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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment" *by* D. Rose et al.

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We would like to thank S. Martin, G. Biskos, D. Topping, S. Clegg, A. Wexler, the three anonymous referees, and the editor A. Nenes for the constructive comments and suggestions we received during the review and interactive public discussion of our manuscript.

We have individually responded to all comments in the public discussion, and we have implemented all suggestions for improvement upon revision of our manuscript. The main changes and additions to our study are outlined below.

Measurement data analysis

In addition to the effect of doubly charged particles, we have investigated and characterized also the influence of the limited size resolution of the differential mobility analyzer (DMA transfer function) used to select monodisperse calibration aerosol particles. Correcting for the DMA transfer function was found to have little influence on the determination of activation diameters and CCNC calibration, but a strong effect on the width of the CCN activation spectra, which is relevant for the analysis and interpretation of laboratory and field measurements of mixed aerosols.

Following up on the exchange with S. Martin and G. Biskos, we have included the approach of Biskos et al. (2006) and DeCarlo et al. (2004) to apply a size dependent dynamic shape factor for cubic particles. As outlined below, however, the applicability of shape correction factors for NaCl aerosol particles was found to depend strongly on the applied techniques of aerosol generation and conditioning, which can change the actual particle shape.

CCNC flow model

We have revised the CCNC flow model section according to the input of Referee #1. For other users of the DMT-CCNC instrument and flow model, we consider it important to explicitly state that the effective thermal resistance, which can be determined as suggested by Lance et al. (2006) and performed in our study, depends on the Köhler model applied for instrument calibration.

As detailed below, different Köhler models can yield substantially different results, and the flow model results can vary accordingly. Moreover, non-linearities and varying temperature difference offsets in the calibration function were found to limit the

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applicability of the CCNC flow model, especially at low water vapor supersaturation (relative deviations up to 40 %).

Köhler theory and models

Following up on the discussion with D. Topping, S. Clegg and A. Wexler, we have included an additional activity parameterization model (AP3) which is based on the Aerosol Inorganics Model (www.aim.env.uea.ac.uk/aim/aim.html; Clegg et al., 1998a,b). According to Clegg and Wexler (2007), the uncertainty of water activity calculations with the AIM for dilute aqueous solutions of ammonium sulfate and sodium chloride is only 10^{-6}–10^{-5}, and the AIM can be regarded as an accurate reference for Köhler model calculations.

Thus we have taken the AIM-based model AP3 as a reference model for the revised manuscript, and we point out that water activity deviations on the order of 10^{-5} (10^{-4}) lead to relative changes of critical supersaturation by up to 2 % (20 %) for the investigated range of dry particle diameters (20–200 nm). Provided that the relative uncertainty of water activities calculated with the AIM is indeed as low as 10^{-6}–10^{-5}, we conclude that only Köhler models that are based on the AIM or that yield very similar results should be used for CCNC calibration and other investigations involving the CCN activation of $(NH_4)_2SO_4$ and NaCl. In any case, we would like to emphasize that CCN studies should always report exactly which Köhler model equations and parameters were used, in order to ensure that results can be properly compared.

Following up on the exchange with S. Martin and G. Biskos and related publications, we have also included an additional osmotic coefficient model that is based on Brechtel and Kreidenweis (2000). Moreover, we have added effective hygroscopicity parameter models based on the parameters proposed by Petters and Kreidenweis (2007; κ) and Wex et al. (2007; ρ_{ion}), and we have included the κ parameter in a simple analytical approximation model (AA2).

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For better readability of the manuscript, we have moved the derivation and description of the different Köhler models into the Appendix. Moreover, we have re-structured the section on Köhler model results and expanded the discussion of temperature effects on CCNC calibration.

CCN activation of ammonium sulfate and sodium chloride particles

Following up on the interactive public discussion of our paper in ACPD, we have performed additional measurements to double-check the consistency of experimental results and model calculations.

We found that the applied techniques of aerosol particle generation and conditioning can have a strong influence on the shape of NaCl particles. Under the assumption that the AIM-based Köhler model can be regarded as accurate and that there is no artificial bias between the calibration experiments with $(NH_4)_2SO_4$ and with NaCl, the results indicate that the NaCl particles generated by rapid quenching of nebulized solution droplets with excess air were more or less compact spheres or had at least very similar dynamic shape factors as the ammonium sulfate particles. Alternatively, the results would indicate an inconsistency between the AIM-based water activity parameterizations for $(NH_4)_2SO_4$ and NaCl.

In any case, the conditions of particle generation and the shape and microstructure of NaCl particles appear to be critical for their application in CCN activation and CCNC calibration experiments. Further systematic investigations of the dependence of NaCl aerosol particle shape and microstructure on particle generation and conditioning are under way and will be presented and discussed in detail elsewhere.

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