Review of Chen et al., "Modeling organic aerosol from the oxidation of a-pinene in a Potential Aerosol mass (PAM) chamber

In this work, the authors modeled the evolution of a-pinene SOA in a flow chamber using the recently developed 2-dimensional volatility basis set (2D-VBS) framework. The experiments cover a wide range of OH exposure, which is a feature of the PAM chamber. Their model agrees qualitatively with measured values of organic aerosol concentrations and O/C, but cannot reproduce the temporal trend of O/C. The authors also examined the sensitivity of the model to each individual oxidation and partitioning parameter using global sensitivity analysis techniques. The paper is well written and the analysis is novel and carefully thought out. I recommend publication in ACP upon considering the following comments.

Major comments:

1. My main take-home message from the results is that although 2D-VBS is a step forward in predicting OA chemical properties by incorporating O/C, the representation is still too simplistic to be able to capture evolution of O/C. This is not simply because of poor estimates of parameter values, as demonstrated by the sensitivity analysis shown in Figs. 4 and 5. The authors did in fact point out that some parameters can change with oxidation state but did not investigate further. I believe the above result points out very clearly that the constant values for some parameters is the major deficiency in the model. I suggest the authors explore this further using the model. For example, I imagine that PO1, PO2 and PO3 changes with oxidation. How would it need to change in order to better fit the data? Is such a trend physically justifiable? How does oligomerization (which has been proven to happen in the a-pinene SOA system) change the picture? In general, I would link the results to more mechanistic understanding of the chemistry.
2. Following on the last point, the authors suggest that O/C depends on kOH_homo more strongly at low OH exposures, and on kOH_hetero more strongly at high OH exposures. It is unclear to me if that was an attempt to explain the poor correlation with measured values. If so, is there some combination of kOH_homo and kOH_hetero that will better fit the measured data?

Minor comments:

1. P. 2761 Line 13 and 20: The term "mass yield" is confusing. It can refer to total SOA/HC reacted. I suggest using the term "stoichiometric yield" or "aerosol mass fraction" (Donahue et al., 2006).
2. Pg. 2763 Line 19: I disagree that 0.14 cm-1 is a "small" S/V ratio. It is much higher than those of smog chambers.
3. Pg. 2763 Line 25: How was OH exposure varied? Was it achieved by varying the residence time in the flow tube, or by changing the UV intensity? If it is the latter, does that affect the particle phase processing of OA (photolytically induced oxidation in the particle phase)? That could be related to the higher O/C increase at higher O/C exposure.
4. Pg. 2764 Lines 11 – 14. Is this temperature correction a function of c*? The heat of vaporization (and hence, temperature dependence of partitioning) should depend on c*.

5. Pg. 2766 Line 5: O3 reacts with alkenes only, so this rate constant should decrease very significantly beyond the first generation. Any products of a-pinene would react with O3 with a kO3 << 1E-17.
6. Figure 1: This is a very busy and confusing figure. First, the subpanel labels should be clearer (larger). Second, the explanation for the legend should appear inside the legend itself, not in the captions. Third, the split x-axis on the right panel should have the same exponent (e.g. 0 – 0.2E12 and 0.2–5E12). Alternatively the authors should consider using log scale. Lastly I think this figure is trying to demonstrate too many messages, such that each is lost behind the myriad of plots. I recommend splitting this into two figures, one demonstrating the correlation between modeled and observed data, and the other looking at detailed evolution of one particular case. I also suggest making all markers the same for the left panels, as it does not seem to depend on RH or starting concentration, and using different markers serve as an unnecessary distraction.