Assessment of ultrafine particles in Portuguese pre-schools: levels and exposure doses

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Practical implications

This study reports information on ultrafine particles in various indoor and outdoor micro-environments (canteens, classrooms, gymnasiums, outdoor) of urban and rural pre-schools. It identifies the potential sources and origins; characterize the influence of meteorological parameters on UFP levels and perform a comparison with other existing international studies. To this date relatively few studies have investigated ultrafine particles (UFP) in pre-schools (none in Portugal) and none assessed exposure dose for different age-groups. The obtained findings showed that levels of UFP in various microenvironments of schools differed significantly. Therefore, in order to obtain an accurate representation of child's overall school exposure profiles, the exposures occurring in these different microenvironments should be always accounted for.

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Abstract

The aim of this work was to assess ultrafine particle (UFP) number concentrations in different microenvironments of Portuguese pre-schools and to estimate the respective exposure doses of UFP for 3-5 years old children (in comparison to adults). UFP were sampled both indoors and outdoors in two urban (US1, US2) and one rural (RS1) pre-school located in north of Portugal for 31 days. Total levels of indoor UFP were significantly higher at the urban pre-schools (mean of 1.82×10^{4} and 1.32×10^{4} particles cm^{-3} at US1 an US2, respectively) than at the rural one (1.15×10^{4} particles cm^{-3}). Canteens were the indoor microenvironment with the highest UFP (mean of 5.17×10^{4}, 3.28×10^{4}, and 4.09×10^{4} particles cm^{-3} at US1, US2, and RS1) whereas the lowest concentrations were observed in classrooms (9.31×10^{3}, 11.3×10^{3}, and 7.14×10^{3} particles cm^{-3} at US1, US2, and RS1). Mean indoor/outdoor ratios (I/O) of UFP at three pre-schools were lower than 1 (0.54–0.93), indicating that outdoor emissions significantly contributed to UFP indoors. Significant correlations were obtained between temperature, wind speed, relative humidity, solar radiation and ambient UFP number concentrations. The estimated exposure doses were higher in children attending urban pre-schools; 3-5 years old children were exposed to 4-6 times higher UFP doses than adults with similar daily schedules.

Keywords (6): ultrafine particles; pre-schools; indoor/outdoor air; children, exposure dose;

Introduction

Up to this date various studies have reported the health risks caused by exposure to particulate matter (Brunekreef et al., 2009; Cassee et al., 2013). In the last years the scientific attention focused on ultrafine particles (UFP), i.e. particles with aerodynamic diameter smaller than 0.1 μm, because evidence indicates that UFP may have a greater potency to cause adverse health effects than large particles (Kumar et al., 2013, 2014; Diapouli et al., 2007). UFP contribute very little to overall particle mass, but they dominate the number concentrations (Morawska et al., 2008). When compared to larger particles, UFP have higher particle number concentration, surface area and larger concentrations of adsorbed (or condensed) toxic pollutants per unit mass (Sioutas et al., 2005). Due to their smaller sizes, UFP can penetrate cell membranes and deposit in the brain tissues and secondary organs (Donaldson et al., 2001; Semmler et al., 2004; Unfried et al., 2007). Combined effects of UFP high surface area and potentially toxic composition may promote physical and chemical reactions inside the organisms that can further result in adverse health outcomes (Kumar et al., 2011; Stone et al., 2007). Studies have shown that exposures to UFP are associated to impaired lung function and pulmonary defense mechanisms, inflammatory responses, asthma, worsening of respiratory diseases and allergic conditions, cardiovascular problems, and even with carcinogenic and genotoxic consequences (Ferreira et al., 2013; Stanek et al., 2011, Terzano et al., 2010).

UFPs are emitted to atmosphere by combustion processes (associated mostly with emission from traffic or industrial sources; Kumar at al., 2010), formed by nucleation and condensation of hot supersaturated vapours while being cooled to ambient temperatures (Sioutas et al., 2005), and by chemical reactions in the atmosphere (Morawska et al., 2008). In addition, indoor UFP may be emitted from indoor combustions (cooking, smoking, candle use) and or result from occupant-related activities (consumer products, paining, cleaning) (Long et al., 2000; Morawska et al., 2003; Wallace, 2006; Bhangar et al., 2011).

The complexity of UFP exposure (spatial variability, indoor sources, infiltration of UPF from various outdoor emission sources, seasonal variability in concentrations and composition; Sioutas et al., 2005) indicates the need to further study this pollutant in order to fully comprehend its impacts on human health. This is especially relevant for sensitive groups. Young children in particular are very susceptible to air pollution (Schwartz, 2004) because they receive a higher dose of airborne particles relative to lung size compared with adults while at the same time their physiological and immunological systems are still developing (Burtscher and Schüepp, 2012; Morawska et al., 2013). In Portugal young children spend approximately 30% of their time (7-8 h per day) at pre-schools, which raises interest in understanding of air pollution in these environments. Nevertheless, as the importance of UFP has been recognized recently, there have been only few studies on UFP levels in schools (Buonanno et al., 2012, 2013a; Clausen et al., 2012; Diapouli et al., 2008; Fromme et al., 2007; Guo et al., 2010; Kim et al., 2011; Morawska et al., 2009; Mullen et al., 2011; Norbäck et al., 2011; Rumchev et al., 2007; Weichenthal et al., 2008; Zhang and Zhu, 2012) and only some of these studies investigated the correlation with outdoor traffic or indoor processes; as far it is known no study was published on UFP levels in Portuguese schools. In addition, during the school time children move between different microenvironments (classroom, gyms, outdoor playgrounds, and etc.) where levels of UFP may vary greatly (Zhang and Zhu, 2012). Therefore, quantification of UFP in these specific microenvironments is essential in order to correctly assess child overall school exposure to UFP.

The aim of this work was to study UFP levels in urban and rural pre-schools in the north of Portugal. The specific objectives of this work were: (i) to quantify UFP number concentrations in different microenvironments of urban and rural pre-schools, and to compare the attained results with other international studies; (ii) to assess the impacts of outdoor UFP to indoor air quality in pre-schools; and (iii) to estimate exposure doses of UFP for 3-5 years old children (in comparison to adults).

Materials and Methods

Characterization of sampling sites

UFP were consecutively measured at three pre-schools in Portugal for 31 days (May-June) of 2013. Pre-schools are educational establishments that provide education for 3-5 old children, prior to the beginning of compulsory attendance at primary schools. Specifically in Portugal "pre-schools" referred to institutions that are directly operated by primary schools. In this work the pre-schools were selected in order to represent different environments. Two pre-schools (US1 and US2) were situated in Oporto Metropolitan Area in Paranhos district (north of Portugal); previously it was demonstrated that vehicular traffic emissions are the main pollution source in this area (Slezakova et al., 2011, 2013). The third pre-school RU1 was located in Xisto also in the north of Portugal but in a rural zone. The detailed characteristic of all three pre-schools are presented in Table 1.

In all three pre-schools, UFP were simultaneously measured at different micro-environments, namely in classrooms (2-3) situated on ground and first floor, and in canteen (1). UFP were also measured in gymnasium or playroom, if existent (Table 1). The characteristics of each studied micro-environment (volume, area, occupancy patterns, number of individuals) as well construction properties (construction materials, ventilation mechanisms, and temperature and relative humidity) are summarized in Tables 1S-3S of the Supplementary material.

In order to better understand the impacts of outdoor UFP emissions to indoor pre-school environments, the levels of UFP were concurrently measured in ambient air (i.e. outdoor).

The traffic densities were estimated for each pre-school (Table 1). During two consecutive days (avoiding Mondays and Fridays) the number of road vehicles was manually counted between 5 a. m. to 12 p. m. during 10 minutes of each hour. These data were used in order to better describe the surroundings of selected pre-schools.

Sample collection

UFP number concentrations in size range 0.02-1 µm were measured by condensation particle counters – TSI P-Trak™ (UPC 8525; TSI Inc., MN, USA). The instrument operates on the principle of condensing 100% grade isopropyl alcohol (Sigma-Aldrich, Steinheim, Germany) onto ultrafine particles in order to increase their dimensions to the size that can be detected. UFP were measured daily between 8:30 a.m. to 5:30 p.m. which corresponded to the period that children were at pre-schools. Intake flow of 0.7 $L.min^{-1}$ was used and UFP logging interval was 60 s accordingly to 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 breathing zone). In each micro-environment, the particles counters were placed as far as possible from windows or doors, and from other probable sources of UFP (heating equipment, blackboards, printers, etc.) in order to minimize direct influence of any source; in canteens the equipment was always positioned in the eating area, as far as possible from the serving area and kitchen where cooking was done. Over the sampling period, the cooking process included boiling, frying and baking; each meal consisted of soup, main dish and desert (typically fruit). All requirements to maintain child safety were fulfilled. During sample collection a researcher was present in order to keep a record of classroom occupancy, ventilation systems (door and window positions), and potential source activities. In addition, teachers and staff were daily inquired regarding the occurrence of additional UFP sources and activities.

The UFP in ambient air were measured at pre-school yards in a safe distance from areas with children intense activity. The samplers were always positioned in open area avoiding any obstacles and barriers (trees, bushes walls, and fences) that could interfere with data collection. The equipment were mounted on support (sampling inlets height at 1.2 m above the ground) and protected from rain. The distance from the main street was 8–42 m.

Indoor temperature and relative humidity were measured by using Testo mini data-logger (174H, Testo; Germany) which operated continuously with a logging interval of 10 min. Information on outdoor meteorological conditions, namely temperature (T), relative humidity (RH), wind speed (WS), and precipitation (P) were retrieved from the local meteorological stations and are summarized in Table 1.

Dose rate exposure analysis

UFP dose rates from inhalation exposure were calculated using Equation 1 (Kalaiarasan et al. 2009, Castro et al., 2009):

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

where D is the age-specific dose rate (particle number kg^{-1}); BR_{WA} is the age-specific weighted average breathing rate (L min^{-1}); BW is age-specific body weight (kg); C_{WA} is the age-specific weighted average concentration (particles L^{-1}); OF is the occupancy factor (considered 1, as children kept their schedules and associated locations tightly); N is the total time per day spent by age-specific children in the pre-school (min day^{-1}). UFP dose rates were estimated for 3-5 years old children that were the common age group in all three pre-schools (Table 1). The daily activity patterns of children were analyzed during each day. Locations in which the different activities happened during the day were identified. Total daily residence time of children spent in each micro-environment 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 timetable and activity patterns is shown in Table 4S of the Supplementary material. As the information concerning the Portuguese population is not available, the age-specific factors were retrieved from U.S. EPA data (U.S. EPA, 2011) considering the mixed population (both male and females). BW of 18.6 kg for 3-5 years old children was used. The BR were selected as the followings: 4.3 L.min^{-1} for rest or sleep; 4.5 L min^{-1} for sedentary or passive activities; 11.0 L min^{-1} for light intense activity, and 37.0 L min^{-1} for highly intense activities (running, etc.). BR_{WA} was estimated then as weighted average, i.e. considering the intensity of performed activities in each microenvironment and the amount of time spent there. The exposure doses were estimated using the UFP average concentrations (weighted by the real time that children spent in each microenvironment). Table 5S of the Supplementary material shows examples of UFP exposure doses calculation. For comparison, dose rates of inhalation exposure to UFP were also estimated for the teachers and pre-school staff (aged 25-64 years). Time schedules of teachers and pre-school staff (i.e. period spent in each micro-environment) were considered the same as of children. Age specific parameters BR_{WA} (12 L min^{-1}; i.e. light physical activity) and BW (77 kg) were used (U.S. EPA, 2011).

Statistical methods

t Student's test was applied ($p < 0.05$; two tailed) in order to establish the statistical significance of the existing differences between the calculated averages. In order to assess the impact of outdoor UFP on indoor environments, the associations between indoor and outdoor UFP number concentrations were estimated by a bivariate linear regression, assuming a linear relationship. Spearman's rank correlation coefficient (*rs*) ($p < 0.05$) was also calculated to assess the influence of meteorological parameters on UFP number concentrations.

Results and discussion

UFP number levels

The UFP number concentrations measured in various micro-environments of the three pre-schools are presented in Table 2, which shows the mean and ranges obtained for each micro-environment, as well as, the total UFP concentrations.

At all three pre-schools, canteens were the micro-environment with the highest levels of UFP particle number concentrations. Examples of representative daily profiles of UFP concentrations in canteens are shown in Figure 1S of the Supplementary material. During the morning UFP concentrations were increasing. When meals were served, typically between 11:30 and 14:00 (i.e. the highest room occupancy), UFP reached the maximal levels. After that, cooking activities stopped, children and staff left the eating area (canteens were vacant for the rest of day), and consequently UFP levels continuously decreased. In all three pre-schools the canteens were directly connected through serving areas (i.e. open spaces) to kitchens equipped with gas fueled stoves. Therefore, cooking emissions could easily penetrate to the eating area and seem to represent the main emission source of UFP in these micro-environments (Buonanno et al., 2013b). The highest levels of UFP were found at canteen of US1 (1.6 and 1.3 times higher than at US2 and RS1, respectively), which was the one with the highest number of enrolled students (Table 1). Similarly, lowest UFP levels were observed at US2 (pre-school with the smallest number of enrolled students).

In all three pre-schools, classrooms were the micro-environment with lower UFP number concentrations. This finding is somewhat reassuring, given that it is the micro-environment where children spend most of their school time. Out of the three pre-schools, classrooms at RS1 exhibited the lowest levels of UFP which might be due to the lack of urbanization and/or anthropogenic sources of this site. At US1 and US2 the concentrations of UFP in classrooms were, approximately 30 and 60% higher, respectively than at RS1; the differences between the means of UFP in classrooms of rural and urban pre-schools were statistically significant ($p < 0.05$). Specifically, the highest mean of UFP was found at classrooms of US2, probably due to the room organizations, sizes and characteristics; classrooms at US2 were the smallest and most cluttered (Table 2S). Furthermore, within each pre-school, the levels of UFP were significantly different in classrooms on 0 and 1 floor ($p < 0.05$). In addition, it is necessary to point out that temporarily (3 up to 120 minutes) UFP concentrations reached high levels in classrooms of all three pre-schools. These increases were associated with specific indoor sources registered in the classrooms of the three pre-schools which included children activities during classes (i.e. painting, sculpturing, and other arts and crafts activities), combustion sources (candles on birthday cake), and classroom cleaning (dusting and wood polishing) (Morawska et al., 2009). Ventilation by open windows and consequent penetration of UFP from outdoors was also identified as an important source of UFP indoors. This specific source was identified based on the comparisons of the daily activity observations (a research and/or teacher registered open windows) and temporarily increases of UFP. The highest maximal levels of UFP were measured at US1 (up to 13 times higher than estimated mean) during candle burning on birthday cake (Figure 2S of the Supplementary material) and during activity that included clay grinding (Figure 1a).

Gymnasium and playroom exist only at one pre-school (US2 and US1, respectively). The levels of UFP in gymnasium were similar to those at classrooms of the respective pre-school. Playroom exhibited approximately twice higher levels of UFP number concentrations than the classrooms. This room was used for multiple purposes: waiting area to drop off and pick up of children (before, during and after school hours), for extra-curricular activities, to eat snacks, or even as classroom or gymnasium for 3-5 years old children. Consequently, levels of UFP varied greatly and concentration profiles exhibited considerable variances every day. Among the identified sources were: children physical activities (dancing, exercising), painting, cleaning and use of chemical products, and ventilations (opened windows and doors).

Total mean UFP concentrations were determined using all measured data for each pre-school despite the inexistence of some micro-environments in some pre-schools (gymnasium, playroom), association with highly specific indoor sources (canteen), and small occupancy times. The highest total mean levels of UFP were found at US1 being 1.4 and 1.6 higher than at US2 and RS1, respectively; the results showed that the total means of UFP at urban pre-schools were statistically different ($p < 0.05$) than at rural one. Nevertheless, these findings need to be interpreted with care once UFP were measured at three pre-schools consequently. The comparisons of UFP particles with other studies are shown in Table 3. The total levels of UFP in the three characterized Portuguese pre-schools were similar to those of Southern Europe, namely Italy (Buonanno et al., 2012, 2013), Greece (Diapouli et al., 2008), Australia (Rumchev et al., 2007) and South Korea (Kim et al., 2011). Other studies from Europe (Germany, Sweden, France), North America (USA and Canada), and Australia reported levels of UFP in pre-schools 3-17 times lower than in the present work. Seasonal influences, meteorological conditions, level of urbanization and overall development of area where the pre-schools were located could account for some of these differences (Morawska et al., 2009; WHO 2006). Other study design (sampling period, duration, number of pre-schools) could also contribute to the obtained differences (Morawska et al., 2013). In addition, differences in the measured particle range, especially in terms of lower cut off size could also account for some of the existent results (Kumar et al., 2010). Finally, with exception to the study by Zhang and Zhu (2012) and Diapouli et al. (2007, 2008) all other works assessed UFP only in classrooms. The total concentrations in the present study also considered various other micro-environments of pre-schools. In that regard, canteens were especially relevant indoor places (Table 2). The levels of UFP in canteens were 3-6 times higher than in classrooms which consequently contributed to higher overall average in the studied schools; absence of these special micro-environments in other studies could also justify the differences between UFP levels.

Indoor/ outdoor UFP

The statistical parameters of average UFP number concentrations outdoors (i.e. pre-school yard) and indoors (in classrooms) at three pre-schools are presented in Figure 2. Examples of UFP concentration profiles in ambient air and in the classrooms of three pre-schools are shown at Figure 1a–c. At the urban pre-schools the mean of UFP concentrations in outdoor air was 1.72×10^4 and 1.21×10^4 particles cm^{-3} at US1 and US2, respectively. The statistical analysis of these results indicated that they are significantly different ($p < 0.05$). The previously conducted study has shown that emissions from vehicular traffic are the main pollution source to ambient air in this area (Slezakova et al., 2013) and the higher traffic density nearby US1 (Table 1) may account for some of the observed differences. At pre-school situated in rural area, mean concentration UFP in ambient air (1.02×10^4 particles cm^{-3}) was significantly lower ($p <$ 0.05) in comparisons with urban ones, probably due to much lower traffic density and lower influence of anthropogenic sources (Table 1); the mean of UFP was at RS1 70 and 20% lower than at US1 and US2, respectively. Natural sources of UFP, namely atmospheric formations and emissions from vegetation (plantations, forests) (Diapouli et al., 2007; Morawska et al., 2008) that surrounded the vicinity of rural pre-school might account for these levels. It is necessary to repeat that UFP at three schools were sampled during different dates, which could also account for some of the observed differences. Furthermore, during the UFP sampling at RS1 soil plowing and other farming activities were registered during three days at several plantations which might contribute to observed levels of UFP in ambient air (Figure 1c). In addition, the results in Figure 2 show that overall levels of UFP in the classrooms of RS1 were lower than in ambient air; this pattern was also observed in the urban pre-schools. On the contrary, maximal levels of UFP outdoors were lower than indoors ones. These occurrences were due to the presence of specific indoor sources (combustion, indoor activities) in the classrooms of three pre-schools that caused during relatively short periods of time high levels of UFP, especially high maxima of UFP were observed at US1.

In order to further evaluate the influence of outdoor emissions to indoor air quality, I/O ratios between the concentrations of UFP in classrooms and in outdoor air were calculated. At US1 the values of I/O ratios ranged between 0.13 and 9.77 (mean of 0.54) whereas it was between 0.31 and 4.72 at US2 (mean of 0.93); the respective ratio range was between 0.35 and 2.59 at RS1 (mean of 0.70). Overall, the mean I/O ratios

PROOF

were similar to those previously reported (Buonanno et al, 2013a; Weichenthal et al., 2008). At all three pre-schools, the mean values of I/O ratios were lower than 1 which indicates that outdoor emissions may influenced UFP levels in classrooms. It is necessary to point out that high values of maximal I/O ratios (9.77 for US1, 4.72 for US2 and 2.59 for RS1) probably indicate contribution of UFP from indoor sources with RS1 being the least influenced by those sources.

The influence of air quality to indoors was also analyzed by bivariate linear regression, assuming a linear relationship (Figure 3a-c). It is possible to observe that at US2 and RS1 (Figure 3 b-c) indoor and outdoor UFP were relatively well associated (with R^2 of 0.82 and 0.58, respectively) which further supports the previous findings concerning the impacts of outdoor air. At US1 (Figure 3a) the linear regression between the indoor and outdoor UFP concentrations was poorer (R^2 of 0.14) due to the much high variance of indoor UFP levels (temporal contribution from specific indoor sources).

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Influence of meteorological parameters

The potential influence of indoor (T, RH) and ambient parameters (T, SR, RH, and WS) on indoor and outdoor UFP number concentrations were analyzed through the calculation of Spearman's correlation coefficient (Table 4). For the analysis of indoor UFP, classrooms were the only considered indoor micro-environment (due to their existence in all three pre-schools); canteens were not considered in order to avoid the specificity of cooking emissions. Positive correlations were found between T and UFP number concentrations both indoors and outdoors. In addition, SR was positively correlated with outdoor UFP number concentrations so the positive correlation between UFP, SR and outdoor T might be due to photochemical activity, leading to an increase in the concentration of UFP (Park et al., 2008). Evaluating different fractions of ultrafine particles, Wang et al. (2010) reported that nucleation mode particles (4–10 nm, 10–30 nm in diameter) are more affected by SR and T, whereas Aitken mode fractions (30–50 nm and 50–70 nm) corresponded closely to RH. The results of this study are somewhat inconclusive concerning RH. Whereas outdoor UFP and RH were inversely correlated at all three pre-schools, the correlation coefficients for indoors were only significant for US2. Finally, WS showed significant inverse correlations with outdoor UFP as reported also by Wang et al. (2011). In agreement with these findings, Weichenthal et al. (2008) also reported inverse correlations of WS and UFP number concentration. High WS might influence the observed UFP number concentration profiles during the sampling, promoting a higher instantaneous variability and oscillation of UFP number concentrations. It is necessary to point out that although the obtained results appear to be consistent with the finding of previous studies, the observed associations between the meteorological parameters and UFP may be influenced by unmeasured confounding factors between these parameters.

Exposure dose analysis

The inhalation exposure dose rates of UFP were estimated for 3-5 years old children that were the common age group at all three pre-schools (Table 5). At both urban pre-schools, 3-5 years old children were divided into the classes according to their age (though differently at both pre-schools). These age-classes had different daily schedules and activities. For example the youngest rested (i.e. napped) after lunch for 2-2.5 hours whereas older children spent daily more times outdoors (0.75-1.75 h). The organization structure of the rural pre-school was simpler and all children between 3-5 years were joined to the same class and they all had the same daily schedule and/or activity patterns. The highest exposure doses of UFP were found for children of US1. At both urban pre-schools, classrooms were the micro-environment where children spend majority of their school time (approximately 70-75% for young ones and 57%-70% for older ones). As previously shown (Table 2) overall levels of UFP in classrooms were the highest at US2. Still, for all age categories the exposure doses of UFP were at US1 1.5 times higher than at US2, mostly due to the exposure to higher levels of UFP in canteen of US1. Although children spend in canteens rather limited period of school time (18 and 19% of their school time at US1 and US2, respectively) the contribution to the total exposure of UFP is relevant. In addition, these findings clearly show that when assessing children exposure to UFP in pre-schools, all potential micro-environments should be considered.

The total estimated dose rates between the different age-groups at the two urban pre-schools were also compared. The results in Table 5 clearly show that at both urban schools exposure doses of UFP were approximately 1.5 times for older children (namely 5 years old at US1 and 4-5 years old at US2) than for younger ones (3-4 years and 3 years at US1 and US2, respectively). At each urban pre-school older children spent approximately twice more outdoors (25 and 7% of their school time at US1 and US2, respectively) than young ones (11% at US1 and 4% at US2). Older children also performed more frequently physical activities such as exercising, running, and playing (both indoors and outdoors) which were associated with the highest breathing rates and consequently led to higher inhalation doses of UFP. In agreement with these findings, the dose rates due to outdoor exposure contributed for older children 48 and 27% of the total UFP school dose at US1 and US2 respectively, whereas for younger ones it was 33% at US1 and 19% at US2. In addition, UFP dose rates due to outdoor exposure were higher at US1 where children spent more time outdoors. On the contrary young children spend more time indoors where overall UFP levels were lower that outdoors which might cause the lower total dose rates of UFP (Table 5). In addition, at both pre-schools the younger children napped (in the classrooms) after the lunch which was an activity with the lowest breathing rates.

The estimated total dose rates of UFP at RS1 were similar to those of US1 (3-4 years old) and US2 (4-5 years old). These exposure doses were higher than expected (in a view of lower indoor UFP concentrations at this pre-school; Table 2) probably due to the considerably longer period spent outdoors. At rural pre-school children spent approximately 40% of their school times outdoors and the UFP dose rates due outdoor exposure accounted for 60% of the total school exposure, thus being at RS1 the highest proportion of all three pre-schools. These findings show that daily activity patterns at the respective schools influenced significantly the overall child exposure dose rates to UFP.

Finally, in order to better understand the magnitude of UFP exposures at the three characterized pre-schools, the dose rates of children were compared to those of adults. The results in Table 5 show that exposure doses for 3-5 years old children in the respective pre-schools were 3.6 to 6.4 times higher than those of adults. Considering the high susceptibility of young children, these results demonstrate that pre-schools are an important environment for child overall particles exposure. Finally, the information on the exposure to UFP in children is limited and therefore the findings on UFP dose rates of 3-5 years old children obtained within this work could not be compared with other studies.

Conclusions

This study fills a gap providing information on the UFP levels and exposure doses in Portuguese pre-schools. The results demonstrated that levels of UFP in various microenvironments of pre-schools differed significantly with the lowest levels of UFP observed in the classrooms (where children spend 70-75% of their school time) and the highest ones found in canteens. Therefore, future population-based studies focusing on the health effects of airborne pollutants need to account for the exposures occurring in these different microenvironments in order to obtain a representation of child's overall pre-school exposure profiles. Furthermore, the results of the present study suggested that children attending urban pre-schools are potentially exposed to higher concentrations of UFP in air, mainly due to the contribution of outdoor traffic-related sources and extra cooking activities (usually due to higher number of enrolled students). Nevertheless, the daily activity patterns at the respective schools influenced significantly the overall child exposure dose rates to UFP.

Children represent one of the most vulnerable groups in society. However, in comparison to adults, the exposure doses for 3-5 years old children in the respective pre-schools were 4 to 6 times higher than those of adult. Therefore, in order to provide information for the protection of public health, the future work should focus on the individual exposure of children.

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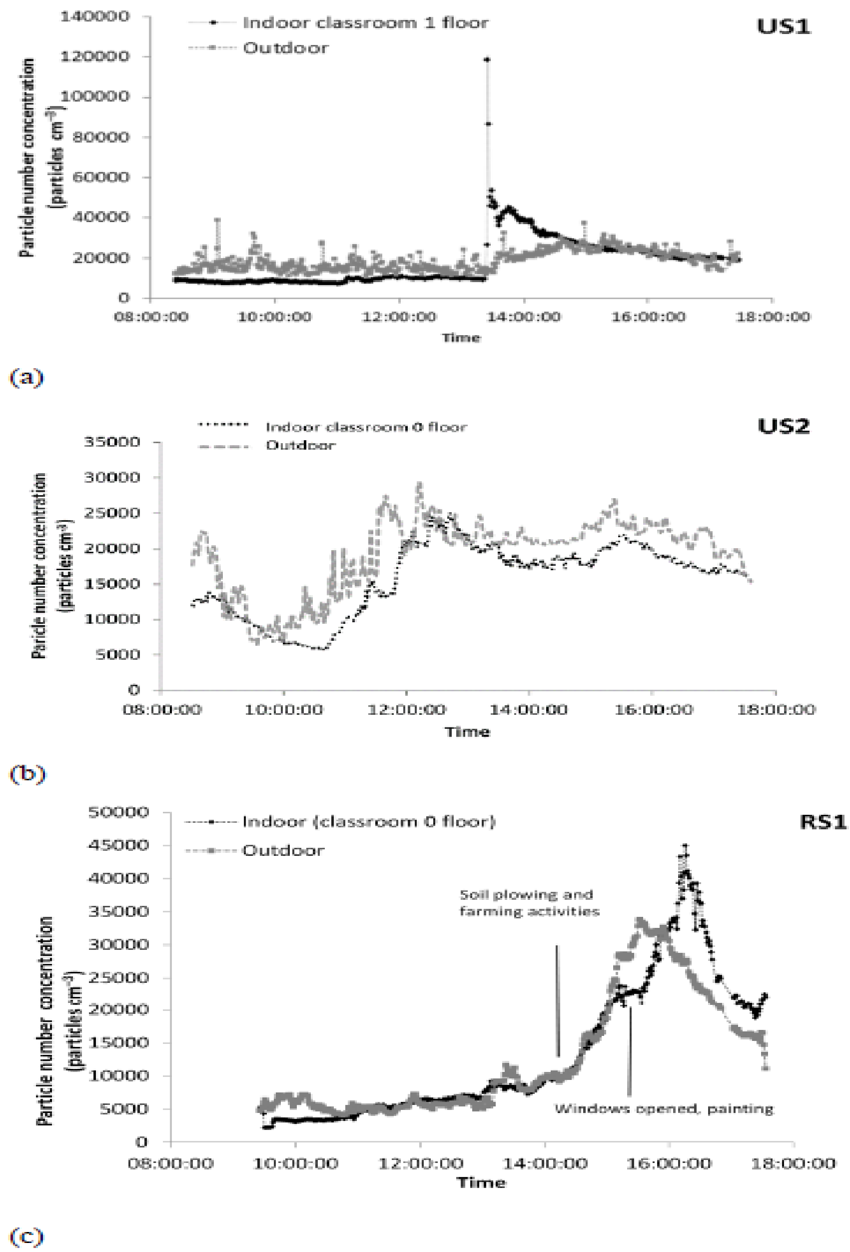


Figure 1. Examples of indoor and outdoor UFP concentrations profiles at three schools: (a, b) urban (US1 and US2), (c) rural (RS1). At urban school US1 the increase of UFP was associated with clay material that was being grinded by teacher for afternoon classes. Concentration trend of UFP at urban school US2 shows the similarity of indoor and outdoor profiles.

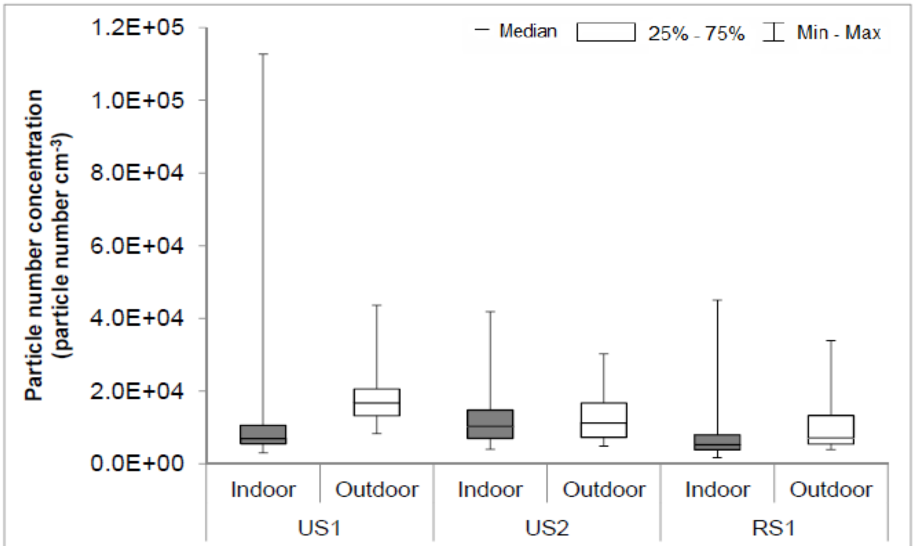
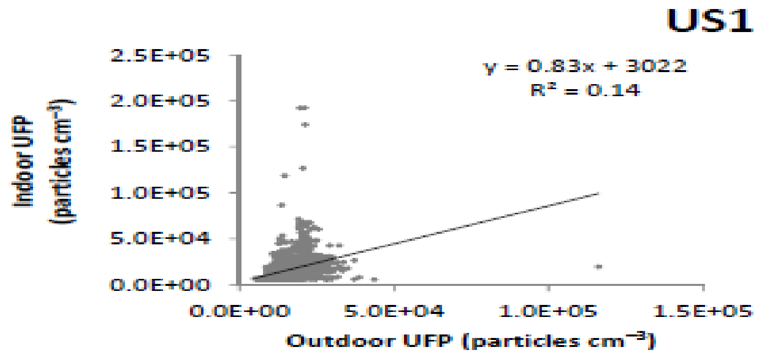
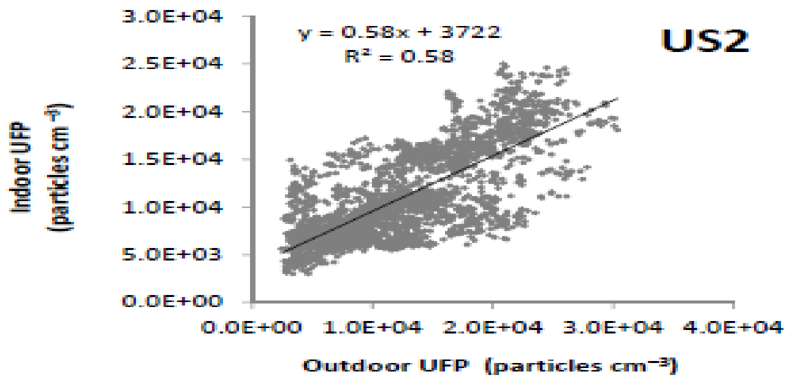


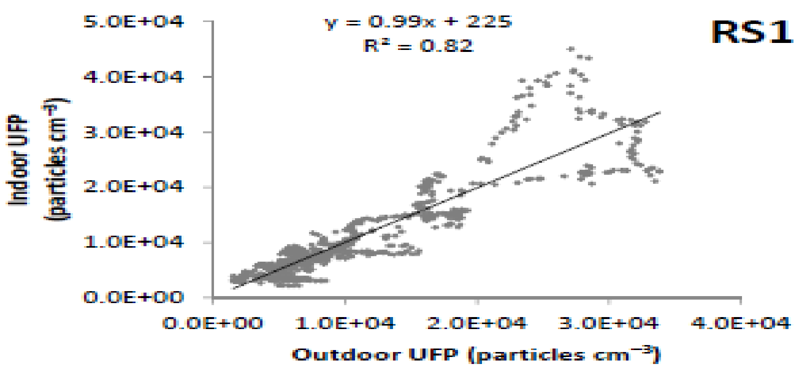
Figure 2. Average UFP number concentrations at two urban (US1 and US2) and one rural (RS1) schools: minimum and maximum values, median, 25th and 75th percentile.



(a)



(b)



(c)

Figure 3. Correlation of indoor and outdoor UFP concentrations at three schools: (a, b) urban (US1 and US2), (c) rural (RS1).

Table 1. Characterization of the studied schools and meteorological conditions during the sampling campaigns

School	Description	Location	Traffic density data	Studied microenvironments	Outdoor meteorological parameters: mean ± SD
US1	Private school Built in 1940 Two-floors building Enrolled children 173: 3-5 years	Urban - traffic Situated on moderately trafficked street	mean: 16 cars/min peak hours: 8:30h (27 cars/min) 18:30h (25 cars/min)	Indoors: classrooms (3) canteen (1) playroom (1) Outdoors: school yard	T: 17.5 ± 2.5 °C RH: 56.3 ± 8.0 % WS: 16.0 ± 5.5 km/h Precipitation: 0.3 ±0.0 mm
US2	Private school Built in 1905 Three-floors building Enrolled students 69: 1-6 years (30: 3-5 years)	Urban - traffic Intersection of moderate and low trafficked street	mean: 13 cars/min peak hours: 8:30h (21 cars/min) 14:30h (18 cars/min) 1 8:30h (18 cars/min)	Indoors: classrooms (3) canteen (1) gymnasium (1) Outdoors: school yard	T: 15.3 ± 1.9 °C RH: 67.0 ± 8.0 % WS: 19.0 ± 6.7 km/h Precipitation: 3.5 ± 0.1 mm
RS1	Public school Built in 1981 Two-floors building Enrolled students 48: 3-11 years (20: 3-5 years) School constructed for children with special needs; 3-5 years old kept in separately from older ones	Rural	mean: < 1 car/min peak hours: 8:30h (1 car/min) 12:00h (1 car/min) 17:30h (2 cars/min)	Indoors: classrooms (2) canteen (1) Outdoors: school yard	T: 16.7 ± 1.1 °C RH: 82.0 ± 5.5 % WS:15.4 ± 5.3 km/h Precipitation: 0.6 ± 0.0 mm

Table 2. UFP number concentrations at the three characterized schools (particle cm^{-3})

		Urban school 1		Urban school 2		Rural school 1	
		Mean	Range	Mean	Range	Mean	Range
Micro-environment	Classrooms[a]	9.31×10^3	$2.51\times10^3 – 1.13\times10^5$	1.13×10^4	$3.01\times10^3 – 4.19\times10^4$	7.14×10^3	$2.24\times10^3 – 4.50\times10^4$
	0 Floor	8.58×10^3	$2.51\times10^3 – 1.04\times10^5$	1.16×10^4	$3.01\times10^3 – 4.19\times10^4$	7.61×10^3	$2.24\times10^3 – 4.50\times10^4$
	1 Floor A	8.80×10^3	$3.14\times10^3 – 1.13\times10^5$	1.24×10^4	$5.87\times10^3 – 2.25\times10^4$	5.91×10^3	$2.53\times10^3 – 1.60\times10^4$
	1 Floor B	1.06×10^4	$4.23\times10^3 – 2.92\times10^4$	8.86×10^3	$3.64\times10^3 – 2.79\times10^4$	n. a.	n. a.
	Canteen	5.17×10^4	$9.28\times10^3 – 1.73\times10^5$	3.28×10^4	$1.05\times10^4 – 2.48\times10^5$	4.09×10^4	$7.18\times10^3 – 1.38\times10^5$
	Gymnasium	n. a.[b]	n. a.	9.72×10^3	$5.46\times10^3 – 1.71\times10^4$	n.a.	n.a.
	Playroom	1.70×10^4	$5.28\times10^3 – 1.93\times10^5$	n. a.	n. a.	n. a.	n. a.
Total		1.82×10^4	$2.51\times10^3 – 1.93\times10^5$	1.32×10^4	$3.01\times10^3 – 2.48\times10^4$	1.15×10^4	$2.24\times10^3 – 1.38\times10^5$

[a] based on the measurements of all the classrooms (0 and 1 floor A, B)

[b] n.a. not available (i.e. inexistent micro-environment)

Table 3. UFP at schools: summary of existent studies

Continent	Country	Mean (range) (particle cm^{-3})	Particle fraction	Study design	Reference
Europe	Portugal	$14.9–19.3\times10^3$ ($2.23\times10^3–1.93\times10^5$)	0.02–1 µm		This study
	Germany	6.5×10^3 ($2.6-12.1\times10^3$)	10-487 nm	64 primary and secondary schools; 36 classrooms; Sampling both in winter and summer; Sample collection during 1 day for 5 h;	Fromme et al., 2007
	Greece	24×10^3 (n.r. -52×10^3)	0.01–1 µm	7 primary schools; Different indoor microenvironments; Samples collected in 2 winter periods; 8 hour sample collection (8:00 to 16:00);	Diapouli et al., 2007, 2008
	Denmark	1.6×10^{3a} ($2.2-36.4\times10^3$)	n.a.	150 day-care facilities (children 1-5 years old)	Clausen et al., 2012
	Italy	$12-40\times10^3$ (n.r.)	10–300 nm	3 schools (2 primary and 1 secondary); Sample collection for two days; Personal exposure assessment: 100 children aged 8-11 years; Various microenvironments/activities;	Buonanno et al., 2012
	Italy	$19.5-20.4\times10^3$ (n.a.)	< 100 nm	3 schools; 2-3 classrooms in each school; 2 weeks sampling in each schools; Sample collection during school hours (8:30 to 13:30 or to 16:30)	Buonanno et al., 2013a
	Sweden	$0.7-4.4\times10^3$ (n.a.)	0.01–1 µm	1 elementary school; Total of 61 classrooms; Sampling repeatedly during 3 weeks; 3 h sample collection;	Norbäck et al., 2011
North America	Canada	5.4×10^3 ($1.1-10.4\times10^3$) 4.6×10^3 ($1.0-11.4\times10^3$)	0.02–1 µm	2 schools: 1 elementary 1 secondary; 37 classrooms; Sampling during three 1-week periods; Sample collection for 7 h (8:30 to 15:30);	Weichenthal et al., 2008
	California, USA	6.9×10^3 ($2.1-21.7\times10^3$)	< 100 nm	1 school; 6 classrooms; Samples collected during 18 days;	Mullen et al., 2011
	Texas, USA	$0.9-3.8\times10^3$ ($0.6-29.3\times10^3$)	7.6–100 nm	5 schools; Various microenvironments; Sample collection during 3-8 days in each school;	Zhang and Zhu, 2012
Australia		$12.1-16.9\times10^{3a}$ (n.a.)	0.01–1 µm	6 primary schools (3 new and 3 old); 4-6 classrooms in each school; 8 hour sample collection;	Rumchev et al., 2007
		5.2×10^3 (n. a. - 140×10^3)	< 100 nm	1 primary school; 3 classrooms; Samples collected in 60 days (2 winter periods); 23 h sample period;	Morawska et al., 2009
		3.19×10^3 (n.a. -110×10^3)	< 100 nm	1 primary school; 1 classroom, 1 pre-school center (children < 6 years old) Sample collection for 10 days continuously;	Guo et al., 2010
Asia	South Korea	18.2×10^3 ($3.7-52.8\times10^3$)	0.02–1 µm	34 schools; Sample collection for 7 day periods for each school;	Kim et al., 2011

[a]-median;

n.a. not available

Table 4. Spearman correlation coefficients between UFP number concentration, indoor (T, RH) and outdoor meteorological parameters (T, RH, WS, SR) at the two urban (US1, US2) and rural (RS1) schools

	Urban school 1		Urban school 2		Rural school 1	
	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
T (° C)	**0.423**	**0.119**	**0.205**	**0.598**	**0.254**	**0.473**
RH (%)	-0.029	**–0.430**	**-0.308**	**–0.478**	-0.070	**–0.630**
WS ($km\ h^{-1}$)	n.a.[a]	**–0.136**	n.a.[a]	**–0.171**	n.a.[a]	**–0.301**
SR ($W\ m^{-2}$)	n.a.[a]	**0.108**	n.a.[a]	**0.178**	n.a.[a]	**0.581**

Note: values in bold are statistically significant for $p<0.05$;

[a]Not available

Table 5. Total age-specific dose rates of UFP at two urban (US1 and US2) and one rural (RS1) school

	Dose rate (particles $kg^{-1}day^{-1}$) Children				
	US1		US2		RS1
	3–4 years	5 years	3 years	4–5 years	3–5 years
Mean	4.60×10^{9}	7.52×10^{9}	2.94×10^{9}	4.48×10^{9}	4.50×10^{9}
Min	1.06×10^{9}	1.84×10^{9}	1.04×10^{9}	1.37×10^{9}	9.50×10^{8}
Max	2.95×10^{10}	4.20×10^{10}	1.48×10^{10}	2.51×10^{10}	1.92×10^{10}

	Adults				
	US1		US2		RS1
Mean	1.12×10^{9}	1.25×10^{9}	8.17×10^{8}	1.01×10^{9}	7.01×10^{8}
Min	2.60×10^{8}	3.05×108	2.90×108	2.87×108	1.48×108
Max	7.21×10^{9}	6.95×10^{9}	4.11×10^{9}	5.25×10^{9}	3.00×10^{9}