

Dependence of α -pinene secondary organic aerosol formation on relative humidity and aerosol surface distribution

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Organic aerosol (OA) represents a major fraction (20-90%) of the submicron aerosol (Jimenez et al., 2009). Potential sources and processes leading to the formation of secondary organic aerosol (SOA) are still rather poorly understood, although SOA contributes significantly to the global OA burden. To determine parameters relevant to SOA yields, in particular volatility and degree of oxygenation, the formation and aging of particles has to be investigated more thoroughly.

Several laboratory studies suggest that aqueous phase SOA formation can be almost as important as gas phase oxidation of volatile organic compounds and their subsequent partitioning to the aerosol phase (Ervens et al., 2011).

In this study, the aerosol chemical composition as a function of the vacuum aerodynamic diameter (d_{va}) was investigated using an Aerodyne aerosol mass spectrometer (AMS) equipped with a $PM_{2.5}$ lens. In the PSI smog chamber, the aerosol precursor α -pinene was irradiated in the presence of low (~30%) or high (~80%) relative humidities (RH) and various NO_x concentrations. NH_4HSO_4 was used as a hygroscopic seed, providing a large water volume at high humidity. A wide diameter range, ~50 to 2500 nm, was covered by the particle phase instruments. The aging time in the chamber was related to the atmospheric oxidation time using OH exposures derived from the decay of an OH tracer injected into the chamber (Barmet et al., 2012).

For a similar NO_x concentration, higher SOA yields were observed at higher RH compared to lower RH. Fig. 1 shows mass distributions of organics, sulfate, ammonium, and nitrate, as well as organic mass fractions f_{44} (m/z 44 / total organics, a surrogate for the degree of oxygenation) and f_{43} (m/z 43 / total organics, a surrogate for less oxygenated compounds like aldehydes, ketones and alcohols) as a function of particle diameter. The data is averaged over the experiment period when aerosol aging dominates. The wet aerosol surface distribution shown in Fig. 1 was calculated using a volume-mixed growth factor and the measured RH ($72 \pm 1\%$). This particular experiment was conducted with 16 ppb α -pinene and 20 ppb NO_x .

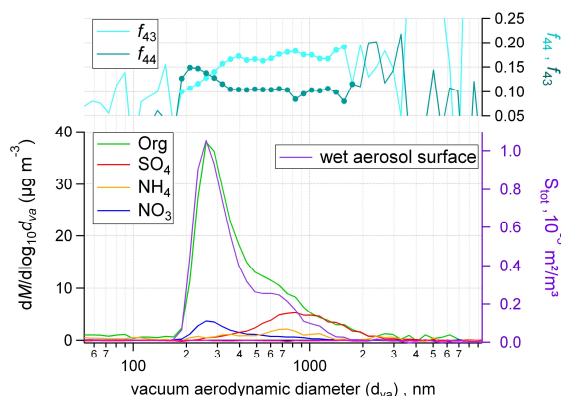


Figure 1. Averaged mass distribution of chemical species, calculated wet surface distribution and organic mass fractions f_{44} and f_{43} as a function of particle diameter. Dots (f_{44} , f_{43}) represent data when $d(org)/dlog_{10}d_{va}$ is above a threshold of $2\mu g \cdot m^{-3}$.

The organic mass distribution follows the wet surface distribution of the aerosol, suggesting a fast, surface dependent, SOA formation process. Secondly, the chemical composition is dependent on particle size, indicated by the increase of f_{44} with diameter. This could be due to the larger available water volume, favouring aqueous phase processes that lead to more highly oxygenated products like acids.

The effect of particle size will also be assessed by analysis of single particle mass spectra acquired using a light scattering module integrated with the AMS. The combined effects of varied relative humidity and NO_x concentration on SOA yields and composition will be investigated and discussed in this study.

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