Response to comments of anonymous referees # 2, acp-2021-577

General comments

This manuscript "On the evolution of sub- and super-saturated water uptake of secondary organic aerosol in chamber experiments from mixed precursors" presents a series of laboratory studies on SOA hygroscopicity at 90% RH and RH above 100 % under the different chamber conditions from single or multi-precursors. Comparison and discussion of hygroscopicity and CCN activity (k) of SOA are very important to understanding the aerosol-cloud interaction in atmosphere. Therefore, the manuscript fit well the scope of ACP. I recommendation its publication in ACP after addressing the following general and minor comments.

Many thanks to the reviewer for the comments and suggestions. We have improved the manuscript accordingly. Please find a point-by-point response below.

Minor comments:

1.Page 2: line 34-38 "With the increase of condensed SOA on seed particles throughout the experiments, the discrepancy of κHTDMA and κCCN became weaker (down to ~ 0 %) and finally the mean κHTDMA was ~ 60% higher than κCCN on average when the SOA mass fraction approached ~ 0.8. This is possibly attributable to the non-ideality of solutes at different RH or the different co-condensation of condensable organic vapours within the two instruments." The explanation is not clear. Also. I would suggest explain "non-ideality" of solutes at different RH.

Thanks for your suggestion. The reviewer 1 came up with the same question. To better understand the role of non-ideality for the κ discrepancy between HTDMA and CCN, we performed thermodynamic model calculations (AIOMFAC) and simulated the particle hygroscopic growth with inorganic/organic mixtures under ideal and non-ideal conditions. A new section 3.5 has been added to the updated manuscript to compare the simulation results with measurements.

2. Page 3: line 47-48 "The reliability of cloud condensation nuclei (CCN) activity predicted from the aerosol hygroscopic growth under sub-saturated condition remains unresolved" I would like to suggest that " the problem or issue of reliability….. remains unresolved"

The manuscript has been updated as suggested.

3. Page 9: line 207-208 "Fig. 1 and Fig. 2" should be "Figure 1 and 2" please check other sentence in the whole of manuscript.

This typo has been checked over the whole manuscript and corrected in the updated manuscript.

4. Page 12 line: 281-283 "As shown in Sec. 3.1-3.2, the aerosol chemical composition is size-dependent. It is essential to ensure the chemical composition are comparable for HTDMA and CCN measurements for the reconciliation study if their measured dry particle sizes are different" "are" should be "is"

This typo has been corrected in the updated manuscript.

5. Page 14 line 341-343: "It is worth noting that the prediction of critical diameter and CCN number concentration from κHTDMA are based on the concurrently measured critical supersaturation and particle number size distribution." I would appreciate more explanation here.

We explained this in the next sentence (also your next comment), but as it did not seem to be sufficiently clear to understand. we have rephrased it for clarification:

"It is worth noting that the prediction of critical diameter and CCN number concentration from κ_{HTDMA} are based on the concurrently measured critical supersaturation and particle number size distribution. This dependence of $\kappa_{HTDMA}/\kappa_{CCN}$ ratio on chemical composition can have a varied impact on the uncertainty of the

predicted CCN activity from sub-saturated κ_{HTDMA} at different supersaturation ratio of water vapour and/or different particle number size distribution as measured above. Because the activated CCN number concentration is determined by all the three factors: the κ_{HTDMA}, water supersaturation ratio and particle size distribution. If at different supersaturation ratio of water vapour and/or different particle number size distribution as measured in this study, the uncertainty of the predicted CCN activity from sub-saturated κ_{HTDMA} can change. Indeed, this discrepancy trend between κ_{HTDMA} and κ_{CCN} could introduce a varied impact on the CCN prediction, which needs further investigations."

6. Page 14 line 343-346: "The broader influences of the observed trend of $\kappa HTDMA/\kappa CCN$ as a function of MRSOA/PM on CCN activity prediction can vary a lot under different conditions of supersaturation and particle size distribution, which need further investigations."

What does author mean "under different conditions" please explain it.

Here, "under different conditions of supersaturation" means different supersaturation ratio of water vapour. This sentence has been rephrased in the updated manuscript as shown in the 5th response above.

General comments:

1. The authors should emphasize the novelty of the paper. The systems they chose have been somewhat studied by others in laboratory experiments. Comparison with the literature results are encouraged when appropriate.

Sentences have been added in the introduction to emphasize the novelty of the paper:

"The novelty of the project is its design to investigate SOA formation from single to mixed precursors whereas previous studies mainly focused a single precursor (Voliotis et al., 2022). The interaction of the mixed precursor could influence SOA properties, therefore this study takes a further step of lab studies towards the real atmosphere where thousands of precursors are existing and reacting at the same time even the chemical regime and complexity of the chamber studies could deviate from the real atmosphere.

In the updated manuscript, a new section on AIOMFAC model simulations and the κ of SOA at sub- and super-saturated conditions based on ZSR method are added. More comparisons with literature and discussions are added to explain the observation. Please see details in section 3.5.

2. What is the residence time for HTDMA system? As author discussed "non-ideality", does mean residence time affect SOA hygroscopicity, especially for formed viscous organic compounds?

Chan, M. N. and Chan, C. K.: Mass transfer effects in hygroscopic measurements of aerosol particles, Atmos. Chem. Phys., 5, 2703–2712, https://doi.org/10.5194/acp-5-2703-2005, 2005.

Explain the non-ideality.

As demonstrated in Chan and Chan (2005), a few seconds of residence time in HTDMA system (30s in this study, (Duplissy et al., 2009)) is possibly not enough for aerosol particles with viscous organic film to equilibrate with water vapour. The inhibited water

uptake then leads to underestimation of aerosol hygroscopic growth. In my recent work, we concurrently recorded the particle rebounding fraction as an indicator of phase state and found that aerosol particles tend to be liquid-like without rebounding when RH is larger than 80% irrespective of the organic mass fraction (0-0.8) in all investigated VOC systems (Wang et al., 2021). Therefore, at 90% RH, the equilibration of water should be fast. Furthermore, the residence time can influence gas-particle partitioning of semi-volatile species. We address the topic of semi-volatility in Sec. 3.5 of the revised manuscript.

The detailed discussion on how non-ideality influences the hygroscopic growth is also shown in the newly added section 3.5.

3. Mixing state effect on hygroscopicity and CCN activity of SOA under the sub-saturated and super-saturated condition are encouraged discussed in the main text. Have author perform experiments on morphology or mixing state of SOA at different RHs (e.g., RH<100% or RH>100%) using an optical microscopy or AFM?

We fully agree that the morphology or mixing state measurement will largely improve our discussion, but unfortunately it is not available in this study. It is a very interesting topic, which is worthwhile to investigate in the future study.

4. Fig. 3 shows that there is a weak size dependence of GF ((κHTDMA) between 100 and 200 nm aerosol particles, with up to ~ 0.2 due to non-uniform size-dependent chemical composition. Could author estimate uncertainty of GF ((κHTDMA) at different sizes in the HTDMA system? or what is GF uncertainty of SOA aerosol nanoparticles with diameters from 75 to 300 nm at 90% RH as well as calculated kHTDMA uncertainty? Please see Table 2 from Mochida and Kawamura (2004).

Good suggestions. An uncertainty estimation is important when comparing GF/κ values

among different particle sizes, experiments and instruments. The uncertainty of κ values from HTDMA and CCN counter are estimated following the method in Irwin et al. (2010). Herein, for HTDMA measurement, the GF uncertainty is calculated with the same method as in Mochida and Kawamura (2004) as the reviewer suggested and then propagated to the calculation of κ. Overall, the κ uncertainty for HTDMA and CCN are within 10%. The κ uncertainty has been added to Fig.5 and the discussion in Sec. 3.5.

5. Page 13 line: 302-304 "A higher κ_{HTDMA} (κ_{CCN}) of the multi-component SOA-inorganic mixtures at the same level of M_{RSOA}/PM indicated a higher κ of the SOA, according to the ZSR mixing rule of κ demonstrated in Petters and Kreidenweis (2007)." It is not clear, could author explain more in details? And could author evaluate $ZSR_{multi-component SOA-inorganic}$ according to Eq. (7) from Petter and Kreidenweis (2007)?

Thanks for your comment. The reviewer 1 also pointed out this (1ˢᵗ question in the general comment). I have calculated the κ of OA with ZSR method as both of you suggested and discussed in the updated manuscript. Please see the added part in Sec. 3.4.

6. Page 13 line: 319-324: I would appreciate more discussion that how these factors (e.g., surface tension, molecular volume) affect the kappa, the following references are recommended:

Wang, J., Shilling, J. E., Liu, J., Zelenyuk, A., Bell, D. M., Petters, M. D., Thalman, R., Mei, F., Zaveri, R. A., and Zheng, G.: Cloud droplet activation of secondary organic aerosol is mainly controlled by molecular weight, not water solubility, Atmos. Chem. Phys., 19, 941–954, https://doi.org/10.5194/acp-19-941-2019, 2019.

Davies, J. F., Zuend, A., and Wilson, K. R.: Technical note: The role of evolving surface tension in the formation of cloud droplets, Atmos. Chem. Phys., 19, 2933–2946, https://doi.org/10.5194/acp-19-2933-2019, 2019.

All impacting factors are discussed in the new added section 3.5, including e.g. surface tension, molecular weight. Your suggested references provide useful evidence and have been cited in the discussions.

References of response:

Chan, M. N. and Chan, C. K.: Mass transfer effects in hygroscopic measurements of aerosol particles, Atmos. Chem. Phys., 5, 2703-2712, 10.5194/acp-5-2703-2005, 2005.

Duplissy, J., Gysel, M., Sjogren, S., Meyer, N., Good, N., Kammermann, L., Michaud, V., Weigel, R., Martins dos Santos, S., Gruening, C., Villani, P., Laj, P., Sellegri, K., Metzger, A., McFiggans, G. B., Wehrle, G., Richter, R., Dommen, J., Ristovski, Z., Baltensperger, U., and Weingartner, E.: Intercomparison study of six HTDMAs: results and recommendations, Atmos. Meas. Tech., 2, 363-378, 10.5194/amt-2-363-2009, 2009.

Irwin, M., Good, N., Crosier, J., Choularton, T. W., and McFiggans, G.: Reconciliation of measurements of hygroscopic growth and critical supersaturation of aerosol particles in central Germany, Atmos. Chem. Phys., 10, 11737-11752, 10.5194/acp-10-11737-2010, 2010.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric chemistry and physics : from air pollution to climate change, Third edition., John Wiley & Sons, Hoboken, New Jersey2016.

Wang, Y., Voliotis, A., Shao, Y., Zong, T., Meng, X., Du, M., Hu, D., Chen, Y., Wu, Z., Alfarra, M. R., and McFiggans, G.: Phase state of secondary organic aerosol in chamber photo-oxidation of mixed precursors, Atmos. Chem. Phys., 21, 11303-11316, 10.5194/acp-21-11303-2021, 2021.