

#### Reviewers Comments:

The authors investigate the phase transition behaviors and resulting mixing state of phase-separated aerosol consisting of 1,2,6-hexanetriol, ammonium sulfate, and surfactants. In order to study on surfactant concentration effect on phase separation of mixture particles, phase separation including core-shell structure, partial organic-phase engulfing, and inorganic-phase engulfing is observed for particles consisting of 1,2,6-hexanetriol and ammonium sulfate by adding surfactant from very low to high concentration. It is found at very low concentrations of hydrocarbon surfactants, the organic phase partially engulfed the aqueous inorganic phase, but the inorganic phase partially or fully spread over the organic domain at higher surfactant levels. These distinctive mixing states may substantially alter the physicochemical properties, and optical properties of organic-inorganic aerosols under real atmospheric conditions, which help to bridge a critical knowledge gap regarding the role of surface tension evolution in the equilibrium particle morphology of internally mixed atmospheric particles and its potential impact on aerosol-chemistry-climate interactions. The topic is interesting and emphasis of surfactant concentration effect on phase separation in the mixed particles. However, there are lots of description on investigating LLPS, driven by the non-ideal thermodynamic behavior of organic-inorganic mixed aerosols, was strongly regulated by hydrocarbon and fluorocarbon surfactants. These surfactants modified the surface and interfacial tensions between organic and inorganic phases. There is a lack of phase separation mechanism for mixed particles consisting of 1,2,6-hexanetriol and ammonium sulfate by adding surfactant from very low to high concentration. In addition, many studies shows that ratio of inorganic: organic results in phase separation. The comparison is typical supposed to further explore this mechanism for inorganic-organic particles.

Specific comments are listed below:

Page 2 line 29-30: “These mixed organic-inorganic aerosols can undergo phase transitions, such as liquid-liquid phase separation (LLPS), efflorescence during dehumidification, or deliquescence upon humidification.” Please distinguish the efflorescence and dehumidification, uniform the “efflorescence” or “dehumidification”

Page 2 line 30-31: “The resulting mixing states, such as liquid-liquid or liquid-solid equilibria,” please rewrite them.

Page 2 line 46: “In addition, several other factors such as the organic fraction (Ciobanu et al., 2009; Bertram et al., 2011; Song et al., 2012a), inorganic species (You et al., 2013), drying rate (Altaf and Freedman, 2017), particle size (Ohno et al., 2023; Freedman, 2020; Kucinski et al., 2019), and bulk viscosity (Ye et al., 2025), can also influence LLPS occurrence.” Please detail these effects on the LLPS occurrence.

Page 2 line 49: “However, limited studies have focused on the mixing states of biphasic organic-inorganic particles.” to data, there are many studies on mixing states of biphasic organic-inorganic particles, please rewrite them.

Page 2-3 line 86-96: it seems that there is no strong motivation. Please rewrite them.

Page 11 Figure 3c, Evolution of SDS concentrations with AS or 1,2,6-hexanetriol in the mixed particles. actually, I am confused about this figure. Would be possible for mixture particles consisting of AS, 1,2,6-hexanetriol, and SDS to directly measure its surface tension?

Page 18 line 385: “high concentrations” to “high concentration” please check grammar, sentence structure, and so on.