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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Liquid-liquid phase separation and viscosity within secondary organic aerosol generated from diesel fuel vapors" *by* Mijung Song et al.

Anonymous Referee #1

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Th manuscript describes the analysis of secondary organic aerosol (SOA) samples generated from diesel fuel vapours, concentrating on observations of liquid-liquid phase separation and viscosity. This continues a sequence of publications on similar topics by the authors examining a variety of SOA systems, expanding the coverage of atmospheric aerosol surrogate systems that can improve our understanding of ambient particles.

The authors should consider my specific comments below in revising their manuscript.

Collection of samples on glass slide for liquid-liquid phase separation and viscosity measurements: It would be helpful if the authors could say something about the possible changes in composition (particularly partitioning of semi-volatile and more volatile components) that result – how reflective of the actual aerosol composition in the Teflon chamber are the particles once deposited and then analysed on the surface? This may be discussed in previous work but should be reiterated here. For example, the viscosity measurements are made in a flow-cell and necessarily this will presumably drive the composition to lower volatility and higher viscosity components over time. These changes in composition are not accounted for in the predictions of viscosity based on mass spectrometry measurements of the SOA composition.

Page 8: The authors state "The increase in the range of RH values over which LLPS occurs is likely related to the spread in O:C values within the organic particles – as the spread in O:C values increases, the RH range for LLPS is also likely to increase." In itself, the spread is not a reason, is it? Can the authors provide some rationale for why increasing chemical complexity leads to an increase in the LLPS RH range?

Figure 1: Are (a-c) just three different particles prepared under the same reaction chamber conditions? Similarly, in Figure 2, the different poke-flow measurements are made on different particles?

Page 8 onwards: Lower and upper limits to viscosity could clearly be dependent on any changes in composition that occur during the conditioning period. It would be helpful if the authors could show how the measured viscosity changes during the conditioning period, does it tend to a limit as the conditioning time increases? Is this conditioning based solely on adjustment to RH or is there some change in composition/partitioning of components into the gas phase during this time period (given the high concentrations in the reactor chamber)?

Page 9: To what extent is it appropriate to even represent the viscosity by a single value given that could be multiple phases/heterogeneities with within the particle with different viscosities? For example, the method for recording the "upper limit" could presumably be a measure of the viscosity of one phase, providing sufficient mobility for the shape recovery, with a more viscous phase moving within the more mobile phase.

Page 12: The authors state “At 30 % RH τmixing is âĽ§0.4 h, and at 38 to 50 % RH τmixing is in the range of $\sim$0.01 h to $\sim$50 h (Fig. 5a). These results provide important constraints on τmixing values within anthropogenic SOA.” Given the viscosity goes down with increasing RH, this sentence is confusing. The scales on Figure 5(a) are confusing, partly because it is very hard to read values due to the extremely large range. I recommend the authors only show a viscosity range from 104 Pa s to $\sim$109 Pa s. Lower and higher viscosities are to a large extend superfluous and there are no data in these arranges.

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