

# ***Interactive comment on “Liquid-liquid phase separation in particles containing secondary organic material free of inorganic salts” by Mijung Song et al.***

**Mijung Song et al.**

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David Topping, Co-Editor of ACP

Dear David Topping,

Listed below are our responses to the comments from the reviewers of our manuscript. We thank the reviewers for carefully reading our manuscript and for their very helpful suggestions! For clarity and visual distinction, the referee comments or questions are listed here in black and are preceded by bracketed, italicized numbers (e.g. [1]). Authors' responses are in red below each referee statement with matching numbers (e.g.

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[A1]).

Sincerely,

Allan Bertram Professor of Chemistry University of British Columbia

Response to Referee #2 (Reviewer comments in black text)

Summary. Previously, it was found that at high RH (>95%),  $\alpha$ -pinene-derived SOM particles free of inorganic salts can undergo LLPS, while isoprene-derived SOM particles free of inorganic salts do not. In this paper, additional SOM particles free of inorganic salts were studied. It was found that SOM generated from ozonolysis of caryophyllene and limonene that are similar to  $\alpha$ -pinene-derived SOM, while particles generated by photo-oxidation of toluene that are similar to isoprene-derived SOM. In addition to the LLPS information, the authors found a relationship between occurrence of LLPS and the average oxygen-to-carbon elemental ratio (O:C) of the organic material. Low O:C ratio resulted in LLPS. Publication is recommended with minor revisions. Discussion paper Concerns.

[1] On page 5, the authors mention that 20 – 80 micron diameter particles are required for LLPS. Is there a reference to that? Why is there a size dependence in the observations?

[A1] The resolution of the microscope used in the current experiments was roughly 1 micron. From experience, detection of LLPS with our microscope setup is the clearest when the size of the particles are roughly 20 - 80 micron, although smaller sizes are possible with optical microscopy. We will re-phrase the sentence referred to by the referee for clarity. We did not observe a size dependence for the LLPS phase transition for the relative narrow range of sizes investigated.

[2] In addition, is there a RH rate of change dependence? More discussion is needed.

[A2] We did not observe a dependence of LLPS on the RH ramp rate, although only a narrow range of rates were used. This information will be added to the revised

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manuscript.

[3] While the observation of LLPS or no LLPS with SOM derived in various ways is interesting and important to the atmospheric chemistry community, the manuscript would be stronger with proposed explanations or discussions on \*why\*. Discussion on differences in  $\alpha$ -pinene-derived SOM and isoprene-derived SOM structures, or the structures of the components studied here might have been helpful.

[A3] We certainly agree that an explanation of “why” LLPS is observed in some cases but not others is important. Previous thermodynamic modelling studies give some explanations for why LLPS occurs in  $\alpha$ -pinene-derived SOM but not isoprene-derived SOM (See Renbaum-Wolff et al., 2016; Rastak et al., 2017). We hope to explore this question in more detail in the future by investigate LLPS in organic particles containing mixtures of commercially available organic compounds.

[4] In Table 1, some MRH values are higher than the SRH values. This seems counterintuitive. Discussion is needed justifying these observations.

[A4] The uncertainty in MRH and SRH values reported in Table 1 is  $\pm 2.0$  % RH, due to the uncertainty in the RH measurement. As a result, the SRH and MRH values agree within the uncertainty of the measurements. To address the referee’s comment, in the caption to Table 1, we will point out that the uncertainty in the MRH and SRH is  $\pm 2.0$  % RH, due to the uncertainty in the RH measurements.

[5] How are the SOM mass concentrations at phase change calculated (figure 2, x-axis)?

[A5] The mass concentrations of SOM were determined from measurements of the number-diameter distribution of SOM particles in the flow tube reactor or OFR. This information will be added to manuscript for clarity.

[6] Is the volume of the droplet known? What is the change in contact angle of the droplet with the surface, as a function of RH?

[A6] The contact angle of the droplets with the surface was not measured in our experiments, and hence an accurate volume of the droplets was not known. From the optical images, we only determined the projected diameter of the droplets.

[7] Typo. Page 8 – 2nd line – “observed”. It should be “observe”. [A7] This change will be made in the revised manuscript.

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