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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 #3

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General Comments:

In this manuscript, authors investigated the liquid-liquid phase separation (LLPS) as a function of average O:C ratio in organic particles free of inorganic species containing one component and binary mixture of α-pinene and β-caryophyllene-derived ozonolysis products and commercially available organic species. Compared to previous studies on this topic, this work used atmospherically relevant SOA products and showed that increased complexity of particulate organic species widen the range of O:C ratios over which LLPS will occur, improving our understanding of the LLPS behavior and

providing better constrain of the O:C range required for LLPS. I am supportive of the publication of this manuscript on Atmospheric Chemistry and Physics with the following comments/suggestions for the authors to consider in their revision.

Specific Comments: 1) Lines 163-171 and Figure 4: As indicated in the Gorkowski et al. (2019), the BAT model was intended for use to represent thermodynamics for with only bulk O:C information rather than a specific single organic system. It is not clear how the BAT model result was generated here. Is it simply a reproduction of the Figure 2 in the original paper (Gorkowski et al., 2019)? If it is, the comparison here doesn't seem to be fair. Or some modifications were made to tailor the model to the organic species studied in this work? If this is the case, could author include a section in the SI to describe the parameters and assumptions chosen when using that BAT model to generate the result shown in Figure 4? Either way, the discussion on Figure 4 doesn't seem to be sufficient. Could the author elaborate more on what implications one could draw from the discrepancies between the BAT model and observations? Especially if the model wasn't used in a system it was designed for the comparison here was potentially misleading. Given the complex composition and matrix effect within the ambient aerosols, it might be more appropriate to compare the observation vs. model comparison for the two component particles compared to one component particles.

2) Figure 3b showed that several points of LLPSlower RH were significantly lower than what the Sigmoid-Boltzmann fit would predict. It is obvious that O:C ratio is not a single determinant for LLPS. Authors should comment on possible explanations (relevant properties of the organic species, functional groups, spread in O:C values, etc.) for the variations of LLPSlower for two component organic particles.

Minor Comments: 1) On lines 132-133 β-caryophyllinic acid was discussed while the labeling on Figure 1e as well as in the caption was β-noncaryophyllininc acid.

2) It is hard to read the black texts of RH on top of the dark optical images. I would suggest either changing the color of the texts or not overlaying the labels and the

images.

3) Authors are recommended to double check the manuscript for grammatical errors. For example, on line 199, “When LLPS was observe” should be “When LLPS was observed”.

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