Dear Editors and Reviewers:

Thanks for your letter and the reviewers' comments concerning our manuscript entitled "Measurement Report: Water diffusion in single suspended phase-separated aerosols" (Manuscript ID: egusphere-2023-1346). Those comments are all valuable and helpful for revising and improving our paper, as well as the important guiding significance to our researches. We have studied comments carefully, and revised the article by supplementing corresponding interpretations according to the reviewers' suggestions. Accordingly, the framework of this manuscript has been adjusted to systematically show the results that fits the theme. The point-by-point responses are as follows, and the corrections excerpted from the main article are highlighted by yellow. We hope the corrections can meet with your approval.

Responds to the reviewers' comments:

Reviewer 1:

#1. I do not agree that the changes in the spectra shown in Figure 3 can be rationalized by evaporation of citric acid. It is much more likely to arise from a change in RH occurring during the H2O/D2O shift, evidenced by the rapid change in peak positions initially (in the first 2 mins), followed by a deceleration of the change over the next few minutes.

Answer:

You are right, the WGM shift may be induced by the RH change experienced by the droplet. While the moisture was switched from D_2O to H_2O, the H_2O needed ~1.6 min (chamber response time) to fill the chamber. However, during this period, dry nitrogen entered the chamber sustainedly while H_2O molecules delayed in the bubbler bottle and gas tubes. The true RH experienced by the droplet may hence decrease, inducing the WGM shift.

We have supplemented these interpretations in the revised manuscript (Section 3.2, Line 160-164).

#2. Regarding diffusion rates in hex-AS-water system - I remain surprised by the observation that any diffusion limitations are observed. However, given some of the observations reported by Richards et al. (DOI: 10.1126/sciadv.abb5643), this may not be totally unexpected if some of the ammonium ions remain in the organic-rich phase and influence the viscosity through ion-molecule interactions.

Answer:

Thanks for providing this enlightening reference. According to Richards et al., the supramolecular ion-organic interactions may exist when aerosols contain organics (specifically those containing vicinal hydroxyl groups) and inorganic divalent ions, which produces internal cross-linking molecular networks. Such ion-organic networks may form in the shell of H_2O+AS+HEX aerosol and thus inhibited water diffusion.

We have supplemented these interpretations in the revised manuscript (Section 3.4, Line 255-258).

If you have any query, please do not hesitate to contact me at the address below.
Thank you and best regards.

Yours sincerely,

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