



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

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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.

References

Laaksonen, A., Kulmala, M., O'Dowd, C. D., Joutsensaari, J., Vaattovaara, P., Mikkonen, S., Lehtinen, K. E. J., Sogacheva, L., Dal Maso, M., Aalto, P., Petaja, T., Sogachev, A., Yoon, Y. J., Lihavainen, H., Nilsson, D., Facchini, M. C., Cavalli, F., Fuzzi, S., Hoffmann, T., Arnold, F., Hanke, M., Sellegri, K., Umann, B., Junkermann, W., Coe, H., Allan, J. D., Alfarra, M. R., Worsnop, D. R., Riekkola, M. L., Hyotylainen, T., and Viisanen, Y.: The role of VOC oxidation products in continental new particle formation, *Atmospheric Chemistry and Physics*, 8, 2657-2665, 2008

Spracklen, D. V., Bonn, B., and Carslaw, K. S.: Boreal forests, aerosols and the impacts on clouds and climate, *Philosophical Transactions of the Royal Society a-Mathematical Physical and Engineering Sciences*, 366,

4613-4626, 10.1098/rsta.2008.0201, 2008

Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, *Atmospheric Chemistry and Physics*, 9, 2891-2918, 2009