Environmental Pollution 266 (2020) 115209

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Influence of methodology on the estimation of the particle surface area dose received by a population in all-day activities☆

Mauro Scungio [a, *], Valeria Rizza [b], Luca Stabile [c], Lidia Morawska [d], Giorgio Buonanno [c, d]

[a] School of Engineering, University of Tuscia, Viterbo, Italy
[b] Institute of Atmospheric Pollution Research (IIA), CNR, Rome, Italy
[c] Department of Civil and Mechanical Engineering, University of Cassino and Southern Lazio, Cassino, Italy
[d] Queensland University of Technology, Brisbane, Australia

ARTICLE INFO

Article history:
Received 13 January 2020
Received in revised form
6 July 2020
Accepted 6 July 2020
Available online 11 July 2020

Keywords:
Surface area dose
Particle dose
Ultrafine particles
Inhalation rates
Personal exposure monitoring

ABSTRACT

In everyday life, people are exposed to different concentrations of airborne particles depending on the microenvironment where they perform their different activities. Such exposure can lead to high sub-micron particle doses. The received dose depends on particle concentration to which people are exposed (typically expressed in terms of number or surface area), time spent in each activity or microenvironment (time activity pattern) and amount of air inhaled (inhalation rate). To estimate an actual value of the received dose, all these parameters should be measured under real-life conditions; in fact, the concentrations should be measured on a personal scale (i.e. through a direct exposure assessment), whereas time activity patterns and inhalation rates specific to the activity performed should be considered. The difficulties in obtaining direct measurements of these parameters usually lead to adopt time activity patterns and inhalation rates already available in scientific literature for typical populations, and local outdoor particle concentrations measured with fixed monitoring stations and extrapolated for all the other microenvironments.

To overcome these limitations, we propose a full-field method for estimating the received dose of a population sample, in which all the parameters (concentration levels, time activity patterns and inhalation rates) are measured under real-life conditions (also including the inhalation rates, that were evaluated on the basis of the measured heart rates). Specifically, 34 volunteers were continuously monitored for seven days and the data of sub-micron particle concentrations, activities performed, and inhalation rates were recorded. The received dose was calculated with the proposed method and compared with those obtained from different simplified methodologies that consider typical data of particle concentrations, time activity patterns and inhalation rates obtained from literature. The results show that, depending on the methodology used, the differences in the received daily dose can be significant, with a general underestimation of the most simplified method.

1. Introduction

Exposure to airborne particles is known to have a negative health impact on the cardiovascular and respiratory systems (Franchini and Mannucci, 2011; Cesaroni, Badaloni et al., 2013; Buteau and Goldberg, 2016). Several studies have suggested a possible association between airborne particle exposure and increased cardiovascular morbidity and mortality (Basagaña, Jacquemin et al., 2015; Xie, Li et al., 2015; Azimi and Stephens, 2018). It has also been found that airborne particle exposure induces changes in heart rate and heart rate variability (Brook, Rajagopalan et al., 2010; Rizza, Stabile et al., 2019), and is responsible for a range of adverse outcomes in the respiratory system (Schmid, Möller et al., 2009; Buonanno et al., 2013b). In addition, particulate matter (PM) has been recognized as a group 1 carcinogen to humans by the International Agency for Research on Cancer (IARC, 2013).

Most of the epidemiological studies focus on the effects of super-micron particles (which are defined in terms of particle mass concentrations, i.e. PM_{10} and $PM_{2.5}$) rather than those due to sub-

☆ This paper has been recommended for acceptance by Payam Dadvand.

* Corresponding author.
E-mail address: mauro.scungio@unitus.it (M. Scungio).

https://doi.org/10.1016/j.envpol.2020.115209
0269-7491/

micron particles (which are expressed in terms of particle number and surface area concentrations). Over the years, attention has extended from PM to sub-micron particles because the latter are characterized by a higher deposition fraction, a larger surface area, and a potential to translocate in the circulatory system (International Commission on Radiological Protection, 1994) while several sub-micron particles exposure assessments were carried out from the scientific community (Qiu, Wang et al., 2019, Slezakova et al. 2019; Correia, Martins et al., 2020; Madureira, Slezakova et al., 2020; Slezakova, Pereira et al., 2020). In addition, some studies have shown that the surface area is the most appropriate parameter for health concerns related to sub-micron particles (Giechaskiel, Alföldy et al., 2009; Rushton, Jiang et al., 2010; Schmid and Stoeger, 2016). Therefore, to investigate the health effects caused by exposure to sub-micron particles, it is crucial to evaluate the received surface area dose, which is calculated from data of the sub-micron particle concentration levels, the time spent in each activity or microenvironment (i.e. the time activity pattern, or TAP), and the inhalation rate (IR), which in turn depend on the specific activity performed (Fuoco, Stabile et al., 2017; Scungio, Stabile et al., 2018).

To evaluate the particle concentration levels to which people are exposed, different exposure assessments can be adopted depending on the spatial scales of investigation (Cattaneo, Taronna et al., 2010). The easiest and cheapest exposure assessments are the indirect exposure assessments, which are based on city scale or outdoor scale approaches. These are the typical approaches applied in cohort studies evaluating epidemiological effects on a large population (Raaschou-Nielsen, Andersen et al., 2013; Chen, Zhang et al., 2016; Stafoggia, Schneider et al., 2017). Nonetheless, these represent oversimplified exposure assessments as they do not consider: (i) the spatial variability of sub-micron particles within different outdoor environments (Kaur, Nieuwenhuijsen et al., 2005; Buonanno, Fuoco et al., 2011; Rizza, Stabile et al., 2017; Puustinen, Hämeri et al., 2007; Buonanno, Lall et al., 2009) and, even worse, (ii) the exposure in the indoor environment, which represents the environment where the population spends most of the day (McGinnity, Russel et al., 2005, Lader, Short et al., 2006, Australian Bureau of Statistics, 2010, Soggiu, Vollono et al., 2010, Statistics Sweden Economic Welfare Statistics Unit, 2012). The most appropriate and detailed scale for exposure assessments is the personal scale, i.e. using portable instruments carried as personal monitors (Cattaneo, Taronna et al., 2010). This approach allows a direct exposure assessment to be performed, thus measuring concentrations with high temporal and spatial resolution and also reflecting the personal exposure of people moving between different microenvironments (Avery, Mills et al., 2010; Manigrasso and Avino, 2012; Steinle, Reis et al., 2013). This is a key aspect, since the exposure to sub-micron particles, even more than super-micron particles, cannot be defined through fixed monitors (Strak, Boogaard et al., 2010). Very few studies evaluating the dose received by a population through direct exposure assessment have been carried out; as an example, Pacitto, Stabile et al. (2018) calculated the surface area doses received by different Western populations by measuring their personal exposure through wearable monitors and adopting TAPs and IRs typical of the investigated populations. The study revealed that high doses are received in indoor environments (including cooking and eating activities) by all populations.

Direct exposure assessment through wearable sensors is becoming quite common in exposure studies (Buonanno, Stabile et al., 2013; Buonanno, Stabile et al., 2014; Mazaheri, Clifford et al., 2014; Clifford, Mazaheri et al., 2018; Pacitto, Stabile et al., 2018; Rizza, Stabile et al., 2019). For a correct estimation of the particle surface area dose, it is crucial to accurately evaluate the actual TAPs that characterize the lifestyle of the specific population under investigation. Such specific TAPs can differ from those characteristic of typical populations provided by the scientific literature (Avino, Scungio et al., 2018; Pacitto et al., 2018b). In addition, it is also important to measure the actual and personal IRs for each microenvironment and activity. In fact, the common approach is using IRs obtained for average populations which, again, may not reflect those of specific populations under their real-life conditions (Buonanno et al., 2014c).

On the basis of the above considerations, in this paper a full-field method is proposed and applied to calculate the received dose of sub-micron particles of a population sample by means of high spatial and temporal resolution particle concentration measurements, and detailed and actual TAPs and IRs. To this end, 34 volunteers were continuously monitored for seven days; concentrations and TAPs were measured/recorded for each participant, while the IRs were determined on the basis of the heart rates (HR) measured for each individual. The received particle surface area dose calculated with the full-field method was then compared with those obtained from different simplified methodologies based on TAPs and IRs obtained from the literature and concentrations measured at the city scale in order to analyse the influence of the calculation methodology.

2. Materials and methods

2.1. Methodology description and data collection

The study was conducted on weekdays and weekend days between November 2016 and November 2017 on 34 participants (12 females and 22 males) aged from 19 to 64 years old equally distributed between the study period. The population sample considered in the study is characterized by full-time workers. In particular, the selected people were non-smokers, non-alcoholics and with good health status. The volunteers were selected between all professional category or socioeconomic status considering only people without a personal history of health diseases (cardiovascular, pulmonary, neurological or endocrine). All the participants lived in Cassino and in the surrounding areas (Central Italy, milder climate, resident population 33 000 inhabitants; surface area 83 km^2, main sources of sub-micron particles from traffic emission and domestic heating systems). The 34 volunteers were equipped with a mobile monitor, brought always with them as they moved between different microenvironments, in order to measure their exposure to sub-micron particle concentrations at personal scale (Cattaneo, Taronna et al., 2010). Particle number (PN) and lung-deposited surface area concentration [LDSA, sum of the particle surface area concentrations deposited in the alveolar and tracheobronchial regions of the lung (Marra, Voetz et al., 2010)] were measured. The participants kept the mobile equipment for seven consecutive days, carrying it with them in all the microenvironments in which they spent their time. The subjects were also asked to transcribe their main indoor and outdoor activities in a diary, reporting the time spent in each microenvironment/activity (transport, working, eating, cooking, outdoor, indoor and sleeping, as detailed in Table 1). Any time their location or activity changed, they were asked to report it in the diary. Examination of the diaries filled out by the subjects provided the time spent (t_j) in each activity. Particular attention should be paid to the “cooking” and “eating” microenvironments as they were recognized in our previous papers as the main microenvironments contributing to the daily dose (Pacitto, Stabile et al., 2018). As clearly stated in Table 1, we considered as cooking activity both cooking their own food and staying in a microenvironment where a family member is cooking, as already adopted in previous papers, e.g. in the evaluation of the

Table 1
Description of the microenvironments/activities provided to the population sample as options for their diary compilation.

Microenvironment	Activities
Transport	Trip and use of time not specified, round-trip to work
Working	All type of activities performed in the workplace environment
Eating	Eating and drinking
Cooking	Cooking; including both cooking their own food and staying in a microenvironment where a family member is cooking
Outdoor	Gardening and animal care, restoration, sport and outdoor activities, physical workout, productive exercise, sports-connected activities, etc.
Indoor	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

dose of children, that clearly do not perform cooking activities per se (Pacitto, Stabile et al., 2020). Since the population under investigation presents mainly full-time workers, a shorter time spent is expected in cooking microenvironment, and consequently, a lower dose received by the population should be estimated with respect to our previous studies. Nonetheless, the authors highlight, once again, that the paper doesn't represent a population study aiming at providing dose data characteristics of a representative population; on the contrary, the exposure data of the population were used to test the different methodologies available to estimate the dose.

The heart rate (HR) of each participant was continuously monitored (with 1 ms of time resolution) during each activity by means of a heart rate monitor (AthenaDiaX Germany, ECG recorder - ARES and ZEUS 2D attached to the subject's chest using adhesive electrodes), as detailed in Rizza, Stabile et al. (2019). The inhalation rate was calculated on the basis of the model proposed by Greenwald, Hayat et al. (2019), in order to estimate the inhalation rates using heart rate, age, sex and forced vital capacity (FVC) as predictors, in the case of HR as the only field-measured parameter. The model is based on the following equation:

$$IR = e^{-9.59} HR^{2.39} age^{0.274} sex^{-0.204} FVC^{0.520}$$

Where the FVC for each subject was evaluated using the Global Lung Function Initiative method (Cooper, Stocks et al., 2017) and sex is 1 for males and 2 for females.

Thus, the daily particle surface area dose for each subject (δ_{SA}) was calculated by multiplying the LDSA measured during the specific activity by the time spent, t_j, and IR_j of each activity (Klepeis, 2006):

$$\delta_{SA} = \sum_{j=1}^{n} (IR_j \cdot LDSA_j \cdot t_j) \quad (1)$$

2.2. Instrumentation and quality assurance

The PN and LDSA concentrations at personal scale were measured with two portable battery-powered diffusion charger particle counters operating in the sub-micron particle range (10−300 nm) already used in previous studies (Pacitto, Stabile et al., 2018; Rizza, Stabile et al., 2019): a NanoTracer, Oxility, Netherlands (10-s sampling time resolution) and a DISCmini, Matter Aerosol AG, Germany (1-s sampling time resolution). The operating principle of the diffusion chargers is based on the particle charging and the consequent measurement of the total particle charge per unit of air volume. The participants were instructed to carry the mobile monitors with them when moving around or doing activities, while when remaining in a confined microenvironment for long time, they were allowed to leave the portable instrument on a very close surface (table or chair). The lung-deposited surface area concentration measured by the instruments is characteristic of a reference worker under light exercise and breathing only through the nose (Asbach, Kaminski et al., 2012). The PN and LDSA concentration data were post-processed as 10-s average values in order to harmonize the amount of data obtained from the two counters. It should be pointed out that the 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 region of the respiratory system through the PN concentration and average particle size measured data as described in details in (Marra, Voetz et al., 2010; Fierz, Houle et al., 2011). Then, the LDSA concentration is the sum of the alveolar and tracheobronchial-deposited contributions. Since we have used the LDSA data provided by the instruments, we will refer to the LDSA data as measured data. The performance of the counters was compared through parallel measurements with an Aerosol Electrometer (TSI Model 3068B) and a Nanoparticle Surface Area Monitor (NSAM, TSI Model 3550) to check them in terms of PN and LDSA concentrations, respectively. The calibration was performed in the European Accredited Laboratory of Industrial Measurements (LAMI) at the University of Cassino and Southern Lazio (Italy) using aged indoor aerosol and freshly emitted aerosol from incense burning by means of laboratory tests. The agreement between the Nanotracer/DiSCmini and the Aerosol Electrometer/NSAM was between ±10%, which represents an acceptable range

Table 2
Combinations of the particle concentrations, time activity patterns (TAPs) and inhalation rates (IRs) adopted for calculation of the received particle surface area dose in the different methodologies adopted in this study.

Parameter	Methodology for dose estimate				
	M0	M1	M2	M3	M4
Particle concentrations	Measured	Measured	Measured	Measured	Not measured
IRs	Measured	Measured	Not measured	Not measured	Not measured
TAPs	Measured	Not measured	Measured	Not measured	Not measured

for such a hand-held instrument.

2.3. Methodologies for calculation of the dose

The doses obtained through the full-field methodology were compared with those estimated through simplified approaches commonly used in the literature which are based on TAPs and IRs not measured directly (but obtained from the literature), and LDSA concentrations not representing the personal exposure experienced by individuals in each microenvironment/activity (but characteristics of more general spatial scales). The combinations of LDSA concentration, IR and TAP data used in the different methodologies for particle surface area dose estimates are summarized in Table 2. Here, the proposed full-field method is referred to as methodology *M0* and, as mentioned above, all the data necessary for the dose calculation (LDSA concentrations, IRs and TAPs) were directly measured in the experimental campaign; thus, they are identified as "measured" in Table 2. For the sake of simplicity, the term "measured" is used also for the IR even if actually this parameter was not directly measured but instead evaluated on the basis of the HR. The other methodologies applied in the study to calculate the dose are indicated in the table as *M1*, *M2*, *M3*, and *M4*: each presents a different combination of concentration, TAP and IR data. LDSA concentrations, TAPs and IRs not directly measured are identified as "not measured" in Table 2. As an example, the most simplified approach (*M4*) was based on typical IR and TAP data obtained from the literature, as representative of the typical Italian population, and LDSA concentrations typical of an exposure assessment performed at outdoor/city scales. In particular, the "not measured" particle concentrations for the *M4* scenario were considered to be the LDSA concentrations measured in the outdoor microenvironment during the experimental campaign. In fact, the outdoor PN concentration was similar to the annual median value measured during a previous experimental campaign performed in a background sampling site in the city (Cassino) and reported in our previous papers (Buonanno, Fuoco et al., 2013; Stabile, Buonanno et al., 2013). Thus, the outdoor concentrations obtained from the present campaign can be considered representative of an exposure assessment at the outdoor/city scale. Therefore, in the *M4* methodology the median value of the outdoor PN and LDSA concentrations were adopted for all the activities performed outdoors (transportation and outdoor), whereas the PN and LDSA concentration of indoor environments were estimated from the outdoor ones by multiplying them by outdoor-to-indoor penetration factors of 0.7 and 0.5 for daily activities (indoor, cooking, eating, working) and nocturnal activities (sleeping), respectively (Zhu, Hinds et al., 2005; Stabile, Buonanno et al., 2019). The authors point out that the outdoor-to-indoor penetration is a function of the airtightness of the building, the ventilation strategy adopted, and the outdoor meteo-climatic conditions (Stabile, Dell'Isola et al., 2016; Stabile, Buonanno et al., 2019), thus a wide range of penetration values were found in the scientific literature (Zhu, Hinds et al., 2005). In the present paper, an average penetration factor equal to 0.7 (and 0.5 during the night) was chosen, nonetheless, this choice doesn't affect the general finding of the paper when comparing the *M4* methodology to the methodologies where the indoor concentration was actually measured. A summary of the definitions for the terms "measured" and "not measured" adopted in Table 2 for the three parameters used for dose calculation (concentrations, TAPs and IRs) is shown in Table 3.

3. Results and discussion

3.1. Particle concentrations, inhalation rates, time activity patterns and surface area doses obtained from the proposed full-field method

In Fig. 1 and Table 4 the statistics of the data concerning the TAPs, the PN and LDSA concentrations, and the IRs measured during the experimental campaign for each activity/microenvironment (considering the entire population sample under investigation) are summarized. In addition, in Table 5 the parameters used as "not measured" (see Tables 1 and 2) are reported.

As can be seen, the prevailing activities relate to working (275 min per day, 20 %), indoor activities (364 min per day, 27 %) and sleeping (453 min per day, 34 %). The figure clearly shows that the sample population spend most of their daily time indoors. When compared with the typical time activity patterns of the Italian population obtained from the literature (Soggiu, Vollono et al., 2010; Pacitto, Stabile et al., 2018), the authors recognize that the population investigated spent a shorter amount of time in cooking activities and a longer period in the transport microenvironment. This is due to the specific population under investigation which is composed of full-time workers typically eating out and not cooking their own food; these differences with respect to the typical population will affect the dose estimates as hereinafter reported.

The sub-micron particle concentrations (in terms of PN and LDSA concentrations) show, as expected, the highest median value for cooking (3.6×10^4 part/cm^3 and 105 μm^2/cm^3) and eating (2.7×10^4 part/cm^3 and 84 μm^2/cm^3), even if such concentrations were lower than those measured in other studies performed through personal monitoring (Buonanno, Stabile et al., 2014; Pacitto, Stabile et al., 2018). This is likely because the population studied, as full-time workers, likely do not cook their own food and

Table 3
Definitions of the terms "measured" and "not measured" adopted for the three parameters used for dose calculation.

Data	Parameters adopted to calculate the dose		
	Particle concentrations	Inhalation rate	Time activity pattern
Measured	Concentration obtained from measurements performed at the personal scale (direct exposure assessment) through personal monitors	Calculated on the basis of the heart rates directly measured for each individual through a personal monitor	Calculated on the basis of the diaries filled out directly by each individual reporting the time spent in each activity
Not measured	Concentrations representative of the outdoor/city scale. Indoor concentrations estimated through an outdoor-to-indoor penetration factor (0.7 and 0.5 for day and night, respectively)	Obtained from the literature (Adams, 1993; Schuda, Moya et al., 2009, U.S. Environmental Protection Agency, 2009) as representative of the typical population	Obtained from the literature (Soggiu, Vollono et al., 2010; Pacitto, Stabile et al., 2018) as representative of the typical Italian population

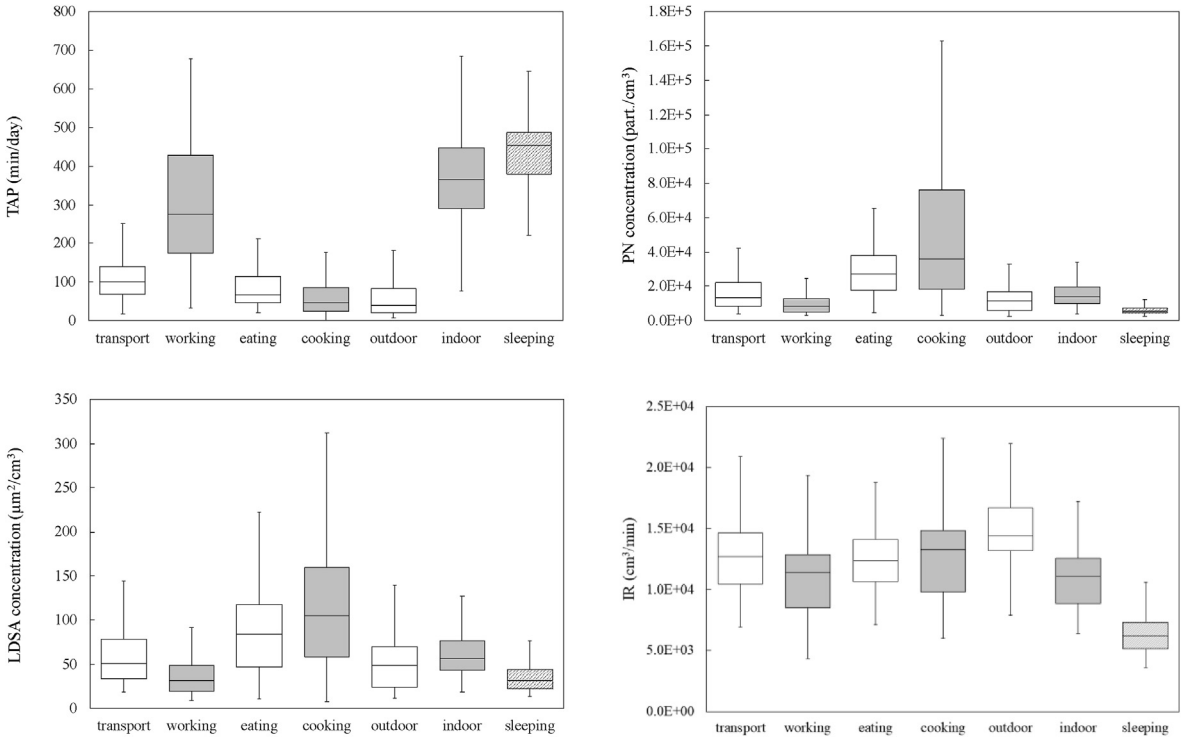

Fig. 1. Statistics of time activity patterns (TAPs, top-left panel), PN concentrations (top-right panel), LDSA concentrations (bottom-left panel) and inhalation rates (IRs, bottom-right panel) as measured from the experimental campaign for the analysed population sample for each microenvironment/activity. Data are reported as box-plots that show minimum, maximum, $25^{th} - 75^{th}$ percentile, and median values.

Table 4
Summary of the data measured in the experimental campaign (time spent, inhalation rate and PN and LDSA concentrations), as well as daily dose, contribution to daily dose, and dose intensity calculated with the proposed full-field method (median values for all subjects).

Microenvironment	Time spent (min/day)	Inhalation rate (cm^3/min)	PN concentration (part/cm^3)	LDSA concentration ($\mu m^2/cm^3$)	Daily dose (mm^2)	Contribution to daily dose (%)	Dose intensity (mm^2/min)
Transport	100	1.3×10^4	1.3×10^4	51	7.7×10^1	11	0.59
Working	275	1.1×10^4	8.2×10^3	31	1.0×10^2	17	0.40
Eating	66	1.2×10^4	2.7×10^4	84	8.0×10^1	10	0.99
Cooking	46	1.3×10^4	3.6×10^4	105	4.1×10^1	8	1.00
Outdoor	40	1.4×10^4	1.1×10^4	49	3.0×10^1	5	0.66
Indoor	364	1.1×10^4	1.4×10^4	56	2.2×10^2	33	0.63
Sleeping	453	6.2×10^3	5.7×10^3	32	9.5×10^1	13	0.20

Table 5
Parameters for the evaluation of the received dose as reported in Tables 1 and 2 used as "not measured".

Microenvironment	Time spent (min/day)	Inhalation rate (cm^3/min)	LDSA concentration ($\mu m^2/cm^3$)
Transport	45	8.3×10^3	35
Working	67	7.5×10^3	35
Eating	429	1.6×10^4	35
Cooking	84	1.6×10^4	50
Outdoor	436	6.0×10^3	25
Indoor	68	1.0×10^4	50
Sleeping	320	7.5×10^3	35

often do not have lunch at home. In contrast, the concentrations recorded in other activities are roughly similar to those found in previous studies. In particular, PN concentrations ranged from 5.7×10^3 part/cm^3 (32 $\mu m^2/cm^3$) for sleeping activities to 1.4×10^4 part/cm^3 (56 $\mu m^2/cm^3$) for indoor activities. The median concentration measured for outdoor activities was 1.1×10^4 part/cm^3 (49 $\mu m^2/cm^3$), which is similar to the median background PN concentration measured in Cassino (6.0×10^3–1.4×10^4 part/cm^3) (Stabile, Dell'Isola et al., 2017; Pacitto, Stabile et al., 2018). This value was adopted, as described in the methodology section, as the concentration representative of the outdoor/city scale in the *M4* simplified methodology. The authors point out that this outdoor PN concentration value (1.1×10^4 part/cm^3) is lower than the indoor value (1.4×10^4 part/cm^3); thus, as hereinafter reported, a simplified methodology only relying on the concentrations measured at the city scale will underestimate the actual dose received by the population. As regards the transportation, even if it is a critical microenvironment due to the high concentration levels, its

contribution to the daily dose results as minor due to the reduced time spent therein.

The IRs calculated from direct measurement of heart rate show an almost constant value for all activities except, of course, for sleeping. Looking at the IRs reported in this paper, some differences can be observed with respect to the typical data commonly used in the literature (Adams, 1993; Schuda, Moya et al., 2009, U.S. Environmental Protection Agency, 2009). In particular, the inhalation rates calculated in the present paper result as follows with respect to the literature values: cooking, 59 % higher; eating, 65 % higher; indoor, 29 % lower; outdoor, 8 % lower, sleeping, 3 % higher; transport, 30 % higher; working, 52 % higher. These differences are likely due to the ability of the applied model to take into account parameter specific for each individual (FVC, HR and age) which cannot be considered by not measured literature values.

The received daily doses for each microenvironment/activity resulting from the full-field method (*M0*) are reported in Table 4 as median values and in Fig. 2 as box-plots (refer to the *M0* data in the figure). The data clearly show that the higher and lower median values obtained with the proposed full-field method result for indoor (2.2×10^2 mm^2) and outdoor (3.0×10^1 mm^2) activities respectively, while all the other activities are characterized by doses ranging between 4.1×10^1 mm^2 (cooking) and 1.0×10^2 mm^2 (working). The total daily dose of 7.5×10^2 mm^2 was quite low with respect to those estimated in previous papers for the typical Italian population (up to 2×10^3 mm^2) (Buonanno et al., 2011b; Pacitto, Stabile et al., 2018). The lower total daily dose estimated for the specific population is likely due to the quite low contribution of eating and cooking activities to the daily dose (10 % and 8 %, respectively, for the specific population investigated) (Table 4), whereas in the abovementioned previous studies these contributions were >50 % of the total daily dose (Buonanno, Giovinco et al., 2011; Pacitto, Stabile et al., 2018). Once again, the lower contribution is due to both the shorter time spent in such activities and the lower particle concentrations; indeed, even if the larger ratios between dose contribution and time fraction (defined as the dose intensities) of the present study are due to eating and cooking activities (0.99 mm^2/min and 1.00 mm^2/min, respectively), such values are lower than those measured in previous studies (Buonanno, Giovinco et al., 2011; Pacitto, Stabile et al., 2018) in which dose intensities for those activities were in the range of 4–14 mm^2/min. The highest contribution to the daily dose for the specific population analysed is due to indoor activity (33 %), while the lowest contribution comes from outdoor activity (5 %). In any case, with the exception of the cooking and eating activities just discussed, the other activities recorded dose intensities <1; for example, sleeping activity, as expected, is characterized by the lowest dose intensity (0.20).

3.2. Comparison of the proposed full-field method with simplified methods

To analyse the influence of the calculation methodology, the daily doses received by the considered population sample were calculated using the simplified methodologies described in Tables 2 and 3, and compared with those obtained through the full-field method. In Fig. 2 the daily doses estimated with all the methodologies are presented as box plots for each activity/microenvironment, while in Table 6 the median values of daily doses, contributions to the daily dose and dose intensities obtained through the different methodologies are summarized. The data reported in Table 6 represent median values between all the population samples for each microenvironment/activity, while the total doses at the bottom of the table represent median values between the total doses of each subject in the sample.

As mentioned above, the median value of the total dose received by the population sample, calculated with the full-field methodology (*M0*), was 7.5×10^2 mm^2. Adopting the *M1* methodology, the total daily dose was 3 % lower (7.3×10^2 mm^2), whereas the *M2* and *M3* methodologies provided total doses 3 % and 5 % higher than *M0*, respectively (7.7×10^2 mm^2 and 7.9×10^2 mm^2). Finally, the total daily dose calculated with *M4* methodology resulted 32 % lower

Fig. 2. Daily doses received by the population sample for each microenvironment/activity as calculated with the different methodologies reported in Tables 1 and 2.

Table 6
Daily dose, contribution to the daily dose, and dose intensity calculated adopting the different methodologies described in Table 1. The data represent median values between all subjects for each activity, while the total dose, at the bottom of the table, represents the median value between the total doses of each subject.

Activity	Daily dose (mm^2)					Contribution to daily dose (%)					Dose intensity (mm^2/min)				
	M0	M1	M2	M3	M4	M0	M1	M2	M3	M4	M0	M1	M2	M3	M4
Transport	**7.7×10^1**	4.5×10^1	5.6×10^1	3.5×10^1	3.4×10^1	**11**	7	9	5	7	**0.59**	0.64	0.50	0.52	0.50
Working	**1.0×10^2**	1.3×10^2	7.7×10^1	7.7×10^1	8.4×10^1	**17**	16	9	12	17	**0.40**	0.40	0.25	0.26	0.26
Eating	**8.0×10^1**	6.7×10^1	5.0×10^1	4.3×10^1	1.8×10^1	**10**	10	6	6	3	**0.99**	1.01	0.66	0.66	0.26
Cooking	**4.1×10^1**	4.9×10^1	2.9×10^1	3.7×10^1	1.3×10^1	**8**	9	6	5	3	**1.00**	1.28	0.94	0.82	0.29
Outdoor	**3.0×10^1**	5.2×10^1	3.0×10^1	6.0×10^1	6.6×10^1	**5**	7	5	8	13	**0.66**	0.62	0.79	0.72	0.78
Indoor	**2.2×10^2**	2.6×10^2	3.7×10^2	4.0×10^2	2.4×10^2	**33**	38	48	52	46	**0.63**	0.63	0.94	0.94	0.55
Sleeping	**9.5×10^1**	8.9×10^1	1.0×10^2	8.3×10^1	6.5×10^1	**13**	13	13	13	13	**0.20**	0.20	0.19	0.19	0.15
Total	**7.5×10^2**	**7.3×10^2**	**7.7×10^2**	**7.9×10^2**	**5.1×10^2**										

than *M0* (5.1×10^2 mm^2). The different dose values obtained need for an in-depth analysis assessing the contribution of each parameter. Looking at the total daily doses reported in Table 6, it is possible to evaluate the influence on the calculation of the single parameters (particle concentration, TAP and IR). In particular, comparing the *M1* methodology with the *M0* methodology, and the *M3* methodology with the *M2* methodology, it is possible to highlight the effect of the TAP: considering standard instead of measured TAP values, the dose results 2.4 % lower in the case of measured IRs (*M1* versus *M0*) and 2.3 % higher in the case of standard IRs (*M3* versus *M2*). The effect of IR itself can be analysed by comparing the total daily doses obtained with *M0* and *M2* methodologies; in particular, the use of IRs not directly measured (*M2*) instead of measured ones (*M0*) results in an overestimation of the received dose of 2.9 %. Anyway, comparing the doses calculated with methodologies *M0*, *M1*, *M2* and *M3*, i.e. considering particle concentrations measured at personal scale, it can be seen that no significant differences exist: the greatest difference in the dose evaluation results from the comparison of *M1* (measured IRs) with *M3* (standard IRs): in this case, the standard IRs lead to higher dose estimation (7.9 %). The behaviour significantly changes when particle concentrations measured at city scale are used (*M4*). In particular, by comparing *M4* with *M3*, it can be seen that using concentrations measured at the city scale (*M4*) instead of a direct exposure assessment through personal monitoring (*M3*) resulted in an underestimation of the total daily dose of 36 %. The activities contributing the most to this difference are those performed in indoor microenvironments (in particular cooking and eating) which are badly estimated when concentrations at the city scale are adopted.

The authors highlight that the differences recognized in the received dose estimates are characteristic of the specific population under investigation. Indeed, from the data shown here, it can likely be inferred that greater differences among the different dose calculation methods would have been detected for a population spending more time in cooking and eating activities at home (e.g. typical Italian TAPs); in other words, the use of simplified methods to evaluate the dose would cause even larger particle dose underestimates with respect to the actual dose quantifiable through a full-field approach. Nonetheless, the specific population considered does not affect the general finding of the paper, i.e. the importance of adopting measured parameters for a proper estimate of the received sub-micron particle dose of a population sample. A further consideration that can be deduced from our results is that the dose received by the typical Italian population [estimated in previous papers as up to 2×10^3 mm^2 (Buonanno, Giovinco et al., 2011; Pacitto, Stabile et al., 2018)] could have been underestimated. Indeed, the higher IRs measured with respect to the not directly measured values would have significantly increased the actual received dose.

3.3. Discussions and limitations of the study

The findings of this study highlight the criticalities in accurately estimating the sub-micron particle dose received by a specific population when no direct measurements of the parameters contributing to the dose are available and, thus, simplified approaches have to be applied. The authors stress that the full-field method itself is not easily applicable and has its own limitations. First of all, it should be pointed out the limitation of self-reported time-activity diaries made out by paper and pencil, and even if direct recording of the TAP will likely be made easier through wrist-worn devices such as trackers (Maher, Ryan et al., 2017), direct measurement of the other two contributors to the daily dose is not easily achievable and presents metrological concerns. In particular, the IRs in this study were obtained on the basis of heart rates directly measured through personal monitors that are not commonly used on a daily basis. Furthermore, a discussion on the metrological performance of the instruments available for personal monitoring of the particle concentration is needed. As described in the methodology section, the wearable instruments typically used to measure the concentrations to which people are exposed during their daily life provide measurements of particle number and lung deposited surface area concentrations. The latter is useful for estimating the surface area dose since it represents the product between the surface area concentration and the alveolar/tracheobronchial deposition fractions size-by-size that otherwise should have been calculated as follows: (i) measuring particle number distribution through differential mobility particle size spectrometers (e.g. a scanning mobility particle sizer); (ii) converting the particle number distribution to a particle surface area distribution (assuming or knowing the shape of the particles); (iii) multiplying the particle surface area distribution by the corresponding lung deposition probability [e.g. applying the above-mentioned ICRP model (International Commission on Radiological Protection, 1994)]. Nonetheless, the diffusion chargers are clearly not able to replace such complex procedures; as mentioned above, they provide a measure of the lung-deposited surface area concentration considering the particle deposition fraction of a specific activity [reference worker under light exercise and breathing only through the nose (Asbach, Kaminski et al., 2012)]; thus, the deposition fractions (and then the LDSA concentrations) of other activities and exercise could be miscalculated. In addition to such oversimplification of particle deposition, the diffusion chargers are also characterized by metrological performances poorer than those typical of non-portable laboratory-based instruments used to measure particle concentrations and size distributions (e.g. condensation particle counters and differential mobility particle size spectrometers). Previous papers have recognized that diffusion chargers can significantly overestimate or underestimate particle number concentrations (and consequently LDSA concentrations)

depending on the type of aerosol under investigation in terms of the mode of the particle under measurement, the presence of pre-charged particles, and the presence of aggregate particles (Fierz, Houle et al., 2011; Asbach, Kaminski et al., 2012; Buonanno, Jayaratne et al., 2014; Vosburgh, Ku et al., 2014; Bau, Payet et al., 2017; Todea, Beckmann et al., 2017). Therefore, adopting a full-field approach to calculate the dose is undoubtedly correct from a methodological point of view, but the application of such an approach is problematic and can hide measurement uncertainties that could nullify the advantage theoretically provided by direct exposure assessment if proper comparisons with reference instruments for the different aerosols investigated are not performed. The limitations described here raise a more essential question that should be addressed in the future: are we really able to measure the particle surface area dose of the population with an acceptable uncertainty? At present, the technological solutions available to measure/obtain necessary data for the dose evaluation are not yet adequate and result in high levels of uncertainty. Future development of this research field should aim to address such limitations and to estimate the uncertainty of the dose.

4. Conclusions

In this paper a new method to calculate the sub-micron particle surface area dose received by a population, named full-field method, was proposed and tested. The method was tested using a population sample made up of full-time workers living in Central Italy. The method is based on direct measurements of the main parameters contributing to the dose that in other simplified approaches are estimated on the basis of data from the literature: the particle concentrations to which a population is exposed, the time spent in each activity (time activity pattern, or TAP), and the air inhaled during such activities (inhalation rate, or IR). Particle concentrations were measured through a direct exposure assessment (i.e. using personal monitors with high spatial and temporal resolution), whereas TAPs were assessed by means of a personal diary, and IRs were evaluated from heart rate values directly measured for each individual through a mobile monitor. The received surface area dose calculated with the full-field method was then compared with those obtained from different simplified methodologies using typical values for particle concentrations, TAPs and IRs in order to analyse the influence of the calculation methodology.

The data obtained through the different approaches highlighted how the methodology applied can affect significantly the estimate of the dose received by the population. Indeed, using the TAPs typical of the Italian population instead of measured values of the population sample under investigation can lead to difference of about ± 3%. The effect of the IR was similar; in fact, the dose estimated through typical IRs was 3 % higher than that obtained through actual IRs of the population sample investigated. Finally, when the exposure assessment to determine the particle concentration was estimated through oversimplified approaches based on concentrations measured at the city scale rather than at the personal scale, an underestimation of 35 % for the specific population investigated was detected. The main contributors to such differences were the indoor activities (due to the long time spent undertaking them), and cooking and eating activities (due to the high concentrations). The authors also highlight that such differences among the different methodologies could have been even larger for a population characterized by a different lifestyle, such as a population spending more time in cooking activities, as is characteristic of the typical Italian population.

Statement of ethics approval

The study obtained ethics approval from all the participant which gave informed consent before taking part to the research program.

CRediT authorship contribution statement

Mauro Scungio: Conceptualization, Methodology, Writing - original draft. **Valeria Rizza:** Investigation, Resources, Data curation. **Luca Stabile:** Formal analysis, Validation, Writing - review & editing. **Lidia Morawska:** Supervision, Writing - review & editing. **Giorgio Buonanno:** Conceptualization, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.115209.

References

Adams, W.C., 1993. Measurement of Breathing Rate and Volume in Routinely Performed Daily Activities. California Environmental Protection Agency.
Asbach, C., et al., 2012. Comparability of portable nanoparticle exposure monitors. Ann. Occup. Hyg. 56 (5), 606–621.
Australian Bureau of Statistics, 2010. Customised Report. Cat. No. 4430.0. Australian Bureau of Statistics, Canberra, AUS. EPA 600/P-99/002aF-bF.
Avery, C.L., et al., 2010. Estimating error in using residential outdoor $PM_{2.5}$ concentrations as proxies for personal exposures: a meta-analysis. Environ. Health Perspect. 118 (5), 673–678.
Avino, P., et al., 2018. Second-hand aerosol from tobacco and electronic cigarettes: evaluation of the smoker emission rates and doses and lung cancer risk of passive smokers and vapers. Sci. Total Environ. 642, 137–147.
Azimi, P., Stephens, B., 2018. A framework for estimating the US mortality burden of fine particulate matter exposure attributable to indoor and outdoor microenvironments. J. Expo. Sci. Environ. Epidemiol. 30 (2), 271–284.
Basagaña, X., et al., 2015. Short-term effects of particulate matter constituents on daily hospitalizations and mortality in five South-European cities: results from the MED-PARTICLES project. Environ. Int. 75, 151–158.
Bau, S., et al., 2017. Performance study of portable devices for the real-time measurement of airborne particle number concentration and size (distribution). J. Phys. Conf. 838, 012001.
Brook, R.D., et al., 2010. Particulate matter air pollution and cardiovascular disease: an update to the scientific statement from the American Heart Association. Circulation 68, 224–230.
Buonanno, G., et al., 2013a. Airborne particle concentrations at schools measured at different spatial scales. Atmos. Environ. 67, 38–45.
Buonanno, G., et al., 2011a. Influential parameters on particle exposure of pedestrians in urban microenvironments. Atmos. Environ. 45 (7), 1434–1443.
Buonanno, G., et al., 2011b. Tracheobronchial and alveolar dose of submicrometer particles for different population age groups in Italy. Atmos. Environ. 45 (34), 6216–6224.
Buonanno, G., et al., 2014a. Metrological performances of a diffusion charger particle counter for personal monitoring. Aerosol and Air Qual. Res. 14, 156–167.
Buonanno, G., et al., 2009. Temporal size distribution and concentration of particles near a major highway. Atmos. Environ. 43 (5), 1100–1105.
Buonanno, G., et al., 2013b. Health effects of daily airborne particle dose in children: direct association between personal dose and respiratory health effects. Environ. Pollut. 180, 246–250.
Buonanno, G., et al., 2014c. Personal exposure to ultrafine particles: the influence of time-activity patterns. Sci. Total Environ. 468–469, 903–907.
Buteau, S., Goldberg, M.S., 2016. A structured review of panel studies used to investigate associations between ambient air pollution and heart rate variability. Environ. Res. 148, 207–247.
Cattaneo, A., et al., 2010. Comparison between personal and individual exposure to urban air pollutants. Aerosol. Sci. Technol. 44 (5), 370–379.
Cesaroni, G., et al., 2013. Long-term exposure to urban air pollution and mortality in a cohort of more than a million adults in Rome. Environ. Health Perspect. 121

(3), 324–331.
Chen, X., et al., 2016. Long-term exposure to urban air pollution and lung cancer mortality: a 12-year cohort study in Northern China. Sci. Total Environ. 571, 855–861.
Clifford, S., et al., 2018. Effects of exposure to ambient ultrafine particles on respiratory health and systemic inflammation in children. Environ. Int. 114, 167–180.
Cooper, B., et al., 2017. The Global Lung Function Initiative (GLI) Network: bringing the world's respiratory reference values together. Breathe 13, e56–e64.
Correia, C., et al., 2020. Particle exposure and inhaled dose while commuting in Lisbon. Environ. Pollut. 257.
Fierz, M., et al., 2011. Design, calibration, and field performance of a miniature diffusion size classifier. Aerosol. Sci. Technol. 45 (1), 1–10.
Franchini, M., Mannucci, P.O., 2011. Thrombogenicity and cardiovascular effects of ambient air pollution. Blood 118 (9), 2405–2412.
Fuoco, F.C., et al., 2017. Tracheobronchial and alveolar particle surface area doses in smokers. Atmosphere 8 (1).
Giechaskiel, B., et al., 2009. A metric for health effects studies of diesel exhaust particles. J. Aerosol Sci. 40 (8), 639–651.
Greenwald, R., et al., 2019. Estimating minute ventilation and air pollution inhaled dose using heart rate, breath frequency, age, sex and forced vital capacity: a pooled-data analysis. PloS One 14 (7), e0218673.
IARC, 2013. Air Pollution and Cancer. International Agency for Research on Cancer, Geneva, Switzerland.
International Commission on Radiological Protection, 1994. Human respiratory tract model for radiological protection. A report of a task group of the international commission on radiological protection. Ann. ICRP 24, 1–482.
Kaur, S., et al., 2005. Pedestrian exposure to air pollution along a major road in Central London, UK. Atmos. Environ. 39 (38), 7307–7320.
Klepeis, N.E., 2006. Modelling human exposure to air pollution. In: Ott, et al. (Eds.), Human Exposure Analisys. CRC Press, pp. 445–470.
Lader, D., et al., 2006. The Time Use Survey, 2005. How We Spend Our Time. Office for National Statistics, London.
Madureira, J., et al., 2020. Assessment of indoor air exposure among newborns and their mothers: levels and sources of PM_{10}, $PM_{2.5}$ and ultrafine particles at 65 home environments. Environ. Pollut. 264.
Maher, C., et al., 2017. Users' experiences of wearable activity trackers: a cross-sectional study. BMC Publ. Health 17 (1), 880.
Manigrasso, M., Avino, P., 2012. Fast evolution of urban ultrafine particles: implications for deposition doses in the human respiratory system. Atmos. Environ. 51, 116–123.
Marra, J., et al., 2010. Monitor for detecting and assessing exposure to airborne nanoparticles. J. Nanoparticle Res. 12 (1), 21–37.
Mazaheri, M., et al., 2014. School children's personal exposure to ultrafine particles in the urban environment. Environ. Sci. Technol. 48 (1), 113–120.
McGinnity, F., et al., 2005. Time Use in Ireland, 2005. Survey Report. The Economic and Social Research Institute, Dublin.
Pacitto, A., et al., 2018a. The influence of lifestyle on airborne particle surface area doses received by different Western populations. Environ. Pollut. 232, 113–122.
Pacitto, A., et al., 2020. Exposure to submicron particles and estimation of the dose received by children in school and non-school environments. Atmosphere 11 (5), 485.
Pacitto, A., et al., 2018b. Particle-related exposure, dose and lung cancer risk of primary school children in two European countries. Sci. Total Environ. 616–617, 720–729.
Puustinen, A., et al., 2007. Spatial variation of particle number and mass over four European cities. Atmos. Environ. 41 (31), 6622–6636.
Qiu, Z., et al., 2019. Exposure assessment of cyclists to UFP and PM on urban routes in Xi'an, China. Environ. Pollut. 250, 241–250.
Raaschou-Nielsen, O., et al., 2013. Air pollution and lung cancer incidence in 17 European cohorts: prospective analyses from the European study of cohorts for air pollution effects (ESCAPE). Lancet Oncol. 14 (9), 813–822.
Rizza, V., et al., 2017. Variability of airborne particle metrics in an urban area. Environ. Pollut. 220 (Part A), 625–635.
Rizza, V., et al., 2019. Effects of the exposure to ultrafine particles on heart rate in a healthy population. Sci. Total Environ. 650, 2403–2410.
Rushton, E.K., et al., 2010. Concept of assessing nanoparticle hazards considering nanoparticle dosemetric and chemical/biological response metrics. J. Toxicol. Environ. Health 73 (5–6), 445–461.
Schmid, O., et al., 2009. Dosimetry and toxicology of inhaled ultrafine particles. Biomarkers 14 (Suppl. 1), 67–73.
Schmid, O., Stoeger, T., 2016. Surface area is the biologically most effective dose metric for acute nanoparticle toxicity in the lung. J. Aerosol Sci. 99, 133–143.
Schuda, L., et al., 2009. Metabolically Derived Human Ventilation Rates: a Revised Approach Based upon Oxygen Consumption Rates. EPA.
Scungio, M., et al., 2018. Measurements of electronic cigarette-generated particles for the evaluation of lung cancer risk of active and passive users. J. Aerosol Sci. 115, 1–11.
Slezakova, K., et al., 2019. Assessment of ultrafine particles in primary schools: emphasis on different indoor microenvironments. Environ. Pollut. 246, 885–895.
Slezakova, K., et al., 2020. Ultrafine particles: levels in ambient air during outdoor sport activities. Environ. Pollut. 258.
Soggiu, M.E., et al., 2010. Human Exposure Assessment to Environmental Contaminants: Exposure Scenarios. Rapporti ISTISAN 10/19. Istituto Superiore di Sanità (Italy).
Stabile, L., et al., 2013. Dimensional and chemical characterization of airborne particles in schools: respiratory effects in children. Aerosol and Air Qual. Res. 13 (3), 887–900.
Stabile, L., et al., 2019. The effect of the ventilation retrofit in a school on CO2, airborne particles, and energy consumptions. Build. Environ. 156, 1–11.
Stabile, L., et al., 2017. The effect of natural ventilation strategy on indoor air quality in schools. Sci. Total Environ. 595, 894–902.
Stabile, L., et al., 2016. Effect of natural ventilation and manual airing on indoor air quality in naturally ventilated Italian classrooms. Build. Environ. 98, 180–189.
Stafoggia, M., et al., 2017. Association between short-term exposure to ultrafine particles and mortality in eight European urban areas. Epidemiology 28 (2), 172–180.
Statistics Sweden Economic Welfare Statistics Unit, 2012. Swedish Time Use Survey 2010/11. Orebro.
Steinle, S., et al., 2013. Quantifying human exposure to air pollution-Moving from static monitoring to spatio-temporally resolved personal exposure assessment. Sci. Total Environ. 443, 184–193.
Strak, M., et al., 2010. Respiratory health effects of ultrafine and fine particle exposure in cyclists. Occup. Environ. Med. 67 (2), 118–124.
Todea, A.M., et al., 2017. Inter-comparison of personal monitors for nanoparticles exposure at workplaces and in the environment. Sci. Total Environ. 605–606, 929–945.
U.S. Environmental Protection Agency, 2009. Metabolically Derived Human Ventilation Rates: A Revised Approach Based upon Oxygen Consumption Rates. National Center for Environmental Assessment, Office of Research and Development. EPA/600/R-06/129F. E. P. Agenct.
Vosburgh, D.J.H., et al., 2014. Evaluation of a diffusion charger for measuring aerosols in a workplace. Ann. Occup. Hyg. 58 (4), 424–436.
Xie, W., et al., 2015. Relationship between fine particulate air pollution and ischaemic heart disease morbidity and mortality. Heart 101 (4), 257.
Zhu, Y., et al., 2005. Penetration of freeway ultrafine particles into indoor environments. J. Aerosol Sci. 36 (3), 303–322.