Response to Reviewers:

Thanks for the reviewer's comments on our manuscript entitled "Observations on hygroscopic growth and phase transitions of mixed 1, 2, 6-hexanetriol/$(NH_4)_2SO_4$ particles: Investigation of liquid-liquid phase separation (LLPS) dynamic process and mechanism and secondary LLPS". The manuscript has been revised according to the reviewer's suggestions. The responses to the comments are given point-to-point below.

1. Line 65: I suggest authors to give a brief introduction about the atmospheric significance of 1, 2, 6-hexanetriol, the organic species investigated in this work. Has it ever been detected in ambient aerosols? Or it was selected as a model species according to the O:C ratio, similar to Bertram et al., (2011)? This should be clarified in Introduction.

   Author reply: Thanks for the reviewer's suggestion. The organic species in the mixed atmospheric aerosols consists of 1000s of different molecules, with only about 10% identified (Hallquist et al., 2009). In the present work, the 1, 2, 6-hexanetriol could act as a model organic species with O:C < 0.7. Thus, the 1, 2, 6-hexanetriol/AS mixed system represents a model system for mixed organic-inorganic aerosols undergoing LLPS. We have clarified it in the Introduction.

2. Line 100: Why not calculate GF using the image areas of particle at different RH and that of dry particle? How to estimate the diameter for the irregular or non-spherical particles in the software, especially for particles in effloresced state? The deviation between these two approximation methods should be estimated in this section. In addition, have the imaging pixel been calibrated in your measurement?

   Author reply: Thanks for the reviewer's suggestion. The hygroscopic growth of aerosol particles is generally expressed as mass growth factors, i.e., the mass of particles at a given RH divided by the mass of dry particles (Ma et al., 2019), or size growth factors, i.e., the diameter of particles at a given RH divided by the diameter of dry particles. In the present work, the size growth factors of mixed particles were determined by the optical images with an image analysing software to explore the hygroscopic behaviours of mixed particles, similar to the treatment of Sun et al. (2018). Indeed, the size of particles after efflorescence was estimated approximatively in the software, but the error of growth factors caused by such the approximation was negligible compared with the large GF values of homogeneous aqueous droplets. The ratio of image area of the OIR = 1:1 particle at different RH to that of the effloresced particle is determined by the same software, as shown in Fig. R1. It is clear that the image area ratio and the size growth factors show similar trends regarding the hygroscopic behaviours of the OIR = 1:1 particle. In addition, we have calibrated the imaging pixel with a fixed pixel and size ratio (1 µm = 10.667 pix).

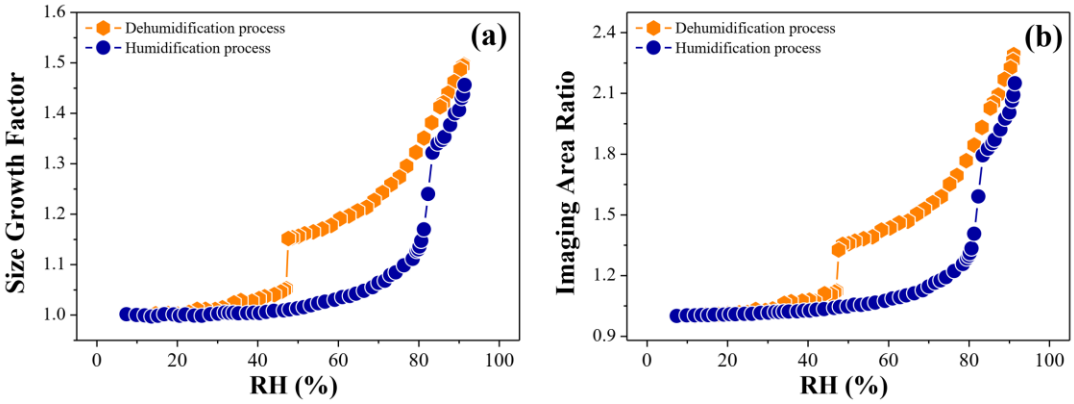

Figure R1: Size growth factors (a) and imaging area ratio (b) of mixed 1, 2, 6-hexanetriol/AS particles with OIR = 1:1 during the RH cycle.

3. Line 149-150: "the continuous water release would cause a gradual increase in sulfate concentration in the inner phase, which ultimately results in the occurrence of secondary LLPS.". What cause the secondary LLPS? Why was it not observed in the study of Bertram et al. (2011)?

Author reply: First of all, the phase separation in the mixed particles can be attributed to the salting out effect, i.e., the decrease in the solubility of organics in an aqueous salt solution. The correlation of the solubility of organics, S, and the concentration of the salt, C_s, can be expressed by the Setchenov equation (Lee, 1997): In $S/S_0 = k_sC_s$, where S_0 is the solubility of organics in water without the salt, k_s is the Setchenov constant. Second, as confirmed by our Raman spectra in the present work, there were a small amount of AS and 1, 2, 6-hexanetriol present in organic-rich and sulfate-rich phases, respectively. Thus, as RH decreased after LLPS, the concentration of AS in the inner phase increased significantly and the solubility of organics decreased, resulting in the formation of more concentrated AS inclusions in the inner phase due to the salting out effect, similar to the occurrence of LLPS. In the study of Bertram et al. (2011), the size of observed particles was around 10-30 µm, while that was 55-80 µm in our work. We speculate that the secondary LLPS may not be clearly observed in the case of smaller particle size.

4. Line 178 and 179: I suggest to first introduce the result of mixed particles with OIR=1:4, followed by that of OIR=1:2. Also suggest for Line 200, Figure 4, 5 and 8 and related statements in main text, in an order with increasing molar fraction of organics.

   Author reply: Thanks for the reviewer's suggestion. We have adopted reviewer's advice and revised our manuscript accordingly.

5. Line 192-194: Please provide appropriate references to support the argument.

   Author reply: Thanks for the reviewer's suggestion. We have adopted the reviewer's advice in the revised manuscript.

6. Line 240: When compared the phase transition RH with different OIR, I suggest to summarize the DRH, ERH and SRH values in a table in the revised manuscript for clear presentation. Of course, the DRH and ERH of AS, the results for the same systems investigated in previous study, i.e., Bertram et al. (2011), should also be included for comparison.

   Author reply: Thanks for the reviewer's suggestion. We have adopted the reviewer's advice. The Table S1 summarized the DRH, ERH and SRH of mixed particles with different OIRs has been placed in the Supplement.

7. Line 250 and Figure 8: Please clearly mention the temporal changes of LLPS dynamic process, not including secondary LLPS in the revised manuscript.

   Author reply: Thanks for the reviewer's suggestion. We have adopted the reviewer's advice in the revised manuscript.

8. Figure 3(c): Please add the error bars.

   Author reply: Thanks for the reviewer's suggestion. We have adopted the reviewer's advice and revised our manuscript accordingly.

Reference:

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Lee, L. L.: A molecular theory of Setchenov's salting-out principle and applications in mixed-solvent electrolyte solutions, Fluid Phase Equilibr., 131, 67-82, 1997.

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