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Comment on acp-2021-17

Anonymous Referee #2

Referee comment on "Estimation of secondary organic aerosol viscosity from explicit modeling of gas-phase oxidation of isoprene and *α*-pinene" by Tommaso Galeazzo et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-17-RC2, 2021

In this manuscript, Galeazzo et al. presented the organic aerosol viscosity estimated using a recently developed glass transition parameterization coupled to an explicit chemical mechanism, GECKO. This approach is used to predict the viscosity of secondary organic aerosol (SOA) produced from isoprene and a-pinene oxidation, and the results are evaluated using the chamber/flow tube measurements in the literature. It is found that the simulated viscosity of isoprene SOA is in reasonable agreement with the measurements, but major bias exists for a-pinene SOA. The authors explored the potential drivers of such discrepancy. This manuscript is fairly compact and generally well prepared, and is in line with the scope of the Journal. I do have several major and specific comments, and I recommend this manuscript for publication in Atmospheric Chemistry and Physics, provided the following major and specific comments are addressed

Major comments:

The approach to estimate the organic aerosol viscosity presented in this work consists of two key components: (1) glass transition temperature parameterization based on elemental composition (DeRieux et al. 2018 and Li et al. 2020). (2) explicit chemical mechanism generator GECKO (Aumont et al. 2005, etc). The glass transition temperature parameterization appears to be robust for both isoprene SOA and a-pinene when combined with volatility basis set which is largely derived based on measurements (e.g., Li et al. 2020). What is missing in the current manuscript is how GECKO performs for isoprene and a-pinene. It will be valuable to show the GECKO predictions for the chamber and flow tube experiments, and compare to key measurements available (e.g., mass loading alone may explain some of the variations in viscosity). Evaluating GECKO against measurements might sound out of the scope. However, I would argue that, a recent study (Gervasi et al. 2020) showed that the viscosity of a-pinene SOA can be reasonably well captured using MCM, a near-explicit gas-phase mechanism that also does not have particle-phase chemistry and underestimates high molecular weight compounds.

Specific comments:

Page 2, Line 24-25. "As gas-phase oxidation is often regarded as the rate-limiting step of SOA formation, there is a strong need for a computational tool enabling to generate exhaustive gas-phase chemical mechanisms." I'm not sure if I follow the logic here. There is a need for a tool that is capable of generating chemical mechanisms based on "first principles", but it has little to do with whether gas-phase oxidation is the rate-limiting step of SOA formation.

Page 4, Line 107-108. Some key technical details are missing here. How is the number concentration determined for each experiment? How does the model handle particle size evolution (e.g., is it only condensation or does it include nucleation/coagulation)? Is the aerosol scheme modal or sectional?

Page 4, Line 112. Please fix the citation throughout this manuscript per the Journal requirements.

Page 4, Line 120-121: "SOA particles were formed under dry conditions and then SOA were exposed to water vapor at different RH for viscosity measurements." This sentence is not well connected with the previous one. The previous sentence explains how the box model is configured for chamber experiments. Yet it is unclear if this sentence describes the same box model configuration, or if it actually describes how experiments were conducted.

Page 5, Table 1: what is the unit of RH? It looks like fractional (for all a-pinene experiments) but then for isoprene RH goes beyond unity.

Page 9, Line 225-226: Could the authors please elaborate the experimental uncertainties, and how these are translated/propagated into the uncertainties of the viscosity? This also calls for a closer look at GECKO, e.g., how does GECKO predicted aerosol loadings in all experiments compare to measurements.

Page 10, Line 233: how does the GECKO modeled overall particle-phase O/C ratio compare to measurements? This may provide key insight, i.e., to what degree can GECKO explain the bias in the predicted viscosity.

Page 10-12, Line 241-265: It is interesting to see the GECKO simulated functional group distribution and all. However, it is unclear what the take-away message is and what the community can really learn from it. The authors did attempt to make a connection between the function group information (-RCHO) and the particle-phase reactivity affecting particle viscosity. This is promising and may point to potential future research. However, I find this paragraph (Line 256-266) not well supported and scratches only the very surface of the issue. MCM does not treat particle-phase chemistry either, yet Gervasi et al. (2020) showed better agreement for a-pinene with measured viscosity using MCM.

This paragraph starts with Zhang et al. experiments yielding the highest –RCHO fraction but no further information is provided– does Zhang et al. also yield highest oligomer or HOM fraction? Whether a particular aldehyde (or a –RCHO group) actually facilitates oligomerization can vary a lot. The GECKO results presented here (Figure 5) do not provide any in-depth information on such potential of forming oligomers.

Lastly, it'll be great if Figure 5 can be evaluated with measurements but I understand this may be difficult at this moment. Please comment on what techniques may provide such information, perhaps HRMS or FTIR?

Page 11, Line 252-253: but isoprene oxidation also produces products with high O/C ratios. Can the authors please provide functional group distribution (similar to Figure 5) for isoprene?

Page 12, Line 261: recent studies suggest that autoxidation may also play a role in isoprene chemistry under certain circumstances (e.g., low NOx). Does GECKO include autoxidation for isoprene?

Page 13, Figure 6: Please also show the calculated alpha_eff, and discuss in the context of recent studies (e.g., Liu et al. 2019)