

Interactive comment on “The effect of adding hydroxyl functional groups and increasing molar mass on the viscosity of organics relevant to secondary organic aerosols” by James W. Grayson et al.

Anonymous Referee #3

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The manuscript describes a systematic comparison of new measurements of viscosity along with a compilation of data appearing in the literature for a series of polyols and saccharides. Measurements are made using the previously developed bead mobility and poke-flow techniques for particles deposited on a substrate. This contributes a set of benchmark data that help to expand the availability of viscosity measurements in the literature, providing greater insight into the factors that govern the phase and viscosity of secondary organic aerosol. As such, it should be published once the authors have had an opportunity to respond and address the following comments.

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Major points:

- Equilibrium phase state of alcohols: The authors do not clearly state the equilibrium phase of any of the polyols studied. From a brief investigation, 1,2,3,4-butanetetrol is a solid at room temperature, it is less clear for some of the others. This should be stated for each compound in Section 2.5. Indeed, if the compounds are liquids, it is not clear what is gained by making the measurements by the approaches described here rather than conventional rheological measurements. In addition, some discussion of the expected phase state in the aerosol phase would be beneficial. For the particles prepared for this work, it is clear that the deposited particles probed are all in a liquid state.

- Volatility of polyols: For some of the shorter chain alcohols presented in this work with the fewest OH groups, volatilization of the alcohol may occur fairly quickly. This could lead to additional temperature gradients driving circulation in the droplets, and compromising the bead mobility measurement, possibly leading to a lower reported viscosity than might be the reality. Has this been considered? It would be helpful if the authors could present data for one of the alcohols that can be compared with previous bulk phase measurements. They should at least discuss the possible effect of volatility and temperature depression, particularly as they are probing pure liquid droplets.

- Relative humidity: Water clearly acts as a plasticizer but there is very little consideration of the role of water, particularly for the systematic studies presented for the polyol systems. I recommend the authors consider including RH dependent data if they can easily be measured or if they have already determined them. The distinction between systems for which RH dependence has been examined and those for which it has not should be made clearly in the abstract. Indeed, there is very little new data in this manuscript, particularly for the polyols, 4 points out of 16 points in Figures 3 and 4.

Minor Points:

- The title leaves some ambiguity as to what is reported. I suggest the authors replace

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" viscosity of organics" by "viscosity of pure liquid phases of organics".

- Abstract: "with the over-prediction being up to 19 orders of magnitude" - is there really any significance to this? Surely this just suggests the model is fundamentally flawed?

- For ease of comparison, it would make sense to combine Figures 3 and 4 into one figure given they both consider sequences of polyol compounds.

- Figure 5: The authors present viscosity data for sucrose in Figure 5(a) but this is the system used to calibrate the bead mobility measurements so I am a little confused if this is really new data? Or is it just the data from the literature? I also recommend the authors remove the shaded areas above 10^8 Pa s as these are just a lower limit - at a first glance, the reader might be deceived into thinking these systems show constant values over these ranges. If it is new data, it could be helpful to see the comparison with previous RH dependent measurements directly in the Figure, particularly if it gives an indication of the level of uncertainty in the current approach. More generally, Figure 5 is poorly produced.

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