General comments

This manuscript explores an important and previously unexplained paradox in secondary organic aerosol (SOA) formation, namely that chamber experiments suggest that the majority of SOA is semi-volatile while recent studies focused on particle nucleation show high mass yields of very low volatility compounds. The authors present a comprehensive modeling analysis of previously published experimental data in an attempt to explain this paradox. They propose and test several possible theories to explain the paradox and conclude by suggesting further experiments needed to fully resolve it. The manuscript fits very well within the scope of ACP and is scientifically sound. I think the manuscript is suitable for publication in ACP, but further clarification/explanation is needed in several parts of the manuscript as well as minor technical corrections.

Major comments

1. Page 6, lines 1-11: Is the rate of vapor wall loss assumed to be the same for both the CMU and CLOUD chamber? Is this a reasonable assumption? What is the size difference between the two chambers? If the surface-to-volume ratio between the two chambers is significantly different, the vapor wall loss rates may also differ substantially. How might a significantly different vapor wall loss rate between the two chambers affect the overall conclusions of the manuscript?

2. Page 6, lines 13-24: I found this paragraph describing particle wall loss vague and somewhat confusing. Please explain more clearly what $k^{s,d}$ is and how it is defined. On line 19, the authors state "we rely on the seed loss rate measured prior to the injection of α-pinene." Is this the total number or total volume loss rate? Is $k^{s,d}$ defined as this loss rate? One line 20, what is meant by "minor discrepancies between the data and the model concerning the mass of seeds in the chamber"? Does the model over- or underestimate the mass of seeds in the chamber? Why does this discrepancy exist since the mass of seeds is not directly measured by the SMPS? If the measured loss rate is used to extrapolate the initial mass of seeds in the chamber and then this loss rate is applied in the model, I would assume that the modeled and 'measured' (which must still be corrected by this loss rate) mass of seeds would be identical.

4. Page 17, line 12-13: What is the significance of a higher condensation rate? Do the authors mean to suggest that there is a higher nucleation rate for a lower mass yield? Wouldn't a higher condensation rate also increase the yield for the CMU experiment?

5. Page 17, section 3.3: How does this section affect the conclusions regarding discrepancies in volatility distributions between the CMU and CLOUD experiments? At the beginning of this section, page 18, line 7-10, the authors state, "if larger particles had a lower effective mass accommodation coefficient (for example driven by slow particle-phase diffusion), that might direct more vapors to the walls and lower the overall observed SOA production. We explore this by varying the particle condensation sink in our simulations." However, the results of these simulations are never connected back to this initial hypothesis. Do these simulations support the idea that the lower yield in the CMU chamber could be due to changes in the particle condensation sink? In its current form, this section does not contribute to the overall attempt to reconcile the CLOUD and CMU experiments.

Minor comments

1. Throughout the manuscript, the authors switch between using the words “dynamic” and “dynamical.” These words are similar but in some instances have slightly different meanings. I would recommend choosing one version and using it throughout the paper.

2. Page 1, line 10: What experiments is the phrase “substantially lower SOA mass yields” referring to? The CMU SOA experiments or the CLOUD experiments? If referring to the CLOUD experiments, I recommend mentioning in the abstract how the second oligomerization model behaves for the CMU experiments.

3. Page 1, lines 22-25: This sentence is very long and somewhat confusing. It would be easier for the reader if it was broken up into two sentences.

4. Page 3, Figure 1: What are the small black arrows drawn between the volatility bins? They are not mentioned in the text or the caption and resemble a reverse aging scheme. I recommend explaining what is meant by the arrows or removing them.

5. Page 5: The organization between the “Dynamic Model” and “Results and Discussion” section could be clearer. Specifically, I would prefer to see a subsection within the “Dynamic Model” section (which could possibly be renamed “Methods”) describing the experimental conditions of both the CLOUD and CMU experiments (i.e. moving the first paragraph from “Results and Discussion” here and adding a brief description of the CLOUD experiments). It will be helpful to the reader to have both experiments described in the same section for later reference.

6. Page 5, line 21: I believe “low volatility” is meant in in place of “semi-volatile”: the volatility bin that is referenced is $10^{-1}$ μg $m^{-3}$ and the corresponding abbreviation in parenthesis is “LVOC”.

7. Page 6, section 2.2: It was unclear if the volatility distribution used was taken directly from Tröstl et al. (2016) or if the authors performed the fit themselves. Furthermore, was the temperature correction done by Tröstl et al. (2016) or the authors?

8. Page 6: lines 31-32: I found the phrasing of this sentence awkward and had to reread it several times to understand it.

9. Page 8: equation (5): What are $\nu_{i,p}$ and $B_{i,p}$? These are not defined in the subsequent paragraph. What value is used for the accommodation coefficient $\alpha_{i,p}$?

10. Page 9, line 4: I believe “(E)LVOC **vapor** wall loss” is meant in place of “(E)LVOC **particle** wall loss.”

11. Page 9, Figure 3: It is unclear how the light blue area, the Precursor Yield Potential, is calculated. The caption states that it is unreacted precursor, but line 7 of the following page states it is “the concentration of organics that have not yet been formed by ozonolysis.” How is this calculated?

12. Page 11, line 9: What is a “perfect correction for the deposited particle mass”? Is this related to the lower or upper limit assumption (i.e., Loza et al., 2012)?

13. Page 15, lines 24-15: What are the “constant HOM experiment” and the “increasing HOM experiment”? These were not described previously.

14. Page 18, line 10: Please describe more explicitly what the base case is. Does the high (E)LVOC simulation include the additional LVOC concentration required to explain the observed growth rates in CLOUD?

Loza, C. L.; Chhabra, P. S.; Yee, L. D.; Craven, J. S.; Flagan, R. C.; Seinfeld, J. H. Chemical aging of m-xylene secondary organic aerosol: laboratory chamber study. Atmos. Chem. Phys. 2012, 12, 151−167.