CLOUD FORMING POTENTIAL OF SECONDARY ORGANIC AEROSOL

Zs. Jurányi¹, M. Gysel¹, J. Duplissy¹, E. Weingartner¹, S. Henning², F. Stratmann², U. Baltensperger¹

¹Laboratory of Atmospheric Chemistry, Paul Scherrer Institut, CH-5232, Villigen PSI, Switzerland ²Leibniz-Institute for Tropospheric Research, Permoser Strasse 15, 04318, Leipzig, Germany

1. INTRODUCTION

Secondary organic aerosol (SOA) contribute an important fraction of the atmospheric organic aerosol. SOA is formed by oxidation of volatile organic compounds (VOC), which results in complex mixtures of organic species in the particle phase. Their hygroscopic and cloud condensation nuclei (CCN) properties are much less understood than those of inorganic mixtures. Even though SOA is to a great extent water soluble, its contribution to cloud formation is still largely unknown.

2. EXPERIMENT

Experiments were conducted on SOA formed through photooxidation of the gaseous precursor species α -pinene –representative of natural emissions– under controlled conditions in a smog chamber. The precursor concentration was varied between atmospherically relevant (10-20 ppb) and an order of magnitude higher values (180-240 ppb). The hygroscopicity and CCN activity of the SOA particles were investigated at different relative humidities (RH) in the sub- and supersaturated region.

A Hygroscopicity Tandem Differential Mobility Analyzer (HTDMA) and the Leipzig Aerosol Cloud Interaction Simulator (LACIS-field) were used to measure diameter growth factors (GF) at 90–98% RH and 97–99% RH, respectively. The CCN activation behavior of SOA at supersaturated RH was followed by two CCN counters (DMT-CCNC) operated behind a differential mobility analyzer (DMA) selecting the same dry size as the HTDMA.

3. THEORY

A simplified one parameter representation of the concentration dependence of the water activity (a_w) [Petters and Kreidenweis, 2006], which shows up in the Köhler equation was used to compare the results that were measured at different relative humidities:

$$a_w^{-1} = 1 + K \frac{V_s}{V_w}$$

where K is the free parameter, V_s is the volume of the solute and V_w is the volume of the water in the aerosol particle.

4. RESULTS

The HTDMA measurements show that the GF of SOA particles at RH≈97% depends on the initial precursor concentration (green symbols in Figures 1 and 2), confirming the results of an earlier study [Duplissy et al., 2008]. The GFs measured by the HTDMA are systematically lower at high precursor concentrations compared to low precursor concentrations, which is reflected in a decrease of the HTDMA dervied K-values. This can be the consequence of the higher partial vapor pressures of the gas phase components at higher precursor concentration thus driving a larger fraction of species with relatively high vapor pressures into the particle phase. The latter compounds are less oxygenated and less hygroscopic and therefore lowering the measured GFs and thus the HT-DMA derived K-values.



Figure 1: K-values retrieved from CCNC, HT-DMA and LACIS during low concentration experiments



Figure 2: K-values retrieved from CCNC, HT-DMA and LACIS during high concentration experiments

Contrary to the HTDMA data no precursor concentration dependence of the critical supersaturation for CCN activation was measured by the two CCNCs, which is reflected in comparable CCN derived K-values shown in Figures 1 and 2. The reason for these different effects of precursor concentration changes on hygroscopicity and CCN activity is not yet clear. Growth factors at subsaturated RH depend mostly on the concentration dependence of the water activity and on the dissolved fraction of SOA if there are solubility limitations. CCN properties are more sensitive to the surface tension due to the Kelvin term in the Köhler equation (all K-values were calculated assuming surface tension of pure water), though surface partitioning effects may compensate surface tension changes to some extent. More sophisticated modeling efforts will be undertaken, in order to explore possible combinations of effects which might explain the observed behavior. However, HTDMA and CCN derived K-values still overlap within experimental uncertainty (colored bands in the Figures), showing that this simple one-parameter model makes approximate predictions of CCN activity from GF measurements possible. Currently it is unclear why the LACIS measures a significant lower hygroscopicity, which is in good agreement with the results of Prenni et al., 2007.

5. BIBLIOGRAHPY

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Acknowledgements

We thank the IFT and the PSI team for performing the experiments, special thanks to M. Ziese, A. Kiselev and T. Tritscher.