

Interactive comment on “Mixing times of organic molecules within secondary organic aerosol particles: a global planetary boundary layer perspective” by Adrian M. Maclean et al.

Anonymous Referee #1

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Maclean et al. has estimated mixing times of organic molecules within secondary organic aerosol particles. In chemical transport models SOA particles are often assumed to be homogeneously well-mixed on the timescale of <1h, which could be in question if SOA particles adopt glassy or amorphous semisolid states. Combining laboratory data, meteorological conditions, and chemical transport modeling, this study predicted that mixing times should be indeed within <1h in the planetary boundary layer. They concluded that the assumption of well-mixed SOA in chemical transport models seems reasonable for biogenic SOA in most locations in the PBL. This is a very interesting study, the method seems reasonable, and the manuscript is clearly written and easy to follow. I have several comments as below, which should be implemented in the revised

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manuscript before publication in ACP.

- The analysis is focused on 200nm-diameter particles and I agree that this may be most frequent size to be observed in ambient environments. Aged particles can have much larger diameters of up to 1 μm , as observed for example in remote areas or Tokyo (see Fig. 7 in Takegawa et al., J. Geophys. Res., 111, D11206, 2006). Thus, I would suggest that the same analysis should be conducted with a larger diameter, say 500 nm-diameter particles. Then same figures of Fig. 3 could be presented and lines can be added in Fig. 4 (if the results are too similar with 200 nm, then they can be placed in the supplement/appendix). Mixing times should be larger for larger particles and I would be curious to know if mixing timescales would be still below 1 h. This should be easy and straightforward to do for authors and it will certainly strengthen their conclusion.

- It is very interesting to compare Fig. 6 in this study with Fig. 3d in Shiraiwa et al. (Nat. Commun., 8:15002, 2017). Shiraiwa et al. predicted the glass transition temperatures of SOA in a global model and estimated mixing timescales using annual average of RH and T for 2005-2009, while this study considers seasonal dependence, but did not simulate T_g or viscosity directly but viscosity was parameterized based on α -pinene viscosity measurements. I think there should be some discussion with a paragraph or two comparing these two studies. General trends seem to be consistent: longer timescales in west US, Sahara, and Mideast and shorter timescales in Europe and higher latitudes (Why there are no information over some places, such as Europe in panel a and over Amazon in both panels?). However, this study seems to estimate mixing timescales shorter in general. Please add some discussions.

- Abstract, L23: "SOA concentrations are significant." is ambiguous. I suggest being specific here ($> 0.5 \mu\text{g m}^{-3}$).

- P2, L4: I suggest replacing "the lowest" to "low". Not only the lowest ones, but low and semivolatile products would also condense.

- P5, L3: “under predict” should be “underpredict”.
- Figure 6 is not very easy to read and I feel this is because of overlapping yellow lines, arrows, and letters. Can you just remove these yellow things, and just put colors for places with SOA concentrations above 0.5 $\mu\text{g m}^{-3}$? This would improve accessibility of this important figure.
- It may be good and helpful for readers to have a summary/conclusion section in the end of the manuscript.
- I suggest combining Section S1 with the main text, or include it as Appendix (particularly bring eq S1 and S2).
- I would suggest moving Fig. S3, S4 (also S5?) in the main text (maybe in Appendix?). There seem to be non-negligible cases with mixing timescales >1 h for anthropogenic SOA (given that sucrose is a good proxy for that).

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