Distribution of gaseous and particulate organic composition during dark α-pinene ozonolysis

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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 phase simultaneously. In this study we investigate SOA formation from dark α-pinene ozonolysis with particular emphasis upon the mass distribution of gaseous and particulate organic species. A detailed model for SOA formation is compared with the results from experiments performed in the EUropean PHOtoREactor (EUPHORE) simulation chamber, including on-line gas-phase composition obtained from Chemical-Ionization-Reaction Time-Of-Flight Mass-Spectrometry measurements, and off-line analysis of SOA samples performed by Ion Trap Mass Spectrometry and Liquid Chromatography. The temporal profile of SOA mass concentration is relatively well reproduced by the model. Sensitivity analysis highlights the importance of the choice of vapour pressure estimation method, and the potential influence of condensed phase chemistry. Comparisons of the simulated gaseous- and condensed-phase mass distributions with those observed show a generally good agreement. The simulated speciation has been used to (i) propose a chemical structure for the principal gaseous semi-volatile organic compounds and condensed monomer organic species, (ii) provide evidence for the occurrence of recently suggested radical isomerisation channels not included in the basic model, and (iii) 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.

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