | BC and UFP measurements and space-time-activities-budget

BC was satisfactorily measured in 95 of 96 children; a technical problem with the MA device prevented the measurement for one child. Of the total of 154 959 theoretical recorded minutes in the 95 children, only 0.05% (83 minutes) had to be removed, because they represented outlier values of BC. UFP were satisfactorily measured in 83 of the 96 participants, and the lack of UFP data for 13 participants was due to technical problems with the DM devices. Of the total of 135 097 theoretical minutes of measurements in the 83 children, 0.76% (1031 minutes) were removed because of measurement "error codes."

The measurements were performed in the four microenvironments: school, home, places of extracurricular activities, and trips. They started for 68 children (70.8%) at school, for 22 children (22.9%) at home, for 5 children (5.2%) in the city while traveling, and for one

TABLE 1 Characteristics and activities of participants in the BC-UFP-PARIS Study (n = 96)

Characteristics		
Children		
		Mean (SD)[a]
Age (years)		9 (0.2)
		Number (%)
Sex	Female	45 (46.9)
	Male	51 (53.1)
School participation ("days" 1 and 2)	Yes	75 (78.1)
	No	21 (21.9)
Extracurricular activities participation ("days" 1 and 2)	Yes	45 (46.9)
	No	51 (53.1)
Transport ("days" 1 and 2)		
Commuting school↔home	Yes[b]	75 (78.1)
	No	21 (21.9)
Journey(s) to another destination[c]	Yes[d]	38 (39.6)
	No	58 (60.4)
Journey to the hospital[e]	Yes	95 (99)
	No	1 (1)
Families		
		Mean (SD)[a]
Number of inhabitants at home		4 (1)
		Number (%)
Geographical location of the home	Paris intramural	53 (55.2)
	Paris suburbs	43 (44.8)
Socioeconomic level[f]	"Low"	2 (2.1)
	"Medium"	36 (37.5)
	"High"	58 (60.4)
Parents' activities during the measurement period		
Smoking ("days" 1 and 2)	Yes[g]	9 (9.4)
	No	79 (82.3)
	Not interviewed	8 (8.3)
Cleaning the house ("days" 1 and 2)	Yes	19 (19.8)
	No	69 (71.9)
	Not interviewed	8 (8.3)
Burning candles, incense, or chimney fire ("days" 1 and 2)	Yes	6 (6.3)
	No	82 (85.4)
	Not interviewed	8 (8.3)
Cooking in the evening of "day 1"	Yes	78 (81.3)
	No	10 (10.4)
	Not interviewed	8 (8.3)
Type of stove used in the evening of "day 1"[h]	Gas	25 (26)
	Electric	53 (55.1)
	Not relevant[i]	10 (10.4)
	Not interviewed	8 (8.3)

(Continues)

TABLE 1 (Continued)

Characteristics		
Using an airing way during the preparation of the meal in the evening of "day 1"	Yes[j]	42 (43.8)
	No	36 (37.5)
	Not relevant	10 (10.4)
	Not interviewed	8 (8.3)

[a]Standard deviation.
[b]The 75 children that commuted (school↔home) during the two days realized a mean of 1.5 trips (SD 0.6 trips, median 1 trip, range 1-3 trips).
[c]Journey(s) to another destination except the journeys to the school and to the hospital.
[d]The 38 children that moved to another destination during the two days realized a mean of 1.9 trips (SD 0.8 trips, median 2 trips, range 1-5 trips).
[e]Journey to the hospital: One participant (1/96) did not make the trip to the hospital with the backpack.
[f]The highest socioeconomic level of parents (mother and father): "low"—unemployed, employed with low income, workers; "medium"—artisans, merchants; "high"—executives or business leaders.
[g]Smoke: electronic cigarettes (*n*=5) and cigarettes filter (*n*=4); smoke outside on the balcony (*n*=8) and inside the flat (*n*=1 e-cigarette).
[h]Of the 78 families who cooked in the evening of "day 1," 53 had used an electrical stove (electric plate, induction, or ceramic stove) and 25 used a gas stove.
[i]Not relevant because they did not cook.
[j]Among the 78 families, 22 kept the window open, 10 used the suction hood, 7 used both modalities at once (open window and suction hood), and 3 switched on the mechanical ventilation.

child (1.1%) at the beginning of an extracurricular activity. On "day 1," the backpack was given to 48 children (50%) in the morning, to 32 children (33.3%) before noon, and to 16 children (16.7%) in the afternoon. On "day 2," it was recovered in the hospital for 12 children (12.5%) in the morning, for 83 children (86.5%) in the afternoon, and for one child (1%) later in the afternoon. The average length of the monitoring periods was nearly 27 hours during which the children spent on average more than two-thirds of the time at home (65.8%) and to a lesser extent, at school (23.7%), in transport (5.2%), and in extracurricular activities (5.3%). Of the 96 children, 49 (51%) attended school the two days, 26 (27.1%) only one day, while 21 (21.9%) did not attend because their participation in the study was planned during holidays. Ten children had extracurricular activities on the two measurement days; 35 participated on a single day (24 on the first day and 11 the second day). Among the 74 activities realized over the two days, 55 (74.3%) were "indoor" activities and 19 (25.7%) were "outdoor" activities. The "indoor" activities consisted of attending courses (*n*=9), practice of physical activity (*n*=6), visits to family members or physicians (*n*=22), attendance at museums or library (*n*=3), eating in restaurants (*n*=11), and supermarket or mall shopping (*n*=4). The "outside" activities corresponded to a walk in a park or forest (*n*=12), gardening (*n*=2), and adult accompaniment in travels (*n*=5).

During the two days, all 96 children made at least one trip with the backpack. A total of 344 trips were carried out: 155 commuting between school and home, 94 performing extracurricular activities ("other" journeys), and 95 journeys going to the hospital for the health examination. Overall, the most frequent transport mode was walking (55.2%), followed by car or taxi (15.1%), subway or train (12.2%), bus (7%), bike or scooter (4.1%), and tramway (2%). A percentage of 4.4% of trips was performed by several modes (Figure 2).

As shown in Figure 2, the transport modes differed depending on their purpose; walking was the predominant mode in all trips except for going to the hospital where participants travelled by public transport or by car. The shortest trips were those corresponding to commuting from home to school and *vice versa* (mean 871 m; SD 1 198 m, median 450 m; range 36-6600 m), while the longest trips were those to the hospital (mean 6779 m; SD 6433 m, median 4600 m; range 800-41 100 m).

3.3 | BC and UFP exposure concentrations

They were log-transformed. BC and UFP exposure concentrations over the total monitoring period were correlated (*r*=.31, *P*=.005, *n*=82; Appendix S3). Descriptive statistics regarding the concentrations of BC (*n*=95) and UFP (*n*=83) are given in Tables 2 and 3, respectively, overall for the entire duration of the measurements and separately by microenvironment. In the same way, descriptive statistics regarding the LDSA are given in Appendix S4. The highest overall geometric means of BC and UFP were measured during journeys of participants (3.02 μg/m^3 and 23.3×10^3 pt/cm^3, respectively) (Tables 2 and 3).

3.4 | BC and UFP exposure concentrations by attended microenvironment

3.4.1 | School

The number of children attending school during the three time slots under consideration varied, almost half being present in at least two of them (*n*=50 for BC measurements and *n*=44 for UFP measurements). At school, BC and UFP were correlated (Pearson coefficient *r*=.60, *P*<.0001, *n*=64). However, the average BC concentrations measured in the morning of "day 1" and "day 2" were significantly higher than those in the afternoon of "day 1" (*P*<.001). No difference between time slots was evidenced for UFP (Appendix S5). Relative to seasonality, only for BC, a difference at the limit of significance (*P*=.048) was observed between levels measured in the cold seasons (GM of 1.61 μg/m^3) than in warm seasons (1.27 μg/m^3). BC was also significantly different (*P*=.03) in schools located in inner Paris (GM of 1.59 μg/m^3) compared with schools located in the suburbs (1.22 μg/m^3).

3.4.2 | Home

BC and UFP at home differed according to the four considered time periods (Tables 2, 3, and Appendix S5). BC exposure concentrations measured between 6:30-9:30 p.m. and 9:31-0:00 p.m. were significantly higher than those of time slots 0:01-6:30 a.m.

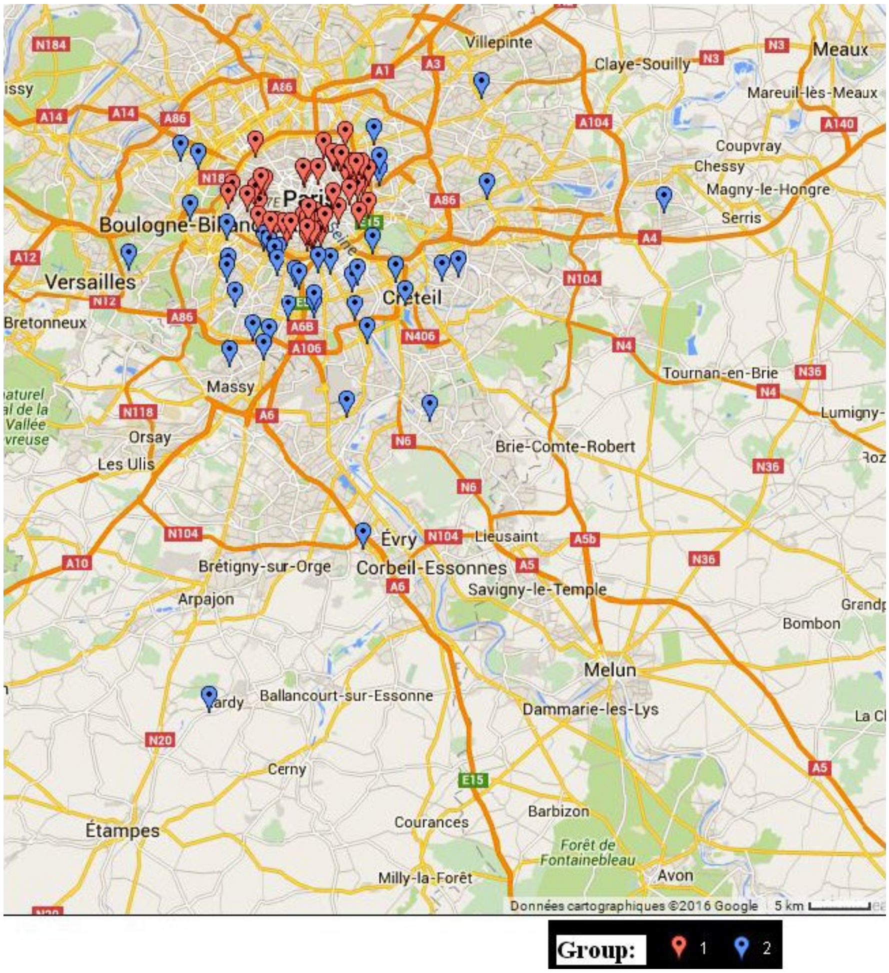

FIGURE 1 Geographical location of home addresses of participants in the BC-UFP-PARIS Study (n=96). Group 1: inside Paris (department 75); Group 2: Paris suburbs (departments 77-78 and 91-94)

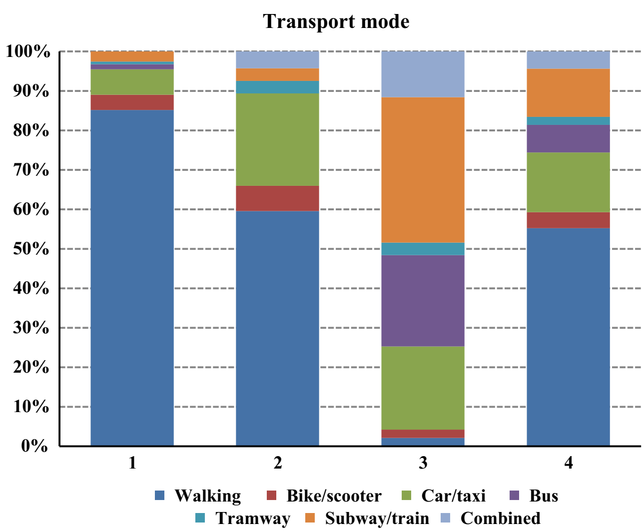

FIGURE 2 Percentage of each transport mode by type of journey for participants in the BC-UFP-PARIS Study (n=96). 1, Commuting school–home; 2, Journeys to another destination (except school and hospital); 3, Journey to the hospital; 4, All journeys

and 6:31-8:30 a.m. (P<.0001 and P<.01, respectively; Appendix S5), the lowest average being that measured in the night, between 0:01 and 6:30 a.m., during sleeping (Figure 3A). For the UFP, the mean values declined drastically from 6:30 p.m. to 6:30 a.m. (Figure 3B), significant differences being measured among all time slots (Figure 3B; Appendix S5). At home, BC and UFP were also correlated (r=.30, P=.007, n=82). UFP between 6:30 p.m. and 8:00 a.m. were correlated with domestic activities, such as preparing dinner (r=.36, P=.002, n=77), using a stove for cooking (r=.35, P=.002), and burning candles, incense, or making a fire in a chimney (r=.25, P=.03), whereas no correlation was observed with BC. Regarding seasonality, only for BC, a significant difference (P<.0001) was observed between the average measured during the hot and the cold season (GM of 0.91 μg/m^3 and 1.44 μg/m^3, respectively).

3.4.3 | Extracurricular activities

Descriptive statistics regarding BC and UFP are shown by groups of activities (indoor or outdoor) in Table 4. Average BC and UFP (pt/cm^3 and LDSA) values were not statistically different between the two groups, although measurements showed a high variability within groups. Among the indoor activities, the highest levels of BC and UFP were measured in restaurants (mostly fast food) attended by participants (Table 4). BC concentrations and number of UFP during extracurricular activities were correlated (r=.42, P=.01, n=37).

TABLE 2 Black carbon exposure concentrations (μg/m^3) measured in the BC-UFP-PARIS Study participants (n=95[a])

Microenvironment and period of measurements	Number of participants	Duration (minutes) of measurements Mean (SD)[b]	Black carbon (μg/m^3) Arithmetic Mean (SD)[b]	Median	Range	Geometric Mean
Total School “day 1” and “day 2”	74	496 (153)	1.62 (0.86)	1.52	0.38-5.14	1.43
School “day 1”	73	374 (138)	1.58 (0.93)	1.46	0.38-5.14	1.35
Day 1: 8:30-11:30 a.m.	38	181	2.13 (1.70)	1.76	0.46-8.26	1.71
Day 1: 2:00-4:30 p.m.	72	151	1.29 (0.61)	1.17	0.40-2.90	1.15
School “day 2”	47	200 (35)	1.77 (0.89)	1.74	0.38-3.82	1.54
Day 2: 8:30-11:30 a.m.	47	181	1.77 (0.92)	1.76	0.38-4.04	1.53
Total Home “day 1” and “day 2”	95	1 105 (227)	1.30 (0.64)	1.15	0.30-3.29	1.15
Day 1: 6:30-9:30 p.m.	94	181	1.50 (0.92)	1.28	0.37-4.62	1.28
Day 1: 9:31-0:00 p.m.	95	150	1.43 (0.89)	1.18	0.25-4.62	1.19
Day 2: 0:01-6:30 a.m.	95	390	1.00 (0.68)	0.82	0.16-3.45	0.79
Day 2: 6:31-8:00 a.m.	95	90	1.19 (0.82)	1.01	0.13-5.99	0.97
Total Activities “day 1” and “day 2”	43	119 (97)	1.86 (1.11)	1.60	0.29-4.75	1.54
Activities “day 1”	32	104 (87)	1.61 (1.01)	1.27	0.29-4.35	1.36
Activities “day 2”	19	98 (75)	2.15 (1.25)	2.04	0.41-4.75	1.78
Total Journeys “day 1” and “day 2”	95	84 (35)	3.23 (1.21)	2.99	1.13-6.71	3.02
Journey “day 1”	83	24 (20)	2.77 (1.71)	2.26	0.62-8.01	2.29
Journey “day 2” (except the journey to hospital)	56	26 (17)	2.90 (1.83)	2.41	0.92-10.15	2.46
Journey to hospital “day 2” (only)	94	48 (20)	3.23 (1.33)	2.93	0.99-6.38	2.96
Total exposure concentration “day 1” and “day 2”	95[a]	1630 (106)	1.50 (0.61)	1.44	0.47-3.27	1.38
Geographical location[c]						
Inner Paris	52	1633 (111)	1.54 (0.61)	1.48	0.52-3.27	1.42
Suburbs	43	1627 (102)	1.46 (0.62)	1.35	0.47-3.06	1.33
Season[d]						
Hot (spring–summer)	47	1628 (106)	1.27 (0.56)	1.16	0.47-3.06	1.16
Cold (autumn–winter)	48	1633 (108)	1.73 (0.58)	1.67	0.56-3.27	1.64
School calendar[e]						
Vacation	21	1630 (96)	1.43 (0.45)	1.49	0.52-2.28	1.35
School period	74	1630 (110)	1.52 (0.65)	1.37	0.47-3.27	1.38

[a]One participant (1/96) did not have BC measurements because of technical problems with the device.
[b]SD: standard deviation.
[c]BC: comparison between groups (Student’s t-test) P=.49.
[d]BC: comparison between groups (Student’s t-test) P<.0001.
[e]BC: comparison between groups (Student’s t-test) P=.84.

3.4.4 | Trips

BC and UFP are shown in Table 5, by type of trips according to their destination and travel modes. The highest exposure concentrations of BC and UFP were measured during journeys to the hospital and to the places of extracurricular activities. On average, BC concentrations during journeys to the hospital were significantly different from those measured when commuting between school and home (P=.0004). The UFP measured during journeys to the hospital and to places of extracurricular activities significantly differed from those corresponding to commuting school–home, in terms of number of pt/cm^3 (P<.0001 and P=.015, respectively) and LDSA (P=.0002 and P=.002, respectively) (Appendix S4 and S5). Among the transport modes, walking offered a low average exposure to BC and UFP, statistically comparable to that of the tramway and bike/scooter (Appendix S5). The highest averages were measured during trips by subway/train, followed by bus and by car/taxi for BC, and by bus and subway/train in terms of number pt/cm^3 (Table 5). Compared to walking, mean exposures in subway/train and bus were statistically different (BC: subway/train P=.0001 and bus P=.036; pt/cm^3:

TABLE 3 Ultrafine particles exposure concentrations (×10³ particles/cm³) measured in the BC-UFP-PARIS Study participants (n=83[a])

Microenvironment and period of measurements	Number of participants	Duration (minutes) of measurements	Ultrafine particles (×10³ pt/cm³)			
		Mean (SD)[b]	Arithmetic Mean (SD)[b]	Median	Range	Geometric Mean
Total School "day 1" and "day 2"	65	492 (149)	9.4 (3.5)	8.8	3.0-16.5	8.7
School "day 1"	64	374 (139)	9.6 (3.6)	8.7	3.1-17.3	8.9
Day 1: 8:30-11:30 a.m.	33	181	9.3 (4.0)	8.1	2.4-22.0	8.5
Day 1: 2:00-4:30 p.m.	63	151	9.7 (4.5)	8.2	3.4-24.5	8.8
School "day 2"	41	196 (30)	8.7 (4.1)	8.4	2.9-19.3	7.8
Day 2: 8:30-11:30 a.m.	41	181	8.7 (4.3)	8.0	2.7-18.9	7.7
Total Home "day 1" and "day 2"	83	1098 (223)	14.6 (8.1)	13.0	2.7-45.4	12.8
Day 1: 6:30-9:30 p.m.	82	181	29.9 (32.0)	18.7	2.8-16.4	19.4
Day 1: 9:31-0:00 p.m.	83	150	13.9 (11.1)	11.3	3.2-79.1	11.2
Day 2: 0:01-6:30 a.m.	83	390	57.1 (2.8)	5.6	0.9-13.4	5.0
Day 2: 6:31-8:00 a.m.	83	150	13.3 (21.1)	7.6	1.4-175.3	8.3
Total Activities "day 1" and "day 2"	37	116 (104)	28.0 (45.6)	15.8	3.4-272.8	17.5
Activities "day 1"	27	107 (93)	16.1 (10.2)	13.4	3.4-47.2	13.5
Activities "day 2"	15	94 (74)	48.9 (69.2)	21.3	4.3-272.8	26.1
Total Journeys "day 1" and "day 2"	83	81 (38)	24.9 (9.7)	24.1	9.8-60.4	23.3
Journey "day 1"	72	25 (21)	20.6 (12.3)	17.9	4.5-74.3	17.5
Journey "day 2" (except the journey to hospital)	49	25 (17)	26.6 (11.6)	24.7	9.9-78.6	17.4
Journey to hospital "day 2"	79	46 (22)	27.9 (12.0)	25.8	10.4-78.6	25.7
Total exposure concentration "day 1" and "day 2"	83	1615 (101)	14.3 (6.3)	12.4	5.3-38.4	13.2
Geographical location[c]						
Inner Paris	46	1609 (100)	15.2 (6.7)	13.7	5.3-38.4	13.9
Suburbs	37	1623 (102)	13.2 (5.6)	11.9	7.1-33.2	12.3
Season[d]						
Hot (spring–summer)	43	1613 (106)	14.6 (7.0)	12.7	5.3-38.4	13.3
Cold (autumn–winter)	40	1618 (96)	14.0 (5.4)	12.3	6.2-29.2	13.1
School calendar[e]						
Vacation	18	1628 (96)	16.1 (8.7)	14.1	6.2-38.4	14.2
School period	65	1612 (102)	13.8 (5.4)	12.3	5.3-29.2	12.9

[a]Thirteen participants (13/96) do not have UFP measurements because of technical problems with the device.
[b]SD: standard deviation.
[c]UFP: comparison between groups (Student's t-test): P=.18.
[d]UFP: comparison between groups (Student's t-test): P=.88.
[e]UFP: comparison between groups (Student's t-test): P=.37.

subway/train and bus, P<.0001, Appendix S5). BC and UFP during trips were correlated (r=.47, P<.0001, n=82).

3.5 | Contribution of microenvironments to the total exposure to BC and UFP

Home and school together accounted for much of the total exposure, 83.8% for BC and 85.3% for UFP. The contribution of transport to total exposure was 12.4% for BC and 9.7% for UFP, while extracurricular activities were responsible for 3.8% and 5% of the total exposure to BC and UFP, respectively (Figure 4A and B).

A different pattern of contributions was observed in children who participated during school *vs.* holiday periods (BC Figure 4C, E and UFP Figure 4D, F). The differences between school and vacation periods were significant for both BC and UFP, at school, home, and during extracurricular activities. During vacation, compared to school periods, children spent more time at home (84.3% vs. 60.8%) and during

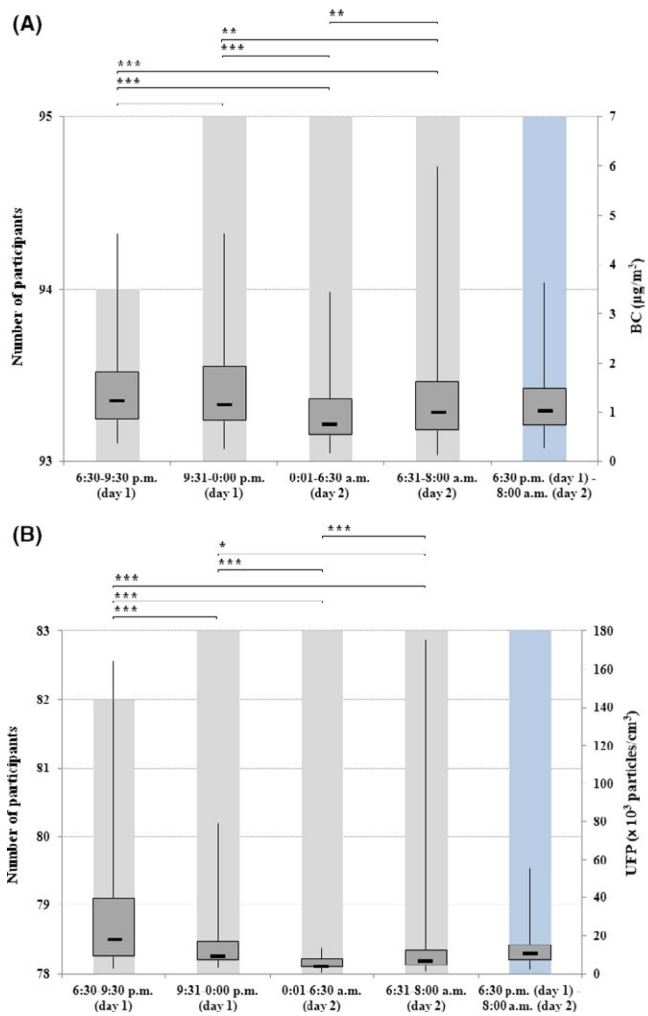

FIGURE 3 Distribution of (A) black carbon (µg/m^3) and (B) ultrafine particles (×10^3 particles/cm^3) exposure concentrations measured at home of participants in the BC-UFP-PARIS Study (between 6:30 p.m. “day 1” and 8:00 p.m. “day 2”). Each box represents the first and third quartile concentrations, the whiskers indicate the minimum and maximum concentration values, and the small lines indicate the geometric means; BC, black carbon; UFP, ultrafine particles. The lines above the graphs represent the side-by-side comparisons between time periods and *P* is the significance level resulting from the comparisons: * *P*<.05; ** *P*<.01; *** *P*<.0001

different activities (10.4% vs. 4.9%), while transport time remained constant (5.3% vs. 4.8%).

4 | DISCUSSION

For the first time in France, this study measured continuously over 24-hour personal exposure concentrations of BC and UFP in children living in Paris area. It showed that children are more exposed to BC and UFP during trips (mainly in metro/train and bus) and to UFP during indoor activities (such as eating at a restaurant). However, the most significant peaks of UFP were measured at home, following family activities and especially during cooking. Home and school together accounted for much of the total exposure (83.8% for BC and 85.3% for UFP), proportional to the long time spent in these microenvironments.

4.1 | Strengths and limitations of the study

Our study has several advantages: (i) It is one of the rare studies on exposure concentrations of BC and UFP conducted in schoolchildren from a population-based birth cohort, and the first one realized in France; (ii) it geographically covered Paris and its suburbs, and in equal proportions both warm (spring–summer) and cold (autumn–winter) periods of the year; (iii) it was conducted in a standardized way by a single epidemiologist who followed standard operating procedures we previously established in a feasibility study in adult volunteers; (iv) the portable devices used to measure continuously personal 24-hour exposure concentrations of BC and UFP were previously tested by our team in a laboratory validation study which showed strong correlations between MA and DM measurements, respectively, and those from the CPC device used as the reference “gold standard.”

This field study did, however, encounter certain difficulties. First, technical problems with measuring instruments sometimes occurred. In the case of MA, the negative false data points were caused by abrupt changes in temperature and/or relative humidity, as mentioned in the literature,[9,22,23] although these problems only concerned 0.05% of recorded data. Regarding DM, although our previous laboratory experiments showed that the quick change in the number of particles per cm^3 of air was not a problem, in the concrete field application, problems occurred a few times, due to (i) a rapid change in aerosol concentrations that generated diffusion stage negative values; (ii) a flow too low, and (iii) a corona voltage too low that generated some error codes in the database. However, these problems did not exceed 0.76% of the recorded data points. Second, BC and UFP measurements were carried out not only in the school period, but also during vacations. This led to different patterns in the timetable of 22% of children, resulting in these cases in exclusion of school and related trips (commuting school–home) and increase in the duration of exposure measurements at home and during other activities. Furthermore, the contribution of the school to total exposure was also underestimated in those children to whom the backpack was provided in late morning or early afternoon, compared with those who started the recordings early in the morning. This was due to the fact that only one person was in charge of the delivery of the measurement equipment to the 2 or 3 children planned to start the study on the same day.

4.2 | Exposure to BC and UFP

In our study, home and to a lesser extent school brought a major contribution to the total exposure to BC and UFP, due to the longer time spent in each of these two microenvironments (in the school periods, on average, around 61% at home and 30% at school). In other studies that included at less 24-hour measurements, it was reported that children 7- to 11-year-old spent 58%[16] or 64%-65%[14,15] of time at home, and at school 24%,[14] 28%,[15] or 31%[16] of time, respectively.

TABLE 4 Exposure concentrations of black carbon (μg/m^3) and ultrafine particles (×10^3 particles/cm^3) measured during indoor and outdoor participants' activities in the BC-UFP-PARIS Study

	Black carbon (μg/m^3)					Ultrafine particles (× 10^3 pt/cm^3)				
Type of activity	Number of activities	Arithmetic Mean (SD)[a]	Median	Range	Geometric Mean	Number of activities	Arithmetic Mean (SD)[a]	Median	Range	Geometric Mean
Indoor	51	1.98 (1.20)	1.66	0.29-4.75	1.63	40	36.3 (78.5)	15.3	3.4-493.0	18.2
Following different courses	9	1.57 (0.81)	1.30	0.86-3.54	1.44	8	9.9 (4.6)	9.7	3.4-19.5	8.9
Doing physical activity	6	1.98 (1.37)	1.80	0.69-4.32	1.60	6	14.7 (6.4)	15.3	7.3-22.4	13.4
Making a visit	21	1.97 (1.29)	1.68	0.29-4.75	1.58	17	34.3 (33.1)	20.1	8.5-118.8	23.9
Eating at restaurant	11	2.40 (1.19)	2.61	0.41-4.55	2.03	7	98.2 (175.8)	26.3	4.3-493.0	36.3
Shopping	4	1.72 (1.34)	1.15	0.85-3.71	1.43	2	7.5 (2.5)	7.5	5.7-9.3	7.3
Outdoor	19	1.67 (1.06)	1.26	0.54-4.26	1.40	16	18.0 (7.9)	17.7	6.1-36.1	16.3
Walking in a park or forest	12	1.60 (1.05)	1.37	0.54-4.26	1.35	10	18.6 (9.5)	18.3	6.1-36.1	16.2
Gardening near home	2	2.75 (0.34)	2.75	2.51-2.99	2.74	2	18.9 (0.4)	18.9	18.6-19.2	18.9
Accompanying an adult in his travels	5	1.41 (1.17)	0.95	0.79-3.49	1.17	4	15.8 (6.1)	13.2	12.0-24.9	15.1

[a]SD, standard deviation.

TABLE 5 Exposure concentrations of black carbon (μg/m^3) and ultrafine particles (×10^3 particles/cm^3) measured during participants' journeys in the BC-UFP-PARIS Study

	Black carbon (μg/m^3)					Ultrafine particles (×10^3 pt/cm^3)				
Journeys	Number of journeys	Arithmetic Mean (SD)[a]	Median	Range	Geometric Mean	Number of journeys	Arithmetic Mean (SD)[a]	Median	Range	Geometric Mean
Direction of journey(s)										
Commuting school↔home	152	2.69 (1.82)	2.17	0.36-9.22	2.18	134	17.6 (10.0)	16.1	3.3-52.4	15.1
Journey(s) to another destination[b]	94	3.27 (2.57)	2.62	0.76-18.40	2.61	74	25.0 (20.7)	18.0	2.8-106.2	19.1
Journey to the hospital	94	3.23 (1.33)	2.93	0.99-6.38	2.96	77	28.0 (12.2)	26.9	10.4-78.6	25.8
Travel mode										
Walking	187	2.68 (2.03)	2.22	0.62-18.40	2.18	160	19.5 (14.2)	16.5	3.3-104.5	16.1
Bike/scooter	14	2.57 (1.06)	2.22	0.85-4.88	2.36	11	21.8 (9.6)	18.4	9.5-41.4	20.0
Car/taxi	51	3.50 (2.27)	2.85	0.36-10.44	2.79	42	21.9 (13.6)	20.3	2.8-55.6	17.5
Bus	24	3.32 (1.25)	2.96	1.44-6.38	3.12	22	32.6 (14.9)	28.4	17.1-78.6	30.2
Tramway	7	2.36 (1.29)	2.25	0.99-5.04	2.11	6	19.4 (8.7)	16.8	9.7-33.1	17.9
Subway/train	42	3.75 (1.77)	3.35	1.01-10.07	3.37	34	29.1 (16.6)	26.6	10.7-106.2	26.1
Combined (several modes)	15	3.36 (1.43)	2.93	1.49-6.58	3.11	10	26.3 (13.9)	20.6	13.8-59.0	23.7

[a]SD: standard deviation.
[b]Journey(s) to another destination, except the journey to the hospital.

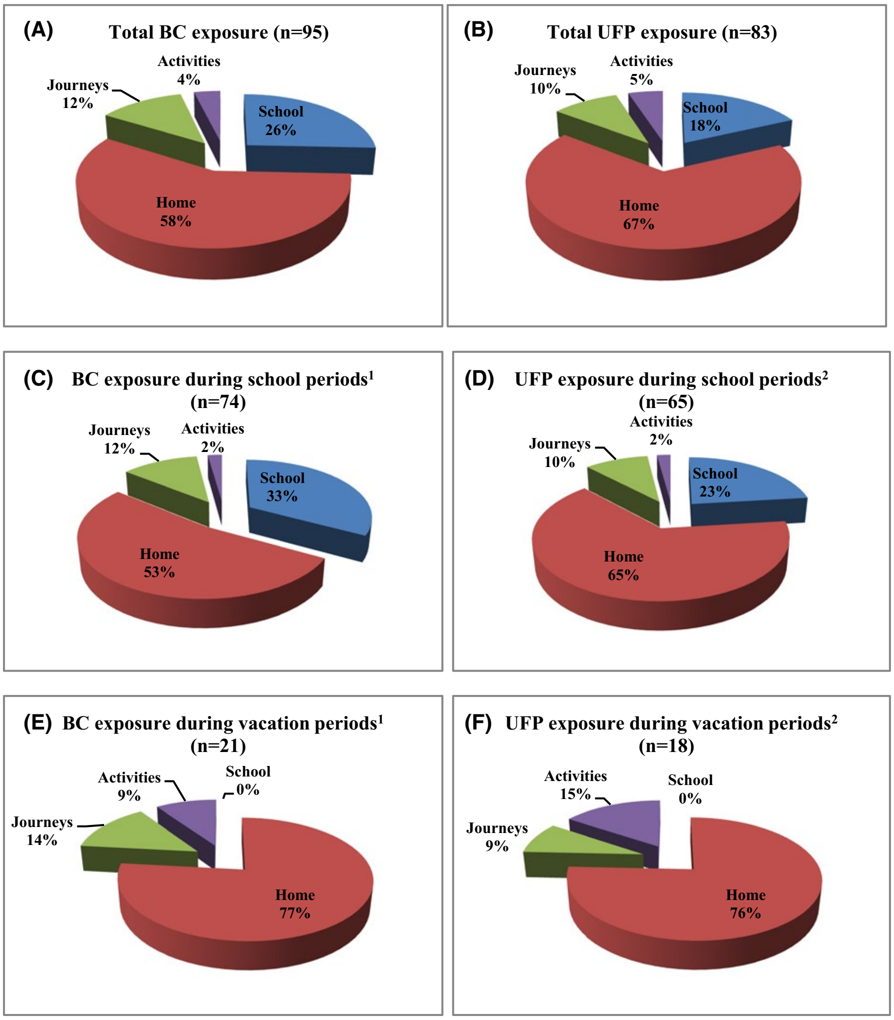

FIGURE 4 Contribution of each microenvironment to the total exposure to black carbon (A, C, E) and ultrafine particles (B, D, F) in the BC-UFP-PARIS Study. [1]Comparison of contribution of different microenvironments to the total exposure to black carbon between measurements during school and vacation periods (Student's *t*-test): school and home $P<.0001$; journeys $P=.48$; extracurricular activities $P=.002$. [2]Comparison of contribution of different microenvironments to the total exposure to ultrafine particles between measurements during school and vacation periods (Student's *t*-test): school $P<.0001$; home $P=.02$; journeys $P=.48$; extracurricular activities $P=.007$

The daily activity pattern of children is thus characterized by longer periods spent at home[24] compared to adults[25] who spend more time in transit.[26] So, in our study, during school terms, home, school, transport, and extracurricular activities contributed to total BC exposure for 53%, 33%, 12%, and 2%, and to total UFP exposure for 65%, 23%, 10%, and 2%, respectively. In comparison, the 15 Parisian adult volunteers that participated in our pilot study (recording weekdays, during 30 consecutive hours) spent less time at home (46%), more time at work (office, 45%) and in transport (7%), and less time in other activities (2%), these microenvironments contributing to total BC exposure for 40%, 35%, 23%, and 2%, and to total UFP exposure for 38%, 49%, 10% and 3%, respectively.

In the literature, few studies have researched personal exposure to BC and/or UFP over a 24-hour period in children of a similar age to ours. This makes it difficult to compare duration of exposure and/or contribution of each microenvironment to total exposure, due to the different methodologies used: for example, measurement of UFP just for 2-5 hour in two consecutive days,[17] calculation of an inhaled dose for each microenvironment/activity,[13] or calculation of proportions of total daily alveolar doses in microenvironments.[15] In addition, different devices have been used to measure the number of pt/cm^3 (such as Philips NanoTracer, CPC Model 3007 or DiSCmini in our case), and no study shows correlations between values recorded with these different devices.

4.2.1 | School microenvironment

Exposure concentrations of BC and UFP in schools vary widely in our study, as described in the literature which attributes this to several sources, both outdoors (traffic-related air pollution) and indoors.

In the school environment, compared to a mean UFP of 9.4×10^3 pt/cm^3 for our participants, other authors have indicated a mean UFP of 8.5×10^3 pt/cm^3 in the Brisbane Metropolitan Area in Australia[15] and 19.8×10^3 pt/cm^3 in Cincinnati, Ohio, USA.[17] Important differences in the mean of UFP were reported by Buonanno et al.[12] between three secondary schools from Cassino (Italy) located: first on an urban street with traffic mostly dominated by light vehicles (40×10^3 pt/cm^3); second, close to the intersection of moderately and heavily trafficked urban street (33×10^3 pt/cm^3); and third, in a rural area, far away from urban traffic (12×10^3 pt/cm^3). Concentrations of BC also greatly varied, especially between those measured in the second and third school (13.9 μg/m^3 vs. 1.9 μg/m^3).[14] Similarly in our study, BC concentrations were significantly different in schools located in inner Paris which are more exposed to traffic, compared with schools located in the suburbs.

Our participants' exposure concentration to BC and UFP was higher in the morning than in the afternoon and was greater for BC than for UFP, possibly due to higher traffic in the morning rush hour. The infiltration of particles (mostly BC) from outside to inside the classroom thus seems to be higher in the morning than in the afternoon, perhaps due to window openings before the start of classes coinciding with the high traffic in the morning rush hour. MacNeill et al.[27] showed that a simple intervention such as adjusting the ventilation system to operate earlier in the morning, prior to high traffic commute periods, significantly reduced traffic-related air pollutants (including UFP and fine particles) in schools located near major roads with heating, ventilating and air-conditioning systems.

According to the literature review of Mejía et al.,[28] it was evident that there is very high exposure to UFP inside classrooms, often exceeding outdoor concentrations. In addition to traffic, there are other sources of exposure at school, such as new furniture, musical instruments, and children's activities.[28] Diapouli et al.[29] reported high mean indoor UFP (51.9×10^3 pt/cm^3) in a primary school in Athens (Greece), in a small library room with a carpet-covered floor and a very high ratio of persons per m^3 of space, compared to a UFP mean of 24×10^3 pt/cm^3 for 8-hour classroom measurements, in seven primary schools. Laiman et al.[30] reported that during school hours, electrical heating and printing in the classroom, as well as the use of kitchen grills in the school "tuckshop" that infiltrate classrooms, are factors that contribute to an elevated level of UFP, and during non-school hours, cleaning events elevated indoor UFP. Tobacco smoking is a major source of fine particles, but in French schools, smoking is not allowed.

4.2.2 | Home microenvironment

UFP exposure concentrations measured in Parisian homes (mean of 14.6×10^3 pt/cm^3) were consistent with those in the literature in urban areas ([10.5×10^3 pt/cm^3][15] and [27×10^3 pt/cm^3][17]). Most of these concentrations occurred in the evening (between 6:30 and 9:30 p.m.), upon return of all family members and in relation to domestic activities, mainly food preparation and use of a gas or an electrical stove, in line with previous studies.[3,26] Kearney et al.[31] observed higher indoor UFP level around 5-7 p.m., suggesting a strong influence of cooking. Cooking on gas or electric stoves, using candles, and ironing clothes were the activities consistently identified by Bhangar et al.[32] to generate UFP in homes. Wallace et al.[9] reported a number of important indoor sources, ranging from cooking on stoves (both gas and electric) and toaster ovens to the use of hair dryers, cigarettes, candles, and noted peak personal exposures often exceeding 100×10^3 pt/cm^3. In our study, smoking was not a source of air pollution: The families of our children did not smoke inside the home. Consistent with Dons et al.[26], Zhu et al.[33] and Weichenthal et al.,[3] we measured the lowest average levels during the night when families were sleeping. Regarding indoor concentrations of BC, we found the same trends with an important seasonal effect: BC concentrations were significantly higher during the cold season than the hot season, mainly due to the use of heating (electric, gas or wood).

4.2.3 | Extracurricular activities microenvironment

Exposure of children to BC and UFP during their extracurricular activities (school, home, and trips excluded) is poorly documented in the literature. Ryan et al.[17] reported in 20 asthmatic children aged between 9.3 and 13.9 years, from Cincinnati (USA), a mean UFP of 4.1×10^3 pt/cm^3, while Mazaheri et al.[15] indicated for 137 children aged 8-11 years from Brisbane (Australia), a mean UFP of 8.7×10^3 pt/cm^3. These levels are much lower than those measured in our study. Indeed, they depend on whether the activity is "indoor" or "outdoor" and on the time devoted to this activity. In our study, among all activities carried out, indoor activities recorded the highest number concentrations per cm^3, the highest average (UFP mean of 98.2×10^3 pt/cm^3) being reached in children who had eaten in restaurants (mostly fast food; maximum of 493×10^3 pt/cm^3). Wallace et al.[9] reported that restaurants maintained consistently high levels of 50×10^3 and 200×10^3 pt/cm^3 for the entire length of the meal (UFP mean of 94. 5×10^3 pt/cm^3 for 22 restaurants visited; highest value of 228×10^3 pt/cm^3).

4.2.4 | Transport microenvironment

In our study, transport is responsible for the highest exposure concentration (GM) of BC and UFP, although travel time only represents 5% of recording time. Indeed, children usually spent only a short time traveling, the longest trips being to the hospital. The averages recorded by Paris children during journeys (mean BC 3.23, range 1.1-6.7 μg/m^3 and mean UFP of 24.9×10^3 pt/cm^3, range between 9.8×10^3 and 60.4×10^3 pt/cm^3) are of the same order as pollution levels recorded in urban traffic sites. Reche et al.[34] reported in seven selected urban sites from southern, Central, and northern Europe that daily average BC concentration measured at roadside sites ranged from 3.5 μg/m^3 in Bern (range 1.3-7.1 μg/m^3) to 7.8 μg/m^3 in Marylebone road in London (range 2.5-14 μg/m^3) and UFP from 22.2×10^3 pt/cm^3 (in London; range 4.8×10^3-58×10^3 pt/cm^3) to 28×10^3 pt/cm^3 (in Bern; range 8.9×10^3-93.1×10^3 pt/cm^3).

In the literature, Buonanno et al.[12] have shown that the UFP during transport in schoolchildren greatly varied according to the area, urban (55×10^3 to 68×10^3 pt/cm^3), or rural (21×10^3 pt/cm^3). The UFP level also depended on the mode of transport. Mazaheri et al.[15] noted that commuting (mostly walking and car travel) represented 3% of the total time of the measurements and an average level of exposure to UFP of 13.7×10^3 pt/cm^3. In the study of Ryan et al.,[17] UFP reached an average level of 21.4×10^3 pt/cm^3 during trips (travel by walking, car and bus). Among Paris schoolchildren, walking was the most frequent mode of transport, especially to commute between the school and home. Several other modes were used, and we found metro/train and bus modes had higher exposure levels than the other modes. In the literature, the exposure to BC and/or UFP by mode of transport has been poorly described in schoolchildren compared to that of adults. Rivas et al.[16] reported for 45 children from Barcelona (Spain) that they spent 6% of the time in the weekday on commuting and received 20% of their daily BC dose. They were also most exposed during trips in the metro and bus. However, in Cincinnati (USA), Ryan et al.[17] reported much higher average levels of UFP for walking (38.1×10^3 pt/cm^3) and lower levels in the school bus (23.4×10^3 pt/cm^3) and car (11.7×10^3 pt/cm^3). It is difficult to compare results between countries since exposure levels depend on the study area, the type of vehicle (diesel or gasoline, light or commercial vehicles), density and fluidity of traffic, distance to traffic, length of the trip, season, time of day, meteorological conditions, etc.[35,36] In an experiment,[37] for journeys through the city of Barcelona, BC was lower in the tram (mean of 3.4 μg/m^3) when compared to the bus (5.5 μg/m^3), subway (7 μg/m^3—values possibly overestimated by interference of light absorption by iron in the microaethalometer measurements), and walking (6.5 μg/m^3 in suburban and 9.6 μg/m^3 in the congested inner city). The mean of UFP was lowest in commuting using subway trains (23×10^3 pt/cm^3), higher during tram travel (30×10^3 pt/cm^3), suburban walking (37×10^3 pt/cm^3), and highest in diesel bus (55×10^3 pt/cm^3) or walking in the city center (59×10^3 pt/cm^3), with extreme transient peaks at busy traffic crossings commonly exceeding 100×10^3 pt/cm^3 and accompanied by peaks in BC and carbon monoxide.[37]

This study measures the personal exposures of children in Paris to two indicators of particulate air pollution, BC and UFP. New portable air monitoring devices that can be carried all day permit accurate measurements of exposure concentrations that previously were not possible. The literature indicates that similar measurements rarely have been performed on children, but our study shows that accurate measurements of exposure in the microenvironments that children occupy during the day can be made with these monitors, especially during children's trips and extracurricular activities. This study demonstrates that small, battery-powered monitors with data logging capability can provide useful measurements of BC and UFP that are complementary indicators of particulate air pollution. Our finding that BC levels are higher outdoors during children's trips is of particular interest given that BC is a very good indicator of combustion sources, especially traffic-related sources. Our measurements of UFP, on the other hand, showed higher levels inside homes and various buildings where different daily activities take place. We observed high UFP levels during indoor activities (mainly eating at restaurants and cooking at home). The results show the need for reduction of air pollutant levels resulting from both indoor and outdoor sources.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

ACKNOWLEDGEMENTS

This study is supported by the *Agence de l'Environnement et de la Maîtrise de l'Energie* (ADEME, Contract no. 1262C0010) and the Research Call *Programme National de Recherche Santé Environnement Travail* (ANSES—French Agency for Food, Environmental and Occupational Health & Safety). The authors thank Dr. Lydia Nikasinovic for her involvement in the development of the study protocol. We thank all the families—children and parents—and volunteers who accepted to participate in the field study. We also thank the school staff, teachers, and principals who allowed the realization of this project in their school.

REFERENCES

1. Adar SD, Adamkiewicz G, Gold DR, Schwartz J, Coull BA, Suh H. Ambient and microenvironmental particles and exhaled nitric oxide before and after a group bus trip. *Environ Health Perspect*. 2007;115:507–512.
2. Rossner P Jr, Svecova V, Milcova A, et al. Oxidative and nitrosative stress markers in bus drivers. *Mutat Res*. 2007;617:23–32.
3. Weichenthal S, Dufresne A, Infante-Rivard C. Indoor ultrafine particles and childhood asthma: exploring a potential public health concern. *Indoor Air*. 2007;17:81–91.
4. Delfino RJ, Sioutas C, Malik S. Potential role of ultrafine particles in associations between airborne particle mass and cardiovascular health. *Environ Health Perspect*. 2005;113:934–946.
5. Duffin R, Tran C, Clouter A, et al. The importance of surface area and specific reactivity in the acute pulmonary inflammatory response to particles. *Ann Occup Hyg*. 2002;46:242–245.
6. Buonanno G, Fuoco FC, Morawska L, Stabile L. Airborne particle concentrations at schools measured at different spatial scales. *Atmos Environ*. 2013a;67:38–45.
7. Nel A. Air pollution-related illness: effects of particles. *Science*. 2005;308:804–806.
8. See SW, Balasubramanian R. Physical characteristics of ultrafine particles emitted from different gas cooking methods. *Aerosol Air Quality Res*. 2006;6:82–92.
9. Wallace LA, Wheeler AJ, Kearney J, et al. Validation of continuous particle monitors for personal, indoor, and outdoor exposures. *J Expo Sci Environ Epidemiol*. 2011;21:49–64.
10. Janssen NAH, Gerlofs-Nijland ME, Lanki T, et al. *Health Effects of Black Carbon*. Copenhagen: World Health Organization. Regional office for Europe; 2012, 96p.
11. Janssen NA, Hoek G, Simic-Lawson M, et al. Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM10 and PM2.5. *Environ Health Perspect*. 2011;119:1691–1699.
12. Buonanno G, Marini S, Morawska L, Fuoco FC. Individual dose and exposure of Italian children to ultrafine particles. *Sci Total Environ*. 2012;438:271–277.
13. Buonanno G, Marks GB, Morawska L. Health effects of daily airborne particle dose in children: direct association between personal dose and respiratory health effects. *Environ Pollut*. 2013b;180:246–250.
14. Buonanno G, Stabile L, Morawska L, Russi A. Children exposure assessment to ultrafine particles and black carbon: the role of transport and cooking activities. *Atmos Environ*. 2013c;79:53–58.
15. Mazaheri M, Clifford S, Jayaratne R, et al. School children's personal exposure to ultrafine particles in the urban environment. *Environ Sci Technol*. 2014;48:113–120.
16. Rivas I, Donaire-Gonzalez D, Bouso L, et al. Spatiotemporally resolved black carbon concentration, schoolchildren's exposure and dose in Barcelona. *Indoor Air*. 2016;26:391–402.
17. Ryan PH, Son SY, Wolfe C, Lockey J, Brokamp C, Lemasters G. A field application of a personal sensor for ultrafine particle exposure in children. *Sci Total Environ*. 2015;508:366–373.
18. Clarisse B, Nikasinovic L, Poinsard R, Momas I. The Paris prospective birth cohort study: which design and who participates? *Eur J Epidemiol*. 2007;22:203–210.
19. AethLabs, San Francisco, USA. Operating Manual – microAeth Model AE51. Rev 00. 32p, June 2011.

20. Fierz M, Houle C, Steigmeier P, Burtscher H. Design, calibration, and field performance of a miniature diffusion size classifier. *Aerosol Sci Technol*. 2011;45:1–10.
21. Matter Aerosol, Wholen, Switzerland. DiSCmini Diffusion Size Classifier miniature Instruction manual. Rev 1.10, January 2012.
22. Cai J, Yan B, Ross J, et al. Validation of MicroAeth® as a black carbon monitor for fixed-site measurement and optimization for personal exposure characterization. *Aerosol Air Qual Res*. 2014;14:1–9.
23. Yu CH, Patton AP, Zhang A, Fan ZH, Weisel CP, Lioy PJ. Evaluation of diesel exhaust continuous monitors in controlled environmental conditions. *J Occup Environ Hyg*. 2015;14:1–9.
24. Xue J, McCurdy T, Spengler J, Ozkaynak H. Understanding variability in time spent in selected locations for 7-12-year old children. *J Expo Sci Environ Epidemiol*. 2004;14:222–233.
25. Branco PTBS, Alvim-Ferraz MCM, Sousa SIV. The microenvironmental modelling approach to assess children's exposure to air pollution - A review. *Environ Res*. 2014;135:317–332.
26. Dons E, Panis LI, van Poppel M, Theunis J, Wets G. Personal exposure to Black Carbon in transport microenvironments. *Atmos Environ*. 2012;55:392–398.
27. MacNeill M, Dobbin N, St-Jean M, et al. Can changing the timing of outdoor air intake reduce indoor concentrations of traffic-related pollutants in schools? *Indoor Air*. 2016;26:687–701.
28. Mejía JF, Choy SL, Mengersen K, Morawska L. Methodology for assessing exposure and impacts of air pollutants in schoolchildren: data collection, analysis and health effects - A literature review. *Atmos Environ*. 2011;45:813–823.
29. Diapouli E, Chaloulakou A, Mihalopoulos N, Spyrellis N. Indoor and outdoor PM mass and number concentrations at schools in the Athens area. *Environ Monit Assess*, 2008;136:13–20.
30. Laiman R, He C, Mazaheri M, et al. Characteristics of ultrafine particle sources and deposition rates in primary school classrooms. *Atmos Environ*. 2014;94:28–35.
31. Kearney J, Wallace L, Macneill M, et al. Residential indoor and outdoor ultrafine particles in Windsor, Ontario. *Atmos Environ*. 2011;45:7583–7593.
32. Bhangar S, Mullen NA, Hering SV, Kreisberg NM, Nazaroff WW. Ultrafine particle concentrations and exposures in seven residences in northern California. *Indoor Air*. 2011;21:132–144.
33. Zhu YF, Hinds WC, Krudysz M, Kuhn T, Froines J, Sioutas C. Penetration of freeway ultrafine particles into indoor environments. *J Aerosol Sci*. 2005;36:303–322.
34. Reche C, Querol X, Alastuey A, et al. New considerations for PM, Black Carbon and particle number concentration for air quality monitoring across different European cities. *Atmos Chem Phys*. 2011;11:6207–6622.
35. Kaur S, Nieuwenhuijsen MJ, Colvile RN. Fine particulate matter and carbon monoxide exposure concentrations in urban street transport microenvironments. *Atmos Environ*. 2007;41:4781–4810.
36. Spinazzè A, Cattaneo A, Scocca DR, Bonzini M, Cavallo DM. Multi-metric measurement of personal exposure to ultrafine particles in selected urban microenvironments. *Atmos Environ*. 2015;110:8–17.
37. Moreno T, Reche C, Rivas I, et al. Urban air quality comparison for bus, tram, subway and pedestrian commutes in Barcelona. *Environ Res*. 2015;142:495–510.

SUPPORTING INFORMATION

Additional Supporting Information may be found online in the supporting information tab for this article.

How to cite this article: Paunescu A-C, Attoui M, Bouallala S, Sunyer J and Momas I. Personal measurement of exposure to black carbon and ultrafine particles in schoolchildren from PARIS cohort (Paris, France). *Indoor Air*. 2017;27:766–779. https://doi.org/10.1111/ina.12358