Geophysical Research Abstracts,
Vol. 11, EGU2009-10864, 2009
EGU General Assembly 2009

OH-Radical initiated ageing of biogenic secondary organic aerosols – A detailed chemical analysis

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The chemical ageing[1] of secondary organic aerosol (SOA) was investigated in two series of experiments using on-line mass spectrometry and off-line high performance liquid chromatography mass spectrometry (HPLC-MS). In a set of photochemical experiments, performed in the large outdoor reaction chamber SAPHIR (Jülich, Germany), SOA was generated from a boreal mixture including mono- and sesquiterpenes (-pinene, -pinene, 3-carene, limonene, caryophyllene). During a long time experiment (30h) the generated SOA was exposed to OH-radicals and the chemical composition was analyzed on-line using atmospheric pressure ionization mass spectrometry (API-MS). The on-line method provides highly time resolved chemical information and therefore a direct insight into the temporal changes of SOA-composition. In parallel, filter samples analysed by HPLC-MS allow the enrichment of trace compounds and finally an unambiguous identification of individual substances. In addition, filter samples allow a direct comparison to samples from field studies. The ageing experiments showed a clear change in SOA composition. The compounds observed can be divided into two groups: A group of first generation SOA-compounds, generated by the OH oxidation of the terpenes and a group of second generation compounds, generated by the reaction of OH with SOA compounds. Among the second generation products, especially a tricarboxylic acid (3-methyl-1,2,3-butanetricarboxylic acid, m/z 203)[2] was observed to be a good marker compound for BSOA ageing.

A further set of experiments was carried out in another large aerosol chamber facility, the AIDA chamber of the Research Centre Karlsruhe. In this dark chamber, the experiments focused on the OH-induced ageing of -pinene SOA and the influence of temperature. The results clearly show that the tricarboxylic acid is a distinctive marker for OH radical induced BSOA ageing and identify cis-pinonic acid as its precursor.

To connect the results of the laboratory measurements with the ambient atmosphere, this paper also compares filter samples taken at the Finnish Forest Research Station in Hyytiälä to the filter samples obtained from SAPHIR/AIDA experiments.

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