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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Liquid–liquid phase separation in organic particles consisting of α-pinene and β-caryophyllene ozonolysis products and mixtures with commercially-available organic compounds" *by* Young-Chul Song et al.

Anonymous Referee #2

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In this work, the authors provide new valuable experimental data related to the liquid-liquid phase separation (LLPS) of aqueous droplets containing single or two components found in the ozonolysis α-pinene- and β-caryophyllene. The findings of works (e.g. relationship between LLPS and O/C) give us greater insights into the phase state of atmospheric aerosols under different environments, which largely govern many important atmospheric processes such as water uptake and CCN activities. I support the publication of this work and have some comments/suggestions for the authors' consideration.

Comments

In the introduction, the authors should provide more information why these classes of compounds are selected for this study. What are the atmospheric significances and abundances of these selected species? What the knowledge gap related to LLPS would like to be filled by investigating these compounds?

Page 3, Line 78, "Seven of the products from the ozonolysis of α-pinene and β-caryophyllene were synthesized. The detailed synthesis methods for these species are described in Bé et al. (2017)." Please provide the purity of these synthesized chemicals used in this study.

Page 5, Line 113, "At the beginning of LLPS experiments, organic particles inside the flow-cell were equilibrated at $\sim$100% RH for 15–20 min.". When the experiment ran at $\sim$ 100%RH, does the condensation of water vapor on the surface of hydrophobic substrate and flow-cell affect the LLPS measurements?

Page 5, Line 119, "Organic particles were selected in the diameter range of 30–100 μm, which was required for LLPS experiments." Could these results be applicable to submicron sized aqueous droplets?

Page 5, Line 125, "Out of the eleven different types of one-component organic particles studied, eight underwent LLPS during humidity cycles (Table S1)." Could the authors comment how the chemical structure of the investigated compounds determine the occurrence of LLPS?

Page 5, Line 133, "Particles of β-caryophyllonic acid and β-nocaryophyllonic acid had a partially engulfed morphology after LLPS (Fig. 1b, d and Movies S2, S4) (Kwamena et al., 2010; Reid et al., 135 2011; Song et al., 2013) while the others particles had a core-shell morphology after LLPS." Could the authors comment how the chemical structure of the investigated compounds determine the morphology of the organic particles after LLPS?

Page 6, Line 164, “For comparison purposes, also included in Fig. 4 is the miscibility boundary of organic compounds based on the BAT model (Gorkowski et al., 2019).” Could the authors elaborate whether the BAT model can predict the RH at which LLPS occurs for the investigated compounds? Could the authors comment how the functional groups of the investigated compounds determine the occurrence of LLPS?

Page 7, Line 192, “In contrast, in experiments with particles containing ozonolysis products mixed with pyruvic acid, phase separation began with the growth of a second phase at the surface of the particle as the RH increased (Figs. 6c, d and Movies S22, 23).” Can the authors elaborate or explain this observation? What are the causes or mechanisms?

Atmospheric implications: can the authors further elaborate how different morphologies of the organic particles after LLPS affect atmospheric processes?

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