

Thank you for your helpful feedback on the manuscript. Detailed responses to the comments and related manuscript revisions will be included following input from other comments and formal reviews. The following minor revisions have been specifically included in the revised manuscript.

- 1. There seems to be a discrepancy between Fig 2A caption and the rest of the text. Fig. 2A caption says the system is glutaric acid and water but in the discussion of the figure in the text (~line 307 page 11) ethanol/water is mentioned. I also see ethanol but not glutaric acid in Table S3.**

The text has been updated to correctly refer to glutaric acid instead of ethanol. The text now reads as follows: “It is shown that all three approaches can reasonably approximate the measured surface tensions of the finite volume droplets, with the volume-fraction-based approach performing better than the mole-fraction-based approach for the water and **glutaric acid** system.”

Table S3 has also been updated with the appropriate molar mass, pure component surface tension, and corresponding reference for glutaric acid.

- 2. It would be helpful to the reader to have a note somewhere (maybe near the top of section 3.1) that the dry aerosol component mass fractions for all systems investigated are in Table S3. Table S3 would also be more clear if it a) had the figure number where the system result is shown or b) was in the same order as systems are introduced in the main text**

The following text has been added to the top of section 3.1:

“Several of3 those methods rely on the assumption that hypothetical gas–liquid surface tensions of the phases involved can be estimated based on a weighted average of the pure-component surface tensions, σ_i° . **A complete list of all σ_i° for the components of the systems examined in this work can be found in Table S3. Also reported in Table S3 are the organic-to-inorganic dry massratio (OIR) for each system.** In order to...”

A column has been added to Table S3 to include the figure panel number that each system in the table refers to and the table has been sorted by the first appearance of each system in the manuscript.

- 3. There seems to be a discrepancy between Fig 4C caption and the rest of the text. Fig 4C caption is labeled dodecane-lithium-chloride, but in Table S3 and main text (~line 333, page 12) dodecane-KCl is mentioned.**

The figure caption for figure 4C has been updated to correctly refer to the dodecane potassium chloride system rather than the dodecane lithium chloride system that was incorrectly referenced. The caption text now reads as follows :

“Predicted $\sigma^{\alpha\beta}$ values (curves) compared to measurements (solid circles). The x-axis scales correspond to those used in the experimental data references. Data and

predictions for all systems are for $T = 298 \text{ K}$. (A) A water–PEG-400–ammonium-sulfate system with experimental data by Song et al. (2013), (B) a water–benzene–sodium-chloride system (Harkins and Humphery, 1915), (C) a water–dodecane–potassium-chloride system (Aveyard and Saleem, 1976), and (D) a water–benzene–methanol system (Pliskin and Treybal, 1966; Paul and De Chazal, 1967). The four distinct parameterizations for interfacial tension from Table 1 are shown (except for the No IFE case). The pure-component surface tensions of organic components can be found in Table S3.”

- 4. page 14 line 399 - during the discussion of Fig. 7, I do not understand the reference to Fig S2 which is for PEG400 - AS (not one of the systems in Fig. 7). OIR is also mentioned on this line but is not defined in the main text.**

The text has been updated to refer to Figure S4, which corresponds to the systems shown in Figure 7. The text now reads as follows:

“The corresponding values of $\sigma^{\alpha\beta}$ at the onset of LLPS are shown in Fig. S4 and the water-free OIRs are listed in Table S3.”

- 5. small typo, there is an extra '-' in 10 - 50 nm on line 527.**

The extra '-' on line 527 has been deleted.