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

Ultrafine particles in domestic environments: Regional doses deposited in the human respiratory system

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ARTICLE INFO

Keywords: Ultrafine particles Indoor Respiratory doses MPPD Chronic exposure Brush electric motors Combustion

ABSTRACT

Based on aerosol measurements carried out in a test room, particle regional respiratory doses have been estimated for both combustion and non-combustion aerosol sources currently encountered in domestic environments. The general population chronically receives doses that, pooled together, are well above those due to urban outdoor pollution. At the end of each source operation, from 75% to 99% and from 27% to 93% of these doses are due to ultrafine particle, respectively on particle number and surface area metrics. Depending on the source, the pattern of exposure may be, for very short time laps, very intense (up to 8.0 \times 10⁹ particles s⁻¹) and involve a fraction of particles with mode at about 10 nm. For appliance operated by brush electric motors, this mode is the major one and is due to the generation of copper nanoparticles. The health relevance of such particles deserves particular attention due to their possible translocation to the brain and in the light of the associations between copper ions and Alzheimer's disease, proposed by several studies.

1. Introduction

Fine Particles (FPs < 2500 nm diameter) and ultrafine particles (UFPs < 100 nm diameter) are well-known air pollutants, present both in outdoor and indoor air. Over the years, they have been associated to a great number of negative health outcomes and diseases involving several systems and apparatus, such as cardiovascular, respiratory and neurodegenerative systems ([Anderson et al., 2012;](#page--1-0) [Noh et al., 2016](#page--1-1); [Wang et al., 2017](#page--1-2)). Besides, Particulate Matter (PM) in outdoor air is classified as group 1 carcinogen to humans by the International Agency for Research on Cancer (IARC) [\(IARC, 2015\)](#page--1-3). These scientific evidences mainly emerged from researches focused on some specific PM fractions (particles diameter with a less than or equal to 10 μm and less than or equal to 2.5 μ m, defined PM₁₀ and PM_{2.5}, respectively). In contrast, in the last years, ultrafine particles (UFPs), i.e. particles with sizes below 10 nm, have caught the attention of the scientific community, promoting further research that highlighted that they play the most significant role in the adverse effects related to PM exposure ([Hoek et al.,](#page--1-4) [2010\)](#page--1-4). Explanation to these evidences can be linked to several reasons. First of all, particles with a diameter $< 1 \mu m$ (PM₁) may persist for longer times and/or may be transported over extensive ranges respect to particles with a larger diameter ([Brauer et al., 1989\)](#page--1-5). Besides,

particles with diameter $> 2.5 \mu m$ seem to be rapidly eliminated from the atmosphere by wet and dry deposition. Secondly, it has been demonstrated an inverse relationship between the PM toxicity per unit mass and its size, that led the researchers to study human exposure in term of particle surface area or particle number instead of the mass concentrations ([Buonanno et al., 2011;](#page--1-6) [Manigrasso and Avino, 2012](#page--1-7); [Stabile et al., 2013;](#page--1-8) [Canepari et al., 2013](#page--1-9); [Manigrasso et al., 2015](#page--1-10); [Marini et al., 2015;](#page--1-11) [Avino et al., 2016](#page--1-12)). The very small size of UFPs, together with their high concentration in term of particles number and high surface area per unit mass, greatly facilitates the absorption of organic molecules onto their surface and their penetration into cellular targets [\(Li et al., 2003](#page--1-13); [Pagano et al., 1996](#page--1-14)). Furthermore, UFP inflammatory effects occur through an oxidative stress mechanism that is correlated not only with their endocytosis but also with their surface area. Sub populations with respiratory diseases having an oxidative stress pathogenesis (e.g. chronic obstructive pulmonary disease) may be particularly susceptible ([Hussain et al., 2009](#page--1-15)). For all these reasons, the evaluation of human exposure to UFPs should be performed considering surface area or particle number rather than the mass concentrations.

Another important consideration about the researches performed to assess the human exposure to UFPs is related to the monitored exposure scenarios. Indeed, most of the human health adverse effects PM-related

<https://doi.org/10.1016/j.envint.2018.05.049> Received 18 March 2018; Received in revised form 20 May 2018; Accepted 29 May 2018 0160-4120/ © 2018 Published by Elsevier Ltd.

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were evidenced considering outdoor PM concentrations. Consequently, at today, negative health outcomes from outdoor PM exposure are well documented, while studies on indoor PM are still lacking respect to the needs of knowledge [\(Morawska and Salthammer, 2015\)](#page--1-16). In contrast, it is well-known that people spend most of their time (up to 90%) in enclosed environments [\(Hubal et al., 2000](#page--1-17); [CalEPA, 2004\)](#page--1-18). Besides, it is important to note that until the 1970s it was generally agreed that the indoor air quality was influenced just by the atmospheric pollution close the enclosed environment (Śmieł[owska et al., 2017](#page--1-19)). Then, it has been recognized that indoor concentrations of many pollutants, including particles, are the sum of outdoor contaminants and those produced directly indoor [\(Diapouli et al., 2013;](#page--1-20) [Morawska et al., 2017](#page--1-21); Śmieł[owska et al., 2017\)](#page--1-19). As regards to FPs and UFPs, several studies demonstrated that there is a great number of particles sources in enclosed environments, mainly represented by combustion activities such as tobacco smoking, heating, cooking activities, burning mosquito coils, burning incense and candles, etc. ([Sarwar et al., 2004](#page--1-22); [Hsu et al., 2012](#page--1-23); [Huang et al., 2012;](#page--1-24) [Liu et al., 2014](#page--1-25); [Protano et al., 2016;](#page--1-26) [Protano et al.,](#page--1-27) [2017\)](#page--1-27). In the recent years several non-combustion sources were evidenced to contribute to the PM indoor pollution: electronic smoking, the use of electric devices such as drills, flat irons, hair dryers, etc. ([Manigrasso et al., 2017b](#page--1-28)). These sources are commonly present in domestic environments, configuring a potential threat for human health; this issue is of great relevance for public health, as risk assessment and management strategies to control indoor air quality, based on official monitoring campaigns and restrictive legislation, cannot find their application in household environments. Thus, an essential research agenda is the evaluation of human exposure to the most dangerous fractions of PM released during common activities in domestic scenarios. The obtained data are necessary to carry out an appropriate risk assessment and, may represent an evidence-based support for health promotion in general population.

The study was performed to assess individual exposure to UFPs released from some major potential sources present and commonly used in domestic environments; for this purpose, we investigated UFP number concentrations and size distributions emitted during the use of combustion and non-combustion aerosol sources. The obtained data were used to estimate the potential human exposure through the estimated doses of particles deposited into the respiratory system for each investigated combustion and non-combustion source.

2. Materials and methods

2.1. Aerosol measurements and quality assurance

Aerosol was released from emission sources currently encountered in domestic environments. Among non-combustion sources, hairdryers, hot flat irons, electric drills, vacuum cleaners and electronic cigarette (e-cigs) were considered. Combustion sources included mosquito coil and incense burning, meat grilling and tobacco cigarette smoke. A detailed aerosol characterization is reported in a previous study, that discussed also the number concentrations and size number distributions of the aerosols emitted by the sources studied [\(Manigrasso et al.,](#page--1-28) [2017b\)](#page--1-28). Briefly, aerosol measurements were carried out in a 52.7 m^3 room where the door and window were both closed. During the aerosol measurements, the room temperature and relative humidity ranged between 24 °C and 26 °C and 25% and 32%, respectively. The air exchange rate (λ) was calculated using the tracer gas technique ([Laussmann and Helm, 2011\)](#page--1-29). $CO₂$ was used as a tracer gas. It was released from a cylinder into the ambient air until a relatively stable concentration was reached; then, the decaying $CO₂$ concentration was measured over time (t). The temporal evolution of the $CO₂$ concentra-tion is described by Eq. [\(1\)](#page-1-0), where λ is the air exchange rate, $C_{in}(t)$, C_0 and C_{out} are CO₂ concentrations respectively indoor (at t = t and t = 0) and outdoor:

$$
\ln(C_{in}(t) - C_{out}) = \ln(C_0 - C_{out}) - \lambda t
$$
\n(1)

 λ equal to 0.67 h⁻¹ was calculated via linear regression analysis.

Due to the fast evolution of the aerosol measured ([Manigrasso and](#page--1-7) [Avino, 2012;](#page--1-7) [Manigrasso et al., 2013\)](#page--1-30), aerosol number size distributions were measured by means of a Fast Mobility Particle Sizer (FMPS, model 3091, TSI, Shoreview, MN, USA). The instrument counts and classifies particles according to their electrical mobility in 32 size channels ranging from 5.6 to 560 nm with a 1 s time resolution. FMPS operates at a high flow rate (10 L min⁻¹) to minimize diffusion losses. It operates at ambient pressure to prevent the evaporation of volatile and semivolatile particles [\(TSI, 2015](#page--1-31)). Before the measuring campaign, the performances of FMPS were checked by comparison with a Scanning Mobility Particle Sizer (SMPS, model 3936, TSI) equipped with an Electrostatic Classifier (model 3080, TSI), a Differential Mobility Analyser (DMA, model 3081, TSI) and a Condensation Particle Counter (model 3775, TSI). The FMPS number concentrations were approximately 15% lower than the diffusion loss corrected SMPS number concentrations, in agreement with the findings of [Jeong and Evans](#page--1-32) [\(2009\).](#page--1-32)

2.2. Dose evaluation

The particle regional deposition fractions per $(F^R(d_i))$ have been estimated using the Multiple-Path Particle Dosimetry model (MPPD v3.01, ARA 2015, ARA, Arlington, VA, USA) ([Asgharian et al., 2001](#page--1-33)). MPPD model includes three airway geometries: a symmetric geometry for the whole lung, a five-lobe model that considers five symmetric different structures for each of the five lung lobes and ten stochastic lung models that take the asymmetry of the airway structure into account. Symmetric models rely on the assumption of a fully symmetrical airway structure and suppose that all the respiratory tracts of the same airway generation have the same geometrical parameters (i.e. length, diameter, branching and gravity angles). Actually, airway branching is not symmetrical and there are variations of geometrical parameters for a given airway generation (intra-subject variation). Due to the great number of possible pathway from the trachea to the alveolar sacs, particles, once inhaled, follow random paths. Relying on the morphometric data of [Raabe et al. \(1976\),](#page--1-34) [Koblinger and Hofmann \(1985\)](#page--1-35) derived statistical probability density functions for the airway parameters in order to describe the airway structure. Then, such structure has been used in a stochastic model to estimate particle deposition through Monte Carlo technique, allowing a random selection of the geometry of the airways along the path of an inhaled particle ([Koblinger and Hofmann, 1990\)](#page--1-36). [Asgharian et al. \(2001\),](#page--1-33) by taking single selections for each parameter from the distribution functions of [Koblinger and Hofmann \(1985\)](#page--1-35), described the bronchial tree of single individuals rather than of the average population. They defined ten five-lobe, asymmetric, structurally different human airway trees, providing so an estimate of inter-subject variability among the human population. In MPPD model, the ten lung models are ordered in size (total number of airways) from smallest to largest and the approximate size percentile of each lung is provided.

In the present study the 60th percentile human stochastic lung was considered along with the following settings: (i) a uniformly expanding flow, (ii) an upright body orientation, and (iii) nasal breathing with a 0.5 inspiratory fraction and no pause fraction. Moreover, the following parameters were used for a Caucasian adult male under light work physical activity, based on the ICRP report ([ICRP, 1994\)](#page--1-37): (i) a functional residual capacity (FRC) of 3300 mL, (ii) an upper respiratory tract (URT) volume equal to 50 mL, (iii) a 20 min^{-1} breathing frequency, and (iv) an air volume inhaled during a single breath (tidal volume, V_t) of 1.25 L.

Since FMPS measures aerosol size number distribution as a function of the electrical mobility diameter (d) , d values have been transformed to aerodynamic diameter (d_a) according to Eq. [\(2\)](#page-1-1) ([Li et al., 2016\)](#page--1-38).

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