**Response to Reviewer #3:**

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

**Line 106. One of the major takeaways of this manuscript is that ELPI particle bounce measurements need to be corrected for variable RH at different impaction stages. The authors should describe the ELPI in greater detail in this section, so the reader can gain a greater understanding of the apparatus, possibly including a diagram in the main text or SI. In particular, the assumption (line 125) that all particles that bounce from any stage end up on the filter stage should be explained in more detail, as it is key to the data acquisition but is not obvious (at least to me). If there is any experimental data to backup this assumption, that should be presented as well.**

We agree, and have provided additional information for the ELPI in the Supplemental text. The reference in the Supplemental text and the cited literature within that reference provide additional information regarding the ELPI operating principle.

**Line 167. Figure 1 seems to show that the deliquescence curve, which should lead to particles exhibiting the same behavior between <40% RH up to the deliquescence point at 80% RH (at which point a discontinuity should occur), actually exhibits an intermediate change in the fractional delta_I between 40 and 80% RH. This is very surprising to me and the authors should offer some physical explanation to why this occurs. Without such an explanation, it is not clear to me how reliable these measurements to probe particle phase state, including when attempting to interpret SOA data later in the manuscript.**

During experiments the chamber RH was increased at a rate of 1% per minute, and a homogenous RH within the chamber could not be guaranteed. ELPI sampling also occurred at roughly 10 Liters per minute. Locally higher concentrations of gaseous water likely allowed for partial deliquescence (when viewing the chamber system as a whole) to occur earlier, prior to reaching 80% RH as measured by the humidity probe in one location of the chamber. Nevertheless, full and clear deliquescence and efflorescence transitions of the AS aerosol were observed utilizing this modified method. Again, the applicability of this method is not intended so much to determine accurately RH or deliquescence/efflorescence of any given aerosol, but rather to be able to infer the general phase state of SOA aerosol during its formation and aging.

**Line 185. I disagree with the statement that the efflorescence and deliquescence RHs are in good agreement with previous reports. While ERHs can vary, DRH should be close to 80%, and 87% is quite high. Some explanation of why this discrepancy might have arisen and how precise the authors believe the RH measurements are in light of this discrepancy should be included.**

This likely occurred due to the relatively higher rate of RH increase (during deliquescence). This has been shown to shift measured glass transition relative humidity values for glassy solids to higher values (Mikhailov et al., 2009). Furthermore, a homogenous RH within the chamber could not be guaranteed as the RH probe only measured one location within the chamber. We have now added a note in the manuscript.

**Line 198. The authors seem to operate their chamber essentially in batch mode where the RH is changed by diluting the particles, which causes an issue with low particle concentrations, as stated here. As a suggestion for future studies, the authors might consider operating their chamber as a continuously mixed flow reactor with particles continuously injected while sampling, and modifying the RH at the inflow of the chamber using a Nafion dryer, which should circumvent this issue.**

We appreciate this suggestion and have begun preliminary work to redesign our experimental setup to convert our chamber into a continuously mixed flow reactor.

**Line 203. Here and at several other points in the manuscript, the authors refer to particle bounce as being "shut down". This terminology seems imprecise and casual to me, and I suggest other terminology such as "eliminated", "nearly eliminated", "greatly reduced", or another term.**

We agree, and have replaced "shut down" with "eliminated".

**Line 231. Here and elsewhere in the manuscript, polydisperse GMD are reported but no measure of the polydispersity is included, which is important for the interpretation. Some measure of the polydispersity, such as a standard deviation, or, even better, actual measured particle distributions, should be included for every polydisperse sample.**

We agree with the reviewer and have edited the manuscript to include typical values of polydispersity (as measured by the geometric standard deviation). Also, typical particle size distributions have been provided in the Supplemental Information (**Figures S4-S6**).

**Section 3.4. The discussion of the Kelvin effect is currently vague and qualitative. I am not sure how much Figure 9 adds to the discussion and would consider removing it. What would be more quantitatively interesting and relevant would be seeing how much the RH changes from the RH a bulk phase or large (>1 micron) particle experiences for each of the particle diameters in the SOA data set. If it is only a few percentage points for the smallest sizes, then this effect should be negligible. However, if it is greater than this, then it would be worthwhile to further correct the ELPI RH for these smaller stages, with this Kelvin effect correction factor as well.**

We included this section to show that even for the case of the highest $C_{SOA}$ studied here (2,420 μg $m^{-3}$), there was no appreciable particle concentration (< 2% of total particle number density) below a diameter of 50 nm, which one might argue would not be of a critical size necessary for water uptake and so would remain solid and bounce. We therefore reason that the persistence of bounce cannot be attributed to the Kelvin effect. Figure 9 shows that there is no correlation between minimum $\Delta i_{fractional, t}$ achieved and percentage of the number concentration of particles below 100 nm (Figure 9), further supporting our suggestion that the Kelvin effect is not responsible for the observed residual bounce. As such, we believe that Figure 9 is important to show. Bulk phase or large (> 1 micron) particles were not the focus here, but certainly warrant their own studies, as they may be more important in the form of organic-coated salt particles in the marine environment. Their behavior with respect to the ELPI and RH may certainly differ from the behavior of the fine particles studied here.

**Section 3.5. This section partially addresses what I consider to be one of the two main takeaways of this manuscript: that the varying RH across ELPI stages could be used to study the kinetics of water uptake/evaporation in aerosol particles. However, this section mostly talks in general about prior literature results. Any relevant information here would fit better in the introduction and should be moved there. What would strengthen this section, and the manuscript as a whole, would be a discussion of how exactly the ELPI could be used to study water uptake/evaporation. Can the residence time in each stage be varied (by flow rate, or by modifying the apparatus itself) to study how long it takes for a particle to effloresce, for instance? Are there other experiments that would be particularly interesting to run in this area? This information would be of great interest to me.**

Unfortunately, our ELPI instrument is commercially produced and adjustments to the residence time via modifications to the flow rate or the apparatus itself are not possible, as this would alter instrumental parameters and affect the internal calculations. However, the ideas presented in this manuscript open the doors to future studies with custom built impactors that allow for modifications of parameters such as flow rate or particle resident times. This would allow for detailed, future studies of water uptake/evaporation. With this manuscript, we aim to show that the Dekati ELPI may not be suitable for measurements under high ambient RH and offer a word of caution for the community. We have edited the manuscript to include this section in the introduction, per the Reviewer’s suggestion.