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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 #2

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In this study the authors investigate liquid-liquid phase separation (LLPS) and viscosity of secondary organic aerosol (SOA) formed in photooxidation of diesel fuel vapors. Such multi-precursors studies on SOA are needed to understand atmospheric SOA and the topic fits well in the scope of Atmospheric Chemistry and Physics. The study is based on experimental observations of LLPS and viscosity and comparison to viscosity parameterizations. Overall the study seem carefully conducted. The manuscript is in most parts clear and carefully written, although I have listed below few points which should be considered before the manuscript is accepted for publication.

Major comment:

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While the authors have overall resented the results carefully considering uncertainties and they discuss some caveats of the technique on page 12 L4-10, one aspect of the experimental technique has been neglected in the discussion of the results. The particles where collected on the glass slides and then conditioned for up to an hour for desired RH. During the conditioning (and the viscosity/LLPS measurements), the particles were surrounded by gas phase free of organics and this should cause some of the organics to evaporate. Therefore, the composition of the particles during viscosity or LLPS measurements would not have been the same as for it was when the particles were suspended in the chamber. This issue should be mentioned and the resulting uncertainty or systematic error in viscosities should be discussed in the text.

Minor comments:

P7 L1-8: Why are different values of coefficient k_GT used?

P7 L13-20: Is the predicted viscosity sensitive for the assumed viscosity of 10ˆ-5 at very high temperature and the assumed value of D_f = 10? Why did the authors chose D_f = 10 which they state is a lower limit from a previous study? Was the viscosity parameterization tuned to match the measured values in this study by selecting these specific values?

P8 L6-7: “two liquid phases may still have been present in the particles, but not in the focus of the microscope” Meaning of this sentence was not clear, probably since the authors have not explained how these optical measurements where actually done. What is the difference between the particles a-c in Fig. 1 and particles a-c in Fig. S2? Where all of these particles generated under same conditions? It seems that there is LLPS at 70 % RH in particles in Fig. 1, but in particles shown in Fig. S2 there is no LLPS always even at ∼82% RH. What causes this? Fig. S2 c shows LLPS at 4.7 % RH but not at the higher RH of 50.2%. This raises a question about the reliability of these results and such issues should be explained in the manuscript.

Technical comments:

P12 L18: “the presence of an organic-rich outer phase at high RH values can lower the barrier to cloud droplet formation” Barrier of what? Please rephrase.

Fig. 3b: Please revise the legend regarding Song et al. results. Currently the legend advises that Song et al. results would be shown with a green solid line. As such line is not present in the figure, I assume these results are the ones shown by the green errorbars.

Fig. 4: The times written on top of the figures do not show well. Please consider writing them with another color.

Fig. S1: “Mass Dp” and “particle Dp” where confusing in labels. The “Mass Dp” refers also to the diameter of particles. E.g. “Mass Dp” and “Number Dp” would be more clear and consistent names.

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