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Multi-metric measurement of personal exposure to ultrafine particles in selected urban microenvironments

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HIGHLIGHTS

- Ultrafine particles (UFP) personal exposure were measured in urban environments.
- UFP were characterized by number, mass concentration, mean diameter and surface-area.
- Appreciable differences among microenvironments and monitoring periods were observed.
- Concentration patterns were related to typical sources of urban pollutants (traffic).
- Temporal and microenvironmental patterns were determinants of UFP exposure.

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ABSTRACT

At the beginning of the study, our hypothesis was that visiting certain microenvironments (MEs) is one of the most important determinants of personal exposure to ultrafine particles (UFP) and that moving between microenvironments significantly differentiates exposure. The overall aim of this study is to perform relevant exposure measurements to extend our knowledge on environmental exposure to UFP in urban environments. The UFP concentrations in different urban MEs were measured by personal monitoring in repeated sampling campaigns along a fixed route. The measurement runs were performed on one-week periods and at different times of day (AM: 08.00–10.30; PM: 16.00–18.30) and repeated in different periods of the year (winter, spring, summer, and autumn) for a total of 56 runs (>110 h). Measurements included on-line monitoring of the UFP particle number concentration (PNC), mean diameter (mean-d) and lung-deposited surface-area (LDSA). Additionally, the PNC, particle mass concentration (PMC) profiles for quasi-ultrafine particles (QUFP; $PM_{0.25}$) were estimated. A significant seasonal difference in the PNC and PMC, mean diameter and surface area was observed as well as between different times of the day and days of the week. In addition, differences in the UFP concentrations were also found in each ME, and there were specific mean-diameter and surface area concentrations. In general, the mean particle diameters showed an inverse relationship with the PNC, while the LDSA had the opposite behaviour. Appreciable differences among all MEs and monitoring periods were observed; the concentration patterns and variations seemed related to the typical sources of urban pollutants (traffic), proximity to sources and time of day. The highest exposures were observed for walking or biking along high-trafficked routes and while using public buses. The UFP exposure levels in modern cars, equipped with high-efficiency filters and in air recirculation mode, were significantly lower.

1. Introduction

Human exposure to ultrafine particles (UFP, particle with diameter < 100 nm) in urban transport microenvironments (MEs) is of particular interest since it has been demonstrated that their levels are particularly high along busy roads, and their peak

Abbreviations: LDSA, Lung-deposited surface area; Mean-d, mean diameter; UFP, ultrafine particles; PNC, particle number concentration; PMC, particle mass concentration.

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concentrations are typically registered during commute hours (Morawska et al., 2008; Moreno et al., 2009). Moreover, the UFP concentrations are characterized by high spatial and temporal variability in urban MEs (Moore et al., 2009; Hudda et al., 2010) and recent studies have documented the dependence of the UFP levels on several urban factors (e.g., traffic volume and mode, street geometry, environmental and meteorological characteristics) (Boogaard et al., 2010; Hoek et al., 2011, Morawska et al., 2008; Buonanno et al., 2011a; Ragettli et al., 2013; Rivera et al., 2012). The highest UFP concentrations were typically found in the vicinity of the primary sources (e.g., near busy roads) where the particle number concentrations are typically between 10^4 and 10^6 particles/cm^3 (Nikolova et al., 2011), while UFP concentrations decrease rapidly with distance from the emission sources (Hagler et al., 2010; Zhu et al., 2008). Furthermore, despite the major contributions to exposure doses for adult population were found to arise from indoor cooking and eating times or working time (Buonanno et al., 2011b; Morawska et al., 2013), transportation MEs are significant in terms of particles dose contribution: in fact, even though the average contribution to particles' alveolar-deposited number and surface area was quite low, the dose intensity was higher than the one received in indoor MEs (Buonanno et al., 2013). Therefore, time spent in transit MEs represents a high-exposure period compared to other daily activities, especially in metropolitan areas, even though individuals usually travel for no more than 6–8% of the day (Kaur et al., 2007). The reason of the significant of the so-called “lifestyle effect” on received particle doses is due to the different particle exposure levels experienced in different micro-environments: highest dose intensities were found during indoor activities (e.g., cooking) and transportations (both indoor, car and bus, and outdoor, pedestrian and bike) (Buonanno et al., 2013). Because of these important temporal and spatial differences of the UFP concentrations between urban MEs, fixed monitoring stations are not capable of depicting the full spatial distribution of air pollution over the extent of an urban area (Kaur et al., 2007), leading to significant underestimations in assessing general exposure (Adams et al., 2001; Gulliver and Briggs, 2004). Therefore, mobile measurements are frequently applied as an efficient tool, capable of describing human exposures with a high spatial and temporal resolution (Berghmans et al., 2009; Cattaneo et al., 2009; Kaur et al., 2005; Peters et al., 2013; Ragettli et al., 2013; Westerdahl et al., 2005), mapping the spatial distribution of air pollutants (Zhu et al., 2008), or characterizing the local source contributions to ambient air pollution and developing exposure models (Ragettli et al., 2014; Zwack et al., 2011). Furthermore, at the present time, there is insufficient information to determine which exposure metrics are the most relevant to human health outcomes. Nevertheless, there is strong toxicity-based evidence that the nano-aerosol surface-area is an appropriate exposure metric for nanoaerosols (Brown et al., 2001; Lison et al., 1997; Oberdörster, 2000, Tran et al., 2000) and that the biological response depends more on the surface-area of particles deposited in the lungs (Brown et al., 2001; Donaldson et al., 1996; Hamoir et al., 2003; Tran et al., 2000) than on the other possible metrics of exposure. However, there are also indications that the PNC within specific particle size ranges might be an important indicator of the health effects of UFP exposures (Beck-Speier et al., 2001, Peter et al., 1997; Oberdörster et al., 1994). Furthermore, the number of sub-100-nm particles dominates the extent of respiratory deposition (ICRP, 1994). This clearly suggests that knowledge of the particle size distributions, which can dramatically change in urban environments (Dall'Osto et al., 2011), is important for accurately estimating UFP uptake (Kumar et al., 2014). Therefore, it is expected that particles within a nanometer size range have a biological behaviour that is more closely associated with the PNC, mean-d and surface area than with the PMC.

In this study, an experimental method to measure the exposure to UFP, at a high resolution is presented. The purpose of this paper is to evaluate the exposure with a study design aimed at analysing space and time variations (different seasons, two daily monitoring periods, urban indoor and outdoor MEs) of UFP concentrations in different urban and commuter MEs. The originality of the present work lies in the continuous, time-resolved personal monitoring of the particle number concentration (PNC), mean diameter (mean-d) and surface area (LDSA: lung deposited surface area) with a study design allowing for measurements of the urban microenvironmental UFP concentrations with high temporal resolution. In addition to the PNC, the particle mass concentration (PMC) profiles were estimated through the use of an estimation method, based on the particulate mass density factors, which were defined by the contextual personal sampling of quasi-ultrafine particles (QUFP; $PM_{0.25}$) and subsequent gravimetrical analysis.

2. Materials and methods

2.1. Study area and monitoring protocol

The measurements were performed in Como, Italy (45°49′00″N, 9°05′00″E). Como is a medium-sized provincial town (84,000 inhabitants, 37.34 inhabitants/km^2) in the northern part of Italy. Experimental data were collected within the central area of the city over four one-week periods in four different seasons during the year 2014. A fixed route (17 km long) was defined and the average travelling time for the entire route was approximately 120 min. The major part of the route was located in the residential areas, but streets with differing configuration and with differing traffic dynamics were included in the designed route. The results presented in this study mainly focus on a selection of urban microenvironments (ME) or modes of transportation, which were assumed to reflect the most relevant variations of exposures with respect to the mode of transportation (walk/bike, car, or bus) and traffic density (High traffic areas – HT; Low- or no-traffic areas – LT). An indoor environment (office) was also included. More specifically, measurements were performed according to a sequential protocol that started and finished at the office. The study design first included a car (2012 Toyota Yaris, petrol-fueled) journey around the city centre (6.3 km); the measurements were collected at the passenger's seat. In-car ventilation settings were set for all runs as follows: windows closed, circulation fan on and recirculation (RC) fan on, and the fan speed was kept low. The study protocol then included a bicycle journey (2.8 km), and a pedestrian route was established, crossing the city centre approximately in the S–N direction for an overall distance of 4 km and considering variations in the mode of transport traffic density with busy roads versus traffic-limited areas and urban parks. Pedestrian and bike journeys were combined into two MEs, the first (qualitatively) related to high traffic condition (Bike/Walk – HT) and the other related to low or no traffic (Bike/Walk – LT). Finally, a bus (diesel-fuel with a diesel particulate filter) journey (2.1 km, N–S direction) was taken to return to the starting point; in-bus ventilation conditions (windows open, ventilation and air conditioning settings) were not standardized. All circumstances in which the operator was exposed in other unspecified ME or situations (e.g., waiting at the bus stop) are classified as ‘undefined ME’ (Other). The monitoring protocol was designed to collect data for at least 5 min in each selected ME and to assess the daily, weekly and seasonal variations in the UFP concentrations and characteristics. Data were collected daily at two times, morning (‘AM’: 08:00–10:30) and afternoon (‘PM’: 16:00–18:30), and measurements were carried out over one-week periods (four repetitions: winter, spring, summer, and autumn).