

This work investigated the hygroscopic growth and dynamic phase evolution processes of mixed 1, 2, 6-hexanetriol/AS particles. The dynamic LLPS processes and LLPS mechanisms for mixed particles with different organic-inorganic mole ratios were depicted quantitatively. More interestingly, the secondary LLPS was first explored when RH continued to decrease after LLPS. This work performs a comprehensive and detailed analysis on the complex LLPS behaviors of mixed organic-inorganic aerosols and provides insights into equilibrium partitioning processes of organic and inorganic components. Thus, I support the publication of this paper in ACP.

Comments:

1. 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.
2. 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?
3. In Fig. 3, b2 represented the intensity ratio of stretching vibration bands of crystalline SO_4^{2-} to C-H, which differed with other intensity ratios, please indicate this in the caption.
4. 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?
5. 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?
6. 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.
7. 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?