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Secondary Aerosol Formation Initiated by Chemical Reactions in Indoor Air – Chamber Studies with Mixtures of Ozone and Emissions from Indoor Terpene Sources

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INTRODUCTION

Terpenes are often added to household products (air fresheners, cleaning products etc.) to improve the fragrance characteristics (e.g. lemon and pine tree smells). Chemical reactions of terpenes with ozone initiate reaction schemes which generate low vapour pressure reaction products (Secondary Organic Aerosol; SOA), which may either condense on the present particle population or form new particles by homogeneous nucleation. Such processes can give a significant contribution to the indoor PM concentration. Reaction products from terpene-ozone mixtures have been shown to be potent irritants in a mouse-bio assay and may hence be of relevance in adverse health effect studies (Wolkoff et al. 1999). Elevated O_3 concentrations indoors may occur during combinations of high outdoor concentrations and high air exchange rates or from indoor sources such as "air cleaners" or laser photo copiers. While several studies have focused on the chemical composition of precursors and reaction products from indoor terpene sources, little attention has been paid to the particle formation and transformation mechanisms. The aim of this work was to determine particle emission levels from selected indoor terpene sources when combined with ozone. Another aim was to investigate under what circumstances particle formation from homogeneous nucleation occurs in indoor air.

METHODS

Common household products which contain or could be expected to contain terpenes (e.g. d-Limonene or α-Pinene) were purchased in local retail-shops. Terpene sources included a plug-in air-freshener using small electrical heaters to volatilize fragrance chemicals, pine-scented cleaning products, spray based air fresheners, citrus fruits and pure terpenes. The experiments were performed in a large air-tight stainless steel room (3*3*2.4 m). The air exchange rate was set to 2.0 h^{-1} and the relative humidity was set to 10, 40 and 80% in the chamber. Conditioned supply air passed a system of HEPA particle filters and active carbon filters before entering the chamber. An ozone generator (model AM-3000, Ozone Tech Systems) was used to generate 40 ppb O_3 in the chamber (in the absence of terpene sources). The O_3 concentration was determined using a UV O_3 photometer (model 49, Thermo Environmental Instruments Inc.). The terpene sources were either episodic or continuous. For episodic sources O_3 was introduced to the chamber 5 min after terpene emissions ceased. Continuous terpene sources were activated throughout the whole experiment, in these cases O_3 was introduced to the chamber for 2 h starting 1 h after the terpene source had been activated. Terpenes and other potential particle forming pre-cursors were sampled using triplicate Tenax sorbents, both before and after addition of O_3. Samples taken after O_3 addition first passed an O_3 scrubber as described by Fick et al. (2001). The samples were analysed using GC-FID and GC-MS. The background indoor particle concentration was simulated by adding NaCl particles generated using a nebuliser. Particle number concentrations and size distributions were characterised using an Electrical Mobility Spectrometer (SMPS 3934). The SOA was separated according to volatility using a Volatility Tandem Differential Mobility Analyzer (V-TDMA).

RESULTS

In experiments without any pre-existing aerosol, several of the studied household products generated new particles through homogeneous nucleation even at the relatively low ozone concentrations used in this study.

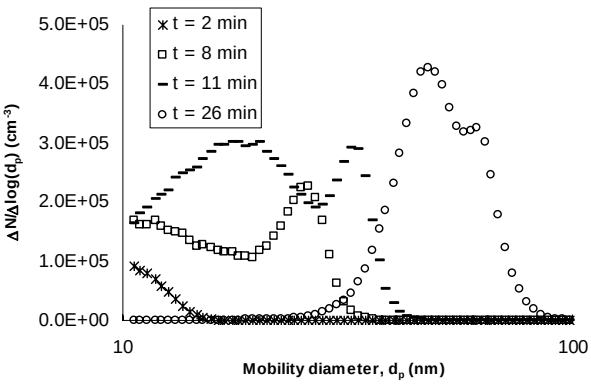

Figure 1. Evaluation of particle number size distribution in an experiment where a plug-in air-freshener was used with a lemon fragrance oil, t refers to the time since ozone was introduced to the chamber.

Figure 1 shows the evolution of the particle size distribution over time for an experiment where a citrus fragrance oil was used in the plug in air-freshener. The homogeneous nucleation stopped after about 15 minutes and further gas-to-particle conversion occurred through condensation, resulting in increased particle sizes and a decreased geometric standard deviation. Assuming spherical unit density particles this corresponded to a mass concentration of 10 $\mu g/m^3$ after 30 min of ozone exposure.

We found that the NaCl aerosol is stable up to 400°C in V-TDMA experiments, while more than 99% of the volume concentration of the SOA reaction products was volatilised at 300° C. Thus the SOA can be effectively separated and quantified also in mixed particles containing simulated background aerosol.

CONCLUSIONS

Chemical reactions between organic emissions from typical indoor terpene sources and ozone can give a substantial contribution to the indoor PM exposure, especially of ultrafine particles. The secondary organic aerosol is mainly volatile below 300° C and can be effectively separated from a NaCl model of the indoor background aerosol.

Keywords: *Terpenes, Ozone, Chemical Reactions, Volatility*

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