

Response to Reviewers:

Thanks for the reviewer's comments on our manuscript entitled "Surfactants regulate the mixing state of organic-inorganic mixed aerosols undergoing liquid-liquid phase separation". The reviewers' comments are helpful for improving the quality of our work. The responses to the comments and the revisions in manuscript are given point-to-point below.

Reviewer #2:

Figure 3: It would be worthwhile to discuss the pure surface tension of 1,2,6-hexanetriol and how it compares to lower-surface-tension organics or typical secondary organic aerosols. This discussion could be added near Figure 3 or incorporated into the main discussion section. My reasoning is that a lower-surface-tension organic compound would likely remain as the particle-engulfing phase for a longer period (as surfactant concentration increases) and may not transition to a core-shell morphology as readily.

Author reply: Thank you for the reviewer's constructive suggestion. The surface tension of pure 1,2,6-hexanetriol was measured to be 52.02 mN m^{-1} . For aqueous 1,2,6-hexanetriol solutions, the surface tension decreased from 67.43 to 56.37 mN m^{-1} as the concentration increased from 0.17 to 2.71 mol L^{-1} . These values are consistent with those of typical secondary organic matter in tropospheric aerosols. For instance, McNeill's group found that hemiacetal oligomers and aldol condensation productions formed from aqueous reactions of formaldehyde, acetaldehyde, glyoxal, and methylglyoxal reduced the surface tensions of pure water and AS solutions to a minimum value of approximately 45 dyn cm^{-1} (Li et al., 2011; Schwier et al., 2010). Hritz et al. (2016) measured the surface tension of ozone-oxidized α -pinene particles using atomic force microscopy and reported values of 27.5 and 44.4 dyn cm^{-1} at $< 10\%$ and $67\% \text{ RH}$, respectively. Similarly, Upshur et al. (2014) reported that aqueous solutions of isoprene oxidation products exhibited the surface tension values around 60 mN m^{-1} at an organic concentration of 30 mM .

Given that the average O:C ratio of atmospheric SOA is around 0.52 , 1,2,6-hexanetriol with an O:C ratio of 0.5 was selected to represent moderately oxidized SOA (Canagaratna et al., 2015; Shen et al., 2022). In contrast, primary organic aerosols with lower O:C ratios and lower surface tensions, such as decane, octanol, and oleic acid, tend to form core-shell or partial engulfing,

rather than inverse core-shell morphologies when mixed with aqueous inorganic phases in the presence of surfactants (Reid et al., 2011). In such systems, the organic-phase surface tension (σ_{23}) is low, while the organic-inorganic interfacial tension (σ_{12}) remains relatively high (Gorkowski et al., 2020), yielding negative spreading coefficients S_1 according to Equation (3) in the manuscript. Consequently, inverse core-shell structures are unlikely to occur, even at high surfactant concentrations.

Lines 313-322: Note that the surface activity of 1,2,6-hexanetriol is comparable to that of secondary organic matter in the real atmosphere (Li et al., 2011; Schwier et al., 2010; Hritz et al., 2016; Upshur et al., 2014). For instance, McNeill's group reported that hemiacetal oligomers and aldol condensation productions formed from aqueous reactions of formaldehyde, acetaldehyde, glyoxal, and methylglyoxal reduced the surface tensions of pure water and AS solutions to approximately 45 mN m⁻¹ (Li et al., 2011; Schwier et al., 2010). Considering that 1,2,6-hexanetriol represents moderately oxidized SOA, primary organic aerosols (e.g., octane and octanal) with lower O:C ratios and lower surface tensions tend to form core-shell or partial engulfing morphologies, rather than inverse core-shell structures, when mixed with aqueous inorganic phases in the presence of surfactants (Reid et al., 2011). In such systems, the σ_{23} is low and σ_{12} remains relatively high (Gorkowski et al., 2020), resulting in negative S_1 values according to Equation (3). Consequently, inverse core-shell structures are unlikely to occur, even at high surfactant concentrations.

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