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Individual dose and exposure of Italian children to ultrafine particles

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Abstract - Time-activity patterns and the airborne pollutant concentrations encountered by children each day are an important determinant of individual exposure to airborne particles. This is demonstrated in this work by using hand-held devices to measure the real-time individual exposure of more than 100 children aged 8-11 years to particle number concentrations and average particle diameter, as well as alveolar and tracheobronchial deposited surface area concentration. A GPS-logger and activity diaries were also used to give explanation to the measurement results. Children were divided in three sample groups: two groups comprised of urban schools (school time from 8:30 am to 1:30 pm) with lunch and dinner at home, and the third group of a rural school with only dinner at home. The mean individual exposure to particle number concentration was found to differ between the three groups, ranging from 6.2×10^4 part. cm^{-3} for children attending one urban school to 1.6×10^4 part. cm^{-3} for the rural school. The corresponding daily alveolar deposited surface area dose varied from about 1.7×10^3 mm^2 for urban schools to 6.0×10^2 mm^2 for the rural school. For all of the children monitored, the lowest particle number concentrations are found during sleeping time and the highest were found during eating time. With regard to alveolar deposited surface area dose, a child's home was the major contributor (about 70%), with school contributing about 17% for urban schools and 27% for the rural school. An important contribution arises from the cooking/eating time spent at home, which accounted for approximately 20% of overall exposure, corresponding to more than 200 mm^2 . These activities represent the highest dose received per time unit, with very high values also encountered by children with a fireplace at home, as well as those that spend considerable time stuck in traffic jams.

Keywords: particle number concentration, individual exposure, children daily dose, deposited surface area, dose intensity.

1. Introduction

Epidemiological studies have demonstrated that exposure to particulate air pollution is associated with several adverse health effects (Pope and Dockery, 2006). Long-term exposure to high

concentrations of particulate matter (PM) increases the risk of lung cancer, respiratory diseases and arteriosclerosis, whereas short-term exposure peaks can cause exacerbation of several forms of respiratory diseases, including bronchitis and asthma, as well as changes in heart rate variability (Sorensen et al., 2003). To date, the majority of these studies have dealt with the relationship between health outcomes and ambient levels of PM₁₀ and PM_{2.5}, which are the mass of particles with an aerodynamic diameter of $\leq 10 \mu\text{m}$ and $2.5 \mu\text{m}$, respectively. Recently, however, interest has focused on ultrafine particles (UFPs, diameter $\leq 100 \text{ nm}$), due to the adverse health effects caused by their high alveolar deposition fraction, large surface area, chemical composition and potential to translocate to the circulation (Donaldson and Tran 2002; Schins et al. 2004; Braniš et al., 2010; Weichenthal, 2012), as well as their ability to induce inflammation, penetrate into cell membranes (Unfried et al., 2007) and deposit in secondary organs (Semmler et al., 2004) and brain tissue (Calderon-Garciduenas et al., 2004). In particular, these effects are much more pronounced in children because they inhale a higher dose of UFPs relative to both lung size (when compared with adults) (Buonanno et al., 2012; Burtcher and Schüepf, 2012) and increased breathing rates, since they are generally more physically active than adults (Bateson and Schwartz, 2008; Pinkerton and Joad, 2006). Physiological research has shown that exposure of developing lungs to particulate matter can permanently affect the lungs themselves, and in particular, exposure to UFPs in early life can result in persistent alterations in distal airway architecture that are characterized by an initial decrease in airway cell proliferation (Lee et al., 2010). While there is considerable toxicological evidence of the potential harmful effects of UFPs on human health, there are still insufficient epidemiological studies to draw conclusions on the dose-response relationship concerning this aerosol fraction, especially with regard to children (WHO, 2005).

1.1 Children's personal exposure

An exposure assessment is defined as the process of estimating or measuring the magnitude, frequency and duration of exposure to an agent, along with the number and characteristics of the population exposed (Ott, 1982). With regard to the exposure assessment of children, five different spatial scales can be carried out: i) “city scale”, the broadest and most common scale used to characterize air quality across several city blocks using remote measurements; ii) “outdoor scale”, which is representative of particle exposure outside school buildings and grounds; iii) “indoor scale”, which reflects indoor-based exposure in classrooms; iv) “individual scale”, where the sampling location is within 3 meters of the person; and v) “personal scale”, using hand-held

instruments carried as a personal monitor, with a distance between the sampling site and the nasal cavities < 30 cm (Cattaneo et al., 2010).

Wallace and Ott (2011) carried out measurements of personal exposure to UFPs by positioning a portable condensation particle counter (CPC 3007, TSI Shoreview, MN, USA) inside homes, cars and restaurants. They identified a number of important indoor sources, ranging from cooking on stoves (both gas and electric) and toaster ovens to the use of hair dryers. Vinzents et al. (2005) studied personal exposure in terms of number concentrations of UFPs in the breathing zone, using hand-held condensation particle counters (CPC 3007, TSI, St. Paul, MN, USA) for 15 healthy non-smoking subjects (who were also determined to have oxidative DNA damage) over six 18-hr periods. They found that biologic effects of UFPs occur from modest exposure, such as that occurring in traffic, which supports the relationship between UFPs and the adverse health effects of air pollution.

In the literature, no studies have investigated the daily personal or individual exposure of children to UFPs and until now, most child exposure assessment studies have used data based on fixed monitoring site measurements inside and/or outside school grounds, and background particle concentrations measured at some central location in the urban area of interest (Janssen, 2001; Kim et al., 2004). PM₁₀ has a smaller spatial and temporal variation in a given location with respect to UFP concentrations, which are primarily generated from gas-to-particle conversion and high-temperature combustion sources present in urban areas (particularly during rush hours), which rapidly decrease with increasing distance from the source (Buonanno et al., 2011a). Because of the different fluid dynamics and sources, UFP exposure evaluations have to be carried out differently from those for coarse particles, and data from fixed monitoring stations are generally not useful in assessing such exposure. Furthermore, ambient concentrations are generally insignificant compared to the concentrations found in other microenvironments, which also depend on the exact whereabouts of individuals (Klepeis, 2006). In fact, each individual person is exposed to pollutant concentrations in a different way, depending on their lifestyle and the different microenvironments which they frequent. When an individual makes a trip from one location to another, their personal exposure can be defined as the weighted average of concentrations present in each single microenvironment through which he or she travels, taking into account the amount of time he or she spends in each one (WHO, 1999).

In general, a child's daily overall exposure is dominated by particle concentration levels in three main microenvironments: at home, on school grounds and aboard transportation (Ashmore and Dimitroulopoulou, 2009; Xue et al., 2004; Hussein et al., 2012). Therefore, children attending the

same school can still receive different doses, depending on the characteristics of their home environment and the mode/s of transport which they use.

1.2 Aims of the work

In previous works (Buonanno et al., 2011b; Buonanno et al., 2012), activity pattern data were combined with micro-environmental data (human activities and particle number size distributions) using an indirect approach, in order to evaluate the dose of alveolar and tracheobronchial-deposited particle number and surface area experienced by different age groups in Italy and Australia. From the Italian study, it was found that the major activities contributing to alveolar and tracheobronchial particle number deposition in children aged 6-10 years were sleeping and resting (13% and 15%, respectively), eating (22% and 18%, respectively) and transportation (19% and 20%, respectively), while the contribution from time spent at school was less than 10%. With regard to surface area, the predominant contribution was from meal times (>54%). This finding confirms the importance of the time spent eating, where high particle concentrations are likely to remain in the air following cooking activities. With regard to transportation microenvironments, the highest dose intensity values, in terms of particle number deposition, were found for children (6-10 years old), indicating that it is very important to consider the exposure of children to UFPs emitted by traffic.

The current work was carried within the international project titled “Ultrafine particle from traffic emission on children health (UPTECH)” (Queensland University of Technology, Brisbane, Australia), which is being undertaken in response to the lack of epidemiological results concerning the effects of exposure to UFPs emitted by motor vehicles on children's health in schools (<http://www.ilqhq.qut.edu.au/Misc/UPTECH%20Study%20Design.htm>). This paper deals with the children's individual exposure to UFPs, in order to identify the activities and microenvironments that make the greatest contribution to a child's average daily dose. In fact, the driving force for this exposure will be the activity pattern of each child and the microenvironments they visit each day. Given that short-term exposure may contribute significantly to average daily exposure, the daily dose of alveolar and tracheobronchial deposited surface area, together with daily exposure to particle number concentration was measured for over 100 children, and a detailed study of each child's daily activity patterns was conducted based on the Global Positioning Systems (GPS) and diaries carried by each child.

2. Methodology

2.1 Study design, sampling sites and population

The measurements were carried out on week days in Cassino, in Central Italy (41°30'0'' N - 13°50'0''E), which can be considered a typical busy Italian middle town (resident population: 33,000 inhabitants; daily commuter workers and students: 20,000; surface area: 83 km²), between October 2011 and March 2012. Personal exposure measurements were performed on children aged 8-11 years who attended three different schools, (S₁, S₂ and S₃):

- S₁ is a primary school located on an urban street with traffic mostly dominated by light vehicles (50% diesel cars). Traffic density on the roads around S₁ was 36 ± 2 vehicles min⁻¹ and traffic peak hours were detected at 8:30am and 1:30pm, which correspond to the times in which pupils enter and exit from school, respectively (Buonanno et al., 2011a). Three school buses were used to transport children to this school.
- S₂ is a secondary school close to the intersection of moderately and heavily trafficked urban streets (7.7% heavy duty vehicles, typically buses). Traffic density and traffic peak hours on the roads around S₂ was comparable to those for S₁ (Buonanno et al., 2011a), where students also entered the school at 8:30am and exited at 1:30pm.
- S₃ is a primary school located in a rural area far away from urban traffic (average traffic density: 4 ± 1 vehicles min⁻¹). At this school, pupils arrived at 8.30am and left the school grounds around 4.15pm, except for one day a week, when children in the fifth grade left the school at 5.45pm.

In order to describe the study population, and measure potential confounders and effect modifiers relevant to the analysis (such as housing conditions, socio-economic status, exposure to environmental tobacco smoke and ethnicity), a questionnaire was developed following the International Study of Asthma and Allergies in Childhood (ISAAC) guidelines. A time-activity diary (which provides information on potential peak exposures in specific locations) was also completed by each child under the supervision of their parents. Overall, 103 children agreed to participate in this project, of which 33 attended S₁, 25 attended S₂ and 45 attended S₃.

2.2 Instrumentation and quality assurance

The mobile experimental apparatus was composed of three hand-held UFP counters (NanoTracer, Philips) equipped with GPS tracking. This device works by diffusion charging, using an electrometer that measures the number particle concentration by means of the current induced by previously charged particles collected on a filter inside a Faraday cage. The NanoTracer is also able to evaluate the different fractions of the lung deposited surface area through a semi-empiric

algorithm implemented by Marra et al. (2010). These personal monitors are equipped with an internal rechargeable lithium-ion battery, which allows them to be used during outdoor trips. The total run time (single battery charge) is about 7 hours. The NanoTracer can operate in two different modes: *fast mode* and *advance mode*. The *fast mode* measures real-time particle number concentrations (in the range 10-300 nm), while the *advance mode* measured both particle size and concentration.

These counters were calibrated at the beginning of the experimental campaign, in order to allow for data quality assurance by comparison with: i) a Condensation Particle Counter (CPC, TSI Model 3775) to measure particle number concentration; ii) a Nanoparticle Surface Area Monitor (NSAM, TSI Model 3550) to assess the human lung-deposited surface area of particles (reported as $\mu\text{m}^2 \text{cm}^{-3}$) corresponding to tracheobronchial (TB) and alveolar (A) regions of the lung; and iii) a Scanning Mobility Particle Sizer Spectrometer (SMPS, TSI Model 3936) to measure the mean diameter of the particle number size distributions.

The calibration was conducted within a closed box (about 16 L), in a uniform and stationary environment, in terms of number concentration. Different polydisperse aerosols were generated from a watery solution of sodium chloride (NaCl) by a TSI 3940N aerosol generator in the 5-300 nm diameter range. Three tests were performed under stationary conditions (30 min of measurements), at low (10 000 part. cm^{-3}), medium (20 000 – 80 000 part. cm^{-3}) and high concentrations (> 120 000 part. cm^{-3}). The correction factors, defined as the ratio between the reference values measured by the reference instrument (CPC for the particle number concentration and NSAM for alveolar-deposited surface area of particles) and the ones obtained by the Nanotracers, varied between 1.3 at high concentrations and 0.9 at low concentrations. Therefore, the Nanotracers were found to under-count at high concentrations, however they were reasonably accurate at medium and low particle number concentrations.

A further CPC TSI 3775 was used to measure total particle number concentration at a background site. This instrument, located on a rooftop at the University of Cassino, was protected from rain and wind, and provided information on airshed exposure. Particle number concentrations were continuously measured with a 30 s time resolution during the experimental campaign. Measurement quality assurance of the CPCs was guaranteed through calibration checks and flow checks conducted at the start of the monitoring periods: each condensation particle counter was calibrated in the European Accredited Laboratory at the University of Cassino and Southern Lazio by comparison with a TSI 3068B Aerosol Electrometer.

2.3. Methodology description

Each child kept the NanoTracer device for two days, carrying it with them in all of the microenvironments where he or she spent their time. The children were also asked to record their main indoor and outdoor activities (such as studying, eating, transportation, sleeping etc), indicating the start and end times for each activity. The temporal resolution of the NanoTracers was set to 16 s (*advance mode*).

Based on the time duration of each activity, the corresponding average particle number concentration, diameter, and deposited alveolar and tracheobronchial surface area concentrations were calculated. The dose (in terms of deposited alveolar or tracheobronchial surface area) received by 8-11 year old children in each microenvironment/activity was determined by multiplying the alveolar and tracheobronchial surface area ($S_{a,tb}$) for the time spent (T) in the j^{th} microenvironment and the inhalation rate ($IR_{activity}$) corresponding to the activity carried out (Klepeis, 2006). Then, we added the partial doses to estimate the daily total deposited alveolar and tracheobronchial surface area (dose), $\overline{S_{a,tb}}$, as reported in eq. (1).

$$\overline{S_{a,tb}} = \sum_{j=1}^n \{ IR_{activity} \cdot S_{a,tb} \cdot T \}_j \quad (1)$$

Inhalation rates for the different activities were adopted on the basis of the US EPA approach (US EPA, 2004), ranging from $0.3 \text{ m}^3 \text{ h}^{-1}$ during sleeping and resting to $1.4 \text{ m}^3 \text{ h}^{-1}$ during sporting activities.

In order to analyze the contributions of each activity/microenvironment in more depth, we determined the “exposure (dose) intensity”, in order to compare exposure (dose) in different microenvironments by linking the daily exposure fraction with the daily time fraction, as described in equation (2) (Wang et al., 2011):

$$\text{Exposure (dose) intensity} = \text{Daily exposure (dose) fraction (\%)} / \text{Daily time fraction (\%)} \quad (2)$$

3. Results and Discussion

3.1 Study population and daily time activity analysis

Table 1 shows the key characteristics of the study population. Data collected from the questionnaires highlight that among children attending the two urban schools (S_1 and S_2), 50% and 44% live in urban areas, 37% in rural areas and more than 13% in suburban areas. Of those children who lived in an urban area, 75% go to school by car and 25% on foot. The data collected for S_3 was quite different, with only 3% of students living in an urban area. While gas cooking was the only cooking method used in the homes in this study, fireplaces were a more common heating method than the use of gas heaters.

Figure 1 shows the relative contributions, C , (minimum, first quartile, median, third quartile and maximum) of each activity/microenvironment analyzed to the daily time activity pattern (grey box plots) for the three schools (S_1 and S_2 , S_3). Children who attended urban schools S_1 and S_2 spent 21% (300 min) of their daily time at school and 71% (1017 min) at home, of which 6% was spent during lunch time and 37% was for sleeping. In contrast, the children at S_3 spent less daily time at home (59%, 868 min), with more time spent at school (35%). Moreover, they spent less time eating (4%), because the children ate pre-cooked lunches at school, and therefore, they were not exposed to the emissions from cooking activities during lunch time. Therefore, the time spent eating by children at S_3 was considered to have the same emissions profile as school time. The daily time contribution for time spent on transportation ranged between 2% (29 min) for children at S_3 and 4% (56 min) for children at S_1 and S_2 .

The values obtained were in good agreement with the data presented in the literature. For example, Chau et al. (2002) reported that individuals from Hong Kong (China) spent an average of 86% of their time indoors, 3-7% in enclosed transit and 3-7% outdoors. Brasche and Bischof (2005) carried out an analysis of the time spent indoors at home, with a mean time equal to 942 min (65%). The overall mean time spent at home is also in good agreement with results from American (940 min, 65%) and Canadian (950 min, 66%) human activity surveys carried out in the nineties, as reported by Leech et al. (2002). Generally, a child's daily activity pattern is characterized by longer periods spent at home (Schweizer et al., 2007; McCurdy et al., 2003; Xue et al., 2004; Hussein et al., 2012) and the daily time spent in different microenvironments is affected by several influential parameters: type of day (weekday, weekend, holiday) ambient temperature, gender. In a previous paper, the authors estimated the time spent by 6-10 years old children at home (73%, 1051 min), as well as during transportation (4.9%, 71 min), sleeping (37%, 537 min) and eating time (4.8%, 69 min), on the basis of the Italian daily activity patterns database (Buonanno et al., 2011b). The good agreement between this and other studies confirms that in Western countries the adopted lifestyles are similar, in terms of daily activity patterns. Different considerations can be withdrawn from

studies carried out in Korea and China (Yang et al., 2011; Jim and Chen, 2009): these population activity patterns are, in fact, substantially different from those in Western countries.

3.2 Exposure to particle number, alveolar and tracheobronchial deposited surface area concentrations.

In Table 2, average particle number, and alveolar and tracheobronchial deposited surface area concentrations are reported for children attending the three schools (S_1 , S_2 , S_3). The daily average particle number concentrations are equal to 6.2×10^4 part. cm^{-3} , 3.9×10^4 part. cm^{-3} and 1.6×10^4 part. cm^{-3} for S_1 , S_2 and S_3 , respectively. Morawska et al. (2008) carried out a meta-analysis of 71 UFP studies performed in several microenvironments. They found mean concentrations of 2.6, 4.8, 7.3, 10.8, 42.1, 48.2, 71.5 and 167.7×10^3 particles cm^{-3} for clean background, rural, urban background, urban, street canyon, roadside, on-road and tunnel environments. Therefore, children attending S_1 and S_2 are exposed to daily UFP exposure typical of on-road and street canyon environments, respectively. For all of the children monitored, the lowest particle number concentrations were found during sleeping time, with average particle number concentrations (and average particle size) equal to $3.8 \times 10^4 \pm 1.7 \times 10^4$ part. cm^{-3} (mode diameter equal to 84 nm) for children attending S_1 , $2.4 \times 10^4 \pm 9.9 \times 10^3$ part. cm^{-3} (101 nm) for S_2 and $7.4 \times 10^3 \pm 3.6 \times 10^3$ part. cm^{-3} (136 nm) for S_3 . Maximum particle number concentrations were detected during eating time, with $2.3 \times 10^5 \pm 2.1 \times 10^5$ part. cm^{-3} (84 nm) for S_1 , $9.2 \times 10^4 \pm 5.1 \times 10^4$ part. cm^{-3} (67 nm) for S_2 and $9.4 \times 10^4 \pm 4.1 \times 10^4$ part. cm^{-3} (79 nm) for S_3 . The sleeping time presents a higher mode diameter range (84-136 nm) in respect to eating time (67-84 nm) because of the presence of more aged particles. The high levels of exposure during eating time are a result of the high emission factors of indoor cooking activities, as well as the reduced use of hoods and forced ventilation systems in Italian kitchens (Buonanno et al., 2009, 2011b; He et al., 2004; Hussein et al., 2006). Children are also exposed to high particle number concentrations during their time spent on transport, with average particle number concentrations equal to $6.8 \times 10^4 \pm 2.8 \times 10^4$ part. cm^{-3} , $5.5 \times 10^4 \pm 2.4 \times 10^4$ part. cm^{-3} and $2.1 \times 10^4 \pm 8.2 \times 10^4$ part. cm^{-3} for children from S_1 , S_2 and S_3 respectively.

The daily average alveolar (and tracheobronchial) deposited surface area concentrations were $1.9 \times 10^2 \mu\text{m}^2 \text{cm}^{-3}$ ($38 \mu\text{m}^2 \text{cm}^{-3}$) for children attending S_1 , $1.5 \times 10^2 \mu\text{m}^2 \text{cm}^{-3}$ ($31 \mu\text{m}^2 \text{cm}^{-3}$) for S_2 and $59 \mu\text{m}^2 \text{cm}^{-3}$ ($12 \mu\text{m}^2 \text{cm}^{-3}$) for S_3 . Minimum alveolar surface area concentrations were detected during sleeping for children from S_1 and S_2 , and during school time for children from S_3 . Once again, maximum values were found, for all children, during eating time (average values of $3.6 \times 10^2 \mu\text{m}^2 \text{cm}^{-3}$ and of $7.2 \times 10^1 \mu\text{m}^2 \text{cm}^{-3}$ for the alveolar and tracheobronchial deposited surface area

concentrations, respectively). These values represent high exposures in comparison to the data reported by other studies. For example, Wilson et al. (2007) measured the alveolar and tracheobronchial deposited surface area in Minneapolis and East St. Louis, USA and found typical values ranging between 10 - 50 $\mu\text{m}^2 \text{cm}^{-3}$ and 5 - 20 $\mu\text{m}^2 \text{cm}^{-3}$, respectively. Moshhammer and Neuberger (2003) found acute asthma-like effects of active particle surfaces on the pulmonary function of elementary school children, with half-hour mean values for deposited active surface area ranging between 4.80 - 343 $\mu\text{m}^2 \text{cm}^{-3}$, and a mean value of 58 $\mu\text{m}^2 \text{cm}^{-3}$.

3.3 Dose, dose intensity and contribution to the daily dose of the different activities/microenvironments of alveolar deposited surface area.

In Table 3, dose, dose intensity and the contribution to the daily dose of the different activities/microenvironments for alveolar deposited surface area are reported for children attending the three schools (S_1 , S_2 , S_3). The daily dose of children attending urban schools was equal to $1.93 \times 10^3 \pm 1.03 \times 10^3 \text{ mm}^2$ and $1.53 \times 10^3 \pm 6.4 \times 10^2 \text{ mm}^2$, respectively, which is in very good agreement with the corresponding value ($1.72 \times 10^3 \text{ mm}^2$) determined by Buonanno et al., (2011b) for 6-10 years old attending schools in Italy from 8.30 am to 1.30 pm on weekdays. The daily dose for children attending S_3 was considerably lower than for S_1 and S_2 , and is similar to the value determined for Australian children, of about $4.0 \times 10^2 \text{ mm}^2$ (Buonanno et al., 2012). The main reasons for this low value are the location of the school and houses (97%) in a rural area and the fact that children only partake in one eating time during the day, which is similar to the activity patterns of Australian children (Buonanno et al., 2012).

Figure 2 reports the average particle number concentration for all children attending a) urban (S_1 and S_2) and b) rural schools (S_3) compared to background levels. Starting from 7.00am, children attending S_1 and S_2 (Figure 2a) were exposed to higher levels of UFP compared to background values until 8:25 am. The main reasons for this finding are due to indoor UFP generation from cooking activities (breakfast) and transportation exposure. From 8:25 am to 1.30 pm (school time), the exposure is lower compared to background levels, which is typical of an indoor microenvironment without any relevant particle number sources. During the afternoon and evening, two major peaks are clearly shown and these refer to cooking activities during lunch and dinner time. The contribution from cooking activities does not only affect UFP levels during eating time, but it also contributes to exposure during other time spent at home. The main difference between children attending the rural school (S_3) is a lower background exposure (due to the location of the school and houses) and the presence of one only peak due to cooking activities at dinner time.

With regard to the alveolar deposited alveolar surface area dose, we found that a child's home was the major contributor (72%, 71% and 65% for children of S_1 , S_2 and S_3 , respectively), even for children attending S_3 , who spent more time at school than at home (see Table 3). School time contributes 16% to the daily time activity patterns for urban schools (with school time from 8.30am to 1.30pm), and 27% for the rural school (with school time from 8.30am to 4.15pm). An important contribution arises from cooking/eating time, with a time fraction of about 15%, corresponding to more than 200 mm^2 . This activity presents the highest dose intensity (greater than 3.7), highlighting the very high dose received per time unit during eating time. The contribution of cooking/eating time is even higher because children are also exposed to high concentrations after the afternoon and the evening eating times. Therefore, even if school and eating time made a similar contribution to the daily dose, the dose intensity is very different, as shown in Figure 1.

In order to better understand the overall population of students attending urban schools (S_1 and S_2), we separated it into several distinct sub-populations according to: gender, smoking parents, presence of fireplaces at home, traffic jams during the school-home route, prevalent type of transportation (car, walking), and the location of the children's houses. Figure 3 shows the relative exposure to particle number concentration, E , and received dose of alveolar deposited surface area, D , for the different sub-populations, with respect to the overall population of students attending urban schools S_1 and S_2 .

The exposure and dose received by females was higher compared to males, with the main additional contribution related to a higher frequency of females in the kitchen and/or adjacent rooms during and post cooking activities. Very high values were also encountered for children with a fireplace at home and for ones that experienced traffic jams on their way to or from school. However, the dose is relatively lower than exposure because of the reduced inhalation rate during these activities. In contrast, children that walk to school generally exhibit a higher inhalation rate and therefore, experience a greater dose compared to exposure. Finally, the location of the houses also seems to be relevant, with children living in urban areas experiencing a higher exposure of about 25%.

4. Conclusions

In this study, children's individual exposure and dose to UFPs were measured during a 6 month experimental campaign, in order to evaluate the contribution of different activities and microenvironments. To this purpose, measurements of particle number concentration, and alveolar

and tracheobronchial deposited surface area were conducted for 8 – 11 year old children attending three different schools in Cassino (Southern Italy). Time activity data were recorded in a diary completed by each child and location data were recorded by a Global Positioning System (GPS).

Our results showed that children attending urban schools, with a higher percentage of subjects living in urban areas than those attending a rural school, experienced higher individual exposure to particle number concentration, mainly due to the extra cooking activities and traffic-related sources they encountered.

In terms of time, a child's home was found to be the most significant microenvironment, contributing to daily exposure to particle number and alveolar deposited surface area dose, accommodating around 70% of a child's daily activities. This contribution mainly comprised of sleeping and eating times, which despite making similar daily contributions to dose, were totally different in terms of exposure. For example, sleeping was characterized by low particle concentrations over a long duration, while eating was characterized by high concentrations over a shorter time period. Children were also exposed to high particle number concentrations during transportation to and from school, with children travelling by car exposed to greater UFP levels than those who walked to school. The exposure and dose received by females was higher when compared to males, and particularly high exposures were experienced by children with fireplaces at home and those that experienced traffic jams on their way to or from school. Finally, the location of the children's houses also seemed to be relevant, with children living in urban areas experiencing a higher exposure of about 25%.

The results of this work show the importance of individual exposure assessment, in order to provide information for the protection of public health, especially for children who represent one of the most vulnerable groups in society. Personal exposure studies should be carried out in developed countries as an essential tool to identify health risks, set and review air quality standards and evaluate effective policy interventions. Future work will focus on the individual exposure of adult females during cooking activities, since they generally experience greater exposure compared to adult men and children.

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Table 1 - Characteristics of Study Population (%). (Total number of children N =103, age = 10.1 ± 1.1 years)

Characteristic	%		
	S ₁	S ₂	S ₃
Male	50	50	59
Living in urban/rural/suburban area	50/37/13	44/37/19	3/97/0
Going to school by car/walking/bus	94/6/0	79/14/0	55/26/19
Urban route /suburban/mixed	59/0/41	34/18/48	0/82/18
Heavy traffic during transport	19	21	3
Smoking in the home	6	13	7
Living in damp home	50	32	36
Pets in the home	31	32	26
Parental education high/medium/low	34/59/7	40/56/5	22/67/11
Gas cooking	100	100	100
Heating gas/fireplace/both	13/62/25	18/50/32	19/33/48
After school care	0	11	13
Living in other house (>50 days/year)	19	18	3

Table 2 - Particle number, alveolar and tracheobronchial surface area deposited concentrations.

Microenvironment or activity	N (part. cm ⁻³)			S _a (μm ² cm ⁻³)			S _{tb} (μm ² cm ⁻³)		
	S ₁	S ₂	S ₃	S ₁	S ₂	S ₃	S ₁	S ₂	S ₃
School	4.0×10 ⁴	3.3×10 ⁴	1.2×10 ⁴	1.5×10 ²	1.5×10 ²	3.5×10 ¹	3.0×10 ¹	3.4×10 ¹	7.0×10 ⁰
Indoor (home)	6.3×10 ⁴	4.5×10 ⁴	2.6×10 ⁴	1.9×10 ²	1.7×10 ²	9.3×10 ¹	3.9×10 ¹	3.3×10 ¹	1.9×10 ¹
Indoor (other)	5.1×10 ⁴	3.8×10 ⁴	2.0×10 ⁴	3.6×10 ²	1.2×10 ²	4.8×10 ¹	7.4×10 ¹	2.5×10 ¹	9.7×10 ⁰
Sleeping	3.8×10 ⁴	2.4×10 ⁴	7.4×10 ³	1.4×10 ²	1.1×10 ²	4.2×10 ¹	2.8×10 ¹	2.2×10 ¹	8.5×10 ⁰
Cooking/Eating	2.3×10 ⁵	9.2×10 ⁴	9.4×10 ⁴	5.6×10 ²	2.8×10 ²	2.4×10 ²	1.1×10 ²	5.7×10 ¹	4.9×10 ¹
Transportation	6.8×10 ⁴	5.5×10 ⁴	2.1×10 ⁴	2.1×10 ²	1.8×10 ²	7.3×10 ¹	4.2×10 ¹	3.7×10 ¹	1.5×10 ¹
Outdoor	2.2×10 ⁴	6.7×10 ⁴	1.8×10 ⁴	8.8×10 ¹	2.4×10 ²	6.9×10 ¹	1.4×10 ¹	4.3×10 ¹	1.4×10 ¹
Daily average	6.2×10⁴	3.9×10⁴	1.6×10⁴	1.9×10²	1.5×10²	5.9×10¹	3.8×10¹	3.1×10¹	1.2×10¹

Table 3 – Dose, dose intensity and contribution to the daily dose of the different activities/microenvironments of alveolar deposited surface area.

Microenvironment or activity	Alveolar deposited surface area dose (mm ²)			Dose Intensity			Daily dose fraction (%)		
	S ₁	S ₂	S ₃	S ₁	S ₂	S ₃	S ₁	S ₂	S ₃
School	287±137	265±58	160±61	0.71	0.91	0.79	15%	17%	27%
Indoor (home)	460±231	407±377	128±73	1.05	1.09	1.55	24%	27%	21%
Indoor (other)	312±318	121±169	43±43	0.95	0.84	1.29	16%	8%	7%
Sleeping	377±150	274±120	110±64	0.66	0.61	0.48	20%	18%	18%
Cooking/Eating	295±290	180±146	89±105	3.58	2.45	5.23	15%	12%	15%
Transportation	102±75	86±73	18±16	1.24	1.52	1.48	5%	6%	3%
Outdoor	94±105	193±171	52±45	0.58	1.48	1.29	5%	13%	9%
Daily dose	1926±1029	1526±635	601±205						

Figure captions

Fig. 1 – Relative contributions (minimum, first quartile, median, third quartile and maximum) of each activity/microenvironment analyzed on the daily alveolar deposited surface area dose for S_1 , S_2 and S_3 (white box plots). Grey box plots represent the corresponding relative contributions to the daily time activity pattern. The time spent by children at S_1 and S_2 was constant. Red line box plots are referred to S_1 , green to S_2 and black to S_3 .

Fig. 2 – Average particle number concentration trends vs. background particle number concentrations for children attending a) urban and b) rural schools.

Fig. 3 – Relative exposure to particle number concentration, E , and received dose of alveolar deposited surface area, D , of sub-populations with respect to the overall population of students attending urban schools S_1 and S_2 .







