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Reply on CC1

Shuaishuai Ma et al.

Author comment on "Observations on hygroscopic growth and phase transitions of mixed 1, 2, 6-hexanetriol/(NH₄)₂SO₄ particles: investigation of the liquid-liquid phase separation (LLPS) dynamic process and mechanism and secondary LLPS during the dehumidification" by Shuaishuai Ma et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-1299-AC1>, 2021

Thanks for the reviewer's comments on our manuscript entitled "Observations on hygroscopic growth and phase transitions of mixed 1, 2, 6-hexanetriol/(NH₄)₂SO₄ particles: Investigation of liquid-liquid phase separation (LLPS) dynamic process and mechanism and secondary LLPS". The reviewers' comments are helpful for improving the quality of our work. The responses to the comments are given point-to-point below.

- Line 37: Mixed organic-sulfate particles can be regarded as model systems for troposphere aerosols? I think it is an inappropriate expression. The 1, 2, 6-hexanetriol/AS may be a model system for mixed aerosols that undergo LLPS, but it isn't a model system for atmospheric aerosols.

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

- The authors suggested that the formation of brighter aqueous phase in the center of inner AS solution phase indicated the occurrence of secondary LLPS. How did the authors determine that this phenomenon come from secondary LLPS, not the optical effect?

Author reply: Thanks for the reviewer's suggestion. As mentioned in the text, the bright globe in the center and the dark ring at the edge were owing to the optical effect of light scattering when the particle existed in homogenous mixed phase. However, the optical effect could not cause any visual errors during the phase transitions, in other words, the dynamic processes of LLPS and secondary LLPS were not affected by the optical effect. Besides, the secondary LLPS processes could be clearly distinguished by the movies in the Supplement.

- In Fig. 3, b2 represented the intensity ratio of stretching vibration bands of crystalline SO₄²⁻ to C-H, which differed with other intensity ratios, please indicate this in the caption.

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

- Line 192: What does the “morphological changes upon crystallization” refer to? How can the authors conclude that aqueous 1, 2, 6-hexanetriol will enter into the veins of the AS crystal and then is enclosed by a crystalline AS crust?

Author reply: As shown in Fig. S4, the morphology of the OIR = 1:4 particle became more irregular and darker as the RH decreased. This indicated that the crystal growth continued at lower RH after efflorescence. Thus, we conclude that aqueous 1, 2, 6-hexanetriol enters into the veins of the AS crystal and then is enclosed by a crystalline AS crust. More importantly, the particle size remained constant until 84.0% RH, differing from the particles with the other OIRs.

- Line 212: The secondary LLPS occurred at ~77.9% RH, almost the same RH as the appearance of LLPS, for the OIR = 1:4 particle with the highest sulfate fraction. Thus, whether the sulfate fraction can determine the RH at which the secondary LLPS occurs?

Author reply: The secondary LLPS occurred at ~68.6%, 69.0%, and 77.9% RH for OIR = 1:1, 1:2 and 1:4 particles, respectively. Moreover, no secondary LLPS was observed for OIR = 2:1 and 4:1 particles. It is clear that the higher sulfate fraction tended to cause the occurrence of secondary LLPS, but the relationship between the sulfate fraction and the RH at which secondary LLPS occurs remains unclear and needs to be further investigated.

- Line 245: The measured SRH in this work was slightly higher than the measurement results (~71.0% RH) by Bertram et al. (2011). Please discuss some of the sources of this discrepancy.

Author reply: Indeed, the measured SRH values in our work were around 75.0%, slightly higher than the measurement results of 71.0% RH by Bertram et al. There are several possible sources of this discrepancy: First, the particle size employed in our work (55-80 μm) was larger than that in Bertram’s work (10-30 μm); second, in both cases, the uncertainty in the measured SRH was 2.5% RH; finally, the rate of RH changes was 0.06-0.07% RH s^{-1} in our work, higher than that of 0.4-0.6 % RH per minute in Bertram’s work, probably causing higher measured SRH values in our work.

- The sulfates are ubiquitous in atmospheric aerosols, but the concentration of 1, 2, 6-hexanetriol is limited. Why did the authors choose the 1, 2, 6-hexanetriol/AS as the research system?

Author reply: In Bertram’s work, LLPS in mixed sulfate-organic particles commonly occurred when the O:C < 0.7, while for O:C > 0.7, no LLPS was observed. Based on this, the 1, 2, 6-hexanetriol can act as a model organic matter with O:C < 0.7 and mixed 1, 2, 6-hexanetriol/AS represents a model system for mixed organic-inorganic aerosols which would undergo LLPS upon the RH fluctuation. Thus, we choose the 1, 2, 6-hexanetriol/AS system to investigate the LLPS dynamic process and mechanism of mixed aerosols.