

Timothy Bertram
Co-Editor of Atmospheric Chemistry and Physics

Dear Tim,

Listed below are our responses to the comments from the reviewers of our manuscript. The reviewers' comments are in black text and our responses are in red text. We thank the reviewers for carefully reading our manuscript and for their helpful suggestions. Note, we would like to add an additional co-author to the manuscript, Y. J. Li, since he contributed significantly to the revised manuscript.

Sincerely,
Allan Bertram
Professor of Chemistry
University of British Columbia

“Relative humidity-dependent viscosities of isoprene-derived secondary organic material and atmospheric implications for isoprene-dominant forests” by M. Song et al.

Response to Referee #1 (Reviewer comments in black text)

This is an important study of viscosity of a mixture of organic compounds that could serve as a model for the organic material in isoprene derived secondary organic aerosol. The measurements have been carried out over a range of relative humidity values, and the results clearly show that the organic material in model isoprene derived aerosols is not as viscous as that in model aerosols produced from alpha-pinene. I have no major suggestions for improving this paper. Minor comments are listed below.

We thank Reviewer 1 for careful reading of the manuscript and the helpful comments and suggestions. Detailed responses are given below with their respective locations in the revised manuscript.

1134, line 1: for the benefit of the readers, please specify a typical size of an organic molecule for which the Stokes–Einstein equation should be applicable.

→ This is a good point. We have now added the following text in Sect. 4.3.

“The Stokes-Einstein relation should give a reasonable estimate of diffusion rates when the viscosity is lower than that of a glass and when the molecules undergoing diffusion are

roughly the same size or larger than the molecules in the SOM matrix. If we assume the SOM matrix is dominated by molecules similar to 2-methyltetrol and 2-methylerythritol ($C_5H_{12}O_4$), which have been identified as key oxidation products of isoprene and have an isoprene skeleton, (Cleaeyns et al., 2004; Carlton et al., 2009; Kleindienst et al., 2009) then the Stokes-Einstein equation should be applicable when the viscosity is lower than that of a glass (10^{12} Pa·s) and for organic molecules with a molecular weight approximately ≥ 136 g mol⁻¹, although additional work is required to confirm these assumptions.”

1136, SOM preparation section: please estimate the contribution of ozone to SOA production. With 15 ppm ozone in the flow and $k = 1.3E-17$ cm³/(molec*s), the lifetime of isoprene should be about 3.6 minutes. The volume of the oxidation flow reactor is not specified in this paper and it is also not specified in the cited papers by Liu et al. (2013 and 2014). However, the Liu et al. (2014) paper quotes a residence time of 110 s at the same flow rate as used in this study. Therefore as much as 50 % of isoprene could potentially be oxidized by ozone instead of OH. Is this an issue considering that ozone-driven oxidation is also capable of producing aerosol from isoprene, e.g., Ref.[1]?

→ To address the referee’s comment we have added the following text to the revised manuscript (Sect. 2.1):

“The volume of OFR was 13.3 L and the OFR was operated at a flow of 7.0 and 9.5 L min⁻¹, resulting in residence times of 114 s and 84 s, respectively. Although the OH concentration was not measured in the OFR in this study, an OH concentration in the OFR in the range of 2×10^8 to 2×10^{10} molec cm⁻³ was expected based on previous experiments under similar conditions (Lambe et al., 2011a). This OH concentration corresponds to a lifetime of isoprene between 0.5 and 50 seconds. For comparison, the O₃ concentrations used in these experiments correspond to a lifetime of isoprene of approximately 3.6 min. The OH concentration in the OFR was adjusted by changing the power of the UV lamps as described in Lambe et al. (2011a). For the experiments in this study, the lamp power was always full; therefore, the OH concentration in this study should have been close to 2×10^{10} molec cm⁻³, and the OH pathway should have dominated the oxidation of isoprene.”

Related to this, the authors should probably mention in this section that the high concentrations used in the oxidation flow reactor results in an unrealistically high contribution of the RO₂+RO₂ reaction products and Criegee intermediate reaction products to the SOA formation. The authors do mention the caveat to the conclusions resulting from high mass concentrations on page 1147; all I am suggesting is to explain it to the readers what could go wrong when using high concentrations.

→ We have now added the following text in Sect. 2.1.

“The concentration of the major oxidants (O_3 , OH, and HO_2) in the OFR is higher than in environmental chambers or the atmosphere, but the ratios of O_3 to OH and OH to HO_2 are similar to those encountered in the atmosphere and in environmental chambers. As a result, the OFR is used to simulate oxidation processes in the atmosphere and environmental chambers. Recent measurements with an aerosol mass spectrometer have shown that the composition of isoprene-derived SOM produced with an OFR is the same, within the uncertainty of the measurements, as isoprene-derived SOM produced with an environmental chamber (Lambe et al., 2015).”

Section 4.2: It may be worth mentioning the study of O’Brien et al. (2014), who looked at how much different types of particles flatten upon impact with an impactor substrate [2]. Their measurements suggest that laboratory models of isoprene derived particles are less viscous than those of alpha-pinene derived particles, and less viscous still than ambient aerosols.

→ We have now mentioned the study of O’Brien et al. in Sect. 4.2 of the revised manuscript as suggested by the referee.

“This conclusion is consistent with work by O’Brien et al. (2014) who also concluded that the viscosity of isoprene-derived particles is lower than the viscosity of α -pinene-derived particles based on the how much the particles flattened after impaction on a substrate.”