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ON-LINE CHARACTERISATION PARTICLE EMISSIONS FROM INDOOR COMBUSTION SOURCES — CHAMBER STUDIES OF PARTICLE VOLATILITY AND HYGROSCOPICITY

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INTRODUCTION

People in industrialised countries spend ~ 90% of their time indoors and often indoor combustion sources give a significant contribution to the personal exposure of e.g. PM$_{2.5}$ or the number of ultrafine particles. Emissions from indoor combustion sources to a large extent consist of organic material. Semi-volatile components may be continuously exchanged between the gas and the particle phase. By varying the dilution thermodynamics e.g. of diesel exhaust, the distribution between the particle and the gas phase can be strongly altered, it is therefore likely that many organic combustion aerosols are produced far from particle-gas phase equilibrium. Conventional filter measurements are prone to gas-phase adsorption and evaporation, which makes it difficult to separate the particle phase. Also off-line methods have poor time-resolution and give little information about evaporation and condensation processes during aerosol ageing. An approach to avoid these limitations is to use on-line aerosol spectrometers and thereby analyse the particles while airborne. The volatility-TDMA in principle gives a measure of the vapour pressure of particle constituents evaporated in the thermodesorber in the instrument. The vapour pressure is the property determining the gas to particle partitioning. It is expected that the volatile fractions evaporated with the V-TDMA for an aerosol far from equilibrium changes over time as the aerosol is aged. Controlled chamber studies are a suitable way to study such effects. Measurements of uptake of water-vapour at high RH can be studied with the Hygroscopic-TDMA. These measurements can be used to predict respiratory deposition and together with the V-TDMA to give size-differentiated insight of the chemical composition and mixing status.

The aim of this work is to use the V-TDMA method during chamber measurements of cigarette and candle aerosols to a) Get insight of the chemical composition b) Study whether the cigarette smoke is in equilibrium at typical indoor concentrations and c) Determine the mixing status of the aerosols. In a few experiments we combined the volatility and hygroscopic methods to gain insight of the water-solubility of constituents with low vapour pressures (Johnson et al. 2004).

METHODS

Indoor combustion sources were generated in a 25 m$^3$ air-tight steel-chamber in the aerosol laboratory at EAT in Lund. The RH was controlled to 25%. A fan was used to ensure mixing in the chamber. Side-stream cigarette smoke was generated from a commercial brand using 2 s long, 30 ml puffs every 30 s during two minutes. The aerosol was stored in the chamber for 3 h at a ventilation rate of 0.5 h$^{-1}$. A commercially available paraffin candle, was continuously burning and studied for 3 h at a ventilation rate of 1 h$^{-1}$.

In the V-TDMA a monodisperse aerosol fraction is selected with DMA1. The particles are conditioned in a thermodesorber (TD) with known temperature profile and a residence-time of around 1 s (Dahl and Pagels 2003). The aerosol is then cooled in a steel-tube and the size shift is determined in DMA2. From the size-shift the evaporated volume-fraction can be assessed assuming spherical particles. In a few experiments we added a third DMA, which was held at RH=93%, after the TD in the V-TDMA. The hygroscopic growth factor in this case is defined by the differences between DMA2 and 3. During
the measurements a conventional SMPS-system (model 3934, TSI Inc., US) was used to study the time-evolution of the particle size-distribution (10-450 nm) in the chamber.

RESULTS AND DISCUSSION

The initial concentration of the cigarette smoke was around 20,000 particles per cm³, with a mass concentration of around 80 µg/m³. The particle size distribution was unimodal and the geometric mean diameter around 120 nm. The candle size-distribution consisted of overlapping modes. A nucleation mode with concentrations higher than 500,000 cm⁻³, with a significant fraction below the detection limit of the SMPS-system and an accumulation mode of around 200 nm with the majority of the particle mass.

Figure 1 shows that the cigarette smoke particles are relatively volatile, as expected for organic constituents from incomplete combustion. The nucleation mode candle particles distinctly decreases their diameters at around 400° C, while the accumulation mode particles are almost stable within up to 500° C, as expected for elemental carbon. In figure 2 it is clearly shown that the volatility decreases when the cigarette smoke ages in the chamber. Almost 50% less material volatilised at 60 °C in the thermo-desorber after 140 min, compared to after 20 min. The difference represents components, which had evaporated during ageing in the chamber. This shows that freshly produced side-stream cigarette smoke is not in equilibrium. In figure 3 the hygroscopic diameter growth (from dry to RH=93%) is shown for the two candle particle diameters as a function of the temperature in the TD. The nucleation mode particles have high water uptake, the growth factor decreases, after particle shrinkage detected in the VTDMA. Further investigation is required to understand the chemical composition and formation mechanisms of these particles. The accumulation mode particles are almost hydrophobic as expected for elemental carbon.

CONCLUSIONS

In this work we have shown how chamber studies with combined volatility and hygroscopic TDMA methods can be used to draw conclusions of the chemical composition and mixing status of indoor combustion aerosols. However nucleation mode candle particles with high hygroscopic growth factors needs to be further studied. Freshly produced side-stream cigarette smoke is not in particle-gas phase equilibrium. A significant fraction of the more volatile components evaporates during ageing.

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