

Interactive comment on “Influence of Seed Aerosol Surface Area and Oxidation Rate on Vapor-Wall Deposition and SOA Mass Yields: A case study with α -pinene Ozonolysis” by T. Nah et al.

Anonymous Referee #1

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This manuscript explores the effects of seed aerosol surface area and oxidant concentrations on chamber wall losses and SOA yields. New chamber experiments are conducted, analyzed and modeled. The topic is important and the manuscript is well written, but I have a few concerns which should be addressed before publication in ACP as detailed below.

Major comments

1. The authors assume in their analysis that coagulation between particles is negligible. I am worried that this assumption may not be appropriate and may be biasing the results,

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and I hence request that they justify this assumption by simulating coagulation of a typical particle size distribution (with or without simultaneous wall losses) and comment on the results. The assumption of “no coagulation” is made explicitly or implicitly in different places in the manuscript, for example:

lines 228-229: The smallest diameter bin is initialized by the total number of particles measured at the end of the experiment to account for the fact that the model does not simulate nucleation. I am especially worried about the assumption in this instance since the smaller particles formed during nucleation are especially likely to coagulate.

lines 251-253: The authors state they are using a model without coagulation because including coagulation showed no change in the predicted SOA mass concentrations. It is not clear to me what is meant by this statement. The model contains several fitting parameters – were the best values for the fitting parameters the same if coagulation was included in the model? Further, even if there is no (large) change in predicted SOA when including coagulation, if a model with coagulation is available, why did the authors not use that model as it is expected to be more accurate?

There is some indication that ignoring coagulation may be biasing results. For example: lines 355-358: In their analysis of the appropriateness of the wall deposition number correction the authors note that the loss-corrected particle number concentrations are 9-17% less than the initial number concentrations for seeded experiments, and that it is unclear why this might be the case. Could this not be due to coagulation which is not accounted for in the calculations?

2. lines 405-407: The authors note that “Higher SOA mass yields are observed in the 500 ppb O₃ experiments, which indicates that the α -pinene oxidation rate controls the absolute amount of SOA formed.” It seems appropriate in this context to comment on why this may be the case – is it due to reduced wall losses when the oxidation rate is higher? This seems inconsistent with the observation that the SOA formation is not kinetically limited.

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3. lines 477-479: I am unconvinced based on the data shown that the vapor-particle mass accommodation coefficient (α_p) equals 1 for two main reasons: 1) as the authors recognized, different combinations of fitting parameters could give similarly good fits (not explored in this manuscript – in the sensitivity analyses shown only one parameter is changed at a time) and 2) based on the data shown in Figure S8, $\alpha_p = 0.1$ seems to yield similar agreement with data as $\alpha_p = 1$. Thus, in my opinion the authors should not base conclusions on the result that $\alpha_p = 1$.

Overall it is not clear how the fitting parameters were chosen. Figures are shown comparing modeled and measured results for different parameter choices. Was the choice of model parameter based on a visual comparison of modeled and measured data? This reminds me of modeling of thermodynamic data (which also includes several fitting parameters), where Karnezi et al. (2014) have updated an evaporation model to explore the parameter space more fully. A similar approach seems appropriate for the model used in this manuscript.

Karnezi, E.; Riipinen, I.; Pandis, S. N. Measuring the atmospheric organic aerosol volatility distribution: a theoretical analysis. *Atmos. Meas. Tech.* 2014, 7, 2953–2965.

4. The overall take-away message from this manuscript is unclear. The authors discuss the effect of seed surface area on vapor-wall deposition and resulting SOA yields, how this effect can be mitigated through the use of additional oxidant. . . but also that high oxidant levels may not be atmospherically relevant. This emphasizes the complexity of these experiments and their evaluation but does not provide guidance on how future chamber experiments should be conducted. It would be useful if the authors could add such recommendations in their discussion.

Minor comments

5. lines 133-134: Please add a comparison of the reactions rates (cyclohexane + OH vs. α -pinene + OH) since that (not the ratio of cyclohexane and α -pinene) determines the effectiveness of the OH scavenger.

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6. lines 149-150: I wonder whether it is appropriate to call this an “initial ratio” since the α -pinene is reacting away while the ozone is injected. Please address how much α -pinene has reacted when ozone injection is completed and to what extent the ratio of VOC/oxidant can truly be controlled in these experiments. Please also discuss the mixing time scale in the chamber and potential effects of α -pinene initially reacting with ozone “hot spots”.

7. Table S1: units should be specified for the particle number concentrations (e.g. particles per cubic centimeter)

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