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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.

D. Rose et al.

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We would like to thank Referee #1 for the positive remarks and constructive comments and suggestions, which are highly appreciated and will be taken into account upon manuscript revision. Responses to individual comments are given below.

General comment 1: Use of Köhler models in the CCNC flow model

Response: In section 2.4 of our discussion paper we described how the CCNC flow model of Lance et al. (2006) can be applied to real calibrations by subtracting a

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Interactive Discussion

Discussion Paper

(constant) temperature difference offset. Model calculations without this adjustment strongly deviated from measurement results.

The determination of the thermal resistance of the CCNC column used in the flow model requires a calibration with particles of known size and composition at prescribed temperature gradients or differences (ΔT), respectively. From the measured CCN efficiency spectra, effective supersaturations in the CCNC can be inferred using Köhler theory.

For these calculations we used the VH4.3 Köhler model, because this had also been used by Lance et al. (2006). In this way we obtained an average thermal resistance of 1.78 K W^{-1} for our CCNC column, which is smaller than but comparable to the value reported in Lance et al. (3.4$\pm$0.5 K W^{-1}). In Figure 5 we showed the thermal resistances (R_T) calculated for different operating conditions of the CCNC; they were in the range of 0 to 2.75 K W^{-1}. The corresponding thermal efficiencies indicating the temperature drop through the walls of the column were in the range of 70 to 100% and similar to Lance et al. (50-85%).

Using the flow model of Lance et al. with R_T = 1.78 K W^{-1} we could simulate the experimentally determined calibration lines under most operating conditions very well, when the VH4.3 Köhler model was applied in these calibrations. With this thermal resistance, however, it was not possible to simulate calibrations based on a different Köhler model (VH4.1), as demonstrated in Figure 6. Accordingly, we agree with the statement of Referee #1, that “the Köhler model used should be consistently applied (for calibration and validation datasets)”. In fact, this was meant to be one of the messages of our study (section 4.4), and will be clarified in the revised manuscript.

When applying the Köhler model VH4.1 in the calibrations used to derive the thermal resistance of our CCNC column, the points displayed in Figure 5 were shifted to lower R_T values, and many of them became negative. For $\Delta T > 3$ K we obtained an average value of R_T = 0.24 K W^{-1}, and with this value we could simulate the experimentally

ACPD

7, S4130–S4134, 2007

Interactive Comment

determined calibration lines when the VH4.1 Köhler model was applied in these calibrations.

With the Köhler model AP1.1 we obtained only negative values for the thermal resistance, and with an average value of R_T = - 0.63 K W^{-1} we could simulate the experimentally determined calibration lines when the AP1.1 Köhler model was applied in these calibrations.

Negative R_T values, however, are physically not realistic. They correspond to thermal efficiencies $>$100%, indicating that the temperature gradient inside the column would be larger than outside. Moreover, a thermal resistance would normally be considered as a "material property" (Lance et al., 2006; p.245) that is expected to vary between instruments rather than between calibrations.

Based on our results, we had assumed that the negative, unphysical thermal resistances obtained with Köhler models other than VH4.3 were caused by the parameters $B_1 - B_5$ in Eq. 16 (and/or maybe also $A_1 - A_{13}$ in Eq. 15) of Lance et al. (2006), which seemed to have been derived by fitting to an experimental calibration using the Köhler model VH4.3. This tentative explanation is obsolete, if indeed no Köhler model was involved in the determination of any of these parameters, as stated by the referee. In this case, we agree that any Köhler model can be used equally well within the framework of Lance et al. (2006) to simulate and extrapolate experimental calibration, as long as the same Köhler model is used throughout. Under these circumstances, however, we think that the parameter R_T has to be regarded as a fit parameter which does not really represent the thermal resistance of the CCNC column. Alternatively, unrealistic R_T values might be taken as an indicator for inaccuracies of the applied Köhler model. We would appreciate the referee's opinion on these issues. In any case, we are planning to elaborate on the above aspects in the revised manuscript.

General comment 2: Terminology

Response: Thanks for pointing out the imprecision on p.8203, l.17, where "critical

ACPD

7, S4130–S4134, 2007

Interactive Comment

diameter" will be replaced by "critical diameter of the dry particles" (or "critical dry particle diameter", D_c), which we have defined and used in analogy to "critical dry diameter" as defined and used, e.g., by Kreidenweis et al. (2005).

Specific comment 1

Referee: Line 21 Page 8198 "The effective water vapor supersaturation (S_{eff}) in the CCNC is determined by $\Delta T = T_3 - T_1$ ..."...

Response: The sentence will be changed into "The effective water vapor supersaturation (S_{eff}) in the CCNC is determined by flow rate, pressure, sample temperature and temperature gradient. The temperature gradient is controlled by the temperature difference ($\Delta T = T_3 - T_1$) between the top...".

Specific comment 2

Referee: Line 23, Page 8220 "It can be seen that R_T varies with ΔT, and differs between the different calibration runs."...

Response: The sentence will be changed to "It can be seen that R_T varies over the investigated range of ΔT, and differs between the different calibration runs."

Referee: ... I suggest plotting this as error bars in Figure 6.

Response: We will consider this suggestion upon revising Fig. 6 according to the final outcome of the discussion regarding general comment 1.

Referee: Although thermal resistance is not expected to change with flow rate, temperature and pressure, it may be sensitive to the liquid water drip rate and length of time that the column is wetted before use, which has not been mentioned.

Response: During all of our measurements, the pump controlling the liquid flow was set to "low flow", inducing a liquid water drip rate of 4 mL h^{-1}. The CCNC column has always been wetted for at least 4 hours before starting a measurement series. This will be clarified in the revised manuscript.

ACPD

7, S4130–S4134, 2007

Specific comment 3

Referee: Line 25, Page 8202 "The input variables to the model are..."...

Response: It will be changed to “Input variables to the model are...” (as formulated in Lance et al., 2006).

Specific comment 4, 5, and 6

Referee: Line 17, Page 8203 "...because the parameters $B_1 - B_5$ of Lance et al. (2006) were based on a van’t Hoff factor model with $i_s = 3$"
Referee: Line 29, Page 8229 "Moreover, the Köhler modeling approach used in the CCNC flow model (constant van’t Hoff factor $i_s = 3$)..."
Referee: Figure 6 The circles representing "S_{eff} values calculated from measured D_{50} using the Köhler model VH4.1"...

Response: These three statements will be removed or adjusted according to the final outcome of the discussion regarding general comment 1.

References

Kreidenweis, S. M., Koehler, K., DeMott, P. J., Prenni, A. J., Carrico, C., and Ervens, B.: Water activity and activation diameters from hygroscopicity data-Part I: Theory and application to inorganic salts, Atmos. Chem. Phys., 5, 1357-1370, 2005.

Lance, S., Medina, J., Smith, J. N., and Nenes, A.: Mapping the Operation of the DMT Continuous Flow CCN Counter, Aerosol Sci. Technol., 40, 242-254, 2006.

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