

Interactive comment on “Droplet activation of wet particles: development of the Wet CCN approach” by S. Nakao et al.

Anonymous Referee #3

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The manuscript “Droplet Activation of Wet Particles: Development of the Wet CCN Approach” describes a new approach to quantify aerosol hygroscopicity based on CCN measurements. Measurements of critical supersaturation are combined with measurements of wet diameter at a range of saturation ratios, instead of measurements of dry particle diameter. The authors demonstrate that this technique is preferable to the traditional dry diameter approach for some chemical systems, and that new, relevant information about these systems can be obtained. This technique represents an advance in CCN measurements, and will be of interest to Atmospheric Measurement Techniques readers. I recommend publication after the following minor issues have been addressed.

General:

C721

One thing I think is missing from the discussion is the fact that growth factor measurements become more useful in predicting CCN activity at high S . This is implied in the caption for Fig. 3 - “especially at higher RH (above $\sim 70\%$)” - as well as associated discussion in the text, but never stated explicitly or quantified. This seems like a critical feature of the Wet CCN approach, and so I suggest the authors discuss it briefly.

Specific:

P 258, L 7-8: “The new approach directly measures Köhler curves under sub-saturated conditions.” This is somewhat confusing because it implies that an entire Köhler curve could be drawn under subsaturated conditions - but then it would no longer be a Köhler curve. I recommend referring to “the subsaturated portion of the Köhler curve”.

Pg 262, Eq. 6: define A (currently defined for Eq. 8)

P 263, L 14-16: The phrase “The possible causes include” suggests that “non-ideality” and “solubility” are the only possible causes. This suggests that surface tension variability is not a possible cause - is this the author’s intent? If not, please remove the word “The”.

P 264, L12: Either state that Fig. 3 is an S vs. D curve, or change “D-axis” to “x-axis.” Somewhat confusing to refer to an unfamiliar “D-axis” without any explanation.

P 265, L 18-20: “Calibration of supersaturation was carried out by observing activation of ammonium sulfate and using $\kappa = 0.6$ based on E-AIM calculation.” If E-AIM is used, doesn’t κ vary slightly, and can’t κ be calculated more precisely? Or are the authors suggesting that AS $\kappa = 0.6$ is adequately precise to calibrate the CCN instrument? Please explain.

P 267, L 1-2: “When a trajectory of data points deviates from an isoline, gf and CCN may be acquired separately by curve fitting as discussed above (Sect. 2.3).” I would say the authors mentioned this but did not really discuss it in 2.3. If the authors would like to add an example of separate gf and CCN curve fitting, that would be helpful. This

C722

is not in my opinion necessary for publication.

P 267, L 23, 26: Please indicate when in the experimental procedure this collapse and/or efflorescence is occurring. I think this will make the discussion more clear.

P 268, L 15-16: I don't understand why irreversible evaporation before size selection by the DMA would cause any deviation. It would cause the particle to shrink immediately after generation, but you would still have the same particle size in the DMA and the CCNC. Please explain the difference between upstream vs. downstream partitioning shifts (L12-13) more clearly.

P 268, L 18-19: "...parameters other than particle composition (e.g., non-ideality, solubility)..." Wouldn't it be better to refer to "particle molar volume?" "[C]omposition" can be used to predict solubility or non-ideality, but molar volume is a distinct parameter.

P 268, L 26: Raoult's Law is typically used for solution components in relatively high concentrations, which at high RH applies only to water (i.e., the solvent). It is potentially confusing to use it as is done here to refer to lowering of solute activity. Please remove or clarify.

P 270, L 14-18: You could also cite $\kappa_{glucose}$ of 0.165 determined at $RH \sim 99\%$ by Ruehl et al. (ACP, 2010).

P 270, L 22: is that κ_{CCN} or κ_{gf} ? Seems relevant to the current topic.

P 271, Eq 10: f_w is used for the molar volume of water, but Eq. (1) uses a different symbol for the "partial" molar volume of water. I assume this is not a relevant distinction, and therefore the same symbol should be used in both equations.

P 271, L 13-15: "The effective κ of ammonium oxalate particles reported here for the Wet CCN technique narrowed down the previously large range in estimates." What κ does this refer to? 0.6? Please give that number here.

P 272, L 17-18: Looking at Fig. 9, it appears that efflorescence occurs as high as $RH \sim$

C723

90%. This seems unusually high and worth of further comment. Has the efflorescence RH for this system been measured previously?

P 275, L 20-21: Again, I don't understand how "Raoult's Effect" would suppress evaporative loss of a dilute solute.

Fig. 3 - Caption does not fully explain figure. Label what panels (a) and (b) are in caption, and I believe this will suffice.

Fig. 7 - I suggest you add arrows to the processes (efflores., collapse, etc.) to indicate the direction in "Wet CCN space" that each process proceeds.

References:

Ruehl, C. R., Chuang, P. Y., and Nenes, A.: Aerosol hygroscopicity at high (99 to 100%) relative humidities, *Atmos. Chem. Phys.*, 10, 1329-1344, doi:10.5194/acp-10-1329-2010, 2010.

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