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Published in:
[Host publication title missing]

2009

Citation for published version (APA):
Fine and Ultrafine Particles in a Supermarket in Sweden

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SUMMARY
The aim of this study was to gain information about fine and ultrafine particles (diameter smaller than 2.5 μm and 100 nm, respectively) loads and their characteristics in a supermarket in Sweden. Assessed particles’ characteristics are: number and mass concentrations and number size distributions. In the studied supermarket, an indoor source of ultrafine particles was observed, most probably due to local emissions of terpenes (from washing powders, cleaning products and air fresheners), which in the presence of ozone from outdoors form particles due to gas-to-particle conversions.

KEYWORDS
Airborne particles, ultrafine particles, terpenes-ozone reactions, supermarket, indoor environments

INTRODUCTION
In numerous epidemiological studies fine and ultrafine particles have been associated with adverse health effects (Pope et al., 2002; Brook et al, 2004). Epidemiological associations between airborne particles and various health outcomes are based on ambient measurements. Even though people on average spend about 90% of their time inside buildings (Leech et al., 2002), the concentration and characteristics of particles in indoor environments are not well known. Indoor particles originate from indoor sources, penetrate from ambient air or are formed indoors as secondary aerosol (e.g. chemical reactions of ozone and terpenes). Studies that investigate continuously changes in particle mass and number in different indoor environments are limited. Scattered information on particle number concentrations and emission factors for indoor sources can be found in several publications, but the available data is far from being concise and complete (Ogulei et al., 2006; Hussein et al., 2006; Morawska et al., 2003; Wallace, 2006; Dennekamp et al., 2001). There is a great need to investigate particle loads and characteristics in various indoor environments. The aims of the study were to assess particle characteristics in a supermarket by means of number and mass concentrations and number size distributions, and to identify indoor sources of particles.

METHODS
Measurements were performed continuously for seven consecutive days in a supermarket in the area with washing powders, cleaning products and air fresheners. Measurements were recorded continuously with the following instruments: an aerodynamic particle sizer (TSI APS 3321), a scanning mobility particle sizer (SMPS), (consisting of differential mobility analyzer (TSI DMA) and condensation particle counter (TSI CPC 3010)), and two DustTracks
Number concentration and size distribution in the range 15 nm to 20 μm were recorded by SMPS and APS. A valve was incorporated into the SMPS sampling system, which enabled alternate measurements of the indoor and supplied ventilation air. Stainless steel tubing with an inner diameter of 0.535 cm and lengths of 1 and 5 m was used to connect the valve to the indoor and supplied air, respectively. During sampling of the supplied ventilation air, the tubing was inserted at the outlet supplying the air, approximately 2 cm into the centre of the duct. A correction for diffusion losses in the DMA and tubing (Karlsson and Martinsson, 2003) was applied for all particles.

Mechanical ventilation supplies heated and filtered air and operates on weekdays between 4:30 and 22:00 and on weekends between 6:30 and 20:30. In the ventilation system there is no recirculation and no dehumidification, supplied air in 100% is composed of filtered outside air, the filter used is a glass-fibre F7 class filter (EN 779: 2002).

The two DustTracks were used for indirect mass concentration measurements of particles smaller than 2.5 μm (PM2.5) to ~0.1 μm, they enabled simultaneous measurement of indoor and outdoor mass concentration. Time schedules and discussions with employees were used to identify various activities which have led to identification of indoor sources of particles.

RESULTS AND DISCUSSION

Series of two day measurements of the total submicrometer particle number concentration (PNC) and number geometric mean diameter (GMD) inside and in the supplied ventilation air in the supermarket are plotted in Figure 1. Submicrometer PNC inside the supermarket (median 2500, min 1100, max 11000) particles cm⁻³ was higher than in the supplied ventilation air (median 850, min 200, max 4000) particles cm⁻³, indicating an indoor source of particles (Wierzbicka et al., 2009). The median GMD inside the supermarket 42 nm was about 20 nm smaller than in the supplied ventilation air. Presence of nucleation mode particles is likely due to local emissions of terpenes (from washing powders, cleaning products and air fresheners), which in the presence of ozone from outdoors (supplied in ventilation air) form particles due to gas-to-particle conversions (Wainman et al., 2000, Sarwar and Corsi, 2007, Leungsakul et al., 2005). The observed concentrations are not high, but considering the high air exchange rate (AER ~ 10 h⁻¹) and supply of air with very low particle loads, these contributions seem to be significant. Ozone levels measured outdoors were about 30 ppb.

Differences were observed between the time when the ventilation was operating and when it was switched off.

**Ventilation on.** Submicrometer PNC inside the supermarket fluctuates, these fluctuations can be due to: incomplete mixing, varying AER due to door opening and draughts, changing source strengths of terpenes (elevated when shelve stocking took place due to movement and opening packages), or influence from other sources of fine particles (Wierzbicka et al., 2009).

**Ventilation off.** When the ventilation was switched off, the removal due to high AER and supply of heated air with low particle loads were eliminated. This might have resulted in local accumulation of terpenes and temperature decrease. Thus, the observed sudden increase in PNC with a number GMD of about 30 nm (Figure 1) can be explained by nucleation via gas-to-particle conversions from terpenes in the presence of ozone. After reaching the peak (within 30 minutes to an hour after ventilation was switched off) a decrease in the PNC was seen, probably due to dilution and spreading within the air in the supermarket and losses to surfaces. Possibly ozone concentrations were depleted by then due to chemical reactions. At
the same time the number GMD increase was observed. This GMD increase could be due to organic vapour condensation, which is in line with Weschler and Shields’ (2000) findings that reduced AER results in more time available for gas phase chemistry and particle growth. The dominant process for the overall PNC decrease was due to diffusional wall/surface deposition, which is an efficient removal process for particles smaller than 100 nm. Large available surface areas (shelves) support this explanation, as the deposition is also dependant on surface area.

Figure 1. Total submicrometer PNC (15–700 nm) and number GMD inside and in the supplied ventilation air in the supermarket example of two days measurements.

Presence of other sources of ultrafine particles during opening hours cannot be excluded although observed different patterns during opening hours (ventilation on) in comparison to night time (ventilation off) confirm likelihood of terpenes-ozone reactions as a source of observed ultrafine particles.

Median approximation of PM2.5 mass concentrations (DustTrak readings) in the supermarket accounted for 6 μg m⁻³ (min 5, max 20 μg m⁻³).

CONCLUSIONS
In the supermarket, an indoor source of ultrafine particles was observed, most probably due to local emissions of terpenes (from washing powders, cleaning products and air fresheners), which in the presence of ozone from outdoors form particles due to gas-to-particle conversions. More data is needed to determine typical levels of ultrafine particles in supermarkets. The results indicate how important adequate ventilation is in places where products containing terpenes are kept in order to minimise the exposure to ultrafine particles.

ACKNOWLEDGEMENT
This work has been supported by the Swedish Research Council FORMAS and the Development Fund of the Swedish Construction Industry (SBUF).
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