Response to Referee comments (comments in black, response in blue)

Anonymous Referee #1

Summary + General Comments

In "Estimation of Secondary Organic Aerosol Viscosity from Explicit Modeling of Gas-Phase Oxidation of Isoprene and α-pinene," Galeazzo et al. present the use of GECKO-A modeling on isoprene and α-pinene SOA to model the chemical composition and calculate the viscosity at various relative humidities. Though experimental samples, either from real-world conditions or from controlled conditions in a chamber, can lead to measurements of viscosity, global climate models will not be accurate solely using viscosity data from a small number of locations or test chambers. Numerous factors, including environmental (e.g. T, RH), chemical (e.g. compound structure, functional group contribution, atomic ratios), and physical (e.g. partitioning) processes, can contribute to large variance in aerosol behavior, e.g. glass transition temperature and viscosity. Recent progress made in parameterizing the prediction of viscosity has reduced the necessary inputs to elemental composition, stopping short of functional group analysis, thus providing a different pathway for viscosity prediction through high-resolution mass spectrometry.

Experimentally, viscosity of aerosol has been determined using a few different methods/instruments, e.g. poke flow mobility, a differential mobility analyzer, a particle impactor coupled with scanning electron microscopy, or a rebound impactor. By taking the priors from those experiments (precursor concentration, ozone concentration, T, RH, reaction time) and running those through GECKO-A, the researchers were able to predict glass transition temperatures and viscosities, which they could then compare to the experimental values.

When comparing modeled to experimental isoprene SOA viscosity with respect to relative humidity, the modeled values were found to be within error to experimental values assuming a hygroscopicity of 0.10. The modeled viscosity values for α-pinene were underestimated by 1 to 4 orders of magnitude, though wide experimental uncertainties led to some experimental values possibly lining up with the model. Better agreement was found between modeled and experimental values when mass loading was varied. A few interesting breakdowns of chemical composition were displayed as well.

Finally, the authors tried varying the mass accommodation coefficient, a property that measures how likely a gas molecule approaching an aerosol will be taken up. They selected two starting experimental conditions from a couple of different papers and observed a stark contrast between the expected viscosity vs. RH curves when predicting using a mass accommodation coefficient from unity to an effective value dependent on penetration depth.

This article is generally well-written, has a logical flow and is well-organized, has a clear description of results, and presents clear and concise graphs. The authors properly point out where future research remains and do well in pointing out subtleties in the models and the data. The section on varying the mass accommodation coefficient is novel, considering the same lab recently published the work on effective mass accommodation coefficients (Shiraiwa and Pöschl, 2020). Some sentences have interesting phrasing, which I will point out in the subsequent "technical corrections" section. I would tentatively recommend this article for publication, provided the authors address the following points.

We thank Referee #1 for the review and positive evaluation of our manuscript.

My biggest concern is a notable similarity of the first few figures in this paper and their corresponding results/discussion paragraphs to the paper Gervasi et al. published in early 2020, also in ACP (Gervasi et al., 2020). Comparing Figures 1 and 2 in Galeazzo et al. (this paper) to Figure 7a and 7c in Gervasi et al., we can note that Gervasi et al. incorporate data from more studies in their figure, specifically from the Bateman et al. study for the isoprene SOA and the Abramson et al. and Pajunoja et al. studies for the α-pinene SOA. Perhaps for the pinene experiments, this is because each of these papers only have one data point each and they use different experimental techniques than the ones mentioned here? These two papers remain different because the Gervasi et al. study uses MCM instead of GECKO-A with box modeling; however, this paper by Galeazzo et al. is missing any reference to Gervasi et al. Additionally, Galeazzo et al.'s paper, by means of chronology, would benefit from a comparison of the effectiveness of their model to the one found in Gervasi et al. While their isoprene SOA models yield fairly similar results in the viscosity vs. RH space, the α-pinene SOA data in Gervasi et al. appear to outperform the Galeazzo et al. model, bringing into question whether this paper represents an improvement on previously published methods. In theory, GECKO-A's model is more detailed and provides many more minor reactions pathways that the MCM does not. However, it is unclear whether this extra information makes the model more accurate or if these results point to some shortcoming in GECKO-A's processing.

We would like to note that the viscosity prediction method in Gervasi et al. (2020) relies on the T_g parametrization and viscosity prediction method that we have developed in our group (DeRieux et al., 2018): Figure 7a and 7c of Gervasi et al. (2020) are based upon and similar to Figure 4a and 4b in DeRieux et al. (2018), respectively. While Figure 4a of DeRieux et al. included Abramson and Bateman studies, we could not simulate Abramson's and Pajunoja's experiments in this study because unfortunately there is insufficient information to reproduce their experimental conditions as several experimental conditions were missing such as reaction time, concentration, temperature, and RH.

Gervasi et al. (2020) analyzed viscosity of α-pinene, isoprene and toluene surrogate mixtures using the AIOMFAC-VISC model. They focused on the impacts of intermolecular effects on the viscosity of organic-water mixtures. The explored solutions were composed by different combinations of water and organic compounds generated during α-pinene and isoprene oxidation in MCM. For predicting the viscosity of SOA, they selected 14-21 representative compounds from the MCM simulations of α-pinene and isoprene oxidation. In order to achieve a better agreement with experimental viscosity measurements, Gervasi et al. (2020) adjusted the composition of the representative α-pinene SOA mixtures to match the O:C ratio found in experimental measurements (Renbaum-Wolff et al., 2013; Grayson et al., 2016). As a result, the model developed by Gervasi et al. (2020) uses surrogate mixtures that cannot explore the effects of different experimental conditions on α-pinene SOA composition and resulting viscosity. Their approximation is valid for the purpose of estimating viscosity once the chemical composition of the particle phase is known, while it cannot explicitly link viscosity measurements with SOA composition generated by specific experimental conditions. Our study distinguishes from Gervasi et al. by providing information on the origins of the variations observed among viscosity measurements by taking into account different experimental conditions of SOA formation in various studies. We highlight how the composition from different experimental setups can impact the final SOA viscosity.

We have clarified this issue and added corresponding discussions in the revised manuscript.

Introduction: "Gervasi et al. (2020) simulated viscosity of aqueous SOA surrogate mixtures derived from oxidation of α-pinene, isoprene and toluene using the AIOMFAC-VISC model combined with the T_g parametrization of DeRieux et al (2018), demonstrating the capability and flexibility of the group-contribution method in predicting the viscosity for organic mixtures of varying degrees of complexity.

There is a strong need for the development of a modelling tool for further elucidation of the impacts of various aerosol properties and processes on particle viscosity. A better understanding of the interplay among gas-phase reactions, chemical composition and viscosity of SOA is essential for further development of model representation of the evolution of SOA in the atmosphere."

One other concern I have with this paper is the current lack of supplementary information. The experimental inputs and/or the model outputs would be useful information to provide for other scientists who wish to investigate such work. Having this more simple data be publicly available, either in the SI or in a repository, would be preferred. The data in Figures 4 and 5 would be helpful in table format for other scientists, while Figure 3 itself may be more efficiently placed in the SI, if it existed.

Following your comment, we compiled the data and model outputs that are used for figures in CSV files and share them in the supplement.

Gervasi et al. paper also builds up validation of the model using various solutions, including pure water, pure single component solutions, and then SOA. This manuscript would benefit from such an analysis, though this is not absolutely necessary for publication.

The purpose of this study is to estimate viscosity of multicomponent mixtures, as generated by GECKO-A. The validation of pure component viscosity and the mixing rule has been explored and validated in a series of our previous studies (Shiraiwa et al., 2017; DeRieux et al., 2018; Li et al., 2020) as well as in Gervasi et al. (2020). We clarify this point in the revised manuscript by adding the below sentence in Sect. 2.3:

"For validation and applicability of T_g parameterizations and the viscosity prediction method, they have been applied to high resolution mass spectrometry data of toluene SOA and biomass burning aerosols (DeRieux et al., 2018), SOA generated by diesel fuels (Song et al., 2019), β-Caryophyllene SOA (Maclean et al., 2021), and surrogate VOC mixtures by healthy and stressed plants (Smith et al., 2021), agreeing well with viscosity measurements."

While GECKO-A provides a plethora of chemical detail, it would be good to know if the researchers, either of this paper or elsewhere, are actively working to overcome the shortcomings and inefficiencies of GECKO-A.

Since the first GECKO-A publication (Aumont et al., 2005), the GECKO-A tool is constantly extended to include new processes, as for example the chemistry of cyclic structures in Valorso et al. (2011), the gas/particle partitioning in Camredon et al. (2008), the gas/aqueous phase partitioning in Mouchel-Vallon et al. (2013), the gas to chamber wall partitioning in La et al. (2016). In the past 5 years the GECKO-A tool has been extended to deal with the structure and chemistry of aromatic organic compounds, all the SAR included in GECKO-A have been updated on the basis of the latest literature (Jenkin et al., 2018a; Jenkin et al., 2018b; Jenkin et al., 2019; Jenkin et al., 2020), and the addition of recently identified chemical pathways, such as autoxidation, is currently under development. Autoxidation is a challenging matter to deal with. Vereecken and Noziere (2020) recently published a SAR to estimate H-shift in peroxy radical reactions. Although it gives estimates for many structures, multifunctional species remain far outside the scope of the SAR. Some thoughts are in progress to define how to extrapolate the SAR to all the species generated by GECKO-A. Another issue with autoxidation is that including such pathways drastically increase the number of species and reactions in the generated mechanisms, leading to unmanageable chemical schemes. Reduction methods are currently under development to overcome this limitation.

We added a paragraph in the manuscript to highlight the research, the progresses and undergoing developments of GECKO-A:

"GECKO-A is frequently updated to include the newly discovered chemical processes in the mechanism generator: recently, the mechanism generator has been extended to treat the chemistry of aromatic organic compounds based on the latest SAR developments (Jenkin et al., 2018a; Jenkin et al., 2018b; Jenkin et al., 2019; Jenkin et al., 2020). Other recent updates include the chemistry of cyclic structures (Valorso et al., 2011), the gas-aqueous phase partitioning (Mouchel-Vallon et al., 2013), and the gas to chamber wall partitioning (La et al., 2016)."

At the end of the introduction, the authors could provide more detail on the overarching objective. The phrase "to expand our understanding on the relationship and interplay among" is vague, and possibly deliberately so. It would be helpful to provide the reader a quick summary clause at the end of the sentence to tie it back into something concrete, such as incorporation into global models.

We revised the end of introduction to be more concrete as below:

"There is a strong need for the development of a modelling tool for further elucidation of the impacts of various aerosol properties and processes on particle viscosity. A better understanding of the interplay among gas-phase reactions, chemical composition and viscosity of SOA is essential for further development of model representation of the evolution of SOA in the atmosphere. … The objective of this work is to develop a useful tool for analysis and investigation of SOA chemical composition and phase state and to expand our fundamental understanding on such properties."

At line 206, the authors wrote "Simulation results for Renbaum-Wolff's and Grayson's experiments fall within uncertainties of experimental measurements for the 40-60 % RH range." This observation is clearly true from Figure 2, but the uncertainties for those data points are quite large, spanning 3-5 orders of magnitude, and the experimental data trend higher than the model data. Such a detail may be worth including.

We note that experimental measurements are subject to relatively large uncertainties.

"Simulation results for Renbaum-Wolff's and Grayson's experiments fall within the relatively large uncertainties of experimental measurements for the 40-60% RH range."

At line 220, the authors assert that the variance of model simulations and experimental measurements is very similar and give a correlation coefficient, but provide no data to support this claim. An SI would be useful to give the reader the option to verify this information.

We show the correlation plot as below, while we believe the inclusion of R^2 value is sufficient in the main text. A tabulated version of the final viscosity vs RH data will be provided as a separate file in SI.

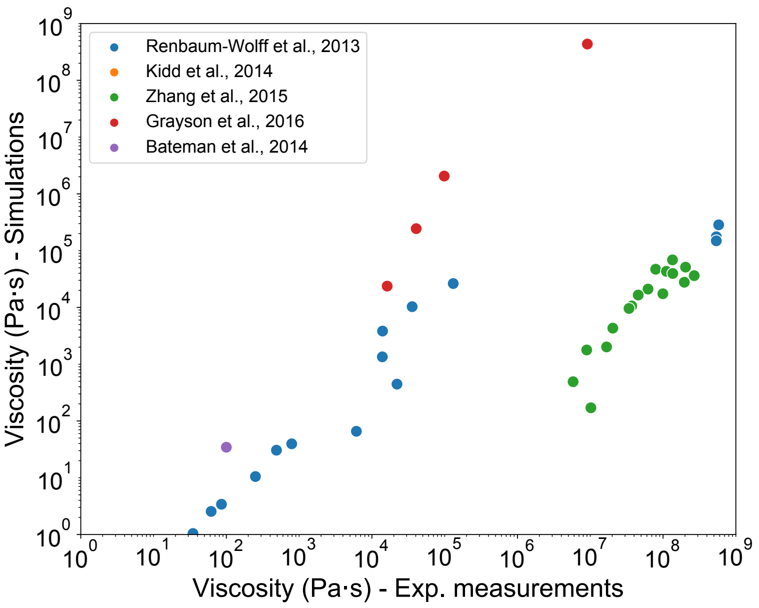

Figure R1: Correlation plot representing the relation between experimental measurements and simulated viscosity of SOA from α-pinene ozonolysis.

Finally, at line 283, the viscosity curves seemed to line up fairly well for the Renbaum-Wolff experiments, but not for the Kidd experiment. Do these results have any bearing on either Kidd et al.'s methodology or results or do they point exclusively toward a general need for more research into how the mass accommodation coefficient should behave over time and chemical composition?

Kidd et al. (2014) provided just a single viscosity estimation point for α-pinene. This viscosity value was not directly measured, but it was inferred from the impaction pattern, which is subject of a large uncertainty. As a result, it is challenging to draw a strong conclusion and further experimental and modeling research is necessary to elucidate impacts of chemical composition and mass accommodation on particle viscosity.

Technical Corrections

This manuscript has a number of sentences that would benefit from a read-through or from reading them out loud. I have also omitted changes I assume will be caught by a copy editor.

Lines 3-6: "In this study, we conduct explicit modeling of isoprene photooxidation and α-pinene ozonolysis and subsequent SOA formation using the GECKO-A (Generator of Explicit Chemistry and Kinetics of Organics in the Atmosphere) model. Our recently-developed parameterizations to predict glass transition temperature of organic compounds are implemented into a box model with explicit gas-phase chemical mechanisms to simulate viscosity of SOA."

These sentences are a tad bit awkward, and the authors used "explicitly" twice in quick succession. A possible fix, though this sentence can be fixed in plenty of different ways, is provided below.

"In this study, we use GECKO-A (Generator of Explicit Chemistry and Kinetics of Organics in the Atmosphere) to conduct explicit chemical modeling of isoprene photooxidation and α-pinene ozonolysis and their subsequent SOA formation. Coupling this level of chemical detail with box modeling and our recently-developed glass transition temperature parameterizations allows us to predict SOA viscosity."

Following your suggestion, we revised the sentences as below:

"In this study, we apply GECKO-A (Generator of Explicit Chemistry and Kinetics of Organics in the Atmosphere) to conduct explicit chemical modeling of isoprene photooxidation and α-pinene ozonolysis and their subsequent SOA formation. The detailed gas-phase chemical schemes generated with GECKO-A are implemented into a box model and coupled to our recently-developed glass transition temperature parameterizations, allowing us to predict SOA viscosity."

Lines 24-25: Change "enabling to generate" to either "capable of generating" or "that can generate."

The sentence has been corrected with "that can generate".

Lines 31-34: It may make sense to combine these two sentences together.
Lines 30-43: Depending on preference, the flow of this paragraph may benefit from lining up the order of each variable as they are first listed with the order of the sentences. To clarify, line 32 states "depending on chemical composition, relative humidity (RH), and temperature." You could move the next two sentences, "Notably, water…" and "It has been observed that…" to after the Petters et al. citation to make sure that subsequent sentences detailing work done with varying chemical composition, then temperature and RH are in closer proximity. Alternatively, or simultaneously, you could make the previous change for lines 31-34, which may make this juxtaposition more compact.

Following your suggestion, it is combined into one sentence.

"The particle phase state is impacted by SOA chemical composition, hygroscopicity, and water content, as water can act as a plasticizer lowering SOA viscosity (Mikhailov et al., 2019)."

Line 44: Change “fast” to “quickly.”

The word has been changed as suggested.

Line 87: Inconsistent spelling of “autooxidation” with “autoxidation” elsewhere.

It is corrected to “autoxidation”.

Table 1: RH in the heading should be RH (%). Put T and RH next to each other. Either convert the Song isoprene study to ppb or convert the pinene experiments to ppm for consistency.
Might be more visually striking to have an extra column on the left with vertical text indicating which precursor is used. As it is, the pinene and isoprene distinction blend in with the table.
In footnote, SEM should be written out, since it was not mentioned previously in the manuscript.
A footnote to explain reaction time would be useful.

We prefer to keep the legend as is for simplicity. The following changes have been implemented in Table 1: 1) SEM is deleted from the footnote; 2) T and RH are moved close to each other; 3) the unit of RH is specified in %.

Study	O_3 (ppm)	a-pinene (ppm)	RH (%)	T (K)	t (min.)	Exp. setup*	$T_{g,org}$ (K)	O:C simulated
Renbaum-Wolff et al., 2013	0.30	0.1	0.5	298	30	Continuous flow. PF-BM	275 K	0.49
Zhang et al., 2015	30	0.7	0.5	293	6	Batch exp., DMA	258 K	0.43
Zhang et al., 2015	30	0.7	0.5	293	1.5	Batch exp., DMA	263 K	0.44
Kidd et al., 2014	0.65	0.8	dry,	297	30	Batch exp., PI	266 K	0.44
Grayson et al., 2016	0.07	0.1	0.5	298	80	Continuous flow, PF-BM	284 K	0.55
Bateman et al., 2015	0.1	0.3	0.5	298	174	Continuous flow, R	275 K	0.48
	O_3 (ppm)	Isoprene (ppm)	RH (%)	T (K)	t (min.)	Exp. setup*	$T_{g,org}$ (K)	
Song et al., 2015	10	4	13.0	293.15	1.4	Continuous flow, PF-BM	260	1.0

* Poke Flow Bead Mobility (PF-BM), Differential Mobility Analyzer (DMA), Particle Impactor (PI), Rebound impactor (R).

Line 183: Change “Results” to “Results and discussion”

The suggested change has been made.

Throughout document, line 187: Copy editor’s job, likely, but the 60 and % are on different lines.

We will make sure on this issue upon proof reading.

Figure 1: Could you make the markers slightly larger?

The markers have been enlarged.

Line 203: In previous literature, a hygroscopicity κ of 0.1 for pinene has been shown to fit quite well (Petters and Kreidenweis, 2007; Prenni et al., 2007).

We used $\kappa = 0.1$ based on previous measurements and we did not use it because it leads to better fitting of the data; hence, we would like to keep the original sentence.

Figure 2: Could you add the line and dashed line to the small legend?

We prefer to keep the legend as is for simplicity, while explaining them in the figure caption.

Lines 278-279: Change "low volatile" to "low volatility" or "lower volatility." Also, add a comma after "unity."

The suggested changes have been implemented in the revised manuscript.

Lines 299-302: Swap "by a few orders of magnitude" and "lower than experimental measurements."

The suggested changes have been implemented in the revised manuscript.

"Simulated viscosity values for isoprene SOA are in good agreement with measurements, while those for α-pinene SOA were lower than experimental measurements by a few orders of magnitude."

Line 308 paragraph: Would it make more sense to have some of this paragraph be a discussion section?

We would like to keep this paragraph in this section, as all of simulated experiments formed SOA under dry conditions and SOA was exposed to water only upon viscosity measurements. Thus, we think that discussing this aspect in a discussion section might confuse readers and would like to keep it separate in the conclusion section.

References

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Anonymous Referee #2

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 α-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 α-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.

We thank Referee #2 for the review and positive evaluation of our manuscript.

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 α-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 α-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 α-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.

Validation of the GECKO-A mechanisms has been conducted extensively for the purpose of SOA formation in previous studies (e.g., Valorso et al. 2011, Denjean et al., 2015, McVay et al., 2016, La et al., 2016). These evaluations have been performed by comparison with chamber experiments of SOA formation from the oxidation of a-pinene, and C7 to C17 alkane and alkenes. The isoprene mechanisms generated with GECKO-A have not been evaluated. The following sentences have been added in Sect 2.1 of the revised manuscript:

"α-pinene GECKO-A mechanisms were evaluated in previous studies by comparisons with chamber experiments of SOA formation from photo-oxidation (Valorso et al. 2011, McVay et al., 2016) and ozonolysis (Denjean et al., 2015). The model captures the qualitative features of SOA formation with the variation of NOx levels during photolysis experiments, and with temperature for ozonolysis experiments. The measured SOA mass was overestimated by the model in Valorso et al. (2011) and Denjean et al. (2015), which could be in part explained by the loss of low volatile organic compounds onto the chamber wall (McVay et al., 2016 ; La et al., 2016). In addition, McVay et al (2016) highlighted a possible overestimation of the simulated contribution of later-generation oxidation products to SOA mass at high OH levels, and an underestimation of the SOA growth at low OH levels that could be due to lack of autoxidation processes in the mechanisms generated with GECKO-A. The influence of these limitations on the results simulated here are discussed along the paper. … Isoprene mechanisms generated with GECKO-A is for the first time indirectly evaluated in this study.”

In addition, we have reported the information needed to compare our model results to experimental O:C ratio in SOA for α-pinene and isoprene.

We cite and discuss Gervasi et al. (2020) in the revised manuscript (please see our response to first comment by Referee #1). We would like to point out that Gervasi et al. (2020) did not explicitly applied MCM for viscosity estimations but selected 14-21 representative compounds based on MCM for viscosity estimations. This study, on the other hand, explicitly couples detailed chemical mechanisms with viscosity estimation, allowing us to investigate the role and impacts of different experimental conditions and SOA composition (O:C ratio, functional group distributions) on resulting particle viscosity. We clarify this point in the revised manuscript.

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.

The sentence has been revised to:

“As gas-phase oxidation is a driving step of SOA formation, there is a strong need for a computational tool that can generate exhaustive gas-phase chemical mechanisms.”

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?

GECKO-A does not treat nucleation (line 104-106). As particle number concentrations were not reported in most studies, the particle number concentration was assumed to be 10^4 particles cm^{-3} following McVay et al., (2016). The particle number concentration was assumed to remain constant during simulations, as coagulation was not treated. We did not resolve evolution of particle size distributions, which are also not reported in experiments. We added some more details in the revised manuscript to clarify this point:

“The particle number concentration is assumed to remain constant during simulations (coagulation is not treated), while the particle radius evolves following the partitioning of organics.”

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

The citation style has been corrected.

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.

We clarify this point in the revised manuscript:

"SOA particles were formed under dry conditions and then exposed to water vapor at different RH for viscosity measurements; the same procedure is applied in our simulations by forming SOA at RH = 0.5% and then viscosity estimations are conducted by considering hygroscopic growth at elevated RH."

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

The unit is % and we make it consistent in the revised manuscript.

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.

Experimental uncertainties were elaborated and discussed in Grayson et al. (2016) and resulting upper and lower bounds of viscosity measurements are shown in Fig. 3. Grayson et al. (2016) did not report the exact precursor concentrations for their flow tube experiments, while they provided a range of precursor concentrations varied to produce samples at 5 different mass concentrations between 100 - 14000 μg m^{-3}. Thus, it is not possible to make comparisons for mass loadings of specific experiments, while the GECKO-A box model successfully captured the observed ranges.

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.

Following your comment, we added the below paragraph in the revised manuscript:

"None of these experimental studies measured average O:C ratio of SOA particles, but some of them reported estimated O:C ratios based on previous experiments with similar experimental conditions. Song et al. (2015) estimated the O:C ratio of isoprene SOA to be 0.64 -1.1, which is consistent with our simulated O:C ratio of 1.0. Renbaum-Wolff et al. (2013) estimated O:C ratio of a-pinene SOA to be 0.3 – 0.4, which is slightly lower than our simulated value of 0.49. Valorso et al. (2011) reported that the GECKO-A box model tends to overestimate the O:C ratio of SOA generated by a-pinene photooxidation. Denjean et al. (2015) showed that the O:C ratio of SOA simulated with GECKO-A from α-pinene ozonolysis was coherent with the one simulated with the Master Chemical Mechanism, and within the uncertainty range of the experimental value. A recent study by Gervasi et al. (2020), which selected 14-21representative reaction products based on the Master Chemical Mechanism to simulate viscosity of the same measurement dataset, estimated the O:C ratio to be 1.1 for isoprene SOA and 0.51 for a-pinene SOA. Overall, the O:C ratios simulated by the GECKO-A box model are reasonable and in line with available measurements and modeling studies, while further studies are warranted for simultaneous measurements of O:C ratio and viscosity along with model applications."

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 α-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?

We have added discussions of Gervasi et al. (2020) in the revised manuscript (see also our response to your comment 1 and also response 1 to Referee 1). Zhang et al. (2015) did not report HOM or particle-phase composition. Thus, we can only speculate that potential reasons for the high viscosity reported in their study. As pointed out, future studies are strongly desired for simultaneous measurements of viscosity and chemical composition by HR-MS or functional groups by FT-IR.

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?

In Fig. 5 we have added the functional group distribution for isoprene SOA simulated for the experimental conditions of Song et al. (2015). We have also added a few sentences comparing our simulation results to experimental measurements:

"Isoprene SOA is characterized by high concentrations of alcohols (–ROH) and hydroperoxides (–ROOH), followed by lower concentrations of ketones (–RC(O)R), and aldehydes (–RCHO). Our results are consistent with experimental measurements showing that polyols and organic peroxides are the primary species formed upon isoprene photooxidation under low NOx conditions (Surratt et al., 2006)."

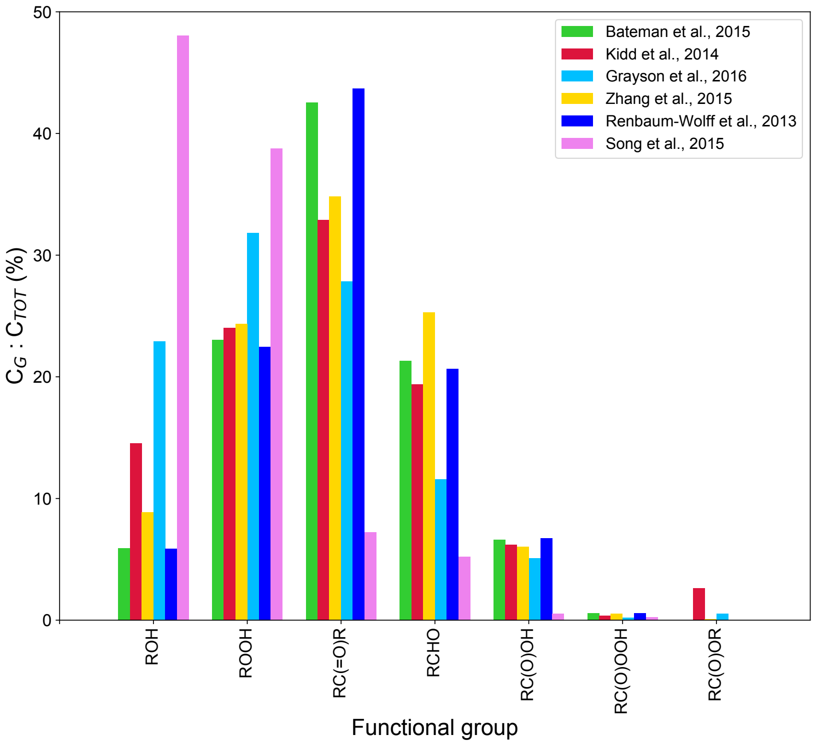

Figure 5: Simulated functional group distributions in particle-phase compounds derived from isoprene photooxidation and α-pinene ozonolysis.

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?

The autoxidation of RO_2 has indeed been recently shown to be an important process under low NO and HO_2 concentrations. Autoxidation processes are currently not included in the GECKO-A tool. The addition of autoxidation processes in GECKO-A is currently under development (see response to Referee #1). We agree that the absence of autoxidation processes could have an impact on the simulated results presented in this study, and the potential influence of autoxidation processes on the model/measure viscosity comparison is discussed along the manuscript.

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)

Following this comment, we have expanded the section of effective mass accommodation by exploring the difference of simulations for Kidd et al. and Renbaum-Wolff et al. (Fig. S1-S4). The simulated α_{eff} is now shown in Fig. S5 in comparison with Liu et al. (2019). Note that it is difficult to make a direct comparison between simulated and measured α_{eff} due to the lack of detailed information on SOA formation conditions in Liu et al. (2019).

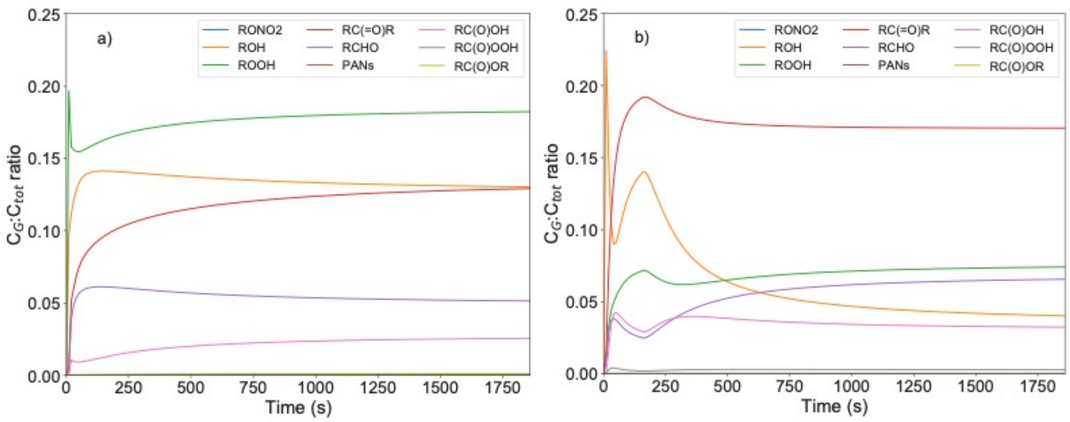

Figure S1: Time evolution of SOA compounds with different functional groups for simulations of experiments by (a) Kidd et al. (2014) and (b) Renbaum-Wolff et al. (2013) by considering effective mass accommodation coefficient α_{eff}. The simulations were conducted at RH = 0.5%.

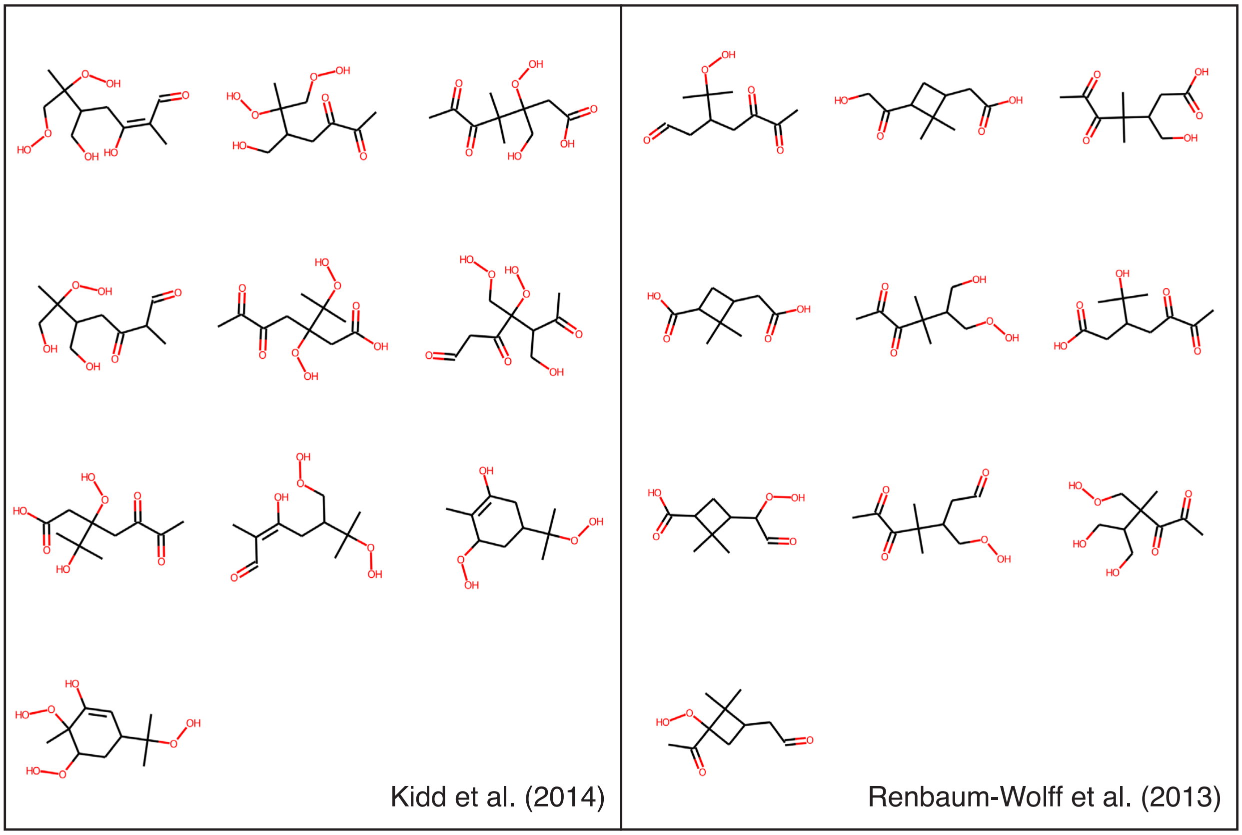

Figure S2: Ten major compounds found in the particle phase at the end of the simulations of Kidd et al. (2014) and Renbaum-Wolff et al. (2013). In both panels species concentrations decreases from the top row to the bottom row and from left to right. The simulations employed the effective mass accommodation coefficient α_{eff}. The simulations were conducted at RH = 0.5%.

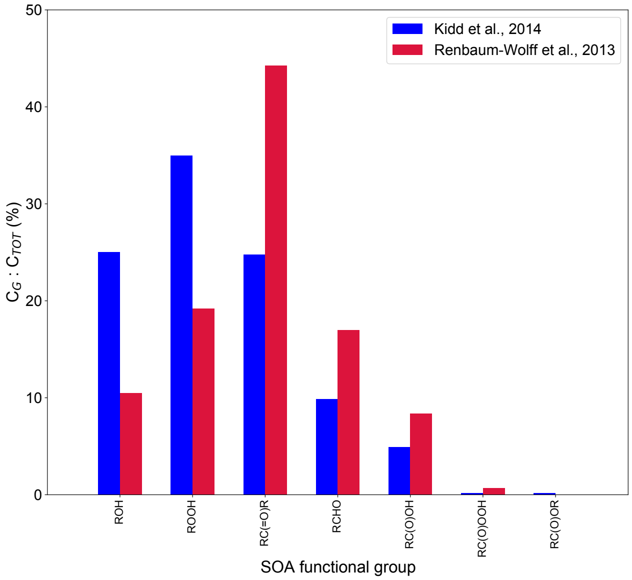

Figure S3: Functional group distributions in particle-phase SOA compounds in simulations of Kidd et al. (2014) (blue bars) and Renbaum-Wolff et al. (2013) (red bars). The simulations employed effective mass accommodation coefficient α_{eff}. The simulations were conducted at RH = 0.5%.

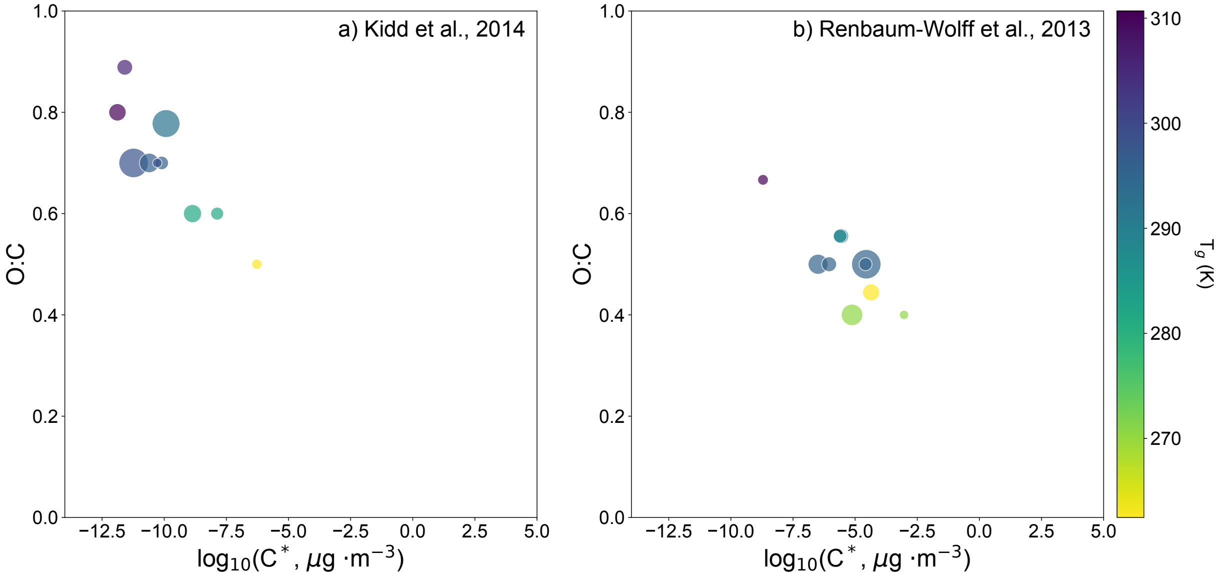

Figure S4: Chemical composition of SOA derived from oxidation of α-pinene from simulations of (a) Renbaum-Wolff et al. (2013) and (b) Kidd et al. (2014) experimental conditions plotted in the 2D-VBS framework of volatility and O:C ratio. The simulations employed effective mass accommodation coefficient α_{eff}. Markers represent the 10 most abundant particle-phase compounds from each simulation. The markers are color-coded with T_g. The marker size is scaled with particle-phase concentration in each simulation. The simulations were conducted at RH = 0.5%.

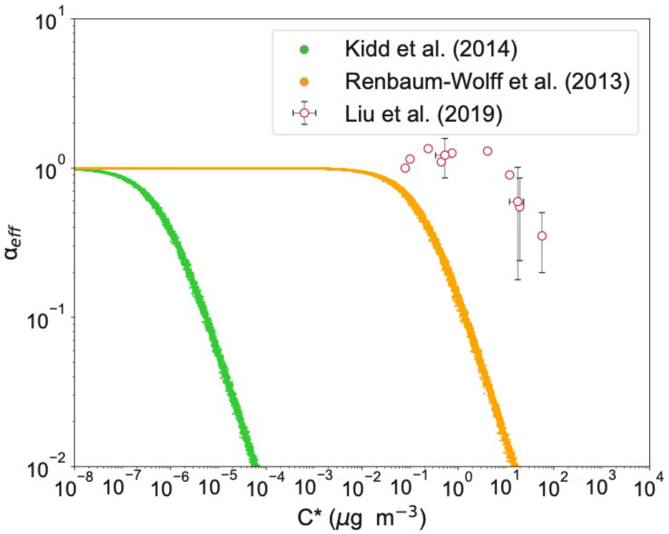

Figure S5: Simulated effective mass accommodation coefficients (α_{eff}) of α-pinene SOA compounds plotted against their effective saturation mass concentration (C^*) for simulations of Renbaum-Wolff et al. (2013) (orange) and Kidd et al. (2014) (green). C^* is assumed to be equal to C^0 with ideal mixing conditions. The markers represent measured mass accommodation coefficients for SOA generated by α-pinene ozonolysis by Liu et al. (2019). The simulations were conducted at RH = 0.5%.

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