

# Formation and growth of indoor air aerosol particles as a result of D-limonene oxidation

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## Abstract

Oxidation of D-limonene, which is a common monoterpene, can lead to new aerosol particle formation in indoor environments. Thus, products containing D-limonene, such as citrus fruits, air refresheners, household cleaning agents, and waxes, can act as indoor air aerosol particle sources. We released D-limonene into the room air by peeling oranges and measured the concentration of aerosol particles of three different size ranges. In addition, we measured the concentration of D-limonene, the oxidant, and the concentration of ozone, the oxidizing gas. Based on the measurements we calculated the growth rate of the small aerosol particles, which were 3–10 nm in diameter, to be about  $6300 \text{ nm h}^{-1}$ , and the losses of the aerosol particles that were due to the coagulation and condensation processes. From these, we further approximated the concentration of the condensable vapour and its source rate and then calculated the formation rate of the small aerosol particles. For the final result, we calculated the nucleation rate and the maximum number of molecules in a critical cluster. The nucleation rate was in the order of  $10^5 \text{ cm}^{-3} \text{ s}^{-1}$  and the number of molecules in a critical-sized cluster became 1.2. The results were in agreement with the activation theory.

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## 1. Introduction

In the absence of aerosol particle sources the aerosol particle concentration indoors typically follows the pattern of outdoor concentration (Raunemaa et al., 1989; Koponen et al., 2001; Hussein et al., 2004). In addition, various indoor

activities such as cooking, dusting, vacuum cleaning, or heating can increase the aerosol particle number and mass concentration indoors (He et al., 2004; Hussein et al., 2005, 2006). The indoor aerosol particles thus come from primary as well as secondary sources with varying source strengths. The strongest reported number-based indoor air aerosol particle sources include cooking, especially frying or grilling, smoking, heating, and burning candles (He et al., 2004; Afshari et al., 2005).

The gas and the particle phase reactions of the terpenes and the oxidants such as ozone in an

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indoor environment can result in high aerosol particle concentrations as shown by e.g. Wainman et al. (2000) and Wescher and Shields (1999). The D-limonene is a monoterpene which is commonly present in indoor air (Brown et al., 1994). In addition to citrus fruits, products such as air fresheners, waxes, and cleaning agents often contain D-limonene (Liu et al., 2004; Nazaroff and Weschler, 2004; Wainman et al., 2000). Hoffmann et al. (1997) reported the high mass-based aerosol yield of D-limonene indoors in the presence of oxidants. In addition, chamber studies conducted by Rohr et al. (2003) indicate a significant increase in ultrafine aerosol particle number concentrations as a result of D-limonene oxidation. Also Nøjgaard et al. (2006) reported the high aerosol number concentration during ozonolysis of D-limonene. These aerosol particles are formed through nucleation and condensation processes. Study by Bonn and Moortgat (2002) shows that reactions with ozone have the biggest impact in nucleation of monoterpenes.

The formation of new aerosol particles through homogeneous or heterogeneous nucleation in the atmosphere affects the earth's radiative balance and is thus an important phenomenon. Recent knowledge on the atmospheric nucleation is summarized by Kulmala et al. (2004). The role of the organic vapours in the aerosol particle formation and growth processes under typically prevailing environmental conditions is a key issue. It is suggested that organic species contribute to the growth of nucleated clusters (etc. O'Dowd et al., 2002; Anttila and Kerminen, 2003). Koch et al. (2000) studied aerosol particle formation as a result of the ozonolysis of several monoterpenes in a laboratory experiment. Their results also showed the high aerosol yield of this process. They concluded that in favourable conditions the monoterpene oxidation can have an effect on atmospheric aerosol particle formation. Despite the intensive study of the subject, the mechanism of aerosol particle formation is yet unsolved.

Currently, knowledge of monoterpene oxidation under realistic environmental conditions is incomplete. The ultrafine aerosol particle production ability of monoterpenes in indoor air, the following health impacts, and the mechanism of outdoor aerosol particle production and its atmospheric relevance are of particular interest.

The first motivation for the study was the observation of exceptionally high aerosol particle production after peeling citrus fruits. As citrus

plants store large amounts of monoterpenes, mainly limonene, in their peels (see e.g. Högnadóttir and Rouseff, 2003; Moufida and Marzouk, 2003), peeling the fruits releases limonene into the air. In addition to aerosol particle number concentrations we measured the concentration of monoterpenes and ozone. Our aim is to increase knowledge of D-limonene containing products as potential indoor air ultrafine and fine aerosol particle sources as well as to clarify the mechanisms of new aerosol particle formation in natural environments.

## 2. Calculation method for vapour concentration and nucleation rate

The theory on the formation and growth of the aerosol particles as a result of the condensing and nucleating organic vapours is explained in more detail by Kulmala et al. (2001). Thus, we will only briefly go through the main steps used in our study.

Nucleation rate (Kerminen and Kulmala, 2002)

$$J_1 = J_3 \exp \left[ \frac{\eta}{d_{p1}} - \frac{\eta}{d_{p3}} \right] \quad (1)$$

is the rate at which the critical-sized clusters of approximately 1 nm vapourize. The  $J_3$  in Eq. (1) is the formation rate of the 3 nm-sized aerosol particles and term  $\eta$  is given by

$$\eta = \frac{\gamma C_S}{2\pi D G_R}. \quad (2)$$

Thus, the  $\eta$  depends on the particle growth rate  $G_R$ , diffusion coefficient  $D$ , factor  $\gamma$ , and the condensation sink

$$C_S = 4\pi D \sum_i \beta_i r_i N_i. \quad (3)$$

Now,  $\beta$  is the transitional correction factor given by Fuchs and Sutugin (1971) and  $r_i$  is the particle radius. Factor  $\gamma$ , which is given by (Kerminen and Kulmala, 2002)

$$\gamma = \gamma_0 \left[ \frac{d_{p1}}{1 \text{ nm}} \right]^{0.2} \left[ \frac{d_{p3}}{3 \text{ nm}} \right]^{0.075} \left[ \frac{T}{293 \text{ K}} \right]^{-0.75} \times \left[ \frac{\rho}{1000 \text{ kg m}^{-3}} \right]^{-0.33} \left[ \frac{d_{\text{mean}}}{150 \text{ nm}} \right]^{0.048}, \quad (4)$$

takes into account environmental conditions such as the initial nuclei size  $d_{p1}$ , the observed particle size  $d_{p3}$ , the temperature  $T$ , the nuclei density  $\rho$ , and the mean diameter of the pre-existing aerosol particle number size distribution  $d_{\text{mean}}$ . Factor  $\gamma_0$  ( $= 0.23 \text{ nm}^2 \text{ m}^2 \text{ h}^{-1}$ ) is a constant.

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