LABORATORY STUDY ON CCN EFFICIENCY OF AEROSOL PARTICLES SIMULATING WOOD COMBUSTION PARTICLES

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1. EXPERIMENTAL

The goal of the LExNo campaign, which was conducted November 2005 at the ACCENT site Leipzig Aerosol Cloud Interaction Simulator (LACIS), was to get further insight in the activation properties of aerosol particles resulting from wood combustion [*Stratmann, et al.*, 2008].

In this laboratory experiment, wood combustion aerosol particles were simulated using spark-generated soot (pure or compacted with propanol) coated with ammonium sulfate (AS, coating temperatures 93°C to 170°C) and/or levoglucosan (LG, coating temperature 80°C to 106°C) using two tube furnaces.

Thermal decomposition of ammonium sulfate is known to take place in this temperature range (e.g. [*Halstead*, 1970; *Kiyoura and Urano*, 1970]). The molar ratio of ammonia to sulfate (resulting for the AMS measurements) leads to the conclusion that the particle coating consisted of ammonium hydrogen sulfate (AHS) independent from the coating temperature (cf. Fig. 1). In addition to AHS an organic substance was detected in the AMS spectra for this coating type, which is most probable propanol. Soot particles passed over AS for coating are therefore considered as soot / ammonium hydrogen sulfate coated including propanol (AHSp).

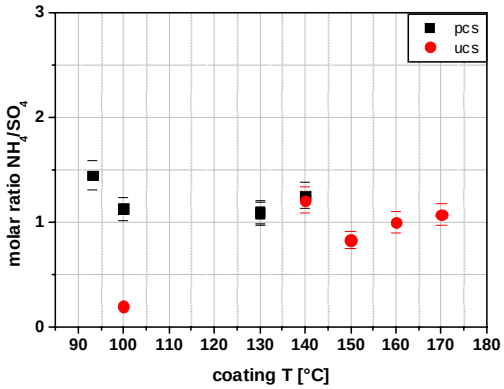

Figure 1 Dependency of coating composition on furnace temperature. The molar ratio of NH4/SO4 should be 2 for AS and 1 for AHS (error bars: 10% error in mass).

For LG decomposition is not expected, because it decomposes above 110°C.

The synthesized particles were analyzed using two aerosol mass spectrometers (AMS), a hygroscopicity tandem differential mobility analyzer (HTDMA), two Wyoming static diffusion cloud condensation nuclei (CCN) instruments, a continuous flow CCN instrument (Droplet Measurement Tech.) and LACIS.

2. RESULTS

Particles were investigated with respect to their critical supersaturation (Sc) and with

respect to their growth factor (GF). Both properties have an impact on the particles cloud-forming potential.

The coating on the soot particles had a clear, hygroscopicity enhancing effect on the particles. The AHSp coating was more efficient in enhancing hygroscopicity than LG. For soot / AHSp coated activation was observed between 0.3% and 0.6% supersaturation for a dry particle diameter of 84.4 nm. For soot / LG coated the critical supersaturation was observed to be between 0.5% and 0.75% for the same particle diameter. Propanol-compacted soot / LG + AHSp coated activated around 0.32%. The range of the critical supersaturation results from the differences in the coating temperature, which influences the coating thickness.

The growth factor (GF) of the particles was clearly related to Sc for all particle types investigated in this humidity range (Fig. 2, RH=98%). The higher the GF was, the more easily the particles were activated. For soot / AHSp coated GFs were found between 1.3 and 2.2. Soot / LG coated particles did not adsorb as much water, and their GF was between 1.25 and 1.5. The soot particles / LG + AHSp coated had a GF of 1.7.

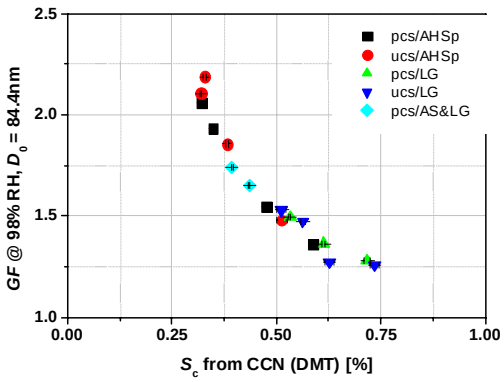

Figure 2 Relationship between growth factor at 98% RH and critical supersaturation of particles (d0=84.4nm).

No influence of propanol-compaction was observed in either growth factor or the critical activation. This is attributed to observed compaction of the particles when coated with AHSp or LG independent from a previous compaction with propanol.

Closure in terms of the prediction of the critical supersaturation was achieved with two different approaches: (1) based on the parameterization of the subsaturated hygroscopicity [*Petters and Kreidenweis*, 2007; *Wex, et al.*, 2007] and (2) based on the chemical composition as determined with the AMS.

Predicted and measured values of the critical supersaturation based on the subsaturated properties were in very good agreement for all investigated particles types (cf. Fig. 3, slope=1.06, R^2=0.98). Growth factors measured at high relative humidities seem to represent the activation properties perfectly for this type of internally mixed particles.

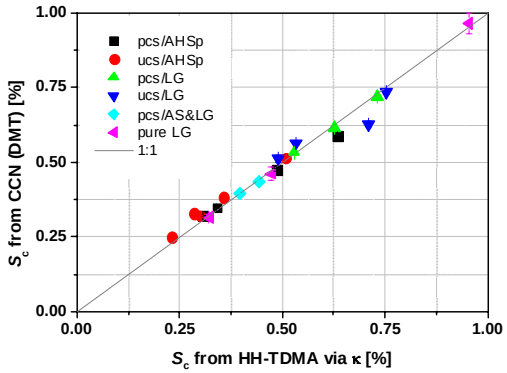

Figure 3 Measured versus predicted critical supersaturation based on subsaturated measurements.

Closure based on the soluble mass as detected with the AMS was not as good, if all particle compositions are taken into account (cf.4, slope=1.26, R2=0.75). However, for soot / AHSp coated predicted and measured Sc are in good agreement (slope=1.13, R2=0.94). Soot particles coated with LG activate at lower Sc than predicted from the measured organic mass from the AMS and scatter more. The reason for this is at this point still under investigation. Possible reasons could be: (1) the high number of doubly charged particles – as observed during the pure LG experiments - could bias mass analysis

from AMS (2) a complex reaction might take place between soot and LG and hinder to regain all the organic material for analysis.

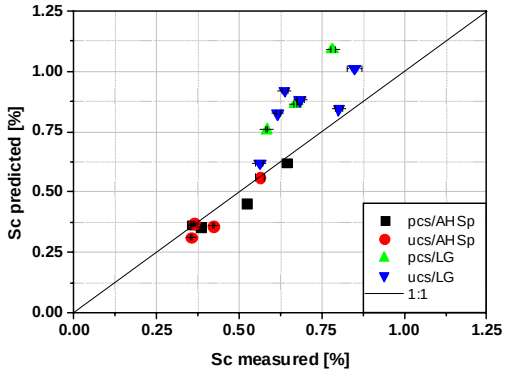

Figure 4 Predicted versus measured critical supersaturation: Prediction is based on soluble particle mass, as detected by the AMS, and Köhler theory (T=20°C, σ(T)=72.8 mN/m).

3. CONCLUSIONS

The particles generated in the laboratory, mimicking biomass-combustion products, were varied in terms of (1) the fractal dimension of the soot core and (2) the applied coating which was either an two component system, levoglucosan or both.
The thermal decomposition of ammonium sulfate in the coating furnace together with a bias from the propanol-compaction unit resulted in a ternary mixed particle, composed of soot / ammonium hydrogen sulfate and propanol (soot / AHSp). The complex coating did not turn out as a disadvantage for the closure experiments, because it could be described explicitly.
Concerning the fractal dimension of the soot particles it was found that initially uncompacted soot particles became compacted when a coating was applied. This effect was more pronounced for the ammonium hydrogen sulfate / propanol coating than for the soot / levoglucosan coated. The differences in the resulting hygroscopic properties were therefore minor between propanol-compacted and initially uncompacted soot cores.
For all particle types a clear correlation between hygroscopic growth at high relative humdities (~98%) and critical supersaturation was observed. This leads to the conclusion, that both of them are dominated by the same particle properties.
Closure between hygroscopicity, activation and soluble mass was achieved by applying two different approaches.
The relationship between hygroscopicity and activation was described via two slightly different one-parameter approaches, which avoid unknown properties (e.g., density, molar mass of mixture) in Köhler theory. The found closure is excellent for all particles types. This might be explained by the fact that the data included in this analysis (HHTDMA and CCN) are measures for the same kind of particles properties.
Closure between soluble mass as measured by the AMS and activation works also very good, although the AMS measures a completely independent particle property.
Between all cumulated data sets closure is achieved and this shows a consistent picture of the activation properties of the investigated particles. It also allows an estimate of the cloud forming potential of combustion particles based on only one of the presented independent measurement data.

4. BIBLOGRAPHY

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Acknowledgments

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