Highly oxidized molecule formation and role in new particles formation and growth

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Introduction

Recent field and laboratory experiments have identified large and rapid formation of extremely low-volatility organic compounds (ELVOC) (Ehn *et al.*, 2014).

Methods

We have evaluated and constrained the proposed ELVOC formation mechanism from Ehn *et al.* (2014) using the Aerosol Dynamics, gas- and particle-phase chemistry kinetic multilayer model for laboratory CHAMber studies (ADCHAM) (Roldin *et al.*, 2014). The mechanism has been implemented into the process-based chemistry transport models ADCHEM (Roldin *et al.*, 2011). We use different datasets to evaluate: (i) the ELVOC formation mechanism, (ii) the role of ELVOC for nano-CN formation (Roldin *et al.*, 2015), (iii) their contribution to particle growth, and (iv) how ELVOCs influence the phase-state and the mass-transfer limited evaporation of SOA particles. ADCHEM was operated as a 1D (vertical column) trajectory model along air mass trajectories reaching the measurement stations Pallas (67.97° N, 24.12° E) and Hyytiälä (61.85° N, 24.28° E). The model results are evaluated using measured particle properties and ELVOC gas-phase concentrations.

Results and conclusions

With the developed ELVOC mechanism ADCHEM captures the main features of the observed particle number size distribution evolution during new particle formation events at Pallas field station in northern Finland (Fig. 1a-c). According to the model about 70 % of the nucleation model particle volume is SOA formed from ELVOCs, 20 % from SVOCs and 10 % from ammonium sulphate (Fig. 1d). The modelled ELVOC mass-spectrum is dominated by C10-monomers with between 8 and 11 O-atoms. The concentrations of ELVOC-dimers are very low during the day (< 1 %). During the night typically about 10 % of the ELVOCs are C20-dimers with 12 to 16 O-atoms (Fig. 1e). A similar diurnal patter is also present in the measured ELVOC spectrum at Hyytiälä. However, in the observations the dimer contribution is about 10 % during the day and 20-30 % during the night. This may indicate that there exists other important ELVOC dimer gas-phase formation mechanism that the model does not take into account.

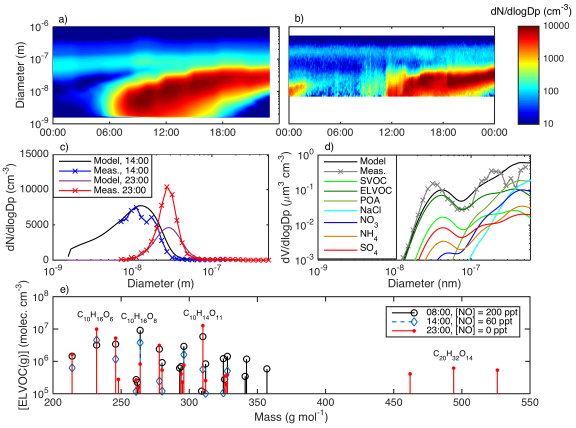

Figure 1. Modelled nano-CN growth at Pallas field station14th of April 2010. Figure a and b shows the modelled and measured particle number size distribution evolution, Fig. c compares the modelled and measured particle number size distributions at 14:00 and 23:00 local wintertime. Figure d shows the modelled and measured particle volume size distribution at 23:00 local wintertime. Figure e shows the modelled ELVOC gas-phase mass spectrum at 08:00, 14:00 and 23:00 local wintertime

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