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Interactive comment on "A predictive group-contribution model for the viscosity of aqueous organic aerosol" *by* Natalie R. Gervasi et al.

Anonymous Referee #2

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The manuscript by Gervasi and co-workers presents a computational estimation tool for the viscosity of aqueous and organic mixtures of chemical substances. The model combines previously published estimation methods for pure compound viscosity (via Tg of organic molecules from DeRieux et al. (2018) and for Tg of water from Dehaoui et al. (2015)), an updated mixing rule based on GC-UNIMOD, and uses the AIOMFAC thermodynamic model for calculation of activity coefficients. The authors demonstrate the need for activity coefficient-based calculation of viscosity with significant deviations between experiment and theory that would be obtained using simpler mixing rules. While there is not much innovation within the utilized substructures, unification of several tools into AIOMFAC-VISC and outlining strengths and weaknesses presents a

significant step forward in the process of understanding the physico-chemical properties of Secondary Organic Aerosol (SOA), which are elusive and highly anticipated by the community. I expect AIOMFAC-VISC to be a well-utilized tool in the atmospheric sciences and a great resource for kinetic modelling applications. The manuscript thus fits well within the scope of ACP and is excellently written. I can hence recommend publication of this manuscript after minor revisions, as detailed in the following comments.

Specific comments

1. My comments mostly address questions about which part of the multi-step process that AIOMFAC-VISC prescribes leads to the significant improvement in estimating viscosities compared to previous simpler methods. This information might help to guide future research, e.g. tools for conversion of volatility into viscosity, or viscosity into diffusivity coefficients. Fig. S3 shows impressively how big the improvement from GC-UNIMOD to AIOMFAC-VISC is, almost to the extent that it might be worthwhile showing such a plot in the main text and presented alongside the discussion of Fig. 4. My question is, can the authors detail which difference in the calculations leads to the discrepancy in output?

2. The authors include a fragile-to-strong crossover (FSC) into their viscosity calculation; however, they do not show how this assumption affects their results. Please indicate the applicability of this model feature. The effect is shown in Fig. 2, but since only glass transition measurements are plotted here, no judgment on the necessity on the FSC is possible. Overall, Fig. 2 strikes me as a figure without much benefit for the reader since, in principal, only the performance of the glass transition estimation tool is evaluated. Can the authors think of any other way to show how temperature-dependence of viscosity is accurately represented by AIOMFAC-VISC?

3. A weakness for the applicability of AIOMFAC-VISC (and any group contribution

method for that matter) is the need for structure formulas of chemical components. For SOA, these are determined in this study through running the Master Chemical Mechanism (MCM) and hence relies on accuracy of the structures provided from this model calculation. The process is a bit convoluted as it demands running the external model and selection of a subset of chemical compounds based on matching to molecular O/C ratio. It would be beneficial for the reader if the authors could outline which parts of the process matter a lot and which are not as important. Hence, what is the sensitivity of the final viscosity output to the assumptions that were made?

4. Fig. 4 is very helpful for the reader. I wonder however, how well a simpler activity coefficient estimation method than AIOMFAC with a simple mixing scheme would fair in this comparison. Which part is more important: accurate activity coefficients or features such as contribution of the residual viscosity?

Technical Comments

- p. 22, l.13 – The word “Figure” seems to be missing before (7b).

References

Dehaoui, A., Issenmann, B., and Caupin, F.: Viscosity of deeply supercooled water and its coupling to molecular diffusion, Proceedings of the National Academy of Sciences, 112, 12020, 10.1073/pnas.1508996112, 2015. DeRieux, W. S. W., Li, Y., Lin, P., Laskin, J., Laskin, A., Bertram, A. K., Nizkorodov, S. A., and Shiraiwa, M.: Predicting the glass transition temperature and viscosity of secondary organic material using molecular composition, Atmos. Chem. Phys., 18, 6331-6351, 10.5194/acp-18-6331-2018, 2018.

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