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Personal day-time exposure to ultrafine particles in different microenvironments

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ABSTRACT

In order to assess the personal exposure to ultrafine particles (UFP) during individual day-time activities and to investigate the impact of different microenvironments on exposure, we measured personal exposure to particle number concentrations (PNC), a surrogate for UFP, among 112 non-smoking participants in Augsburg, Germany over a nearly two-year period from March 2007 to December 2008. We obtained 337 personal PNC measurements from 112 participants together with dairies of their activities and locations. The measurements lasted on average 5.5 h and contained on average 330 observations. In addition, ambient PNC were measured at an urban background stationary monitoring site. Personal PNC were highly variable between measurements (IQR of mean: 11 780–24 650 cm^{-3}) and also within a single measurement. Outdoor personal PNC in traffic environments were about two times higher than in non-traffic environments. Higher indoor personal PNC were associated with activities like cooking, being in a bistro or exposure to passive smoking. Overall, personal and stationary PNC were weakly to moderately correlated ($r < 0.41$). Personal PNC were much higher than stationary PNC in traffic (ratio: 1.5), when shopping (ratio: 2.4), and indoors with water vapor (ratio: 2.5). Additive mixed models were applied to predict personal PNC by participants' activities and locations. Traffic microenvironments were significant determinants for outdoor personal PNC. Being in a bistro, passive smoking, and cooking contributed significantly to an increased indoor personal PNC.

Introduction

Epidemiological studies have suggested that ultrafine particles (UFP, particles with diameter smaller than 100 nanometers) are associated with pulmonary and cardiovascular diseases (Andersen et al., 2010; Delfino et al., 2005; Ibald-Mulli et al., 2002; Ruckerl et al., 2011). UFP dominate particle number concentrations (PNC) but contribute very little to particle mass concentrations (PMC). UFP have been found to have health effects of similar magnitude of larger particles, but the effects are suggested to be independent of effects of larger particles (Pekkanen et al., 2002; Stolzel et al., 2007).

UFP show greater spatial variability (Cyrys et al., 2008; Puustinen et al., 2007) than particle mass, which are generally well correlated within an urban area (Cyrys et al., 1998; DeGaetano and Doherty, 2004; Gu et al., 2013; Wilson et al., 2005). Major sources of ambient UFP include vehicles which emit a large amount of particles in the ultrafine mode. Vehicle exhaust (containing both gas vapor and UFP) goes through a rapid physical transformation including dispersion, coagulation and condensation right after being emitted (Kumar et al., 2011). Number concentrations show a decreasing gradient within a few hundred meters downwind of a road/freeway (Sturm et al., 2003; Zhu et al., 2002). Therefore, exposure to UFP can differ greatly between urban microenvironments. Many studies have measured the exposure to air pollution while commuting including car driving, public transport and cycling, as reviewed by Knibbs et al. (2011). These studies indicated that in spite of the limited time spent on commuting, it contributed to a significant amount of the total daily exposure (Berghmans et al., 2009; Dons et al., 2012; Kaur and Nieuwenhuijsen, 2009; Knibbs et al.,

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2011). High UFP concentrations were also found to be related to activities like cooking, smoking, dining in a restaurant (Wallace and Ott, 2011) and vacuum cleaning (Knibbs et al., 2012). Weichenthal et al. (2006) found electric oven use, indoor relative humidity and smoking to be major determinants of mean indoor UFP exposure. When no indoor sources were present, the indoor/outdoor concentrations were found to be well correlated (Cyrys et al., 2004; Diapouli et al., 2007). However, parameters like outdoor concentration, ventilation condition and particle size distribution can affect the indoor exposure (Cyrys et al., 2004; Rim et al., 2013; Zhu et al., 2005). Therefore, assessment of personal exposure to UFP should consider a variety of microenvironments and respective sources.

Individual's exposure to UFP is also closely associated with time-activity patterns, i.e., where and how he/she spends the time. To accurately quantify the personal exposure to UFP, one needs to take both concentrations variability between microenvironments and individual's time-activity pattern into account. However, the direct measurement of personal exposure to UFP and the assessment of the association with microenvironments as well as time-activity patterns have been limited up to now (Buonanno et al., 2012; Cattaneo et al., 2009).

Epidemiological studies on short-term health effects of UFP often relied on data measured from the central monitoring station, which is considered to represent the average population exposure. Due to the highly variable nature of UFP, the relationship between personal exposure and ambient concentrations should be evaluated in depth (HEI, 2013). Such personal measurement will also provide a more accurate estimate on exposure for subsequent epidemiological panel studies.

As part of the Rochester Particulate Matter Center investigations, an epidemiological study focusing on the health effects of UFP was conducted in the city of Augsburg as well as the administrative districts of Augsburg and Aichach-Friedberg, Germany, between March 19th 2007 and December 17th 2008 (Kraus et al., 2013; Rückerl et al., 2014). In the framework of this study, we measured personal exposure to UFP among 112 participants, as well as UFP levels at a fixed monitoring station located at the urban background of Augsburg. Each participant also filled in time-activity diaries which provided an excellent opportunity for an exposure study.

The aim of the analysis is to assess the personal exposure to UFP during individual day-time activities and to investigate the impact of different microenvironments i.e. locations or human activities on personal UFP concentrations.

Methods

Study overview

In this study, we measured personal exposure to UFP among 112 participants within a period of nearly two years between March 19th 2007 and December 17th 2008. Each participant was invited to participate in the exposure measurements scheduled every four to six weeks on the same weekday. The measurement was conducted on average 21.1% in the spring, 32.3% in summer, 28.5% in autumn and 18.1% in winter. Participants were recruited from the follow-up examination of the KORA (Cooperative Health Research in the Region of Augsburg) survey (Holle et al., 2005) and were between 32 and 82 years old, with a mean age of 61.7 years. Out of 71 male and 41 female participants, 41 were employed. They were equipped with a bag containing portable devices and were asked to keep a diary for recording their activities and whereabouts whenever their activities or locations changed. All measurements started in the morning at the KORA study center located in the city center of Augsburg. Each measurement lasted about 5 to 6 h. During this time, the participants were able to follow either their normal daily routines or to choose any other activities they liked within the Augsburg area. At the end of the measurement, the participants went back to the KORA study center where the study nurses reviewed the diary with the participants and downloaded the data from the instruments.

Measurements

We measured personal exposures to PNC with three portable, real-time Condensation Particle Counters (CPC, model 3007, TSI Inc., Shoreview, MN, USA). It measures PNC in the size range of 10 nm to 1 μm in diameter. The zero point was checked on CPC 3007 before and after each measurement by applying a High Efficiency Particulate Air (HEPA) filter. Personally measured PNC were all obtained in 5-second resolution. One-minute averages were calculated if at least two thirds of the values in a 1-minute segment were available. CPC 3007 is sensitive to tilt as the optical chamber may be flooded by the 2-Propanol and the instrument may shut down and stop collecting data. There were 45 measurements with personal PNC data totally missing mainly due to tilt of CPC 3007. These measurements were not included in the study.

In addition, PNC was measured simultaneously with CPC 3025 (TSI Inc, USA, measuring particles 3–2000 nm) at an urban background measurement station. The monitoring site is located at an urban background setting south of the city centre of Augsburg with the nearest busy street about 100 m away. Details regarding the location of the measurement site can be found in Birmili et al. (2010) and Pitz et al. (2008). As shown by Cyrys et al. (2008) this measurement station could be considered representative for urban background PNC in Augsburg, where most of the study participants are living. Stationary PNC was measured in 1-minute resolution.

The CPC 3007s were serviced before the start of the study and during the measurement period by the manufacturer. Intercomparisons between all portable CPCs and a quality assured Twin Differential Mobility Particle Sizer (TDMPS, measuring particles 3–800 nm) at the measurement station were conducted at regular intervals. Stationary PNC data were also corrected by the TDMPS, which allows a direct comparison between personal and stationary PNC. A detailed description of the comparisons is provided in the Supplementary material.

Air temperature and relative humidity were recorded by data loggers (model Tinytalk 2, Gemini Data Loggers Ltd., Chichester, UK) on a 5-second resolution. One-minute averages were calculated if at least two thirds of the values in a 1-minute segment were available.

Data preparation

A structured/open-ended participant diary (paper-and-pencil diary) was used to collect information on the activities, locations, and transport modes that the participants were using during the measurement period. We were also interested in whether the participants were in a smoking environment or were physically active. Whenever their location or activity changed, the participants were asked to indicate it in the diary. The diary was reviewed by the nurses right after each measurement. In the following data management process, different categorical variables were built. Besides describing whereabouts, transport modes, household duties and passive smoking, a variable named "activity pattern" was created to examine the exposure to UFP under some common daily activities. It comprises the following nine categories: (1) indoors, no exact activity (the participant did not clearly indicate their activities); (2) indoors, with dust lifting activities (e.g., making the bed and dusting); (3) indoors, with water vapor (related to activities producing water vapor such as ironing, coffee making, and cooking etc.); (4) indoors, without dust lifting; (5) shopping (e.g., in the supermarket