We thank the Editor and reviewers for taking the time to review the manuscript and provide their insight, which has allowed us to greatly improve the manuscript. We have responded to the new comments below in blue text.

Editor Comments:

Dear authors,

Many thanks for your revised manuscript.

Both reviewers and I have evaluated your revisions. Overall, the manuscript has greatly improved. Reviewer #2 raised the issue that a few technical details are still missing (see comments below). Concerning the mini-CCNC: Although you write that it follows the same physical principles as the standard CCNC, I miss some more technical details (e.g. on instrument flow and column properties (i.e. is is the same as for the standard one or are there differences?), etc.). It would be great if you could clarify on these aspects as well (you could add this to the supplement if needed).

Last but not least, please add to your manuscript the statement on data availability. The data should be stored on a public/open data repository (if possible with DOI), see guidelines here: https://www.atmospheric-chemistry-and-physics.net/policies/data_policy.html.

Thanks and kind regards

Paul.

Technical details are discussed below in response to the reviewer's comments. The data availability statement is now included.

Reviewer 1 Comments:

The authors have thoroughly answered all remarks and revised their manuscript accordingly. I have no further suggestions.

Reviewer 2 Comments:

N/A

Reviewer 3 Comments:

First of all, the manuscript has improved much. Thank you for addressing these issues.
Albeit this, I still have two minor issues that I hope the authors will address.

#1: CCN counter calibration (author response to 2 (b))
I still have some concerns with respect to how Köhler theory is used to estimate the critical supersaturation. I will try to make myself clearer. When Köhler theory (saturation ratio = some water activity representation * Kelvin effect) is used, there are many models that can represent the water activity (E.g. see paper by Rose et al., 2008). If you are "assuming a water activity of 1.0" (as you now

state in the methods), then only the Kelvin term (curvature effect) is left and you cannot derived a critical supersaturation. I am sure that this is not your case.

On the other hand, if the "procedure for calibrating the miniature CCN counter is identical to the method described by Roberts and Nenes (2005)", then they assume a van't Hoff factor = 3 (stated in Roberts and Nenes, 2005). This water activity parameterisation has be shown to perform poorly compared to the E-AIM model (see Figure 11 (VH4b vs AP3), Rose et al, 2008).

To sum up, I miss the details on how the authors represent the water activity and estimate the theoretical critical supersaturation of monodisperse ammonium sulfate used for calibrating the CCN counter. Please address this issue.

In addition, I find it odd that there is no technical descriptions of this miniature CCN.

First, the statement of "assuming a water activity of 1.0" is not correct and was mistakenly included. This statement was actually intended for another manuscript that was being written at the same time as this manuscript. This statement has been removed from the text.

As the miniature CCN instrument follows the same principle as described in Roberts and Nenes, 2005 – albeit more optimized, technical details on instrument design have not been published. The flow rates and temperature gradients of the scanning CCN instrument vary from 0.09 lpm to 0.22 lpm and 8 K to 12 K, respectively. The length of the CCN wetted column surface is 138 mm. We used a sinusoidal pattern from high flow/high temperature gradient to low flow/low temperature gradient with a period of 10 minute to generate continuous CCN spectra every five minutes. We have updated the following text in the manuscript:

"An instrument model, discussed in Roberts and Nenes (2005) showed a standard deviation in the supersaturation estimate of about +/-0.01%. The supersaturation range of the scanning CCN counter flow rates and temperature gradients vary from 0.09 lpm to 0.22 lpm and 8 K to 12 K respectively. A sinusoidal pattern from high flow and high temperature gradient to low flow and low temperature gradient with a period of 10 minutes generated a continuous CCN spectra every 5 minutes that spanned from 0.06 % to 0.87 % supersaturation. The constant supersaturation CCN counter operated at constant flow and temperature gradient of 150 lpm and 9 K for a 0.43 % supersaturation (referred to as CCN0.43), at 1 Hz and was used to identify CCN gradients in vertical profiles (Section 3.6)."

In this manuscript, we used an upper limit (van't Hoff = 3). We recognize that the accepted values for kappa are 0.61 from CCN-derived values reported by Petters and Kreidenweis (2007) -- which lead to a van't Hoff factor of 2.52 (also reported in Rose et al., 2008). Using a van't Hoff factor of 2.52 implies that the CCN spectra would shift (< 10%) to larger supersaturation, and well within measurement uncertainties. There are no changes to the conclusions described in the manuscript. We have added the following text in the manuscript:

"The critical supersaturation in this study was derived by Kohler theory using a van't Hoff factor of 3.0 as an upper limit for ammonium sulfate. Using a van't Hoff factor of 2.52 (Petters and Kreidenweis, 2007; Rose et al., 2008), would shift the CCN spectra to larger supersaturations by less than 10%."

#2: Kappa derived from CCN spectra.
You write now in lines 154-156: “The CCN spectra and UHSAS number concentrations on the GV were used to estimate the hygroscopicity parameter at 0.07 µm diameter (κ70) for each MBL leg. For this calculation, the critical supersaturation is derived from the CCN spectra, where the UHSAS concentration at 0.07 µm diameter is equivalent to the CCN concentration”

It is still not clear for me how to follow this calculation. Could the authors please clarify this to the reader?
E.g. in supplement, using an example or by pointing to another example elsewhere in litterature.

The following equation is used to derive the hygroscopicity parameter for ambient particles.

$$\kappa = \frac{4A^3}{27D_{UHSAS}^3(\ln S_c)^2}, A = \frac{4\sigma M_w}{RT\rho_w}$$

Where ρ_w is the density of water, M_w is the molar mass of water, σ is the surface tension of water, R is universal gas constant and T is the absolute temperature (273.15 K), D_{UHSAS} is the smallest diameter measured by the UHSAS (0.07 µm) and S_c is the critical supersaturation in which the CCN concentration is equivalent to the total UHSAS concentration (see Figure S1 below for example). This equation is similar to that of Petters and Kreidenweis (2007), which uses the dry diameter at which the CCN reaches 50% of the total particle number concentration at a specified supersaturation as the activation diameter. Here, instead of determining the activation diameter from a specified supersaturation we are determining the critical supersaturation for a specified activation diameter (0.07 µm).

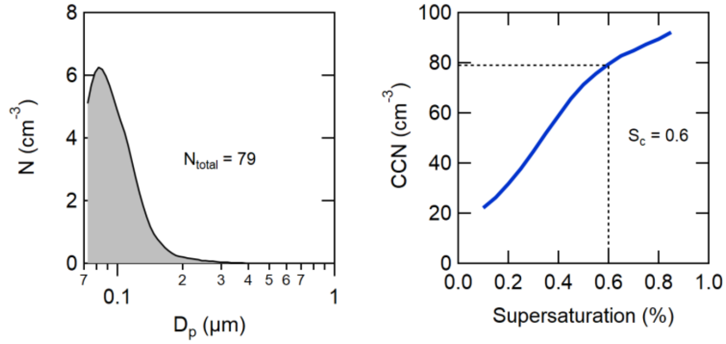

Figure S1: The UHSAS number distribution (left) is integrated to determine the number of particles greater than 0.07 µm (79 cm-3). Then the CCN critical supersaturation is determined with the CCN spectra (right), by identifying when the total UHSAS concentration was equivalent to the CCN concentration.

Reference:

Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O., & Pöschl, U. (2008). 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. Atmos. Chem. Phys., 8(5), 1153–1179. https://doi.org/10.5194/acp-8-1153-2008