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Exposure to Submicron Particles and Estimation of the Dose Received by Children in School and Non-School Environments

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Abstract: In the present study, the daily dose in terms of submicron particle surface area received by children attending schools located in three different areas (rural, suburban, and urban), characterized by different outdoor concentrations, was evaluated. For this purpose, the exposure to submicron particle concentration levels of the children were measured through a direct exposure assessment approach. In particular, measurements of particle number and lung-deposited surface area concentrations at “personal scale” of 60 children were performed through a handheld particle counter to obtain exposure data in the different microenvironments they resided. Such data were combined with the time–activity pattern data, characteristics of each child, and inhalation rates (related to the activity performed) to obtain the total daily dose in terms of particle surface area. The highest daily dose was estimated for children attending the schools located in the urban and suburban areas ($>1000 \text{ mm}^2$), whereas the lowest value was estimated for children attending the school located in a rural area (646 mm^2). Non-school indoor environments were recognized as the most influential in terms of children’s exposure and, thus, of received dose ($>70\%$), whereas school environments contribute not significantly to the children daily dose, with dose fractions of 15–19% for schools located in urban and suburban areas and just 6% for the rural one. Therefore, the study clearly demonstrates that, whatever the school location, the children daily dose cannot be determined on the basis of the exposures in outdoor or school environments, but a direct assessment able to investigate the exposure of children during indoor environment is essential.

Keywords: exposure assessment; school; children; number concentration; lung-deposited surface area; dose

1. Introduction

Many studies highlighted the link between the exposure to airborne particles and health effects, such as respiratory diseases and inflammation [1], cardiovascular diseases [2,3], diabetes [4], higher systolic blood pressure and pulse pressure [5], and decreased cognitive function in older men [6]; in particular, the World Health Organization (WHO) estimated that the overexposure to particulate matter (PM) causes about 4.2 million deaths per year worldwide [7]. Moreover, the WHO has recently classified PM, referred to as outdoor pollution, as a carcinogenic pollutant for humans (group 1) [8–10]. The harmful potential of airborne particles is related to their ability to penetrate and deposit in the deepest areas of human respiratory tract (i.e., alveolar region), causing irritation, inflammation and possible translocation into the blood system, carrying with them carcinogenic

and toxic compounds [11–14]. The inhalation and consequent deposition of these compounds are strictly related to the size of the carrying particles: higher deposition fractions in the lungs are characteristics of submicron and ultrafine particles [15]. Moreover, smaller particles are also recognized to translocate from lungs to the cardiovascular system and from there to other organs (liver, spleen, kidneys, brain) [16–18].

In the last years, the attention of scientific studies has shifted from super-micron particles (whose contribution is expressed in terms of mass concentrations of particles smaller than 10 and 2.5 μm , i.e., PM_{10} and $\text{PM}_{2.5}$) [19,20] to submicron and ultrafine particles (UFPs, particles smaller than 100 nm) whose contribution is better related to particle number [21,22] and surface area concentrations [23,24] than mass concentration. In fact, many studies highlighted that dose-response correlation in terms of human health effects is better related to surface area of particles deposited in the lungs than other metrics of exposure. To summarize, particle surface area is the most relevant dose metric for acute submicron particle lung toxicity [1,25–32].

In light of this, to evaluate the health effect of the exposure to airborne particles, a critical factor that should be assessed and provided to medical experts is the dose of submicron particles received by individuals [33–35]. Moreover, the airborne particle dose is the main input data for human health risk model [36–39]. Airborne particle doses received by people can be evaluated on the basis of measurements obtained from ad-hoc exposure assessment research. Nonetheless, even though the scientific community is moving from particle mass-based (PM) to number- and surface area-based metrics (submicron particles), the current legislation is still limited to the outdoor concentration of PM_{10} and $\text{PM}_{2.5}$; such measurements are limited to some outdoor fixed sampling points (FSPs) placed in specific points classified as a function of the type of site (rural, urban, suburban) and the type of station, i.e., proximity to main sources (background, industrial, or traffic) [40–42]. Moreover, PM_{10} and $\text{PM}_{2.5}$ measurements at FSPs cannot be considered proxies for exposure to submicron and ultrafine particles since they present different dynamics (e.g., dilution, deposition) and origins/sources [43–50]. Indeed, differently from PM_{10} concentrations that are typically quite homogeneously distributed around the city, the concentrations of submicron particle metrics (number and surface area) are strongly affected by the proximity to the source [51,52]. Finally, the measurement at an outdoor FSP cannot take into account for the exposure in indoor environments; therefore, a proper evaluation of the overall human exposure to submicron and ultrafine particles can be only obtained through personal monitoring able to measure the exposure at a personal scale and also to include the exposure in indoor microenvironments [53–55].

One of the most vulnerable populations in terms of air pollution exposure is represented by children [56,57]. This is due, amongst other things, to their high inhalation rates, resulting in larger specific doses than adults [58–61]. Children use to spend a large part of their day in indoor environments, such as schools and homes. In our previous studies involving adults, we found that some environments and activities affect the total daily dose more than other ones: in particular, the indoor environments were recognized to contribute up to 90% of the total daily dose in terms of particle surface area, with cooking and eating activities alone accounting up to 50% [53,62,63]. Schools as well may be considered a critical indoor environment under certain circumstances, in fact, the long exposure time in schools (children spend from 175 to 220 days and from 5 to 8 hours at school [64]) could significantly affect the overall dose received by children. Actually, the exposure (and then the dose) in school environments is not affected by the presence of submicron particle sources (smoking is typically not allowed and cooking activities are in most of the cases no longer performed in the school) but mainly by the outdoor-to-indoor penetration of submicron particles produced outdoors, which depends on (i) airtightness of the building, (ii) type of ventilation and (iii) particle physical-chemical properties (e.g., size) [65–71]. Therefore, the location of the school, as highlighted in few previous studies [39,72–74], is the main parameter affecting the students' exposure to submicron particles leading to critical exposure scenarios for those attending schools located near highly trafficked urban roads. To the best of the authors' knowledge the dose of submicron particles received by children in school and non-school environments was investigated just in one (our) previous paper [55], but in this study the

investigated schools (both in the rural and urban areas) were placed in the same city, thus the possible contribution of the outdoor concentration levels to the daily dose was not adequately deepened.

Within this context, the aim of the present research is to evaluate the actual exposure to submicron particles of children attending schools located in different urban contexts and cities (urban, suburban rural sites) and to estimate the corresponding doses received both in schools and in other non-school environments where they spend time. To this end, an extensive experimental campaign was performed by measuring the personal exposure of 60 children (for 48 h each) attending three different schools in Italy, characterized by different outdoor concentration levels, using wearable monitors able to measure particle number and lung-deposited surface area concentrations.

2. Methodology

2.1. Study Area and Monitoring Site

Children considered in the experimental campaign attended three naturally ventilated schools located in three different cities in Italy (Salerno, South of Italy, Roma, Central Italy, and Parma, North of Italy); the locations of the three schools within the urban contexts are completely different. In particular, the school in Salerno (S1) is placed in a suburban area as it is 1.6 km outside of the city centre, but quite close to a highway. The school in Rome (S2) is located in the urban area, and, in particular, in the proximity of highly trafficked roads, whereas, the school located in Parma (S3) is in the rural area, about 5 km from the city centre, and quite far from trafficked roads. The experimental campaigns in the three schools were performed from November 2018 to May 2019 for about two months in each school as summarized Table 1.

Table 1. School sites, sampling periods and summary of the meteo-climatic conditions (temperature, T, relative humidity, RH) and air quality parameters (NO₂, PM₁₀ and PM_{2.5}) measured by the closest fixed sampling stations of the Italian environmental protection agency. The data related to every single period of the campaign are expressed as daily average values and their ranges (min–max).

School	City	Sampling Period	T (°C)	RH (%)	NO ₂ (µg·m ⁻³)	PM ₁₀ (µg·m ⁻³)	PM _{2.5} (µg·m ⁻³)	Distance of the Closest Fsp to the School and Definition According to the Standard
S1	Salerno	November–December 2018	15 (13–17)	71 (n.a.)	34 (8–66)	21 (8–56)	13 (5–44)	Distance: <100 m type of site: suburban type of station: background
S2	Roma	February–March 2019	11 (6–16)	66 (n.a.)	42 (26–122)	29 (7–63)	16 (12–18)	Distance: 1 km type of site: urban type of station: traffic
S3	Parma	April–May 2019	15 (10–18)	71 (n.a.)	12 (10–42)	12 (<5–27)	7.5 (<5–21)	Distance: 12 km type of site: rural type of station: background

In order to better describe the three sampling sites in terms of outdoor air quality, in Table 1, the distance of the closest fixed sampling point (FSP) installed by the Italian environmental protection agency to the schools are reported, as well as its definition in terms of type of station (background, industrial, or traffic) and type of site (urban, suburban rural sites). The closest FSPs to school S1 (100 m), S2 (1 km), and S3 (12 km), are defined as suburban/background station, urban/traffic station, and rural/background station, respectively. The parameters measured by the three FSPs during the three different sampling periods (November–December 2018, February–March 2019, and April–May 2019 for school S1, S2 and S3, respectively) clearly highlight the different outdoor air quality of the locations investigated: indeed, the highest NO₂, PM₁₀ and PM_{2.5} values were measured by the FSP close to the school in Rome (S2) (average values of 42, 29, and 16 µg·m⁻³, respectively) whereas the lowest, as expected, were measured by the FSP close to the school in Parma (S3) (average values of 12, 12, and 7.5 µg·m⁻³ for NO₂, PM₁₀ and PM_{2.5}, respectively). The authors, once again, point out that

the concentration of the different PM fractions cannot be considered as a good proxy for ultrafine or submicron particles. Indeed, the latter, along with NO₂, are good markers of the tailpipe emissions of the vehicular traffic, whereas PM₁₀ is only partially due to tailpipe emissions of vehicles (a significant fraction is due to the traffic-induced particle resuspension) and it is a good marker, amongst others, of biomass combustion for residential heating [75]. Therefore, an overall correlation between outdoor concentrations of PM₁₀ and submicron particles can be found, but, in some conditions (e.g., co-presence of other sources) these two metrics could be poorly correlated. Actually, since the FSPs close to S1 and S2 are strongly affected by traffic sources, a good correlation between PM₁₀ and submicron particles is somehow expected; this is partially confirmed by the fact that NO₂ and PM fractions data shown in Table 1 present very good correlations (linear regressions with r² equal to 0.95 and 0.99 for PM₁₀ and PM_{2.5}, respectively). Finally, regarding the meteo-climatic parameters, temperature and relative humidity values were found to be roughly similar in the three sites during the three measurement periods. This is a not trivial aspect—indeed, generally, the time of the year (e.g., season) can affect the children’s exposure and doses both in terms of time-activity patterns and ventilation of the microenvironments since warmer conditions would have increased the time spent outdoor and the manual ventilation in indoor environments (e.g., schools and homes). Thus, the similar outdoor meteo-climatic conditions had a relatively negligible effect on the time of the year on the results.

2.2. Study Design

To evaluate the surface area dose received by children attending the three schools considered in the present study, particle number (PN) and lung-deposited surface area (LDSA) concentrations and average particle sizes (D_p) were measured by means of a personal monitor, which is a handheld diffusion charger particle counter (NanoTracer, Philips). The children were equipped with the mobile monitor fixed to a belt at the hip for 48 h.

During the campaign, 20 children for each school (60 children in total) were monitored. In particular, children aged 6–10 years were monitored (both males and females). Measurements were performed only on school days; weekends were not considered in the study. The authors monitored such high number of children in each school in order to obtain sufficient data that could be representative of the exposure level in each microenvironment where they live/reside. Indeed, the exposure of the children in each microenvironment and during each activity was affected by several parameters, such as the outdoor concentration levels, the volume of the indoor environments, and the presence and the strength of indoor sources (e.g., cooking, smokers, incense, candles etc.). As an example, the children’s exposure when they stay in the kitchen during parents’ cooking activities is strongly affected by the kitchen volume and the different types of foods and stoves ([76–80]), thus having performed different measurements (on different children) allowed the authors averaging amongst all these influencing parameters. Similarly, the exposure during transport can vary significantly as a function of the transportation modes (i.e., car, walking, bus, etc.; [43,81]), thus, once again, multiple measurements allowed to take into account for all these conditions.

In order to estimate the dose, the children, with the support of their parents, were asked to fill in an activity diary to take note about the place, time, and activity performed. A pre-compiled form of the activity diary was prepared by the authors and given to the children along with the portable instrument; the form was prepared considering 15-min time slots (e.g., 00:00–00:15, 00:15–00:30, etc.) in order to make it easy to fill in the forms with the required information. The diary was then used during the data post-processing in order to evaluate the time spent in each activity (i.e., the time-activity pattern) and to determine the exposure during each activity and in each microenvironment. The daily dose of the children under investigation in terms of particle surface area in the tracheobronchial and alveolar regions of the lungs (δ), was calculated as sum of the dose received during the activities performed in the *j* microenvironments:

$$\delta = \sum_{j=1}^n \{IR_{activity,j} \cdot LDSA_j \cdot T_j\} \text{ (mm}^2\text{)} \quad (1)$$

where IR_{activity} ($\text{m}^3 \cdot \text{h}^{-1}$) is the inhalation rate of the child, LDSA is the Lung-Deposited Surface Area concentration ($\mu\text{m}^2 \cdot \text{cm}^{-3}$), and T_j (h) is time spent in each microenvironment. The IR_{activity} is a function of the age and activity performed by the children; in particular, we have considered the IR data for 6–10-year-old children summarized in Buonanno et al., 2012 [63]. In Equation (1) the term “microenvironment” is used for the sake of simplicity: the activities performed by the children, obtained based on the time-activity patterns, were grouped in six main microenvironments, summarized in Table 2. Particular attention should be paid to the “Cooking & Eating” microenvironment; indeed, children do not perform cooking activities per se, thus, the exposure related to this microenvironment is due to cooking activities performed by the parents. To compare the received dose of the children in different microenvironments, the dose-intensity ratio (i_{δ} , $\text{mm}^2 \cdot \text{min}^{-1}$), i.e., the ratio between the daily dose fraction and the daily time fraction characteristics of each microenvironment, was also evaluated [53].

Table 2. Classification of the activities performed by the citizens in seven main microenvironments.

Microenvironment	Activities
Transportation	Trip and use of time not specified, round-trip to work
School	All type of activities performed in school environments
Cooking & eating	Cooking, eating and drinking
Outdoor day	Gardening and animal care, restoration, sport and outdoor activities, physical workout, Productive exercise, Sports-connected activities
Indoor day	Personal care, studying not specified, studying in the free time, activities for home and family not specified, housework, purchasing goods and services, helping adult family members, helping other family members, active activities, social activities and entertainment, social life, entertainment and culture, inactivity, hobbies and computer science, art and hobbies, computing, playing, media, reading, watching TV, DVD or videos, listening to the radio or recording
Sleeping	Sleeping

2.3. Instrumentation and Its Quality Assurance

As mentioned above particle number (PN) and lung-deposited surface area (LDSA) concentrations and average particle sizes (D_p) were measured by means of a hand-held diffusion charger particle counter (NanoTracer Philips). It measures the particle number concentration and the average particle size in the range 10–300 nm, with a sampling time of 10 seconds. The operating principle of this instrument is based on the diffusion charging technique. In particular, the sampled aerosol is charged in a standard positive unipolar diffusion charger imparting an average known charge on the particles that is approximately proportional to the particle diameter of the aerosol. The number of charges, and thus the number of particles, is then detected by an electrometer [82–84]. Since over 99% of total particle number concentrations in urban environments are due to particles below 300 nm in diameter [85,86], the instrument was considered adequate for the experimental campaign. Actually, the lung-deposited surface area (LDSA) concentration cannot be considered, strictly speaking, a direct measurement, since it is provided by the instrument on the basis of built-in semi-empiric relationships allowing calculating the particle surface area deposited in the alveolar and tracheobronchial through the PN concentration and average particle size (D_p) measured data as described in details in Marra, et al. [87] and Fierz, Houle, Steigmeier and Burtscher [82]. Then, the LDSA concentration was evaluated as sum of the alveolar- and tracheobronchial-deposited contributions. Nonetheless, in order to take into account for calibrated PN concentrations and D_p values, we have used the semi-empiric relationships to calculate the LDSA concentrations on the basis of the calibrated values. In particular, the calibration of the device was performed before and after each experimental campaign. To this end, both a Condensation Particle Counter (CPC 3775, TSI Inc., Shoreview, MN, USA) and a Scanning Mobility

Particle Sizer (SMPS 3936, TSI Inc.) were used to compare the devices in terms of number concentration and particle size, respectively. The SMPS consisted of an Electrostatic Classifier (EC 3080, TSI Inc.), a Differential Mobility Analyzer (DMA 3081, TSI Inc.), and a CPC 3775. The SMPS 3936 was used, with an aerosol/sheath flow ratio of 0.3/3.0 L·min⁻¹, thus measuring particle number distributions in the range 14–700 nm. The calibration was carried out at the European Accredited Laboratory of Industrial Measurements (LaMI) of the University of Cassino and Southern Lazio (Italy) in a 150 m³-room, with a conventional mechanical ventilation system guaranteeing constant thermo-hygrometric conditions (20 ± 2 °C and 50 ± 5% RH). Comparisons were performed for two different aerosols: aged indoor aerosol and freshly emitted aerosol produced by incense burning. Tests were conducted for 2 h performing simultaneous measurements with the Nanotracer, the CPC 3775, and the SMPS 3936. CPC and SMPS sampling times were set at 1 s and 135 s, respectively. SMPS measurements were corrected for multiple charge and diffusion losses. The correction factors obtained by averaging the results of the two aerosols investigated before and after each experimental campaign were applied as correction factors for each campaign. The differences in correction factors measured before and after the campaigns were found lower than 10%.

2.4. Statistical Analysis of the Data

In order to perform a statistical analysis of the concentrations experienced by the children in the different microenvironments (in terms of PN and LDSA) a preliminary normality test (Shapiro–Wilk test) was performed to check for the statistical distribution of the data. Since the data did not meet the assumptions of Gaussian distribution, non-parametric tests and further post-hoc tests (Kruskal–Wallis test [88]) were considered in the analysis. The statistically significant result was referred to a significance level of 99% (a *p*-value < 0.01). In particular, the Kruskal–Wallis tests were performed (a) amongst the six different microenvironments for each group of children separately (S1, S2, and S3; thus 3 non-parametric tests and further post-hoc tests) and (b) amongst the three groups of children for each microenvironment separately (six microenvironments plus the whole day data, thus seven non-parametric tests and further post-hoc tests).

The PN and LDSA concentration data considered in the statistical analysis, and then shown in the result section, included all the data provided by the instrument (roughly 48 h of total sampling per each child with a sampling frequency of 10 s), thus, a huge number of values were available for each microenvironment of each children group.

On the contrary, the dose values reported and discussed in the results represented the median values (and corresponding ranges) obtained from the 20-dose data (i.e., 20 children) per each microenvironment per each children group. Thus, due to the limited number of dose data, the statistical analysis on such values was not performed as it could lead to misleading results.

3. Results

3.1. Time Activity Patterns

In Table 3, data on time-activity patterns of the children under investigation are reported, which were obtained from the activity diaries filled in by children and parents during the measurements. The median data demonstrate that children spend the most significant time fraction performing indoor activities in indoor microenvironments: indeed, the median time spent by the children indoor, as sum of the microenvironments labelled as “sleeping”, “indoor day”, “cooking & eating”, resulted equal to 68–69%, to which must be added the time spent at school (25%). On the contrary, the time fraction spent in “outdoor day” (2–3%) and “transport” (3–4%) microenvironments resulted very limited, likely due to the fact that just school days were included in the experimental analysis, thus, the time spent in “transport” microenvironment is mostly limited to the time to take children to school. The huge time spent in indoor environments is consistent with our previous studies analyzing western populations, in which emerged that also adults spend a significant time fraction (roughly 90%)

performing indoor activities [53,62,63]. Amongst the indoor activities, the time spent in “cooking & eating” microenvironment (here 8%) is of particular concern since these activities were recognized in our previous papers as the most influencing in terms of exposure and health risk [36,89].

Table 3. Time activity pattern, particle concentrations (PN and LDSA) and dose received by children of the three schools in the different microenvironments expressed as median values and range (5th and 95th percentile). Total daily doses as sum of the median doses received in the different microenvironments are also reported as well as daily dose fractions and intensity–dose ratios.

Microenvironment	School	Time (min)	Time Fraction	PN conc. (104 part. cm ³)	LDSA conc. (µm ² .cm ⁻³)	δ (mm ²)	Daily Dose Fraction	i_δ (mm ² .min ⁻¹)
Sleeping	S1	540 (480–590)	38%	1.11 (0.62–2.04)	66 (37–123)	182 (101–340)	17%	0.34
	S2	530 (485–560)	37%	1.12 (0.61–1.93)	66 (37–117)	180 (100–319)	15%	0.34
	S3	597 (540–620)	41%	0.62 (0.33–1.08)	36 (21–67)	111 (63–204)	17%	0.19
Indoor	S1	320 (135–365)	22%	1.85 (0.59–7.70)	84 (27–356)	407 (128–1714)	38%	1.27
	S2	345 (140–395)	24%	1.79 (0.58–7.36)	81 (26–342)	426 (136–1777)	36%	1.23
	S3	268 (110–320)	19%	1.32 (0.43–5.26)	60 (20–248)	245 (80–1002)	38%	0.91
Outdoor	S1	40 (18–68)	3%	1.91 (0.68–5.20)	79 (28–220)	48 (17–133)	5%	1.20
	S2	35 (13–58)	2%	2.58 (0.86–7.12)	106 (36–304)	57 (119–162)	5%	1.62
	S3	36 (15–60)	3%	0.42 (0.36–1.18)	17 (15–49)	10 (8–27)	2%	0.027
School	S1	360 (295–375)	25%	1.57 (0.54–3.38)	66 (22–144)	163 (56–362)	15%	0.45
	S2	360 (290–385)	25%	2.13 (0.76–4.77)	89 (33–205)	222 (81–515)	19%	0.62
	S3	360 (300–390)	25%	0.34 (0.28–0.76)	14 (12–33)	36 (28–82)	6%	0.10
Transport	S1	60 (16–138)	4%	2.38 (1.29–6.38)	106 (57–274)	62 (33–160)	6%	1.04
	S2	50 (12–106)	3%	1.93(0.91–5.55)	86 (41–254)	41 (20–122)	4%	0.82
	S3	62 (15–131)	4%	0.66 (0.36–1.84)	29 (17–84)	17 (10–50)	3%	0.28
Cooking & Eating	S1	120 (97–148)	8%	4.20 (1.44–15.3)	112 (38–412)	200 (69–744)	19%	1.66
	S2	120 (93–156)	8%	5.11 (1.69–16.6)	136 (46–453)	244 (83–823)	21%	2.03
	S3	117 (90–160)	8%	4.91 (1.67–19.3)	130 (45–525)	227 (80–923)	35%	1.94
Day	S1	1440		1.44 (0.61–5.99)	71 (28–248)	1062		0.74
	S2	1440		1.55 (0.65–6.33)	77 (34–261)	1169		0.81
	S3	1440		0.62 (0.30–5.07)	34 (12–169)	646		0.45

3.2. Exposure to Submicron Particles

In Table 3 and Figure 1, the submicron particle concentrations, in terms of particle number and lung-deposited surface area, to which the children attending the three different schools (S1, S2, and S3) were exposed to in the different microenvironments (sleeping, indoor day, outdoor day, school, transport, cooking & eating) are shown. In the box plots of Figure 1, exposure data not statistically different amongst the six different microenvironments for each group of children separately (S1, S2, S3) and amongst the three groups of children for each microenvironment separately are also indicated ($p > 0.01$) as resulting from the statistical analysis explained in Section 2.4 (Kruskal–Wallis test). Due to the huge amount of data available for each microenvironment, most of the exposure received in the six microenvironments by the same group of children as well as those received in the same microenvironment by the three groups of children resulted in statistically different results.

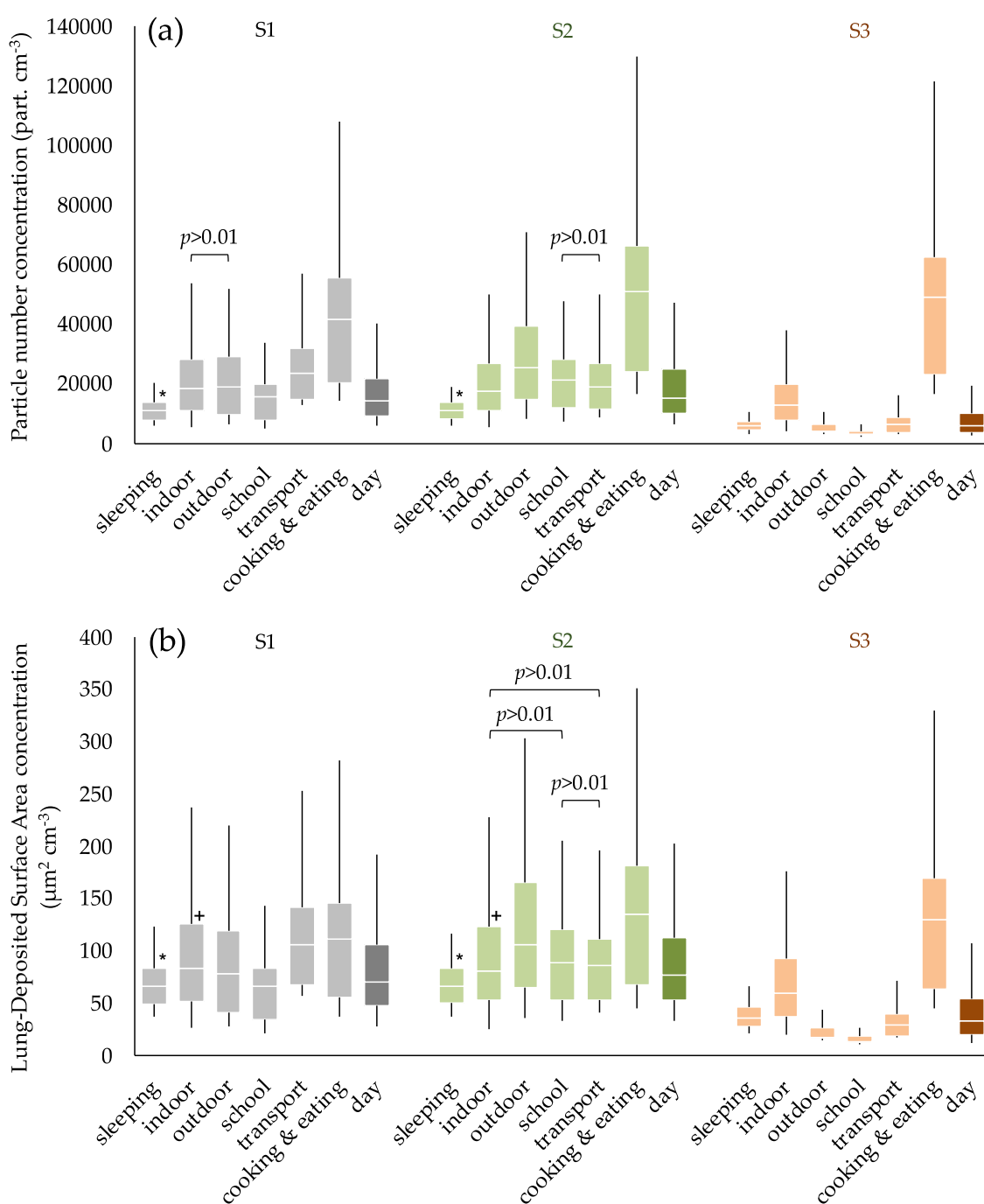


Figure 1. Statistics of (a) particle number and (b) lung-deposited surface area concentrations experienced by three groups of children (attending school S1, S2, and S3) in each microenvironment. Data was not statistically different within each group of children (S1, S2, S3) and amongst the same microenvironments of different groups (*,+) are also indicated ($p > 0.01$).

The children’s exposure to submicron particles in the “school” microenvironment presents a significant deviation amongst the three schools. Indeed, children attending school S1, S2 and S3 were exposed to median PN and LDSA concentrations of 1.57×10^4 part. $\text{cm}^{-3}/66 \mu\text{m}^2 \cdot \text{cm}^{-3}$, 2.13×10^4 part. $\text{cm}^{-3}/89 \mu\text{m}^2 \cdot \text{cm}^{-3}$, and 3.39×10^3 part. $\text{cm}^{-3}/14 \mu\text{m}^2 \cdot \text{cm}^{-3}$, respectively. In particular, the concentration levels in the school S3 were much lower than S1 and S2 ones. This is due to the different outdoor concentrations, indeed, if no indoor submicron particle sources are in operation in the schools (as mentioned in the methodology section), the indoor

concentrations are just affected by the outdoor-to-indoor penetration factors [65,90,91]. Thus, the low concentrations measured in school S3 are just related to the low outdoor concentrations typical of the rural site under investigation and discussed in the methodological section (Table 1). Indeed, the median particle number and lung-deposited surface area concentrations in the “outdoor day” microenvironment were equal to 1.91×10^4 part. $\text{cm}^{-3}/79 \mu\text{m}^2 \cdot \text{cm}^{-3}$, 2.58×10^4 part. $\text{cm}^{-3}/106 \mu\text{m}^2 \cdot \text{cm}^{-3}$, and 4.22×10^3 part. $\text{cm}^{-3}/17 \mu\text{m}^2 \cdot \text{cm}^{-3}$, for children attending school S1, S2 and S3, respectively. The resulting “school”/“outdoor day” concentration ratios (considering the median concentrations) were equal to 0.80–0.83 and 0.82–0.86 in terms of PN and LDSA concentrations, respectively, then consistent with the typical penetration factors reported in the scientific literature for naturally ventilated schools [65,90,91]. The location of the children’s schools and homes is then the most influencing parameters in their exposure to submicron particles in “outdoor day” and “school” microenvironments, in fact the highest correlations between average outdoor NO_2 concentrations measured at the FSPs (Table 1) and PN concentrations measured during the experimental campaigns were determined for these two microenvironments (linear regressions with $r^2 > 0.99$). The correlation between outdoor and indoor concentrations gets weaker when it comes to non-school environments, indeed, here the possible presence of indoor sources (cooking, incense, candles, heating systems) can lead to high indoor concentrations. In this context, as expected, the most critical microenvironment is “cooking & eating” which presents median values of PN and LDSA concentrations of 4.20×10^4 part. $\text{cm}^{-3}/112 \mu\text{m}^2 \cdot \text{cm}^{-3}$, 5.11×10^4 part. $\text{cm}^{-3}/136 \mu\text{m}^2 \cdot \text{cm}^{-3}$, and 4.91×10^4 part. $\text{cm}^{-3}/130 \mu\text{m}^2 \cdot \text{cm}^{-3}$, for children attending school S1, S2 and S3, respectively. The correlation with the average outdoor NO_2 concentrations measured by the FSPs barely doesn’t exist, indeed the concentrations are much larger than the outdoor ones, and also children attending school S3 are exposed to very high submicron concentrations in “cooking & eating” microenvironment and roughly comparable to the S1 and S2 ones despite the much lower outdoor concentrations.

In regard to the other indoor environments labelled as “indoor day” microenvironment, the children’s exposure resulted in lower statistical rates than the “cooking & eating” ones for all the three children groups. Nonetheless, the exposure in the “indoor day” microenvironment, when compared to the “outdoor day” one, varied amongst the different children groups. Indeed, the exposure in the “indoor day” microenvironment resulted statistically similar results, slightly lower, and much larger than the “outdoor day” environment for S1 (1.85×10^4 part. $\text{cm}^{-3}/84 \mu\text{m}^2 \cdot \text{cm}^{-3}$), S2 (1.79×10^4 part. $\text{cm}^{-3}/81 \mu\text{m}^2 \cdot \text{cm}^{-3}$), and S3 group of children (1.32×10^4 part. $\text{cm}^{-3}/60 \mu\text{m}^2 \cdot \text{cm}^{-3}$), respectively. The huge “indoor day”-“outdoor day” difference in the exposure detected for S3 group of children is related to the very low outdoor concentration level; thus, even a minor indoor source can easily increase the indoor concentration to values higher than the outdoor ones. Regarding the exposure in the “sleeping” microenvironment, the concentrations resulted in 0.5–0.6-fold of the “indoor day” microenvironment for all the three groups of children. Finally, during the “transport” microenvironment, higher concentrations were measured for children attending school S1 (2.38×10^4 part. $\text{cm}^{-3}/106 \mu\text{m}^2 \cdot \text{cm}^{-3}$) and S2 (1.93×10^4 part. $\text{cm}^{-3}/86 \mu\text{m}^2 \cdot \text{cm}^{-3}$), which are close to trafficked roads. On the contrary, children attending school S3 were exposed to quite low concentrations (6.58×10^3 part. $\text{cm}^{-3}/29 \mu\text{m}^2 \cdot \text{cm}^{-3}$), likely due to the location of the schools (rural area).

In summary, the daily exposure of the children is not only affected by the location of schools and homes, i.e., the proximity to outdoor sources, but also by the presence of indoor sources (mainly cooking); therefore, using outdoor concentration values as proxies of the daily exposure of the children could lead to serious under- or overestimation of the exposure. This is clearly highlighted by the daily median exposure data reported in Table 3; the concentrations, in terms of PN and LDSA, were equal to 1.44×10^4 part. $\text{cm}^{-3}/71 \mu\text{m}^2 \cdot \text{cm}^{-3}$, 1.55×10^4 part. $\text{cm}^{-3}/77 \mu\text{m}^2 \cdot \text{cm}^{-3}$, and 0.62×10^4 part. $\text{cm}^{-3}/34 \mu\text{m}^2 \cdot \text{cm}^{-3}$, for children attending school S1, S2 and S3, respectively. Indeed, such values were 0.75-, 0.60-, and 1.48-fold the outdoor PN concentration values and 0.90-, 0.73-, and 2.00-fold the outdoor LDSA concentration values for S1, S2 and S3 children groups, respectively.

3.3. Particle Doses Received by Children

Median values (and corresponding 5th–95th percentile ranges) of particle surface area doses received by the three groups of children investigated (attending school S1, S2 and S3) in each microenvironment are shown in Table 3, here the daily doses are also reported. The doses received in the different microenvironments were calculated through Equation (1) considering the above mentioned and discussed (i) time–activity patterns and (ii) exposure data, as well as the (iii) inhalation rates characteristics of the children age and activity as resulting from the activity diaries, whereas the total daily doses here reported represent the sum of the median doses received in the different microenvironments.

The total daily doses for children attending school S1, S2 and S3 resulted equal to 1062, 1169 and 646 mm², respectively. The higher doses received by children of schools S1 and S2 are mostly due to their higher median daily exposures discussed in Section 3.1, while the time activity patterns (and then the inhalation rates) were quite similar amongst the three children groups.

The dose received in “school” microenvironment resulted equal to 163, 222 and 36 mm² for school S1, S2 and S3, respectively; with contributions of 15%, 19%, and 6% to daily dose. The dose received by children in school S3 is extremely low due to the low outdoor concentration of that rural area, whereas, the more polluted outdoor environments of S1 and S2 lead to higher doses. Anyway, such doses can be considered not extremely high if compared to the important time fraction of the day spent in such environments (25% of the day): this is clearly confirmed by the dose-intensity ratio (i_{δ}) summarized in Table 3; such ratios were lower than 1 for all the schools and, apart from “sleeping”, they were the lowest values (0.45, 0.62, and 0.10 mm²·min⁻¹ for S1, S2, and S3, respectively) amongst the microenvironments investigated.

Regarding the non-school environments, the contributions of “outdoor day” (2–5% of the daily dose) and “transportation” (3–6% of the daily dose) microenvironments are very limited due to the reduced time spent therein. As mentioned above, children attending school S1 and S2 were exposed to quite high concentrations in these two microenvironments then leading to dose-intensity ratios >1: this suggests that higher doses would be received in days and seasons characterized by different time-activity patterns with longer periods spent in such environments.

The main contribution to the daily dose is obviously received in non-school indoor environments, indeed summing up the doses received by children in “sleeping”, “indoor day” and “cooking & eating” microenvironments, total contributions of 74%, 73%, and 90% were estimated for children attending school S1, S2 and S3, respectively. The most important contribution is due to the “indoor day” environment (36–38%) due to the both the significant time fraction (19–24%) and the possible presence of other sources leading to concentrations higher than the outdoor ones: indeed, dose-intensity ratios close or larger than 1 were measured for that environment. The contribution of the “sleeping” microenvironment is quite low (15–17%) if compared to the huge time spent in such activities (dose-intensity ratios extremely low), whereas an important dose fraction is received by children in “cooking & eating” microenvironments due to the high concentrations to which children are exposed to. Indeed, despite the time fraction spent in “cooking & eating” microenvironment is about 8% for all the three children groups, the contributions to the daily dose resulted equal to 19%, 21%, and 35% for children attending school S1, S2 and S3, respectively. In fact, such microenvironment resulted the one with the highest dose-intensity ratios (1.66, 2.03, 1.94), then consistently exceeding the “transportation” and “outdoor” microenvironments typically affected by outdoor sources.

In conclusion, the results on exposure levels in the different microenvironments confirm that indirect exposure assessments based on measurements at city scale or outdoor scale, typically adopted in cohort studies evaluating epidemiological effects on large populations [92,93] due to their easiness and cheapness, cannot provide a good estimate of the dose received by children whatever the location of their homes and schools. Thus, direct exposure assessment based on measurements at a personal scale, i.e., sampling aerosol from the breathing zone of the person using wearable instruments carried as personal monitors, is the only accurate experimental approach allowing proper dose estimates as it takes

into account the different personal exposure of people moving between different microenvironments also including the indoor ones.

Regarding the exposure assessment results shown here, some broader implications can be drawn from the paper. In particular, concerning the exposure in outdoor-driven microenvironments (e.g., schools, outdoors), it can be reduced just building the schools and performing outdoor activities as far as possible from main outdoor sources (e.g., vehicular traffic). The reduction of the exposure (and then the dose) in indoor microenvironments can be reached (i) mitigating the particle sources (e.g., using ad-hoc hoods during kitchen activities, avoiding the use combustion sources such as biomass burning, candles, etc.) and/or (ii) reducing the exposure (e.g., increasing the air exchange rates through proper ventilation approaches, using air purifiers).

4. Conclusions

In the present study, an assessment of the total daily dose in terms of submicron particle surface area received by children living in different Italian areas and attending different schools located in different urban contexts (rural, suburban and urban area), was performed. The study aimed at investigating the children daily doses received in different microenvironments (both school and non-school environments) also taking into account the impact of the outdoor concentration levels on the received dose. To this end, an experimental analysis using portable instruments able to measure the concentrations at personal scale of the children was performed.

The findings of the study shown that the contribution of the school environment to the overall daily dose of the children is quite limited although they spent a significant time fraction of the day therein. Such dose is mainly affected by the outdoor concentrations; thus, schools placed close to main outdoor sources (e.g., trafficked roads) may results in higher rates of exposure and related doses than rural ones.

Outdoor and transport microenvironments present an almost negligible contribution to the children daily doses, whatever the investigated sites, due to the reduced exposure time in such environments. Therefore, a child's daily dose is mainly affected by indoor non-school environments, e.g., homes. In particular, the contribution of non-school indoor microenvironments to the children's daily dose account for more than 70% of the data from the children and school locations. Such a high contribution is led by "cooking & eating" and other "indoor day" microenvironments. Indeed, the "cooking & eating" microenvironment contributes up to 36% of the daily dose despite the reduced time spent therein: this is due to the high levels of exposure from high-emitting cooking activities.

In conclusion, the results of the study demonstrate that a proper evaluation of the submicron particle dose received by children cannot be performed only relying upon outdoor concentration data and that despite the location of the school and home, the contribution of indoor non-school environments is essential to properly assess the dose received by children.

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