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Comment on acp-2021-577

Anonymous Referee #1

Referee comment on "On the evolution of sub- and super-saturated water uptake of secondary organic aerosol in chamber experiments from mixed precursors" by Yu Wang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-577-RC1, 2021

General comments:
In this paper, authors conducted a series of laboratory studies on the evolution of sub- and super-saturated water uptake of secondary organic aerosol in chamber experiments from mixed precursors. The comparison of aerosol hygroscopicity under sub- and super-saturated conditions involves complex SOA characteristics are important for CCN predictions and understanding the RH effects (from sub-saturation to super saturation) on aerosol hygroscopicity and further improve aerosol hygroscopicity parameterizations. Therefore, the theme of this paper is scientifically meaningful, and the designed laboratory studies are also valid. However, more insightful analysis is required to better interpret the laboratory results.
Two main conclusions are drawn in this study.
The first one is that SOA composition played a second role in the kappa variations. I agree with the authors, and this is obvious considering that the well-known fact that inorganic aerosol is much more hygroscopic than SOA, thus, of course the SOA composition should play the second role. However, at the same MR_SOA/PM, considerable variations can be found, for example, at MR_SOA/PM ~0.4, the k_HTDMA varies from less than 0.2 to about 0.4, which demonstrates that the SOA compositions played significant roles in determining the apparent kappa. The authors should quantitatively derive the apparent/effective hygroscopicity parameter Kappa_OA using the volume mixing rule under sub-and super-saturated conditions and discuss their differences with previous results (Kuang et al., 2020b), quantitively present the relationships between Kappa_OA and SOA oxidation levels under sub- and super- saturated conditions and discuss more about the controlling factors of Kappa_OA variations based on AMS signals and VOC precursors (Wang et al., 2019). Otherwise, the authors are just talking about the sub- and super-saturated water uptake of mixed aerosol system includes organic and inorganic aerosols but not that of secondary organic aerosol, and is not consistent with the theme of the title.
The second one is "K_HTDMA/k_CCN increased as a function of SOA mass fraction, independent of initial VOC concentrations and sources, the mean k_HTDMA can be 60% higher than k_CCN on average when aerosol fraction approached 0.8". This finding is quite interesting but the explanation is not very convincing. The authors conclude that this finding is possibly attributable to the non-ideality of solutes at different RH or different co-condensation of condensable organic vapors within the two instruments. At L397, the authors claim that water increase in CCN set-up is not favorable for co-condensation of semi-volatile vapors. However, the condensation of these vapors such as HNO3 is influenced by both water vapor content and temperature. Although the temperature in CCN is higher than the temperature in HTDMA which seems not favorable for co-

condensation, however, the aerosol water content in the CCN is much higher than in the HTDMA due to the super-saturated conditions. Thus, I cannot agree upon this argument. As to the possible role of no-ideality, the authors cited two papers published by Brechtel and Kreidenweis (2000a) and Brechtel and Kreidenweis (2000b) and said "the interactions of inorganic ions and organic molecules can exert both positive and negative effects in the water uptake, depending on the organic fraction and inorganic species". The organic fraction and inorganic species are clear in this study, can authors perform some quantitative analysis? or at least deliver a clear qualitative result that the interactions of inorganic ions and organic molecules exerted a positive or a negative effect on the water uptake.

Specific comments:
L45-L50, just a suggestion: the first two sentences of the introduction lack continuity in logic, the authors jumped from aerosol-cloud interaction to the reliability of CCN prediction, I suggest a sentence that claim the accurate CCN prediction is essential for investigating aerosol-cloud interactions in climate models might be needed between the original two sentences.
L53, the SOA con be formed not just through gas-phase partitioning, but also aqueous-phase reactions (Ervens et al., 2011;Kuang et al., 2020a).
L58, about the role of organic aerosol hygroscopicity in climate and aerosol cloud-interactions, references such as (Liu and Wang, 2010;Rastak et al., 2017) might be better choices.
L164 more details about the DMA-CCN set-p should be given, for example, the detailed supersaturation points and time schedule
L175 Please report how the AMS vacuum aerodynamic diameter is converted to the mobility diameter, and estimate potential MR_SOA/PM uncertainties associated with this respect due to that obvious size-dependent chemical composition is observed.
L226, a smaller particle size is not specific (compare to which size range), please presents diameter range directly

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