Review of the paper amt-2018-164

Instrument Artifacts Lead to Uncertainties in Parameterizations of Cloud Condensation Nucleation by Jessica A. Mirrielees and Sarah D. Brooks

Possible artifacts in different types measurements of cloud condensation nuclei need to be discussed much more than has been done previously. This paper could thus be of large value for the scientific community. It is however important to make sure that the discussion is done in a way that is relevant to how CCN measurements normally are made, considering e.g. particle concentrations and data evaluation procedures. I recommend publication after major revision and the authors carefully considering the comments bellow. Since I think this paper needs major revision, I will not comment on small details, but focus on major issues and things that are repeated.

Even though more discussion about artifacts and good practice in CCN measurements are needed, this is not the first paper dealing with the issue and with the role of DMA flow ratio in determining the precision in fitting step functions for evaluating number of CCN as a function of supersaturation. I would recommend that this literature is summarized and that results in this paper are discussed in perspective of this literature.

Also, the use of hygroscopicity parameterizations goes much further back in time than indicated in this manuscript. The parameter epsilon is originally adopted from the work by Fitzgerald (1975) and in 1982 Fitzgerald et al. suggested a single hygroscopicity parameter (Bc). There might be even older literature.

I also recommend that the authors state the limitations of the paper clearly: 1) All CCN measurements are not done with the set up indicated in figure 1. Lab studies can be performed with an SMPS instead of CPC if evaporation from the particles is suspected. 2) Field studies at low particle concentrations are made without a DMA upstream the CCNC. These measurements have there own issues.

Another limitation is that the paper does not treat all the important artifacts. An example is uncertainty in sizing due to evaporation of particle material or residual water in the particles while sized in the DMA. I can understand if this is out of scope for this article, but they could be mentioned. Other sources of errors are closer to the focus of this paper and could be included or at least references made to papers discussing them. I am mainly thinking about three effects: 1) the role of doubly charged particles, especially in lab studies in which atomized aerosols can be overcharged in comparison to equilibrium charge distribution and radioactive sources normally used are not strong enough to neutralize the aerosol. 2) Voltage offset in the DMA is sometimes an issue, especially when working with high supersaturations and small aerosol particle sizes. 3) The role of counting variability due to sampling statistics at low concentrations and how it influences the determination of SSc in different cases (for example different flow ratios).

I also have a comment that might sound nerdy, but I find it important that we stick to the definition of an aerosol as a population of solid and/or liquid particles and the

surrounding gas. Thus, we should not talk about aerosol size, when referring to the size of the particles. Please check the manuscript in line with this.

It is not clear how the critical supersaturation is determined: is it defined as the supersaturation at which #CCN/#CN is 0.5 or when its value is 50% of the level reached at high supersaturation (the later often being used in experimental work)? This will in some of the examples make a large difference, and need to be discussed. An example is line 346-348.

Please check the plots with particle size distributions. The y-axis should be dN/dDp (with the unit cm⁻³ nm⁻¹) if a linear diameter scale is used. Why are you using a linear diameter scale and not a logarithmic? Also, see my comments to figure 6 and 7 below. I think that these are critical for the quality of the paper and the conclusions!

Also, make sure that the figure captions and legends are sufficient.

Figure 6 b. As I understand it, the CPC counting limitations relevant here relate to the number concentration after the DMA. Are the DMA transfer function and the charge distribution taken into account when determining these curves? And if so, for which aerosol to sheath flow ratio are they made? Is it just a coincidence that the size distribution is cut at the same value of dN/dDp (in cm⁻³ nm⁻¹?) as the CPC concentration saturates (in the unit cm⁻³). An how can the "saturated size distribution" be a horizontal line? Both charging probability and transfer function width (in a linear scale) are size dependent.

Figure 7 and the calculations behind them: How is dN/dDp transferred into a concentration after the DMA? Which flows are used?

You use both saturation ratio and supersaturation in the theoretical discussion. As I can see you are using them correctly, but sometimes you use only saturation for saturation ration. I would recommend that you stick to saturation ratio in order to avoid confusing the readers.

Line 184. With a truly monodispers aerosol the concentration would also be 0.

The discussion and the conclusion section is mainly a repetition of the results (which might well be a part of these sections), but I would have liked to see a discussion on what should be considered good practice in CCN measurements, based on this work and the literature.

-Fitzgerald, J.W (1975) Approximation Formulas for the Equilibrium Size of an Aerosol Particle as a Function of Its Dry Size and Composition and the Ambient Relative Humidity J. Appl. Meteorol. 14, 1044-1049

-Fitzgerald, J.W.,Hoppel, W.A., and Vietti, M.A. (1982) The Size and Scattering Coeffi- cient of Urban Aerosol Particles at Washington, DC as a function of Relative Humidity J. Atmos. Sci. 39 1838-1852