Generation of Nano Size Particles from Limonene/Ozone Reactions for Controlled Human Exposures in a Chamber

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Generation of nano size particles from limonene/ozone reactions, for controlled human exposures in a chamber

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An aim of this study has been to develop a method to generate a stable, reproducable terpene/ozone aerosol and deliver it to the exposure chamber while the aerosol still is fresh. An additional aim has been to study the detailed and complex reaction chemistry. The aerosol generated in this study is utilized for exposure of healthy human test subjects in a controlled chamber setting, during and after which various medical responses are being investigated.

Limonene is a common terpene constituent in many consumer products used in indoor settings. On reaction with ambient ozone the oxidation species rapidly form condensed-phase products, which significantly elevate the indoor levels of ultra fine aerosol particles. The size range and the complex chemical composition of the various reaction products and intermediaries suggest that they are likely to cause adverse effects human health. (Rohr et al 2003).

Terpene vapor is generated continuously by passing pure nitrogen (2 lpm) through a glass bottle containing 6.7 ml of commercial essential oil (lemon oil, oleum citri, Interlam ab), consisting of 60-95 % *d*-limonene. The VOC level is monitored with a photoinization detector (Photovac 2020) at the outlet of the glass bottle. Ozone is generated by a spark discharge generator using filtered dry air, and is added to the ventilation air flow just downstream the inlet for terpene vapours. The ventilation air passes HEPA and active carbon filter before vapor flow and ozone are added, just prior to entering the 21.6 m^3 stainless steel exposure chamber.

The behaviour of pure limonene in the exposure chamber is studied in separate experiments. During the exposures particle mass concentration in the chamber is monitored with a Tapered Element Oscillating Microbalance (TEOM, Rupprecht & Patashnic Co inc.) and particle number concentration and size distribution by a Scanning Mobility Particle Sizer system (consisting of a CPC 3010, TSI Inc and a long column Hauke DMA). An aerosol mass spectrometer (AMS, Aerodyne research inc.) is used for investigating the oxidation states and the elemental composition, with regards to carbon, hydrogen and oxygen, of the reactants.

Upon reaction with ozone there is an immediate burst of nucleation particles (5-25 nm), which due to condensation and coagulation processes grow in size to 150 nm (air exchange rate of 4.5 h^{-1}). In 2-3 hours the system reaches steady state with mainly a single mode of particles in the mean diameter range of 95-105 nm. Particles are slowly generated by nucleation at steady state. Prior to reacting with limonene vapor the concentration of ozone is 40 ppb. The ozone is almost completely consumed by the terpenes, leaving a residual ozone level in the exposure chamber of 5-8 ppb. Since ozone itself is an airway irritant, a low ozone level inside the chamber is of importance in the exposure studies. After reaching steady state, the generation system delivers a stable aerosol with regards to particle size, number and mass concentration, as shown in figure 1.

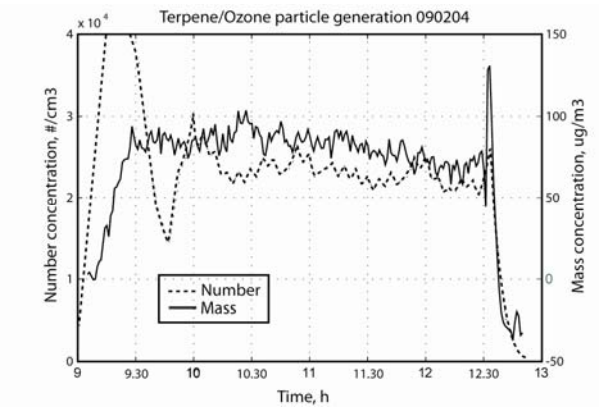

Figure 1. Number and mass concentration during an exposure event. At 12.30 the exposure chamber is evacuated.

The generation method developed resulted in a stable and reproducible terpene/ozone aerosol, and has been successfully used in human exposure studies.

Rohr A C, Weschler C J, Koutrakis P, Spengler J D, *Generation and Quantification of Ultrafine Particles through Terpeen/Ozone Reaction in a Chamber Setting*, Aerosol Science and Technology 37:65-78, 2003

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