

On the connection between new particle formation rate, sulphuric acid and organic compounds in Hohenpeissenberg (Germany)

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Field measurements have shown that the coupling of the new particle formation rate and the sulphuric acid concentration can be described with power-law functions with exponent between one and two (Riipinen et al. 2007, Kuang et al. 2008). Here we analyse this coupling from two large data sets measured in rural southern Germany. We also examine the connection between the particle formation rate and concentrations of oxidation products of volatile organic compounds (VOC).

The data sets were measured at the Hohenpeissenberg Meteorological Observatory, operated by the German National Meteorological Service (DWD), the first in 1998-2000 during the Hohenpeissenberg Aerosol Formation Experiment (HAFEX, Birmili et al. 2003) and the second in 2007-2008 during the EUCAARI-campaign. The particle number size distributions were measured with Differential Mobility Particle Sizer (DMPS) during HAFEX and Neutral Air Ion Spectrometer (NAIS) during EUCAARI. The monoterpene and aromatic hydrocarbon concentrations were measured with Gas Chromatography ion-trap Mass Spectrometer (GC-MS), and the sulphuric acid concentration with Chemical Ionisation Mass Spectrometer (CIMS, Berresheim et al. 2000).

The particle formation rate J was calculated from the size distributions on the days on which a regional particle formation event was observed. J was compared to the sulphuric acid concentration $[H_2SO_4]$ in order to determine the daily nucleation coefficients A and K connecting J and $[H_2SO_4]$ in the activation theory ($J=A[H_2SO_4]$, Kulmala et al. 2006) and in the kinetic theory ($J=K[H_2SO_4]^2$). The momentary values of A and K ($A=J/[H_2SO_4]$ and $K=J/[H_2SO_4]^2$) were compared to meteorological variables, trace gas concentrations and estimated concentrations of VOC oxidation products.

In the HAFEX data set we found a clear correlation between the nucleation rate J and the sulphuric acid concentration to the power from one to two. Linear coupling between J and $[H_2SO_4]$ was slightly more frequent than squared. The medians of the daily nucleation coefficients were $1.8 \times 10^{-7} \text{ s}^{-1}$ for A and $3.3 \times 10^{-14} \text{ cm}^3 \text{ s}^{-1}$ for K . However, the daily

values of A and K varied within two orders of magnitude and even during one day the observed formation rates fluctuated around the rates predicted by the theories. Additionally, the daily kinetic coefficient correlated negatively with median $[H_2SO_4]$, although according to the kinetic theory they should be independent.

We observed a correlation between the momentary values of A and K and the oxidation product concentrations of VOCs originating from coniferous trees (see Fig. 1). This correlation could be the reason for the variations in both daily and momentary values of the nucleation coefficients. Our results suggest that both sulphuric acid and VOCs take part in atmospheric particle formation.

The results from the EUCAARI 2007-2008 campaign will be presented as well.

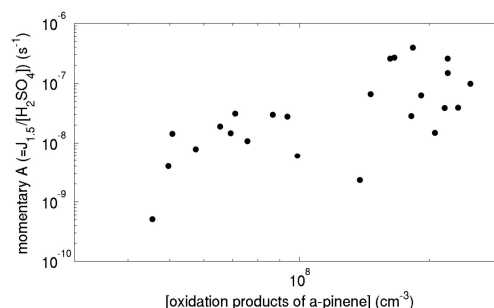


Figure 1. Momentary values of the activation coefficient A versus the estimated oxidation product concentration of α -pinene (HAFEX campaign).

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Birmili, W. et al. (2003). *Atmos. Chem. Phys.*, 3, 361-376.

Berresheim, H. et al. (2000). *Int. J. Mass Spectrom.*, 202, 91-109.

Kuang C. et al (2008). *J. Geophys. Res.*, 113, D10209.

Kulmala, M. et al. (2006). *Atmos. Chem. Phys.*, 6, 787-793.

Riipinen, I. et al. (2007). *Atmos. Chem. Phys.*, 7, 1899-1914.