

The paper investigates the liquid-liquid phase separation (LLPS) in organic particles. LLPS was observed when the O:C ratios of the particles were ≤ 0.44 for single organic species and ≤ 0.58 for mixtures containing two organic compounds. Most of the LLPS can be observed at RH close to 100%. The authors demonstrate that the LLPS may have important implications on the CCN properties of organic particles. The manuscript is well written, and the results are clearly presented. I would like to recommend this manuscript to be published in *Atmos. Chem. Phys.* if my following concerns are fully addressed.

1. Page 4, Line 8

The dimensions of the particles were measured around 30-80 μm . Depending on the particle generation methods, sizes of the particles used in the LLPS studies in the literature vary from 1 to 80 μm . 80 μm used in this study is at the higher end. Does particle size affect the results of LLPS?

2. Page 5, Line 29

- a. Have the authors tried nonanedioic acid ($\text{C}_9\text{H}_{16}\text{O}_4$) and tetradecanedioic acid ($\text{C}_{14}\text{H}_{26}\text{O}_4$)? These are the isomers of suberic acid monomethyl ester and diethyl sebacate with the same O:C ratio.

One may expect different results with these isomers. For example, due to the stronger polarity and hydrogen bonding of acids as compared to esters, the acid compounds may have stronger interactions with water molecules, leading to higher SRHs. Or no LLPS may be observed. These effects may actually be helpful to explain why the authors did not observe LLPS with poly (propylene glycol) and PEG-400. Can you comment on whether/how the intermolecular interactions between organic compounds and water affect LLPS?

- b. In contrast to the experiments with organic + ammonium sulfate conducted in the same group (e.g., Bertram et al.¹ and You et al.²), no LLPS was observed in this study using the same organic compounds (i.e., polyethylene glycol, PEG-400, and diethyl-L-tartrate). I wonder if the authors have any explanation regarding the different observations between these two studies.

3. Page 6, Line 2 and Table 3

What is the molar/mass ratio between the two organic compounds in the mixtures in Table 1? Is it 1:1? Have you tried any other mixing ratios? When nebulizing the mixture, how can the authors be sure that the composition of individual particle was the same as its original mixture?

4. Page 6, Line 6

- a. It is interesting that adding PEG-400 which itself did not give LLPS, expanded the LLPS RH range of other compounds. Does it simply due to the change of O:C ratio? If not, please explain it in more detail.
- b. For polypropylene glycerol/PEG-400, no LLPS was observed in the single organic species scenarios. But mixing them together yielded LLPS with a low SRH (~75%). Have the authors tried any other mixtures of the following: diethyl sebacate, glyceryl tributyrate and suberic acid monomethyl ester? I wonder if the SRH and LLPS RH range will also change? Can you comment on this?

5. Page 8, Line 7

SOM is a bulk mixture. Why the results of SOM are similar to those from single organic components (i.e., LLPS occurred with O:C ratio ≤ 0.44 ; narrower LLPS RH range was observed)? Is it just a coincidence? It seems that O:C ratio solely cannot fully explain the results. Can the authors comment on this?

Minor comments

1. Page 4, Line 10: “deposing”. Should it be “depositing”?
2. Page 17, Table 1: For diethyl-L-tartrate, I don’t think there are ether group and carboxylic acid group.

Reference

- (1) Bertram, A. K.; Martin, S. T.; Hanna, S. J.; Smith, M. L.; Bodsworth, A.; Chen, Q.; Kuwata, M.; Liu, A.; You, Y.; Zorn, S. R. Predicting the relative humidities of liquid-liquid phase separation, efflorescence, and deliquescence of mixed particles of ammonium sulfate, organic material, and water using the organic-to-sulfate mass ratio of the particle and the oxygen-to-carbon elemental ratio of the organic component. *Atmos. Chem. Phys.* **2011**, *11* (21), 10995–11006.
- (2) You, Y.; Renbaum-Wolff, L.; Bertram, A. K. Liquid-liquid phase separation in particles containing organics mixed with ammonium sulfate, ammonium bisulfate, ammonium nitrate or sodium chloride. *Atmos. Chem. Phys.* **2013**, *13* (23), 11723–11734.