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Comment on acp-2021-488

Anonymous Referee #2

Referee comment on "Molecular-scale description of interfacial mass transfer in phase-separated aqueous secondary organic aerosol" by Mária Lbadaoui-Darvas et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-488-RC2>, 2021

This manuscript reports on the MD simulation results of water uptake on LLPS media containing an outer shell of organic material and an inner aqueous core. These simulations show a temperature dependence, indicating entropic factors are important, shows that water accumulation is energetically favored at the in the organic phase at the organic/aqueous interface at 300K. The uptake of water from the organic phase to the core is limited at 300K due to the entropic cost of the orientational ordering of water molecules. No such free energy barrier exists at 200K. Both diffusion effects and uptake effects can cause water to accumulate in the organic shell, with a concentration maximum at the core/shell interface, leaving the vapor/shell interface depleted in water and thus able to maintain a reduced surface tension. The authors take these observations and apply them generally to explore the influence of these effects on cloud droplet formation via Kohler curve simulations.

Overall, the paper is well-written, and the manuscript discusses topics of broad interest that are relevant to the readership of the journal. My expertise does not allow me to robustly comment on the specific details of the simulations, but assuming the results can be trusted, this manuscript presents a compelling molecular-level description for the effects of LLPS in CCN activation. My comments below relate primarily to the configuration of the system interrogated in the simulations, and how this is representative of real aerosol systems.

1) The authors define the interface-normal which is the path along which impinging gas phase water will follow to interact with the system. It is unclear to me if only trajectories following a surface normal were simulated, or if angled trajectories and glancing impacts were considered. Would these show different results to those presented? How much does the momentum of the molecule impact its adsorption into the system?

2) There is a region of low density between the shell and the core, where both water and

hCPA drop to $\sim 5\%$ of their bulk densities. Does this imply there is a void in the system at this interface? Can the authors comment on and/or clarify this?

3) What factors determine the thickness of material required for it to appear "bulk-like"? The water shows bulk density within $< 0.5\text{nm}$, but the hCPA does not show bulk density across the full $\sim 2\text{ nm}$ shell.

4) What is the phase of water in the system at 200 K? In the atmosphere, this would presumably be ice, but there is no mention here as to whether the water is liquid or solid. How would a solid ice core affect the observations / conclusions? Are these simulations actually representative of the phases of matter that would be encountered at 200 K?

5) The presentation of data in figure 4(b) is somewhat confusing (I assume it's a bar graph, but this seems like a poor graph format when the bars for most of the data span the full height of the axis). Would suggest a data table in place of a graph here.