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Secondary organic aerosols - formation and ageing studies in the SAPHIR chamber

Christian Spindler (1), Lars Müller (2), Achim Trimborn (3), Thomas Mentel (1), Thorsten Hoffmann (2), and the EUCAARI-SAPHIR-08 Team

(1) Institut für Chemie und Dynamik der Geosphäre (ICG), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany (t.mentel@fz-juelich.de), (2) Institut für Anorganische Chemie und Analytische Chemie Johannes Gutenberg Universität Mainz, Germany, (3) Aerodyne Res Inc, Billerica, MA 01821 USA, (4) Division of Atmospheric Sciences, Dept. of Physical Sciences, 00014 University of Helsinki, Finland, (5) Institute of Atmospheric Sciences and Climate Research Council, Bologna, I-40129, Italy, (6) Air Chemistry Group of Hung. Acad. Sci. at University of Pannonia, Egyetem 10, 8200 Veszprém, Hungary, (7) Leibniz-Institut für Troposphärenforschung

Secondary organic aerosol (SOA) formation from oxidation products of biogenic volatile organic compounds (BVOC) constitutes an important coupling between vegetation, atmospheric chemistry, and climate change. Such secondary organic aerosol components play an important role in particle formation in Boreal regions ((Laaksonen et al., 2008)), where biogenic secondary organic aerosols contribute to an overall negative radiative forcing, thus a negative feed back between vegetation and climate warming (Spracklen et al., 2008).
Within the EUCAARI project we investigated SOA formation from mixtures of monoterpenes (and sesquiterpenes) as emitted typically from Boreal tree species in Southern Finland. The experiments were performed in the large photochemical reactor SAPHIR in Juelich at natural light and oxidant levels. Oxidation of the BVOC mixtures and SOA formation was induced by OH radicals and O3. The SOA was formed on the first day and then aged for another day. The resulting SOA was characterized by HR-ToF-AMS, APCI-MS, and filter samples with subsequent H-NMR, GC-MS and HPLC-MS analysis.
The chemical evolution of the SOA is characterized by a fast increase of the O/C ratio during the formation process on the first day, stable O/C ratio during night, and a distinctive increase of O/C ratio at the second day. The increase of the O/C ratio on the second day is highly correlated to the OH dose and is accompanied by condensational growth of the particles. We will present simultaneous factor analysis of AMS times series (PMF, Ulbrich et al., 2009) and direct measurements of individual chemical species. We found that four factors were needed to represent the time evolution of the SOA composition (in the mass spectra) if oxidation by OH plays a mayor role. Corresponding to these factors we observed individual, representative molecules with very similar time behaviour. The correlation between tracers and AMS factors is astonishingly good as the molecular tracers represented only a very small mass fraction of the factors. There is indication that some factors grow at the cost of the other suggesting a set of successive generations of oxidation products. This conversion could proceed either by direct condensed phase processes or by an evaporation-oxidation-recondensation mechanism. On the other hand it seems that the factors evolve in parallel, representing products of multiple oxidation which appear on different time scales in the particulate phase. These findings will be discussed with respect to their importance for ageing processes of atmospheric organic aerosols.

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