Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-746-AC1, 2017

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Multi-generation Chemical Aging of α-Pinene Ozonolysis Products by Reactions with OH" *by* Ningxin Wang et al.

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Received and published: 5 December 2017

(1) *In this manuscript the authors present results of a laboratory study in which they investigated the chemical aging of SOA formed from the ozonolysis of a-pinene. The aging was conducted by photolyzing HONO (to form OH radicals) that was added at two different times following completion of the ozonolysis reaction. The effects of aging were determined by monitoring the SOA mass using an SMPS and the composition using an AMS. Careful corrections were made for the effects of particle and vapor wall loss on the SOA yields. The results demonstrate that oxidation comparable to a few days of atmospheric aging lead to the formation of a significant amount of additional SOA mass, as well as small changes in composition as measured by the O/C ratio. The results are consistent with previous studies of this type, but are of to SOA yield*

corrections that are important in aging experiments. The experiments and data analysis were carefully done, and the manuscript is well written. I think the manuscript will be suitable for publication in ACP once the following comments have been addressed.

We address the various comments of the reviewer below. Our responses and corresponding changes in the paper follow each comment.

Specific:

(2) *Page 14, Lines 407–410: It seems that this estimate of the effect of gas-wall partitioning of vapors on SOA formation assumes that all the first-generation products are either fully volatile or non-volatile with respect gas-particle partitioning. But for semi-volatile compounds the 15 min condensation time scale is also the upper limit to the time scale to achieve gas-particle partitioning equilibrium. In this case, some vapor will remain in the gas phase and continue to be lost to the chamber walls throughout the experiment because of the large effective particle mass of the walls. I think the approach used here thus provides only a lower-limit estimate to loss of vapors to the wall.*

The reviewer is correct. Our approach yields the lower-limit estimate to vapor wall loss, and yet this is consistent with what we observed from the measurements. As indicated in Fig. 6, the organics to sulfate ratio stayed practically constant after its first peak at t=0.7 h until the introduction of OH. This is consistent with the fact that the SVOCs formed in our system only accounted for a small fraction of the products. This is also consistent with what Ye et al. (2015) observed for the α-pinene ozonolysis system. With a moderate precursor concentration (α-pinene <90 ppb), the SVOCs formed represented 20 percent of the products. We have added this discussion to the paper.

(3) *Page 14, Lines 408–410: It is not clear how the estimated loss of vapors to the walls is converted to an SOA yield correction.*

Our zeroth-order correction simply assumes that the yields increase also by 6.3 percent. This results in an absolute increase of the yields of 1-3 percent. We have added this clarification to the paper.

(4) *Page 15, Line 435–437: It would be straightforward to estimate the potential increase in O/C ratio due to heterogeneous oxidation by OH radicals and thus test this hypothesis.*

Heterogeneous oxidation here refers to the uptake of OH by the SVOCs and the LVOCs in the condensed phase and the corresponding reactions that take place. Given that these reactions can be quite complex probably involving both functionalization and fragmentation the estimation of the expected O/C ratio is not straightforward.

(5) *Page 15, Line 437: Why can't the observed changes in O/C ratio with aging be the result of gas-phase oxidation of semi-volatile compounds coupled to gas-particle partitioning, as proposed by Robinson et al. (2007), rather than heterogeneous oxidation?*

Gas-phase reactions could of course contribute to the observed O/C changes. However, the corresponding condensation of the products should result in a detectable increase in SOA concentration during the same period. We could not observe such a change; therefore, the contribution of gas-phase oxidation is probably small. Heterogeneous reactions can explain this significant change of O/C without a corresponding increase in SOA mass concentration. We added this explanation to the paper.

(6) *Page 16: The Conclusions section as is really just a brief summary of the results, written solely within the context of these experiments. I suggest the authors provide a broader discussion of the relevance of these results to studies of atmospheric SOA and what they contribute to knowledge of the formation, composition, and properties of SOA.*

We have followed the suggestion of the reviewer and added a discussion of the relevance of these results for atmospheric SOA.

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