Geophysical Research Abstracts
Vol. 12, EGU2010-5173, 2010
EGU General Assembly 2010

Distribution of gaseous and particulate organic composition during dark ozonolysis of monoterpenes

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Secondary organic aerosol (SOA) affects atmospheric composition, air quality and radiative transfer. However, major difficulties are encountered in the development of reliable models for SOA formation. Constraints on processes involved in SOA formation can be obtained by interpreting the speciation and evolution of organics in the gaseous and condensed phases simultaneously. In this study we investigate SOA formation from dark ozonolysis of -pinene with particular emphasis upon the mass distribution of gaseous and particulate organic species. Experiments were performed in the EUropean PHOtoREactor (EUPHORE) simulation chamber, including on-line gas-phase composition obtained from Chemical-Ionization-Reaction Time-Of-Flight Mass-Spectrometry (CIR-TOF-MS) measurements, and off-line analysis of SOA samples performed by Electrospray Ionisation Ion Trap Mass Spectrometry (ESI-MS). A detailed chamber box model designed to simulate SOA formation was developed for comparison with the experimental results, coupling an equilibrium gas/particle partitioning module to the -pinene oxidation scheme extracted from the Master Chemical Mechanism (MCMv3.1).

The simulated temporal profile of SOA mass is in reasonable agreement with the observations, both in terms of the shape and magnitude. Sensitivity analysis highlights the importance of the choice of vapour pressure estimation method. Comparisons of the simulated mass spectrum with the CIR-TOF-MS mass spectrum show a similar mass distribution for gaseous organics. The ESI-MS measurements show the presence in the condensed phase of monomer and oligomer type organics. Comparisons of the simulated condensed phase mass spectrum with the ESI-MS mass spectrum show that most of the peaks in the monomer region are in reasonable agreement, i.e. explained in the simulation by the gas/particle partitioning of semi-volatile organic compounds (SVOC) formed during the -pinene gas-phase oxidation. The simulated composition is used to (i) propose chemical structures for the principal gaseous SVOC and condensed monomer organic species and (ii) explore the possible contribution of a range of accretion reactions occurring in the condensed phase. We find that oligomer formation through esterification reactions gives the best agreement between the observed and simulated mass spectra.

Extending this approach to characterise the gaseous and particulate organic composition from other organic precursors, under various experimental conditions, will help greatly our understanding SOA formation, and therefore assist in the development of reliable SOA schemes for incorporation into 3D chemical transport models.