

CHILDREN'S INDOOR EXPOSURES TO (ULTRA)FINE PARTICLES IN AN URBAN AREA: COMPARISON BETWEEN SCHOOL AND HOME ENVIRONMENTS

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Due to their detrimental effects on human health, scientific interest in ultrafine particles (UFP), has been increasing but available information is far from comprehensive. Children, who represent one of the most susceptible subpopulation, spend the majority of time in schools and homes. Thus, the aim of this study is to (1) assess indoor levels of particle number concentrations (PNC) in ultrafine and fine (20–1000 nm) range at school and home environments and (2) compare indoor respective dose rates for 3- to 5-yr-old children. Indoor particle number concentrations in range of 20–1000 nm were consecutively measured during 56 d at two preschools (S1 and S2) and three homes (H1–H3) situated in Porto, Portugal. At both preschools different indoor microenvironments, such as classrooms and canteens, were evaluated. The results showed that total mean indoor PNC as determined for all indoor microenvironments were significantly higher at S1 than S2. At homes, indoor levels of PNC with means ranging between 1.09×10^4 and 1.24×10^4 particles/cm³ were 10–70% lower than total indoor means of preschools (1.32×10^4 to 1.84×10^4 particles/cm³). Nevertheless, estimated dose rates of particles were 1.3- to 2.1-fold higher at homes than preschools, mainly due to longer period of time spent at home. Daily activity patterns of 3- to 5-yr-old children significantly influenced overall dose rates of particles. Therefore, future studies focusing on health effects of airborne pollutants always need to account for children's exposures in different microenvironments such as homes, schools, and transportation modes in order to obtain an accurate representation of children overall exposure.

During the last two decades, there has been considerable interest in the health effects of exposure to airborne particulate matter (PM) (Krewski et al., 2003; Krewski and Rainham, 2007; Samet and Krewski, 2007; Brunekreef et al., 2009). As knowledge regarding adverse health effects being correlated with size dependency of particles has grown (Kelly and Fussel, 2012), ongoing research has focused attention on ultrafine particles (UFP), which represent a fraction of particulate matter (PM) with particles of aerodynamic diameter smaller than 0.1 μm (Morawska et al., 2013; Carosino et al., 2015).

Unlike coarse particles, UFP contribute little to PM mass, but these dominate number concentrations. Due to UFP small size, high number concentrations, high surface area, and ability to penetrate into the interstitial spaces of the lungs (Bakand et al., 2012; Pereira Gomes et al., 2012), there are various adverse health effects. Clinical and epidemiological studies linked exposure to ambient UFP with adverse respiratory outcomes, including impaired lung function and pulmonary defense mechanisms, inflammatory responses and worsening of respiratory diseases, and possibly cardiovascular

dysfunction (Ibald-Mulli et al., 2002; Bakand et al., 2012; Heal et al., 2012; Tsai and Yang, 2014; Chang et al., 2015), although findings are not consistent (Rückerl et al., 2011). While more epidemiological studies on the UFP fraction are needed, exposure assessment issues such as spatial variability, indoor sources, infiltration of UFP from various outdoor emission sources, and seasonal variability in concentrations and composition are currently being addressed (Wang et al., 2013; Azarmi et al., 2014; Bekö et al., 2013; Viana et al., 2014, 2015; Rivas et al., 2015).

In view of the evidence of negative health impacts of UFP, studies focused on investigation of main sources and processes affecting levels and size distributions of these particles in ambient air of urban areas (Morawska et al., 2008; Kumar et al., 2010; Solomon, 2012). UFP may be formed by condensation of semivolatile organic aerosols, photochemically induced nucleation, and/or nucleation through gas-to-particle conversion (Morawska et al., 2008, 2013). Concerning indoor air, UFP originate from combustion processes that include cooking, namely, boiling, stewing, frying, baking, and grilling, smoking, and use of candles (Bekö et al., 2013; Morawska et al., 2013), and as result of occupant-related activities such as use of consumer products and use of painting and cleaning products (Long et al., 2000; Bhangar et al., 2011).

Young children represent one of the most susceptible subpopulations with regard to potentially harmful effects induced by airborne PM exposure (Schüepp and Sly, 2012; Annesi-Maesano et al., 2013). As their physiological and immunological systems are still developing, young children receive a higher dose of airborne particles relative to lung size compared to adults (Foos et al., 2008; Burtcher and Schüepp, 2012; Morawska et al., 2013; Laiman et al., 2014; Mazaheri et al., 2014). Children spend a significant percent of time at schools and at homes. Specifically, in Portugal, young children spend at school approximately 30% of their time (8–9 h/d). Therefore, knowledge and understanding of indoor air pollution in these specific environments are important in order to

improve child health. As a pollutant of both indoors and in ambient air, UFP possess the potential to harm children's health (Burtcher and Schüepp, 2012; Schüepp and Sly, 2012; Moreno et al., 2014; Reche et al., 2014; Rivas et al., 2014; Viana et al., 2014), yet information concerning children's exposure to UFP is limited.

The aim of this study was to assess indoor exposure to particles in the (ultra)fine range (20–1000 nm) in 3- to 5-yr-old children living in urban areas. The specific objectives were to (i) measure levels of indoor particle number concentrations (PNC) in two preschools and three homes situated in urban low to moderate traffic zones of Oporto Metropolitan Area (Portugal), and (ii) compare dose rates of indoor (ultra)fine particles at schools and home environments.

MATERIALS AND METHODS

Characterization of Sampling Sites

Particle number concentrations in ultrafine (20–100 nm) and fine (>100–1000 nm) ranges were consecutively measured at two preschools and three homes, all of them situated in urban low to moderate traffic zones of Oporto Metropolitan Area in Paranhos district (north of Portugal). The sample collection was conducted for 56 d. Both the preschools (S1 and S2) and homes (H1–H3) were situated in an urban zone; previously studies that evaluated ambient air pollution demonstrated that emissions from vehicular traffic are the main pollution source in these areas (Slezakova et al., 2011, 2013).

In each preschool, PNC were simultaneously measured in different indoor microenvironments, including classrooms, canteens, and, if present, gymnasium or playroom; all microenvironments were assessed using identical sampling methodology and for the same amount of time. At homes, sampling of (ultra)fine particles was conducted in living rooms that were used also as dining rooms; all meals/snacks were served there.

TABLE 1. Characterization of the Studied Environments (Preschools and Homes) and Obtained Concentrations of (Ultra)Fine Particles

Site	Description	Location	Traffic density data ^a	Studied indoor microenvironments	Sampled period	Particle number concentration (particles/cm ³)	
						Mean	Standard deviation
S1	Two-floors building 173 students 3–5 yr old	Situated on moderately trafficked street	Mean: 16 vehicles/min	Classrooms (3) Canteen (1) Playroom (1)	13 d	Classrooms 9.31×10^3 Canteen 5.17×10^4 Playroom 1.70×10^4 Total 1.82×10^4	8.23×10^3 3.41×10^4 1.25×10^4 2.16×10^4
S2	Three-floors building 30 students 3–5 yr old	Situated on intersection of moderate and low trafficked street	Mean: 13 vehicles/min	Classrooms (3) Canteen (1) Gymnasium (1)	13 d	Classrooms 1.13×10^4 Canteen 3.28×10^4 Gymnasium 9.72×10^3 Total 1.32×10^4	5.24×10^4 3.21×10^4 2.36×10^3 1.25×10^4
H1	Multiunit apartment building Situated on fourth floor, 4 occupants (2 children, 3 and 5 yr old)	Situated on intersection of two low trafficked street	Mean: 3 vehicles/min	Living room	10 d	1.24×10^4	1.28×10^4
H2	Multiunit apartment building Situated on fourth floor, 4 occupants (1 child 5 yr old),	Situated nearby highly trafficked road	Not available	Living room	9 d	1.11×10^4	1.15×10^4
H3	Two-floor house 4 occupants (1 child, 5 yr old)	Situated in suburban zone with moderate traffic	Mean: 4 vehicles/min	Living room	11 d	1.09×10^4	1.11×10^4

^aData were obtained by manual counts during 10 min of each hour (between 5 a.m. to 12 p.m.) on two consecutive days (avoiding Mondays and Fridays). The distance between the counting point and main entrance/building outside wall was 5 and 8 m at S1 and S2, respectively, and 3–4 m at H1 and H3.

All indoor locations were naturally ventilated through open windows. The characteristics of the studied preschools and homes, traffic density data, and duration of sampling at each place are summarized in Table 1.

Sample Collection

Particle number concentrations in size range 0.02–1 μm were measured by condensation particle counters, TSI P-TrakTM (UPC 8525; TSI, Inc., Shoreview, MN). The instrument operates on the principle of condensing 100% grade isopropyl alcohol (Sigma-Aldrich, Steinheim, Germany) onto UFP in order to increase their dimensions to a detectable size. At preschools, PNC were measured daily between 8:30 a.m. and 5:30 p.m., which corresponded to the period that children were at preschools, whereas at homes PNC of (ultra)fine particles were measured continuously over 24 h. Intake flow of 0.7 L/min was

used and logging interval was 60 s, according with previous studies (Diapouli et al., 2007; Norbäck et al., 2011; Zhang and Zhu, 2012). Instruments were mounted onto supports so that air was sampled from a height of 0.8 to 1.1 m (in order to simulate children's breathing zone). In each indoor environment, particles counters were placed as far as possible from windows or doors, and from other probable sources of particles such as heating equipment, blackboards, and printers, in order to minimize direct influence of any source. All requirements to maintain child safety were fulfilled.

At both preschools a researcher was present during sample collection in order to keep a record of room occupancy, ventilation systems (door and window positions), and potential source activities; information concerning child activities and schedules at preschools was also registered by an investigator. At homes, all information including child activities was recorded by the parents/child responsible. In addition,

teachers, staff, and parents were asked daily about the occurrence of additional sources and activities. Further, detailed questionnaires were used daily for better description of the studied indoor environments for both preschools and homes. The first questionnaire was dedicated to registering potential sources of particles, where the occupants marked time when these sources/activities were used/conducted in order to cross-reference them with concentration levels. The second questionnaire focused on occupancy/activities of room where sampling equipment was placed. The last questionnaire focused on the schedule of children's activities and their physical activity during sampling. All necessary permissions were obtained from administrative boards of each preschool and directly from parents.

Dose Rate Analysis

Particle dose rates for children were calculated using Eq. (1) (Castro et al., 2011; Slezakova et al., 2014):

$$\text{Dose rate (D)} = (\text{BR}_{\text{WA}}/\text{BW}) \times C_{\text{WA}} \times \text{OF} \times N \quad (1)$$

where D is the age-specific dose rate (particle number/kg/day); BR_{WA} is the age-specific weighted average breathing rate (L/min); BW is age-specific body weight (kg); C_{WA} is the age-specific weighted average concentration of particles (number of particles/L); OF is the occupancy factor (i.e., percentage of residents likely to be in the microenvironment at a given interval; it was considered 1, as children kept to their schedules and associated locations tightly); and N is the total time per day spent by age-specific children in the respective indoor environment (min/d). Particle dose rates were estimated for 3- to 4-yr-old and 5-yr-old children. The daily activity patterns of children were analyzed throughout each day. Locations in which the different activities occurred during the day were identified. Total daily residence time of children spent in each microenvironment (home, preschool) and the types of performed activities were registered.

Each activity was characterized in terms of intensity level in order to assess the corresponding BR. An example of children's timetable and activity patterns is shown in Table 2. As the information concerning the Portuguese population is not available, age-specific factors of body weight (BW) and breathing rate (BR) were retrieved from U.S. Environmental Protection Agency (EPA) data (U.S. EPA, 2011) considering the mixed population (both male and females). BW of 18.6 kg for children 3–5 years old was used. The values of BR were selected as the followings: 4.3 L/min for rest or sleep; 4.5 L/min for sedentary or passive activities; 11 L/min for light intense activity; and 37 L/min for highly intense activities such as running. BR_{WA} was estimated as a weighted average, that is, considering the intensity of each performed activity and the amount of time. The dose rates were then estimated using the average indoor concentrations of each microenvironment and considering the real amount of time that children spent in each place.

Statistical Analysis

For the data treatment, Student's *t*-test was applied to determine the statistical significance ($p < .05$, two-tailed) of the differences between the determined means. All statistical analyses were performed using IBM SPSS Statistics software.

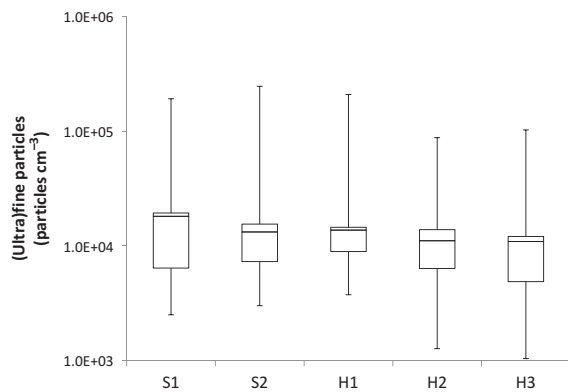
RESULTS

Particle Number Concentrations

Total means of particle number concentrations and statistical parameters (minimum and maximum values, 25th and 75th percentiles) at the two preschools and three homes are shown in Figure 1. These parameters of (ultra)fine particles were determined using all measured data of all examined indoor environments. Concerning the two preschools, mean of indoor PNC was significantly 1.4-fold higher at S1 (1.84×10^4 particles/cm³) than at S2 (mean of 1.32×10^4 particles/cm³).

TABLE 2. Timetable and Child Activity Patterns During a Weekday: An Example for Children 3–4 yr Old at School and a Home

Time		Environment	Observed activities	Activity intensity
School				
8:30–9:00	Arrival at school	Indoor	Playing (calm, seated, TV)	Sedentary
9:03–10:29	Classes/education	Indoor	Seated only (talking)	Sedentary
10:30–11:15	Recess	Playground	Running, jumping, swings	High intensity
11:17–11:40	Classes/education	Indoor	Sedentary and other (painting, walking)	Sedentary
11:45–13:00	Lunch	Indoor	Seated (eating, drinking, talking)	Light
13:05–15:00	Rest	Indoor	Sleeping	Sleep
15:04–16:00	Classes/education	Indoor	Seated, and other	Sedentary
16:00–17:30	Leaving school	Indoor	Organized activities (singing dancing), running	High intensity
Home				
18:00–19:20	Living room		Home works, school preparation, studying	Sedentary
19:25–20:00	Living room		Seated (eating, drinking, talking)	Sedentary
20:05–22:00	Living room		Playing games, painting, walking	Light
22:00–6:50	Bedroom		Sleeping	Sleep
7:00–8:00	Various		Morning routine, breakfast	Light

**FIGURE 1.** Levels of (ultra)fine particles at two schools (S1, S2) and three homes (H1–H3): minimum and maximum values, average, and 25th and 75th percentiles. Particle number concentrations were determined considering the measured levels in all indoor microenvironments existent in each school and home.

At all three homes, obtained means of indoor (ultra)fine particles (Table 1) were rather similar; the results showed that the total indoor means of PNC at the three homes were not statistically different. Overall, the highest mean and ranges of PNC were observed at H1 with mean concentration 1.1-fold higher than at H2 and H3.

Dose Rates

The activities that children conducted during their school time were similar at both

preschools. However, dose rates of indoor particles were estimated for 2 age categories, namely, children 3–4 yr old and 5 yr old, because their daily schedules differed slightly. Children spent the majority of their preschool time in classrooms (approximately 70–75% for 3–4 yr old, and 57–70% for 5 yr old). The younger children rested (i.e., slept, which was an activity associated with the lowest breathing rates) after lunch for 2–2.5 h, whereas older children performed indoors more frequently physical activities such as running, playing, exercising, and use of climbers, swings, and slide. In addition, the 5-yr-old children spent less time (0.75–1.75 h) indoors. Overall, the daily activity patterns of children at the three homes were remarkably similar. On average, children spent 13 h at home, out of which 3 h occurred in a living room (sedentary or light activities; studying, games playing, drawing, or eating). Morning and evening routines (breakfast, bath, etc.) took approximately 1 h, whereas children slept for about 9 h.

Dose rates associated with inhalation exposure to (ultra)fine particles (20–1000 nm) number concentrations at the two preschools and three homes were estimated for two different age categories of children. The results are shown in Table 3. Concerning preschools, the data demonstrate that (i) for both age categories

TABLE 3. Age-Specific Dose Rates (particles/kg/d) to UFP for Children (3–4 yr and 5 yr Old) at Two Preschools (S1 and S2) and Three Homes (H1–H3)

	S1		S2		H1	H2	H3
Dose rate (particles/kg/d)	3–4 yr old	5 yr old	3–4 yr old	5 yr old	3–5 yr old	3–5 yr	3–5 yr old
	1.99×10^9	2.02×10^9	1.49×10^9	1.92×10^9	3.06×10^9	2.74×10^9	2.69×10^9

the highest dose rates of PNC were found at S1; and (ii) for both schools the highest values of PNC total dose rates were observed for 5-yr-old children. Further, the results in Table 3 illustrate that for children 3–4 yr and 5 yr old, dose rates at homes were 1.3- to 2.1-fold higher than at schools.

DISCUSSION

As children represent one of the most susceptible subpopulations in society, more information concerning air pollutants to which they are exposed in schools and home environments is needed. Overall, levels of (ultra)fine particles at the two Portuguese preschools were similar in range to those reported for indoor air of schools in Greece (2.4×10^4 particles/cm³; Diapouli et al., 2008), Italy (1.95 – 2.04×10^4 particles/cm³; Buonanno et al., 2012, 2013a), Spain (1.56×10^4 particles/cm³; Reche et al., 2014; Rivas et al., 2014), South Korea (1.82×10^4 particles/cm³; Kim et al., 2011), and Australia (1.21 – 1.69×10^4 particles/cm³; Rumchev et al., 2007). In addition, a large ongoing epidemiological study of UFP in schools was conducted in Melbourne (Australia) (Mazaheri et al., 2014). Other studies from Europe, namely, in Denmark, Germany, and Sweden (Clausen et al., 2012; Fromme et al., 2007; Norbäck et al., 2011), noted lower levels of UFP (0.7×10^3 – 6.5×10^3 particles/cm³) than in the present study. Different levels of urbanization and development of the area surrounding schools, meteorological conditions, or seasonal influences might account for some of these differences (Morawska et al., 2009). It is also necessary to indicate that the majority of the existing studies on UFP in educational settings focused on assessments in classrooms (Clausen et al., 2012; Fromme et al., 2007;

Guo et al., 2010; Mullen et al., 2011; Norbäck et al., 2011; Weichenthal et al., 2008). Zhang and Zhu (2012) also reported the information on UFP in other school microenvironments: gymnasium, canteen, and libraries. In this investigation, classrooms were the microenvironment associated with lower particle number concentrations at both preschools (mean of 9.31×10^3 and 1.13×10^4 particles/cm³ at S1 and S2, respectively), which is reassuring, considering that these are the sites where children spend the majority of their school time. The major identified sources of (ultra)fine particles, based on daily registered information, were classroom cleaning children's activities during classes, such as sculpting, and combustion sources; levels of (ultra)fine particles in ambient air ranged from 2.4×10^3 to 4.3×10^4 (Slezakova et al., 2014). In contrast, at both preschools the highest PNC were observed in canteens (mean of 5.17×10^4 and 3.28×10^4 particles/cm³ at S1 and S2, respectively). Although children spend short periods of time (18 and 19% of their school time at S1 and S2, respectively) in canteens, exposure in this type of indoor microenvironment might be relevant for overall child school exposure. In addition, exposure to high levels of UFP numbers, even if during a limited period of time, may pose some risks to child health (Burtcher and Schüepf, 2012). In agreement with these findings, Mullen et al. (2011) previously reported that cooking events were the most significant indoor sources (during normal occupancy) at six schools in California. The importance of cooking and eating activities was also demonstrated in more recent studies evaluating particle deposition in the alveolar and tracheobronchial region (Buonanno et al., 2011, 2012, 2013b; Mazaheri et al., 2013).

At the three homes the mean concentrations of particles number ranged between 1.09×10^4 and 1.24×10^4 particles/cm³. These levels of PNC were similar to mean concentrations reported in literature for homes in Germany (0.9×10^4 particles/cm³; Fittschen et al., 2013), Greece ($1.3\text{--}1.4 \times 10^4$ particles/cm³; Diapouli et al., 2011), Canada ($0.8\text{--}1.03 \times 10^4$ particles/cm³; Kearney et al., 2011; Wheeler et al., 2011), and Australia (1.24×10^4 particles/cm³; Morawska et al., 2003). However, recently Bekö et al. (2013) conducted a large study that assessed UFP in 56 residences in Denmark and found levels approximately threefold higher than in Portuguese homes (mean of 2.91×10^4 particles/cm³; Bekö et al., 2013). Different study design (sampling period, duration, number of homes) and/or different particle size ranges of measured ultrafine fraction might also contribute to these differences (Morawska et al., 2013).

Overall, the highest mean of PNC and the maximal levels (2.1×10^5 particles/cm³) were observed at H1. Based on the analysis of information available from the questionnaires, indoor sources of UFP at H1 included cooking (boiling and frying), use of toaster and oven, use of cleaning products, vacuuming, and ironing. Certainly the frequency and durations of these indoor activities might have influenced the respective levels. However, it is also necessary to indicate that, contrary to the other two homes, at H1 the room where sampling was conducted was directly connected with a kitchen. In addition, occupants of this home maintained doors between kitchen and living room almost constantly open. Thus, PNC from cooking emissions easily penetrated the sampling area (Bordado et al., 2012; Buonanno et al., 2013b), and accounted for high concentrations at this home. The variation of time and location (room type) might account for the obtained differences of (ultra)fine particles (Bekö, et al., 2013).

Overall, the levels of PNC at the three homes were 10–70% lower than at preschools. However, activities (and the levels of their physical intensity) that are typically performed in an

educational institution vary greatly from those of homes. Therefore, the dose rates resulting from a stay in these two environments might differ considerably.

The highest doses of PNC at preschools were found for children of S1 (Table 3). Although levels of PNC in classrooms were highest at S2, doses of UFP resulting from school exposure were higher (up to 50%) for children at S1, probably due to the higher levels of PNC in the canteen of respective preschool. These findings thus demonstrate that all potential microenvironments need to be considered when assessing children's exposure to PNC in preschools and schools.

The estimated dose rates of indoor PNC at both schools were compared between both age groups of children. The results in Table 3 showed that at S2 dose rates were higher for 5-yr-old children. As mentioned previously, older children performed more frequently physical activities that were associated with the highest BR and consequently led to higher inhalation doses of particles. In contrast, children 3–4 yr old spent more time in classrooms, where levels of PNC were lowest. Further, after lunch children 3–4 yr old slept in classrooms, which was an activity associated with the lowest BR. At S1, the estimated dose rates were not statistically different between children 3–4 yr old and 5 yr old, which was probably due to different activity patterns; older children spent 1.75 h less indoors, and contributions resulting from the outdoor exposure were not considered in this study. Therefore, in future investigations when assessing children, a period spent during school daytime outdoors needs to be considered, as it might be relevant to child overall school exposure.

When evaluating the three homes (Table 3), the highest dose rates of particles were observed for children at H1 due to the highest levels of UFP at this home. When in use, particles samplers make minor noise. Therefore, in order to maintain soundless rest of children it was not possible to conduct measurements directly in children's bedrooms. The obtained dose rates of PNC at H1–H3 thus represent an approximation of child

home exposure and need to be interpreted carefully.

Finally, dose rates of particles in the (ultra)fine range at homes were higher than those of preschools. Although number concentrations of (ultra)fine particles at the three homes were lower than total levels at both preschools (Figure 1), children spent approximately 13 h at homes (opposed to 9 h at preschools). The longer exposure time might account for the obtained values. Data thus indicate that daily activity patterns significantly influenced overall doses to PNC in children 3–5 yr old.

The dose rates of (ultra)fine particles estimated in this study were due to study indoor exposure at preschools and homes. However, on a daily basis children spend some of their time in other microenvironments such as transportation modes and extracurricular activities, where they are exposed to UFP from additional sources. Therefore, characterization of the respective exposures to UFP for children in these microenvironments is of utmost importance. Future studies focusing on health effects of airborne pollutants always need to account for children's exposures in different microenvironments including homes, schools, and transportation modes in order to obtain a correct representation of children's overall exposure.

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