

## Review Comments

After carefully reviewing the revised version of the manuscript, it is apparent that the author spent a decent amount of effort to improve the clarity and readability of the manuscript, as well as discussing the assumptions and limitations of the method in greater detail.

Even though this manuscript is a continuation of the previously published work of the author, there are several parts of the manuscript that are innovative and contain enough scientific significance to be published on ACP, including (1) using a new (and improved) formula to estimate the viscosity; (2) a comparison of the estimated viscosity with measurements from laboratory generated SOAs; (3) an estimation of the viscosity with different mass loadings; and (4) an estimation of the viscosity of the organic component of the biomass burning aerosols based on molecular composition. Just like any other models, this model has to make certain assumptions and is not perfect (also as reviewer 1 mentioned), but it is still important because it is one of the few so far that can estimate the viscosity of small organic mixtures (such as SOA), which will benefit the atmospheric community. Readers will be able to use the model from this manuscript to estimate viscosity of SOA-like mixtures, and some readers may even be able to further improve the model in the future. This paper certainly meets the publication standard for ACP.

After reviewing the manuscript, there are a few parts that should be modified to be clearer to readers.

Line 87-88. The author mentioned a piece of negative experimental evidence: “Gorkowski et al. (2017) did not observe significant diffusion limitations for glycerol and squalene in  $\alpha$ -pinene SOA. Quasi-equilibrium versus kinetically-limited or non-equilibrium SOA growth remains an open issue and warrants further investigations.” However, in a few other papers that the author mentioned (such as Abramson et al. *PCCP*, 2013, Zhang et al. *ES&T Letters*, 2018), there is positive experimental evidence showing that there are kinetic limitations regarding SOA evaporation, diffusion, and formation from multiphase reactions. It is better to mention these positive findings together with the Gorkowski et al. results to provide a more balanced viewpoint.

Line 318, 523. As previously discussed, the author agrees that the isoprene SOA generated from the PAM might be different from the ambient isoprene-derived SOA due to different reaction mechanisms. So I would recommend to change “isoprene SOA” to “PAM-

generated isoprene SOA” in line 318, 523, and other parts of the manuscript, so as to avoid generalization of the isoprene SOA.

The Batemen et al. results in Figure 4 and Figure 6 are not consistent with the same result in Figure 7. As review #1 mentioned, bounce results are not accurate in measuring viscosity, but the transition from bounce to non-bounce can provide a small viscosity range where a particle changes its phase state from semi-solid to liquid. If a particle does not bounce, it should be liquid. But if a particle does bounce, it may still have a relative low viscosity that will not cause diffusion limitation. According to Bateman et al. (JPC A, 2015) and Reid et al. (*Nature Communication*, 2018), the bounce results shown in Bateman et al. (*Nature Geoscience*, 2015) should be in the range of  $10^0$ - $10^2$  Pa s, corresponding to RH 60-80%. In Figure 4 and 6, the author assigned the RH to be 40-60%, which is different from Figure 7, where the RH range was 60-80%. Please make the RH in all Figures to 60-80%. The text that contains 40-60% needs to be changed to 60-80%, too.

Line 444-446: the author says the O:C ratios based on nano-DESI-HERM was lower than the O:C reported by Song et al. (2016). So why a lower O:C ratio results in higher viscosity estimation then? In line 493-494, the author states that higher O:C ratio will result in a higher viscosity estimation, which is contrary to how the author explained the data in line 444-446. The part may need to be revised to properly explain the difference between the model and the measurement.

Line 500-501: the author should add “providing the upper limit of the viscosity of the organic component, while APPI MS gives the lower limit”, to make it more clear that the predictions are only applicable to organic component rather than the whole biomass burning aerosols.

Line 908, the table and the text overlap with each other. Please fix that.

One general suggestion I have is about the figures. Figure 1 looks very much like the author’s other paper on *Nature Communications*. It could give people the wrong impression that this was a similar paper compared with the old one, but actually this paper contains a decent amount of new information. In the future it is probably better to modify the figures not to be so similar to previous published ones when the author writes a follow up paper, so as to avoid confusion for the readers. But I understand why the author uses this style this time and no changes are needed from me for this draft.

One last comment I have is the using of the fragility parameter  $D$ . I think the current model is fine using different  $D$  values to predict the viscosity. But more recent physical chemistry theory about liquid/glass relaxation has been improved so that all  $D$  values could collapse into one single uniformed equation Elmatad et al. (*JPC B*, 2009). All the experimental data agree nicely with the new theory without introducing the  $D$  value. Maybe the author can consider that applying this theory to the viscosity estimation in the future.

Overall, this article provides an improved model of estimating glass transition temperature and viscosity of organic components that can extend to molar mass of up to 1100 g mol<sup>-1</sup>. The study cross compares with literature data and molecular measurement of the biomass burning aerosols to show the validity of the model. The manuscript reads well especially after the second revision, and is suitable to be published on ACP.

## References

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