The role of organic condensation on ultrafine particle growth during nucleation events

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New particles are introduced in the atmosphere by direct emission from a variety of sources and nucleation (in-situ formation). These ultrafine particles can grow to larger sizes and become cloud condensation nuclei (CCN) affecting cloud droplet concentrations, cloud reflectivity and lifetime. This indirect effect of aerosols on climate is thus perturbing the energy balance of the planet (Twomey, 1974; Albrecht, 1989).

In this work, we applied an updated box model version of the Dynamic Model for Aerosol Nucleation (Jung et al., 2006) in order to simulate the nucleation and subsequent growth in a remote continental location, Hyytiala, Finland. The individual effects of various processes and parameters on aerosol growth were quantified. DMAN uses the Two-Moment Aerosol Sectional algorithm (Adams and Seinfeld, 2002) to track both aerosol number and mass concentration using a sectional approach. The model describes both coagulation and condensation (sulfate, ammonium, organics, etc.) by using the TOMAS algorithm. Our approach combines the SAPRC-99 mechanism for the simulation of gas-phase chemistry, and the 1-D the volatility basis set framework (VBS) for secondary organic aerosol formation and OA chemical aging.

The surface energy of the condensing organics has as a significant effect on the ultrafine particle rate. For a typical spring nucleation episode, an increase of the assumed surface energy results in a decrease of the particle number concentration above 3 nm (N_3) but it increases the number concentration above 100 nm (N_{100}). On the other hand the different parameterizations of the secondary OA chemical aging have a small effect on N_3 but can have a major effect on N_{100}.

Fig. 1 shows a comparison of predicted vs. observed size distribution for 6 May 2008 at Hyytiala. The model reproduces the observed onset of nucleation and the qualitative features of the event. The particle growth is reproduced well with the new particles reaching a diameter of around 50 nm. The nucleation starts at 9.00 am and these new particles grow during the day. The predicted composition of the newly formed particles is shown at Fig 1c. During the nucleation event from 9.00 to 13:00, the new particles consist of sulfate, ammonium and low volatility SOA. After the end of the nucleation, these new particles continue to grow mainly due to the condensation of the semi-volatile SOA. At the end of the day, the new particles consist mainly of organics (95%).

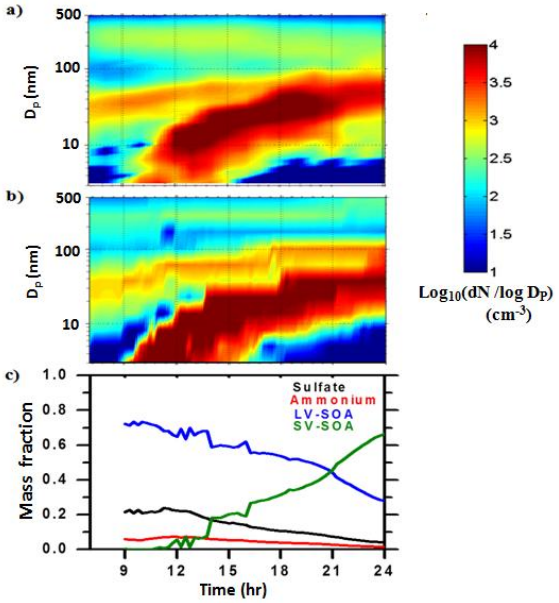

Figure 1. Comparison of (a) measured and (b) predicted dry size distribution as a function of time for 6 May 2008 at Hyytiala. Particle number concentration (z-axis) is plotted against time of day (x-axis) and particle diameter (y-axis). (c) Predicted composition of new particles during the growth.

The simulation suggests that while sulfuric acid plays an important role of nucleation of new particles, it is not responsible for most of their growth, a finding consistent with the analysis of field observations in the site. In contrast, the low-volatility SOA components represent 75% of the new particle mass during the event and 30% at the end of the day. The semivolatile-SOA produced upon oxidation of biogenic volatile organic compounds (BVOCs) is a key player for the later stages of growth.

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