Response to RC2: 'Comment on amt-2021-35', Anonymous Referee #3, 02 Jun 2021:

We thank anonymous referee #3 for taking their time to carefully review our manuscript and provide detailed feedback.

"The authors examine the use of an electrical low pressure impactor (ELPI) to study phase changes in aerosol, focusing on efflorescence, deliquescence, and the formation of viscous amorphous liquid phases. They demonstrate results for ammonium sulfate that are consistent with expectations based on hygroscopic growth and phase change RH's. For sucrose, they show that the starting size is important due to the fact that the RH in the impactor decreases from the inlet to the lowest stage. This results in smaller particles showing more bounce, indicating the formation of a solid or semi-solid phase. Given the RH changes in the impactor, it seems that the observations can be explained in terms of RH-dependent size and viscosity for all these systems, along with size itself for the SOA system."

We would also like to emphasize the D_{50} diameters for each stage, which decrease as particles move down stream within the ELPI.

"Overall, the observations appear to make sense based on the described experiments. However, unless the RH can be made uniform across the impactor stages, it is not clear to me that this method could be reliably used for exploring RH-dependent phase. The authors do a good job at stressing the problems relating to the variable RH, but who is this work intended for? As far as I can tell, the only researchers using the ELPI for viscosity and phase measurements are the authors of this manuscript (please correct me if I'm wrong!)."

Due to the low pressure environment of the lower impaction stages, RH drops are inherent. The atmospheric science community is continuously developing and refining techniques to probe aerosol phase, and this method (or a similar method involving a low pressure impactor setup) has been used by (Järvinen et al., 2014), (Kidd et al., 2014), (Saukko et al., 2012; Saukko et al., 2015), (Virtanen et al., 2010), (Jain and Petrucci, 2015), (Slade et al., 2019), to name a few. One purpose of this manuscript is to highlight potential RH artefacts, which can appear when conducting experiments under high RH conditions.

"Generally, I think this manuscript should be published with some revision, but the authors need to be more clear about the aims of the work, and describe the broader implications. Are other impactor measurments of phase affected in similar ways? Can the problematic aspects of this experiment be harnessed for new insights?"

Other impactor measurements utilizing impactors that do not feature a pressure drop do not appear to be affected in similar ways, as discussed in the manuscript with references to (Tang et al., 2019) and (Bateman et al., 2015). Though, as noted, these methods have different limitations (such as the requirement for monodisperse aerosol populations only.)

"DRH of 87% is quite a lot higher than the literature - this is not an insignificant difference. Can the authors explain this difference in terms of uncertainty in RH probes or some other factors? 1% RH change per minute seems very fast for such a large chamber - likely this is the source. Where was the RH measured? Why not vary the RH more slowly to better equilibrate the chamber?"

We agree that the RH change per minute is relatively fast and likely accounts for the higher DRH, and this is noted in the manuscript. The RH was measured with a RH probe located inside the chamber (HMT 130, Vaisala Corp., Helsinki, Finland), as noted in the manuscript. Ideally, the RH change can be adjusted more slowly. However, our humidification and drying setup does not allow for this. Furthermore, wall losses within the chamber cause a decrease in aerosol population, meaning there is a limited time to probe our aerosol population.

"The impact design and change in RH means that small particles may enter viscous states before reaching the filter. A small aqueous particle that should splat on stage 4, for example, may instead become viscous and bounce down to stage 1. Authors should try to estimate the amount of expected water loss over 50ms in each stage to estimate impact of this RH gradient. For very small particles, 50 ms can be plenty long enough for gas-phase diffusion controlled evaporation to occur."

We agree that gas phase diffusion controlled evaporation may be occurring. Estimating the expected water loss over 50 ms in each impactor stage would involve many assumptions and approximations. We do not think that this estimation would add any information of value, as we are not attempting to quantify the water loss in this manuscript.

"In the discussion of the Kelvin effect, providing some estimates of the water activity in the smaller particles as a function of RH would be insightful to see the potential magnitude of this effect."

The purpose of this section is to highlight that the persistence of particle bounce is NOT likely due to the Kelvin effect. Therefore, we do not feel it is appropriate to estimate the water activity of the smaller particles as a function of RH in this work.

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