Atmospheric Environment 106 (2015) 458–466

Contents lists available at ScienceDirect

Atmospheric Environment

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

Contribution of indoor-generated particles to residential exposure

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HIGHLIGHTS

- Several high time resolution instruments were operated for seven days in 22 homes.
- Concentrations above 10^4 cm^{-3} almost only occur during active periods of occupancy.
- Known and unknown indoor activities were 86% of the total integrated daily residential exposure.
- Source strengths of specific activities ranged from $1.6 \cdot 10^{12}$ to $4.5 \cdot 10^{12}$ min^{-1}.
- Correlation between UFP and mass conc of soot in total dust was on average 56%.

ARTICLE INFO

Article history:
Received 4 April 2014
Received in revised form
18 July 2014
Accepted 30 July 2014
Available online 1 August 2014

Keywords:
Ultrafine particles
Indoor sources
Exposure
Indoor measurements
Soot
Source strength

ABSTRACT

The majority of airborne particles in residences, when expressed as number concentrations, are generated by the residents themselves, through combustion/thermal related activities. These particles have a considerably smaller diameter than 2.5 μm and, due to the combination of their small size, chemical composition (e.g. soot) and intermittently very high concentrations, should be regarded as having potential to cause adverse health effects.

In this study, time resolved airborne particle measurements were conducted for seven consecutive days in 22 randomly selected homes in the urban area of Lund in southern Sweden. The main purpose of the study was to analyze the influence of human activities on the concentration of particles in indoor air. Focus was on number concentrations of particles with diameters <300 nm generated by indoor activities, and how these contribute to the integrated daily residential exposure. Correlations between these particles and soot mass concentration in total dust were also investigated.

It was found that candle burning and activities related to cooking (using a frying pan, oven, toaster, and their combinations) were the major particle sources.

The frequency of occurrence of a given concentration indoors and outdoors was compared for ultrafine particles. Indoor data was sorted into non-occupancy and occupancy time, and the occupancy time was further divided into non-activity and activity influenced time. It was found that high levels (above 10^4 cm^{-3}) indoors mainly occur during active periods of occupancy, while the concentration during non-activity influenced time differs very little from non-occupancy time.

Total integrated daily residential exposure of ultrafine particles was calculated for 22 homes, the contribution from known activities was 66%, from unknown activities 20%, and from background/non-activity 14%.

The collected data also allowed for estimates of particle source strengths for specific activities, and for some activities it was possible to estimate correlations between the number concentration of ultrafine particles and the mass concentration of soot in total dust in 10 homes. Particle source strengths (for 7 specific activities) ranged from $1.6 \cdot 10^{12}$ to $4.5 \cdot 10^{12}$ min^{-1}.

The correlation between ultrafine particles and mass concentration of soot in total dust varied between 0.37 and 0.85, with an average of 0.56 (Pearson correlation coefficient).

This study clearly shows that due to the importance of indoor sources, residential exposure to ultrafine particles cannot be characterized by ambient measurements alone.

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http://dx.doi.org/10.1016/j.atmosenv.2014.07.053

1. Introduction

The main aims of this study were to analyze the influence of human activities on particles in indoor air, with focus on determining the differences between residential and outdoor number concentrations of ultrafine particles; to estimate contribution of indoor sources to integrated daily residential exposure; and to check correlation between mass concentration of soot in total dust and number concentration of ultrafine particles in residences in general as well as during different activities.

In the industrialized part of the world, we spend approximately 65% of our lives in our homes (Leech et al., 2002; Brasche and Bischof, 2005). Hereby we are subjected to various indoor-generated airborne particles as well as to background particles originating from outdoors. It is widely known that outdoor particles contribute to the indoor aerosol concentration levels, and in epidemiology, exposure is often determined based on the outdoor particle concentration. However the outdoor contribution to the indoor aerosol size distribution is modified compared to the distribution outdoors, due to size-specific differences in penetration efficiency (Thatcher et al., 2003; Liu and Nazaroff, 2003; Nazaroff, 2004). Accumulation mode particles have the highest penetration (Nazaroff, 2004) since these particles have a rather low diffusivity and are too small to be significantly affected by sedimentation or impaction. To a far greater extent than outdoor generated particles, the particle concentration and size distribution indoors are dominated by particles generated by the residents' activities. For naturally ventilated buildings, Morawska and Salthammer (2003) summarized indoor/outdoor ratios (I/O) for PM_{10} and $PM_{2.5}$ (particulate matter smaller than 10 μm and 2.5 μm, respectively) from 0.5 to 0.98 and 0.54 to 1.08, respectively, in the absence of indoor sources. However, when indoor sources were present, I/O ratios for PM_{10} and $PM_{2.5}$ ranged from 1.14 to 3.91 and 1 to 2.4, respectively, which stress the significance of indoor source contributions (Morawska et al., 2013).

It has been shown (Hussein et al., 2006; Wallace, 2006; Turpin et al., 2007 Wierzbicka, 2008) that the major sources contributing to indoor air concentrations are combustion related or related to thermal processes (e.g. cooking, smoking and candle burning) and electric appliances. Peak number concentrations from cooking have been found to be higher than reported outdoor peak concentrations (Dennekamp et al., 2001; He et al., 2004; Wan et al., 2011), by at least an order of magnitude. It is well known that combustion generated particles generally are considerably smaller than 2.5 μm, often smaller than 300 nm (Géhin et al., 2008), which justifies that number concentration of ultrafine particles would be a more relevant metric than mass concentration to determine residential exposure to combustion-related particles. Ultrafine (<100 nm) particles are of special interest from a health perspective, since they, due to their small size, can penetrate deep into the respiratory system and cause inflammatory effects (Long et al., 2001). Particles <100 nm also have a higher deposition rate in both the upper airways and the alveolar tract, which in itself can cause adverse effects (Hirano, 2009; Araujo et al., 2008; Oberdörster et al., 2005). Studies (Wang et al., 2009; Kennedy, 2007) have shown that the number concentration of ultrafine particles induce more adverse health effects than the mass concentrations of PM_{10} and $PM_{2.5}$ (to which the ultrafine particles are a poor contributor).

Soot (black or elemental carbon), is a primary component in several emissions known to affect human health indoors, such as wood burning, cigarette smoke, cooking with poor ventilation but also e.g. diesel exhaust, which can infiltrate the building from outdoors. Soot particles are often carriers of elements such as polycyclic aromatic hydrocarbons (PAH) (Pagels et al., 2009), but are of health interest also due to their small size and because of the large surface area available for adsorption of gases and other particle phase toxins. Since the major sources of ultrafine particles indoors have been shown to be combustion/thermal related, it is likely that soot is a major component of this aerosol and it is therefore of interest to monitor the soot concentration indoors. The importance of the potential health effects of indoor generated soot has been pointed out by e.g. Buonanno et al. (2013). Monitoring soot concentrations in parallel to ultrafine particle number concentrations allows determination of their correlation, and, hypothetically, as a consequence gives a possibility of predicting the soot component on the basis of monitoring ultrafine particles.

2. Methods

For seven consecutive days, time resolved stationary air measurements were conducted in 22 randomly selected homes in the urban area of Lund in southern Sweden. The measurements were conducted during wintertime. Home characteristics, including ventilation type, air exchange rate, volume, indoor temperature and RH, and instruments used are summarized in Table 1. Initially measurements were conducted in 39 homes; however mainly due to technical reasons (see data processing for details) results from only 22 homes were included in this article.

The habitants filled in detailed activity log books specifying conducted activities and made notes of when they were present or absent in the dwelling.

2.1. Measurements and instrumentation

Number concentrations, and mean diameter, of ultrafine particles were monitored with Mini Diffusion Size Classifier (DiSC) (University of Applied Sciences, Windisch, CH) or Nanotracer (Philips Aerasense). These instruments are based on an electrical measurement technique, with a diffusion charger, by which the average acquired charge of a particle depends on its size, followed by a diffusion stage. This stage consists of a stainless steel grid, where the particles will deposit due to diffusion, connected to electrometers that detect the current carried by the deposited particles. The current is dependent on flow rate, number concentration and particle size. Both instruments have an upper size limit of 300 nm. Since over 80% of particles measured by these two instruments were below 120 nm, and the majority were below 100 nm, with a minimal contribution of particles between 100 and 300 nm, the measurements of the Nanotracer and DiSC can be considered as good approximations of ultrafine particles (number concentrations measured with Nanotracer and DiSC will in this article be entitled ultrafine particles). The accuracy of both the Nanotracer and DiSC are ±30% (Asbach et al., 2012).

The measurements of particles larger than 0.3 μm were performed by an optical particle counter, Indoor Air Quality monitor (IAQ3016, Lighthouse). Particles passing a laser beam inside the instrument scatter light. A photo detector converts the scattered light to an electrical pulse. For a single particle, the light intensity of the generated pulse depends on the size, refractive index and color of the particle. According to a reference aerosol of monodisperse PSL particles, the pulse height is transferred to six particle size intervals. The pulse intensity thresholds in the IAQ3016 correspond to particle sizes of 0.3, 0.5, 1.0, 2.5, 5.0 and 10 μm.

Black carbon (soot) levels were monitored by microAeth (AE51, Magee Scientific) in 10 of the homes, to investigate the correlation between soot measurements and indoor (particularly combustion