

This work provided a modeling framework based on 2D-VBS to simulate SOA formation from a-pinene photooxidation/ozonolysis in the PAM flow reactor. The modeled and observed Coa and O/C are compared. It is found that the modeled Coa and observed Coa agreed within uncertainties, but the modeled O/C has a different trajectory (with increasing OH exposure) compared to the observed O/C. The sensitivities of Coa and O/C to different model parameters are investigated.

I think the paper is well-written and the results are interesting and original. One of the main concerns I have is regarding the global sensitivity analysis. As shown in the manuscript, even when the 13 parameters are simultaneously perturbed, the authors still cannot reproduce the trajectory of changes in O/C with OH exposure. It is not very clear what this implies. Does this mean the formulation is “wrong” and that none of the 13 parameters is as important in predicting O/C as the authors might have suggested? Or, perhaps not enough simulations have been performed? Or, perhaps the range that the parameters are allowed to vary is not large enough? If so, the authors need to perform further simulations with a larger range of parameter values. All these need to be carefully addressed to support the usefulness of the modeling framework proposed in this work.

Secondly, one of the main features of the PAM reactor is the very high gas-phase OH concentration (many orders of magnitudes higher than ambient) used in the experiments. On one hand, this allows for the study of aerosol aging. On the other hand, heterogeneous reactions would be more prevalent in the PAM flow reactors than in ambient environments and this introduces the concern of whether these results are relevant to ambient environments. Coincidentally, the results in this manuscript showed that heterogeneous reaction is the *only* parameter that becomes progressively more important for *both* Coa and O/C with increasing OH exposure. Is this a direct result of the high OH concentration (and enhanced heterogeneous reactions) used in PAM reactor? The authors need to discuss this in more details and provide justifications on how their modeling framework is applicable to ambient conditions where heterogeneous reactions are probably not as prevalent compared to the PAM flow reactor conditions.

Overall, I recommend the manuscript to be published once these issues are resolved. The authors can find more specific comments below.

Specific comments:

1. Page 2765, line 15. The experiments performed by Pathak et al used 2 butanol as OH scavenger to exclude the OH chemistry. Since both ozone and OH are present in the PAM experiments, what is the relative importance of OH vs ozone reaction with a-pinene in these experiments? Are the authors making the assumption that the yields from Pathak et al are applicable to their systems? If so, this needs to be stated clearly.
2. Page 2767, line 20. The authors chose a specific experiment (281 ug/m3 of a-pinene) as a “typical case”. Is this typical case representative of all the PAM a-pinene experiments that have been performed? For instance, do all other PAM a-pinene experiments exhibit the same behavior in O/C, i.e., increases slowly at low OH exposure, and rapidly at higher OH exposure? The authors need to clarify this and provide more information regarding why the particular experiment (281 ug/m3) is chosen as a “typical case” and how it is “typical”.

3. Page 2767, line 20. For the parameters in Table 2, is the effect of RH explicitly accounted for in the model? Many previous studies have found that RH affects SOA formation from  $\alpha$ -pinene ozonolysis (e.g., Jonsson et al., 2006). How does the difference in RH affect the results of this study and are they consistent with literature? Please address this in the revised manuscript.
4. Page 2768, line 3. How is the uncertainty of 15% determined?
5. Page 2769, discussion of Figure 1.
  - a. Lines 14-16. Do the authors have any insights why the model-measured discrepancy of these three experiments is much larger than other cases? Please comment.
  - b. Lines 19-20. The authors noted that "...the model tends to over-predict the SOA concentration at high OH exposure by a factor of  $\sim 2$ ". This is not obvious from Figure 1. Please explain. I suggest the authors to color the points in Figure 1a by OH exposures (and use different symbols for different  $\alpha$ -pinene initial concentrations).
  - c. In Figure 1a, there is a wide range of observed Coa even for experiments with similar initial  $\alpha$ -pinene concentrations (e.g., the two pink diamonds to the right, and the two blue diamonds in the middle). Why? Also, while the difference in the observed Coa is about 100  $\mu\text{g}/\text{m}^3$  for these cases, it is curious that the modeled Coa is actually in better agreement. (e.g., the modeled Coa values for the blue diamonds in the middle are both around 200  $\mu\text{g}/\text{m}^3$ ). Please comment and address this in the revised manuscript.
6. Page 2770, description of CIT results. The modeled CIT results (both Coa and O/C) appear to be extremely similar to the modeled PAM results, even though the CIT and PAM experiments are performed under very different conditions. Are all the parameter inputs (Table 2) the same for CIT and PAM data, perhaps except for the initial  $\alpha$ -pinene concentration?
7. Page 2770 and 2771. The authors stated that the differences between modeled and observed Coa for both CIT and PAM results could arise from experimental uncertainties. What "experimental uncertainties" are the authors referring to? When taking experimental conditions into account (Page 2771 line 3), it appears that the discrepancy between modeled vs. observed Coa is even worse. Please explain.
8. Page 2771, line 19. The typical case chosen is " $\alpha$ -pinene = 281  $\mu\text{g}/\text{m}^3$ ", is there a reason why this "typical case" is not used to examine the trends in SOA volatility distributions as a function of OH exposure? (instead, an experiment with 443  $\mu\text{g}/\text{m}^3$  reacted  $\alpha$ -pinene is chosen here).
9. Page 2773 and 2774. Both the modified models and global sensitivity analysis do not reproduce the trajectory of evolution of O/C as a function of OH exposure. Does this mean that the parameters included in this study, even when all are allowed to vary, cannot capture this trend? If so, how do the authors justify the usefulness of all these parameters in predicting O/C?
10. Page 2776, line 3. The authors stated that "...gas reaction rate has limited impact on Coa". This appears to be contradictory with the results shown in Figure 1c, where changing the OH reaction rate in the gas-phase obviously leads to changes in Coa. Please clarify.
11. Page 2776, line 19. Heterogeneous reaction rate. One of the main differences between PAM reactors and laboratory chambers is the OH concentration in the gas-phase, in which the OH concentration in PAM is many orders of magnitude higher than ambient OH concentration. Among all the parameters, the KOH-hetero is the only parameter that becomes progressively more important for both Coa and O/C (Figure 6). With the very high OH concentration in the

PAM reactors, is it possible that the role of heterogeneous reactions becomes more important than what it would be in the ambient environment? If so, would the results from this study be representative of ambient conditions?