

We thank the reviewers for their insightful comments and feedback. We have provided our point by point responses in blue text.

Hybrid Water Adsorption and Solubility Partitioning for Aerosol Hygroscopicity and Droplet Growth

Responses to Referees

Referee 1:

This manuscript studied the hygroscopicity and CCN activity of the organic compounds with low solubility under sub-/super-saturation conditions, and the authors developed a new analytical framework, Hybrid Activity Model (HAM), to simulate the CCN activities of compounds with varying chemical structures and aqueous solubility. This article is interesting and worthy of publication.

Comment 1: Is HAM also performance good for simulating the mixture of insoluble compounds (such as black carbon) with the low solubility compounds? Give some discussion regarding that?

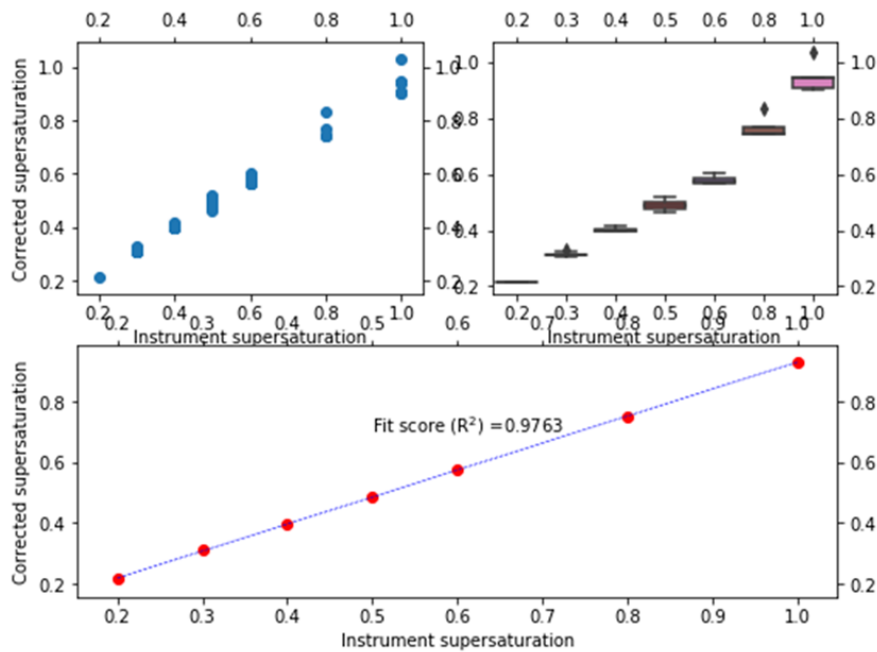
Response: This is a great point and a question we have already begun to address. Our future work includes looking into the application and effectiveness of HAM for the water uptake and droplet growth predictions for extremely low water solubility species, particularly black carbon. In our soon to be submitted work, we have used the Cabot Vulcan XC72R type black carbon to explore the mixtures of effectively insoluble compounds. HAM is applied to black carbon in pure state as well as to black carbon mixed with structurally isomeric compounds with varying aqueous solubility. The structural isomers from Gohil et al. (2022) are chosen to create mixtures and study their water uptake characteristics. Thus we have evidence that HAM also works well for mixtures. In this manuscript we have added a few sentences to provide additional discussion as follows:

“The next step is to evaluate the application of HAM for the CCN analysis of aerosol mixtures for a wider range in aerosol species and compositions. The shift from volume to surface based absorption principles may be more appropriate for significantly water-insoluble compounds. Specifically, the application of HAM can be examined for the hygroscopic growth and water uptake on black carbon agglomerates.”

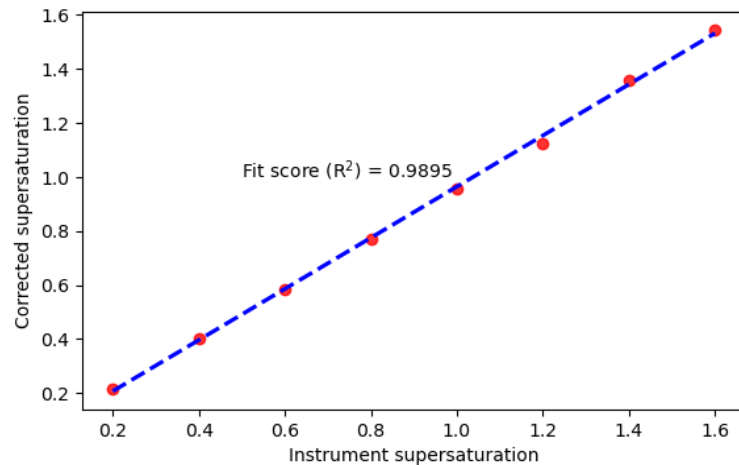
Comment 2: The SS set to 0.6-1.6% in this study, while the SS calibration with the AS only covers from 0.2-1%, what's the uncertainty of the SS above 1%?

Response: We have made a correction. The calibration of the CCNC was performed over supersaturations in the range of 0.2%-1.6%. The CCNC can perform measurements of number concentration over the a supersaturation range of 0.2%-1.6% with minimum uncertainties in the particle counts accompanied by low fluctuations in the temperature gradient across the CCNC column. The calibration plot and table have been updated in the revised supplemental information to contain the supersaturations between 0.2% and 1.6% supersaturations.

Old plot:



New plot:



Old table:

Table S1. Sample CCN Counter (CCNC) calibration data using $(\text{NH}_4)_2\text{SO}_4$

Supersaturation Setting (%)	Calibrated Supersaturation (%)	Critical Dry Diameter (nm)
0.2	0.215	75.6 ± 2
0.3	0.308	61.7 ± 0.6
0.4	0.402	52.3 ± 0.6
0.5	0.493	45.5 ± 1
0.6	0.586	41.2 ± 0.4
0.8	0.771	34.7 ± 0.7
1.0	0.957	29.6 ± 0.6

New table:

Table S1. Sample CCN Counter (CCNC) calibration data using $(\text{NH}_4)_2\text{SO}_4$

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0.8	0.771	34.7 ± 0.7
1.0	0.957	29.6 ± 0.6
1.2	1.125	26.1 ± 0.9
1.4	1.357	23.1 ± 1.1
1.6	1.546	$21.2 \pm 1.$

Comment 3: Humidifier should be added between 2 DMAs in Fig. 1(b). Can you also please specify the residence time of the particles inside the humidifier, and prove this time is enough for the low-solubility (low hygroscopicity) compounds to reach the equilibrium?

Response: This is correct - the humidifier was attached between the 2 DMAs. The nafion tube membrane used for humidification is now shown in the revised Fig. 1(b). The nafion tube was 50cm long and was calibrated using $(\text{NH}_4)_2\text{SO}_4$ before measurements for any of the pure or mixed aromatic acid sample was performed. It is estimated that particles are exposed to elevated RH for ~30 seconds, longer than typical droplet formation and sufficient to establish equilibrium.

Secondly, it should also be noted that the FHH empirical parameters derived from the supersaturated measurements were applied for the hygroscopic growth analysis of the H-TDMA data obtained in the subsaturated regime. This means that in the process of growth under the supersaturated conditions is consistent with the predicted growth and measurement extended to the subsaturated regime. The implication here is that empirical parameters obtained from the supersaturated measurements consider equilibrium droplet growth, and should be able to predict deliquescence under subsaturated conditions when equilibrium is reached.