

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2020-1299

Anonymous Referee #2

Referee 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-RC2>, 2021

The work by Ma et al. investigates phase transitions and hygroscopic growth of mixed 1,2,6-hexanetriol/ammonium sulfate particles with different mixing ratios using an optical microscope and Raman spectroscopy. They found liquid-liquid phase separation (LLPS) behaviors of the mixed particles driven by non-ideal thermodynamics. Interestingly, the authors suggest a secondary LLPS formed inside the inner ammonium sulfate solution phase, possibly because of the formation of more concentrated AS inclusions. Measurements of LLPS behaviors for inorganic-organic species have important implications for understanding the morphology, gas-particle partitioning, and CCN activity of the tropospheric aerosols. The manuscript is in general clearly written and within the scope of ACP. I would support publication once authors address the following comments/concerns.

Major comment:

The authors claimed that more concentrated AS inclusions form inside the inner sulfate-rich core, and this is defined as a secondary LLPS. Optical images indeed indicate heterogeneity in the inner core. The Raman spectra showing different sulfate/organic ratios in Fig 3, however, were actually taken from different RH values. I am not fully convinced that these Raman spectra indicate a secondary LLPS within the inner core. Do Raman spectra within the inner core measured at the same RH show different intensity ratios? What is the thermodynamics explanation of the proposed secondary LLPS? Is the secondary LLPS an equilibrium state or a transitional state?

Minor comments:

- Introduction: it might be worth to mention that LLPS can also occur for organic mixtures, such as in secondary organic aerosol (e.g.,

<https://acp.copernicus.org/articles/16/7969/2016/>
<https://acp.copernicus.org/articles/17/11261/2017/>;
<https://www.nature.com/articles/s41467-018-06622-2>)

- 2 state in the caption that LLPS and secondary LLPS are measured during the dehumidification
- Page 4 Line 119-120: slowly -> gradually, rapidly-> abruptly. Changes of GF values with RH are thermodynamic processes, rather than kinetic processes.