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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.

Anonymous Referee #1

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Summary:

This is a very useful paper detailing the precise calibration of a DMT Cloud Condensation Nuclei Counter (CCNC). The authors provide a thorough analysis of the parameters used in Kohler theory for determining the water vapor supersaturation in the instrument, and experimentally show how the supersaturation varies under different instrument operating conditions. It is shown that assumptions about the water activity of dilute NaCl and ammonium sulfate solutions (typically used to calibrate CCN counters) contribute the most to the uncertainty of the calibrated supersaturation, and that all studies should report the water activity parameterization used. The authors also evaluate a parameterization of supersaturation and thermal efficiency from Lance et al (2006), and show that with inclusion of a nearly constant delta_T offset the parameterizations accurately predict the instrument supersaturation at different operating conditions.

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General Comments:

There seems to be a misunderstanding regarding the framework of the Lance et al. (2006) paper. A key point of confusion is the idea that the parameterizations have inherent within them a specific (simplified) form of Kohler theory. This is not true. There are no inherent assumptions about Kohler theory in either the thermal efficiency or supersaturation parameterizations in Lance et al, (2006). The parameterizations were derived from detailed computational fluid dynamics simulations of the instrument. The only invocation of Kohler theory is in the usage of the calibration experiments (as described below); Lance et al. (2006) used a simplified treatment as an example but any of the numerous forms in Table 5 could have been used without ANY change in the parameterizations.

Provided that the supersaturation is known (from calibrations), the supersaturation parameterization can be used to infer the lengthwise temperature gradient within the CCNC column. Using the thermal efficiency parameterization, the thermal resistance of the instrument may then be inferred. The most sophisticated Kohler model may be used to calibrate the supersaturation and thereby constrain the thermal resistance of the instrument, as long as the same Kohler model is applied in subsequent verification experiments.

ACPD
7, S3777–S3781, 2007

Interactive Comment

With the above said, this paper shows that the parameterizations do capture the physics of supersaturation generation within the CCNC, in many cases with less uncertainty than the parameters used in Kohler theory! The parameterizations have reported standard errors of 0.03% (absolute) and 0.8% (absolute) for supersaturation and thermal efficiency, respectively, as compared to the detailed instrument model. Thus, for experiments in which the supersaturation must be known to greater precision (and calibrations are not available under the experimental operating conditions), it is recommended (as stated in the Lance et al., 2006 paper) that the instrument model be used directly rather than the parameterizations.

Care should be taken to correctly label terms, such as "critical diameter" (Line 17, Page 8203). This term is traditionally used to describe the wet particle diameter in equilibrium with water vapor supersaturation at the point of activation (the maximum of the Kohler curve), while what is meant in the paper is the dry particle diameter (and at other times, the dry particle diameter with a critical supersaturation equal to the instrument supersaturation, usually referred to as Dp50).

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Specific Comments:

Line 21 Page 8198 "The effective water vapor supersaturation (Seff) in the CCNC is determined by delta_T = T3 - T1..."

This is an incomplete statement. Supersaturation is also determined by pressure, temperature and flow rate (all of which can be independently controlled).

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Line 23, Page 8220 "It can be seen that RT varies with delta_T, and differs between the different calibration runs."

Saying that "RT varies with delta_T " implies that these two variables covary, which is not the case. There is a lot of variability in the thermal resistance, but there is not a clear

ACPD

7, S3777–S3781, 2007

Interactive Comment

trend with respect to delta_T for all the calibration runs. The lack of an obvious trend indicates that the parameterization of the instrument model is adequately capturing the physics of heat and mass transfer within the CCNC column (once the delta_T offset is accounted for).

It would be interesting if you plotted in a subsequent figure the effect this variability in RT has on the comparison of modeled and observed supersaturation (as a function of delta_T). For instance, in Figure 6, how different would the modeled line be if a thermal resistance of 1 or 2 K/W were used? I suggest plotting this as error bars in Figure 6.

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.

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

This is a partial listing. Separate inlet sheath and aerosol temperatures may be specified in the instrument model, and these may be different from the internal temperature at the top of the column (T1'). Furthermore, any imposed lengthwise temperature profile may be used (it does not have to be a linear profile).

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Line 17, Page 8203 "...because the parameters B1 - B5 of Lance et al. (2006) were based on a van't Hoff factor model with is= 3"

This statement is incorrect. Please see the general comments.

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Line 29, Page 8229 "Moreover, the Kohler modeling approach used in the CCNC flow model (constant van't Hoff factor is= 3)..."

ACPD

7, S3777–S3781, 2007

Interactive Comment

This statement is incorrect. A van't hoff factor of 3 was used in Lance et al (2006), but this is not inherent in the model or parameterizations. See the general comments.

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Figure 6 The circles representing "Seff values calculated from measured D50 using the Kohler model VH4.1" should not be included, since they are inconsistent with the calibrated thermal resistance (which was derived using a different Kohler model). Any Kohler model may be used to calibrate the instrument supersaturation (and thermal resistance), but the Kohler model used should be consistently applied (for calibration and validation datasets). See the general comments.

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

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 8193, 2007.

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7, S3777–S3781, 2007

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