Reply to editor comments on acp-2017-860-R2

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General comment:

The authors have largely adequately addressed this last round of reviewer comments, for which they are to be commended. However, there are two specific responses that are not satisfactorily addressed or appropriately worded.

Thank you for reading the manuscript carefully and providing useful comments. We revised the manuscript following the comments.

Specific comment 1:

The first is:

"The mass accommodation coefficient subsumes all resistances to gas−particle partitioning other than gas phase diffusion; and a mass accommodation coefficient smaller than 1 would indicate that the condensed phase is highly viscous and exhibits substantial kinetic limitations."

A mass accommodation coefficient smaller than unity by no means indicates a highly viscous condensed phase with substantial kinetic limitation. I would refer the authors to Kolb et al., ACP, doi:10.5194/acp-10-10561-2010, 2010 for a thorough discussion of the meaning of mass accommodation coefficient. It is perfectly possible for mass accommodation coefficient to be very much lower than unity without a particle being highly viscous - for example the mass accommodation coefficient of water vapour to a lubricating oil particle would be extremely low, but the particle would be quite inviscid. Any particle exhibiting immiscibility with the condensing vapour (or with a tendency to outgas or "salt-out" the component because of strong non-ideality) could behave the same whilst being quite fluid. I would agree with the original reviewer's request not to conflate thermodynamics and kinetic inhibition in the absence of a mechanism but find the proposed mechanism unconvincing. I'd invite the authors to tighten up the argument.

Reply:

I agree with the comments. I discussed potential reasons suppressing the accommodation coefficient, and added the reference suggested in the comments. I toned down our suggestion from the results of mass accommodation coefficient. I revised the sentences in lines 357-365 as follows:

The theoretical equilibration scaling time was also evaluated using eq. 3 in Saleh et al. (2013), where the accommodation coefficient was set to a recommended value of 0.1 for α-pinene SOAs. The theoretical equilibration scaling time was determined to be 24–41 min, which was similar to the experimental results: 24–46 min. The mass accommodation coefficient subsumes all resistances to gas–particle partitioning other than gas phase diffusion, for example, surface accommodation, deviation from Maxwell-Boltzmann molecular velocity distribution near the particle surface, and diffusion limitations in the condensed phase (Kolb et al., 2010; Saleh et al., 2013). The mass accommodation coefficient was determined to be less than unity, suggesting that the existence of low-volatility materials in SOAs, kinetic inhibition, or some combined effect may explain the equilibration time scale measured in this study.

Furthermore, I revised the following sentences in the abstract in lines 18-23:

The volume fraction remaining of SOAs decreased with time and the equilibration time scale was determined to be 24 – 46 min for SOA evaporation. The experimental results of equilibration time scale could be explained when the mass accommodation coefficient is assumed to be 0.1, suggesting that the existence of low-volatility materials in SOAs, kinetic inhibition, or some combined effect may explain the equilibration time scale measured in this study.

Specific comment 2:

The second difficulty with the response is the repeated statement that there is a "standard VBS approach". There are a multitude of VBS approaches and the authors should be specific. One way they could do this is to anchor the statement on a reference that distinguishes the approach to which they are referring from other "non-standard" VBS treatments (by stating "standard VBS approach (e.g. xyz et al., 2005)").

Reply:

I added references for “the standard VBS approach” and also discussed non-standard treatments citing references. I revised the sentences in lines 385-389 as follows:

In the standard VBS approach, the product volatility distributions determined by SOA yield curves are employed and no reactions are assumed to occur in the particle phase (e.g., Robinson et al., 2007; Lane et al.; 2008). Currently, only a limited number of non-standard treatments are available; e.g., Trump and Donahue (2014) took into account oligomer formation in the particle phase, and Yli-juuti et al. (2017) employed the product volatility distribution determined from dilution data.

References

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