We greatly value the careful reading and the detailed comments provided by the referees. The responses to the comments of the two referees in our direct reply (shown below) and within the revised manuscript (see marked copy) are provided below. The pages and lines indicated below correspond to those in the marked copy.

Response to Referee 1 (Referees' comments are italicized)

1. Referee comment: "*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, 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?"

Author response: We agree with the reviewer that coagulation may potentially be important and in fact are currently performing experiments to analyze the relationship between coagulation and particle wall loss. We expect to publish these results soon. In the meantime, we have added a brief analysis of coagulation to the current paper. As requested, we reanalyzed the AS seed-only particle wall loss data and α-pinene ozonolysis SOA growth data, this time accounting for coagulation. We applied the Aerosol Parameter Estimation (APE) model (detailed by Pierce et al. (2008)) to the AS seed-only particle wall loss experimental datasets to derive particle wall loss coefficients that correct for particle coagulation. This procedure was performed for both the low and high AS seed-only experiments. As described in Pierce et al. (2008), the coagulation-corrected particle wall loss coefficients were determined by simulating the decay of the initial size distribution due to coagulation and then attributing the difference in this decay and the observed decay to particle wall loss. All nucleation and low AS data were then reanalyzed using modeled particle wall loss coefficients that correct for coagulation derived from the low AS seed-only experiments Similarly, all high AS data were reanalyzed using modeled particle wall loss coefficients that correct for coagulation derived from the high AS seed-only experiments.

Based on our reanalyzed α-pinene ozonolysis SOA growth data, the SOA mass yields stay roughly constant despite the increase in AS seed surface area for both O_3 concentrations. Higher SOA mass yields are observed in the 500 ppb O_3 experiments. These trends are similar to those presented in the original manuscript where we assumed that coagulation is negligible (and therefore not corrected for) in all our experiments. In addition, these newly obtained SOA mass yields obtained at peak SOA growth (where coagulation is corrected for) are generally consistent with those of previous studies.

These newly obtained absolute SOA mass concentrations and mass yields (where coagulation is corrected for) are higher than those reported in the original manuscript (where coagulation is not corrected for) by < 2 % (absolute values). Therefore, we do not anticipate these results will affect our main conclusions that (1) SOA formation in the α-pinene ozonolysis system is governed by quasi-equilibrium growth since the SOA mass yields stay roughly constant despite the increase in AS seed surface area for both O_3 concentrations, and (2) there is an "oxidation rate effect" since higher SOA mass yields are observed in the 500 ppb O_3 experiments.

We have added a discussion on the potential effect of coagulation on the SOA mass yields measured in this study to the revised manuscript:

Page 14 line 377: "Red and blue solid lines in Fig. 1 shows the size-dependent particle wall deposition coefficients measured in the low AS seed-only and high AS-seed only deposition experiments. In these measurements, we assume that the number concentration is low enough such that the effect of coagulation is small and only particle wall deposition affects the particle size distribution, thus allowing for the direct measurement of size-dependent particle wall deposition coefficients. The initial total AS seed surface area concentration in the low AS-seed only and high AS-seed only experiments (which are conducted using 0.015 M AS and 0.05 M AS solutions, respectively) are similar to those used in the α-pinene ozonolysis experiments (i.e., ~1000 and ~3000 μm^2 cm^{-3}, respectively). As shown in Fig. 1, the measured particle wall deposition coefficients from the low AS-seed only and high AS-seed only experiments generally fall within the range of those measured in routine monthly AS-seed only experiments conducted in the chamber. Figure 1 also shows the size-dependent particle wall deposition coefficients corrected for coagulation, shown using dashed lines, which are obtained as described in Pierce et al., 2008 using the data from the low AS-seed only and high AS-seed only experiments. A comprehensive description of the relationship between coagulation and particle wall deposition will be provided in a forthcoming publication. Briefly, as described in Pierce et al. (2008), the coagulation-corrected particle wall loss coefficients are determined by simulating the decay of the initial size distribution due to coagulation and then attributing the difference in this decay and the observed

decay to particle wall loss. For both the measured and coagulation-corrected particle wall deposition coefficients, the minimum coefficient for the low AS-seed only experiment is different from that of the high AS-seed only experiments. The cause of this difference is currently under investigation but may be due in part to uncertainties arising from the low particle number concentrations for the larger particles in the low AS-seed only experiment. To study how coagulation can potentially affect SOA mass yields in this study, both the measured and coagulation-corrected size-dependent particle wall deposition coefficients are used to correct for particle wall deposition in the α-pinene ozonolysis experiments.

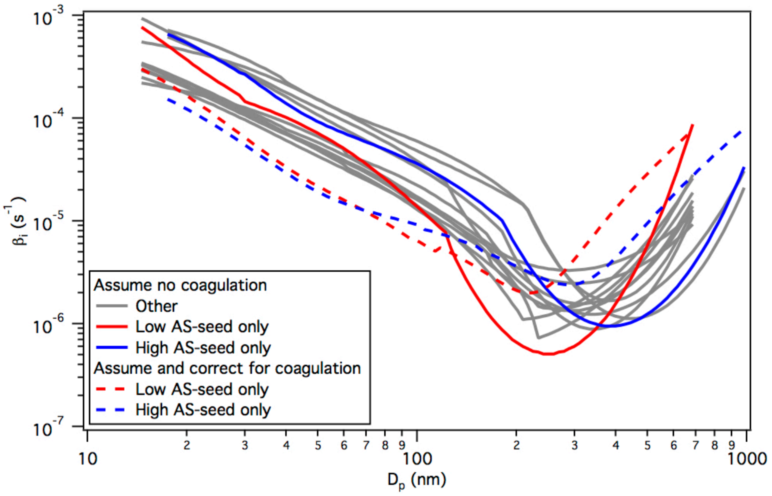


Figure 1: Particle wall deposition coefficients (β_i) measured during the low AS-seed only and high AS-seed only experiments in GTEC. Also shown are the particle wall deposition coefficients (labeled "Other") measured in previous routine monthly AS-seed only experiments in the chamber. These previous routine monthly AS-seed only experiments were performed using either a 0.008 M AS or a 0.1 M AS solution. Coagulation-corrected particle wall deposition coefficients (see Pierce et al. (2008) and main text for details) are also shown, using dashed lines.

Assuming that the effect of coagulation is small, the particle wall deposition corrected number concentration data provide a test of the appropriateness of the particle wall deposition correction. The corrected number concentration should level off at a constant value (i.e., the initial particle number concentration), assuming no significant coagulation, when particle wall deposition is properly accounted for since the wall-deposited particle number distribution is added to the suspended particle number distribution during particle wall loss correction. Neglecting coagulation, we account for particle wall deposition in nucleation and low AS experiments using deposition coefficients measured from the low AS-seed only experiments, while particle deposition in high AS experiments are accounted for using coefficients measured from the high AS-seed only experiments. Figures S1 and S2 show the particle wall deposition-corrected aerosol number and volume concentrations. Over all experiments, the particle wall deposition-corrected final particle number concentration (i.e., at the end of the reaction) is 9 to 17 % less than the initial particle number concentration for the low AS and high AS experiments

(Table S1), respectively, indicating that the particle wall deposition-corrected volume concentrations are slightly underestimated. The fact that the particle wall deposition-corrected final particle number concentrations are somewhat smaller than the initial particle number concentrations may be due to variations in particle wall deposition rates in the AS-seed only and α-pinene ozonolysis experiments or to coagulation. To first examine variations in particle wall deposition rates, we used the average of the measured low AS-seed only and high AS-seed only particle wall deposition coefficients to account for particle wall deposition in all the experiments (Figs. S3 and S4). While there is a negligible difference in the particle wall deposition corrected volume concentrations (Figs. S3 and S4 vs. Figs. S1 and S2), a larger spread (1 to 22 %) exists in the difference between the initial and final particle number concentrations when the average measured particle wall deposition coefficients are used (Table S1). Therefore, all subsequent nucleation and low AS data presented here are particle wall deposition-corrected using coefficients measured from the low AS-seed only experiments, and all high AS data are corrected using particle wall deposition coefficients measured from the high AS-seed only experiments. We furthermore conclude that variations in particle wall deposition rates do not cause the decrease in the particle wall deposition-corrected final number concentration and is most likely due to coagulation. Thus, the SOA data are also corrected using the coagulation-corrected particle wall deposition coefficients (discussed below). We show subsequently the relatively minor difference that correcting for coagulation has on overall SOA mass yields. Therefore, we use SOA concentrations corrected using the measured particle wall deposition coefficients for the bulk of the analysis in this study."

Page 18 line 493: "To investigate the influence of coagulation on the SOA mass yields, the coagulation-corrected size-dependent particle wall deposition coefficients are also used to correct for particle wall deposition in the α-pinene ozonolysis experiments. Specifically, all nucleation and low AS data are particle wall deposition-corrected using coagulation-corrected coefficients derived from the low AS-seed only experiments, and all high AS data are corrected using coagulation-corrected particle wall deposition coefficients derived from the high AS-seed only experiments. Figure S7 shows the time-dependent SOA mass yields (obtained using the coagulation-corrected and measured particle wall deposition coefficients) as a function of initial total AS seed surface area. SOA mass yields obtained using the coagulation-corrected particle wall deposition coefficients are < 2 % (absolute values) higher than those using the measured particle wall deposition coefficients. Similar to the SOA mass yields obtained using the measured particle wall deposition coefficients (Figs. 4, S7c and S7d), SOA mass yields obtained using the coagulation-corrected particle wall deposition coefficients stay roughly constant despite the increase in AS seed surface area for both O_3 concentrations, and the SOA mass yields are higher in the 500 ppb O_3 experiments (Figs. S7a and S7b). The mass yields obtained at peak SOA growth are also generally consistent with those of previous studies (Fig. S8). Taken together, this suggests that the effect of coagulation on the SOA mass yields is likely minor for the aerosol concentrations used in this study. Therefore, only data that have been particle wall deposition-corrected using coefficients measured in the low AS-seed only and high AS-seed only experiments

are fitted to determine model parameters for the vapor-particle dynamics model described in Section 3."

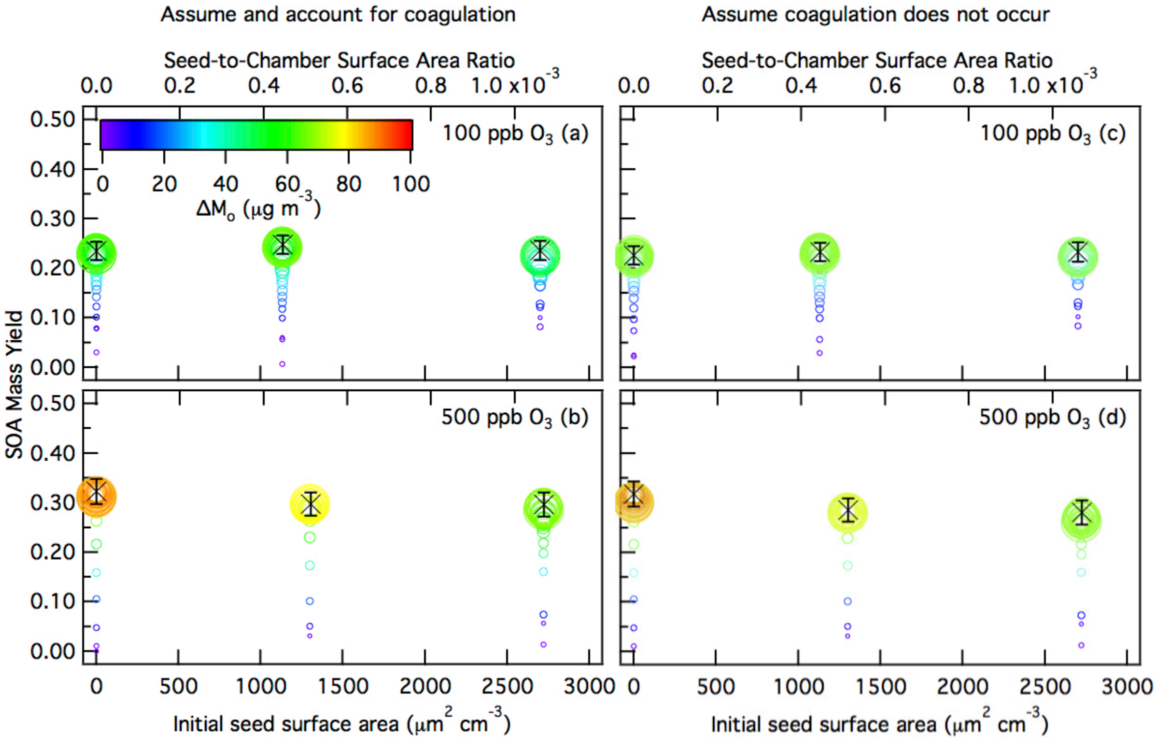


Figure S7: 10 min-averaged SOA mass yields over the course of an α-pinene ozonolysis experiment as a function of initial total AS seed surface area concentration. Panels (a) and (b) show the SOA mass yields obtained using the coagulation-corrected size-dependent particle wall deposition coefficients for the 100 and 500 ppb O_3 experiments, respectively. Panels (c) and (d) show the SOA mass yields obtained using the measured size-dependent particle wall deposition coefficients (that account for coagulation) for the 100 and 500 ppb O_3 experiments, respectively (also shown in Fig. 4 of the main text). Symbol color indicates the SOA mass concentration and symbol size indicates the time after O_3 is injected into the chamber. The × symbols are the SOA mass yields at peak SOA growth. The y-axis error bars represent the uncertainty in the peak SOA mass yield, which originates from the α-pinene injection and the aerosol volume concentration measured by the SMPS at peak SOA growth (one standard deviation). As discussed in the main text, the use of coagulation-corrected particle wall deposition coefficients for particle wall loss correction does not change the conclusions of this work: 1) SOA mass yields are enhanced at higher O_3 concentrations, and 2) there is a lack of a SOA mass yield dependence on the seed surface area within the range of AS seed surface area concentration used in this study.

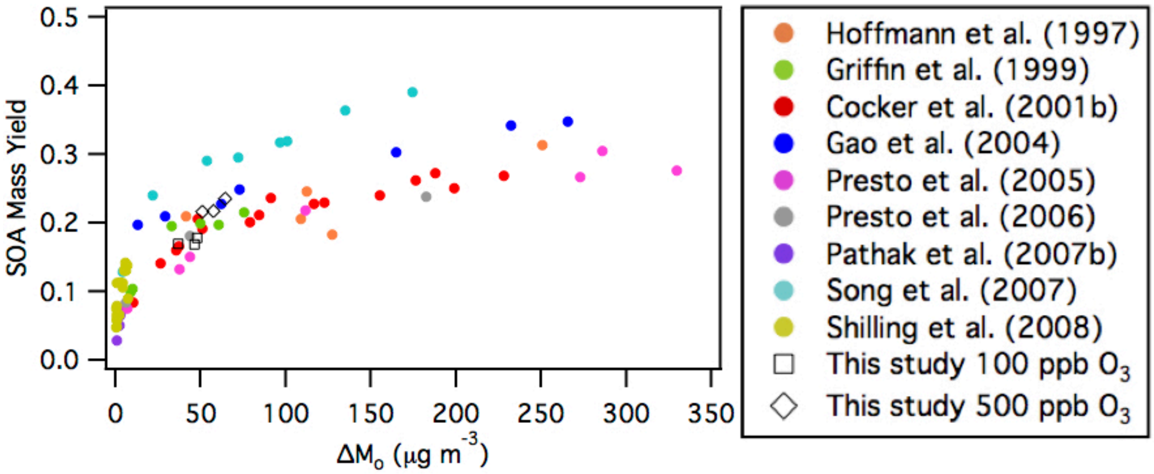


Figure S8: Comparison of SOA mass yields obtained using the coagulation-corrected size-dependent particle wall deposition coefficients to those of previous dark α-pinene ozonolysis studies (Table S2). The SOA mass yields and concentrations of majority of these previous studies (Hoffmann et al., 1997; Griffin et al., 1999; Cocker et al., 2001b; Gao et al., 2004; Presto et al., 2005; Presto et al. 2006; Pathak et al., 2007b; Song et al., 2007) were previously compiled by Shilling et al. (2008). Similar to Shilling et al. (2008), all the data shown here (including those reported in this study) have been adjusted using an organic density of 1.0 g cm^{-3}, and to 298 K using a temperature correction of 1.6 % per K, as recommended by Pathak et al. (2007b) to facilitate easier comparison among the different studies.

2. Referee comment: "*lines 405-407: The authors note that "Higher SOA mass yields are observed in the 500 ppb O3 experiments, which indicates that the a-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.*"

Author response: We believe that the higher SOA mass yields measured in the 500 ppb O$_3$ experiments is due to reduced vapor-wall deposition, even though SOA formation in the α-pinene ozonolysis system is governed by quasi-equilibrium growth. We direct the referee to sections 5.2 and 5.3 for a detailed discussion on why that is the case.

3. Referee comment: "*lines 477-479: I am unconvinced based on the data shown that the vapor-particle mass accommodation coefficient (alpha_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 thermodenuder 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."

Author response: We thank the reviewer for the helpful reference. We have followed the methodology of Karnezi et al. (2014) and conducted a wide parameter sweep. We report both the parameters determined as optimal using the Karnezi et al. inverse error-weighting approach, as well as the combination of parameters with the lowest percentage error. We find that the lowest-error parameters give the best fit to the data, so we use these parameters for our analysis. The Karnezi et al. parameters and the lowest-error parameters are similar to each other, and to the parameters originally chosen in the manuscript, validating that these parameters are relatively robust. The following discussion was added to the paper:

Page 12, line 337: "In order to determine the parameters for α_w, α_p, τ_{olig} and the branching ratios between the oxidation products that provide the best fit to measured SOA data, the parameter space was discretized and all possible combinations of parameters were simulated, following Karnezi et al. (2014). In order to restrict the number of combinations required, only parameter values judged to be physically realistic were chosen. Because the branching ratios in this model are mole-based, they must sum to one; therefore only combinations of parameters summing to one were allowed. The discretization is shown in Table S1 and results in roughly 10,000 different combinations of parameters. All six experiments were simulated with each parameter combination, and simulations were run using GNU Parallel (Tange 2011). For each combination of parameters, the percentage error was calculated from equation 10 of Karnezi et al. (2014):

$$\boldsymbol{E_i = \frac{100}{n}\sqrt{\textstyle\sum_i^n (Moa_{i,guess} - Moa_{i,meas})^2}} \qquad \textbf{(13)}$$

where $Moa_{i,guess}$ is the model-predicted SOA mass concentration at a particular timestep i for one of the experiments, $Moa_{i,meas}$ is the measured SOA mass concentration at a particular timestep i for one of the experiments, and n is the number of timesteps summed over all experiments. The best-fit combination of parameters is defined as the combination of parameters with the lowest percentage error. This lowest-error combination of parameters was compared to the "best estimate" parameters determined from the inverse error weighting factor of Karnezi et al. (2014):

$$\boldsymbol{\bar{x} = \frac{\sum_j^N \left[x_j \frac{1}{E_j}\right]}{\sum_j^N \frac{1}{E_j}}} \qquad \textbf{(14)}$$

where x_j is a value of one of the parameters (α_w, α_p, τ_{olig} or a branching ratio

between the oxidation products), with N different possible combinations of parameters, and E_j is the percent error for that particular combination of parameters. The lowest-error combination of parameters and the Karnezi et al. (2014) best estimate parameters are both reported, but the lowest-error combination of parameters resulted in a lower percentage error than the Karnezi et al. (2014) best estimate parameters. The lowest-error combination of parameters is used for the modeling analysis."

Table S2: Discretization of parameters

Parameter	Discretization
α_p	1, 0.1, 0.01, 0.001
α_w	10^{-7}, 10^{-6}, 10^{-5}
τ_{olig}	4, 6, 8
>10^3 branching ratio[a]	0.5, 0.6, 0.7, 0.8, 0.9, 1
10^2 branching ratio[a]	0, 0.1, 0.2, 0.3, 0.4, 0.5
10 branching ratio[a]	0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.1, 0.15, 0.2
1 branching ratio[a]	0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.1, 0.15, 0.2
0.1 branching ratio[a]	0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.1, 0.15, 0.2

[a]Only combinations of parameters summing to one were allowed.

Page 19, line 519: "As noted earlier, optimal model values for α_p, α_w, τ_{olig} and the branching ratios between the oxidation products were determined by calculating the error between the observed and modeled time-dependent SOA concentrations for all possible combinations of model parameters. The combination of parameters with the lowest percent error is $\alpha_w = 10^{-6}$, $\alpha_p = 0.1$, $\tau_{olig} = 4$ h, branching ratios = 0.6, 0.3, 0.05, 0.05 and 0 for oxidation products with vapor pressures >10^3, 10^2, 10, 1 and 0.1 µg m^{-3}, respectively. This combination of parameters results in a percent error of 21% (Table S4). It is important to note that predictions using $\alpha_p = 0.1$ or 1 resulted in very similar errors; with the same combination of parameters and $\alpha_p = 1$, the percent error only increased to 22%. The "best estimate" parameters determined following the Karnezi et al. (2014) method are as follows: $\alpha_w = 3.6\times10^{-6}$, $\alpha_p = 0.35$, $\tau_{olig} = 6$ h, and branching ratios = 0.66, 0.16, 0.06, 0.06, and 0.06 for oxidation products with vapor pressures >10^3, 10^2, 10, 1 and 0.1 µg m^{-3}, respectively. This combination of parameters results in an error of 37% (Table S4). Model predictions using both sets of parameters are compared to measured SOA concentrations in Fig. S9. The lowest-error parameters are used for the analysis in the remainder of this study."

Table S4: Best-fit parameters, using lowest percentage error and Karnezi et al. (2014) method

Parameter	Lowest percentage error	Karnezi et al. (2014) method
α_p	0.1	0.35
α_w	10^{-6}	$3.6\text{x}10^{-6}$
τ_{olig} (h)	4	6
>10^3 branching ratio	0.6	0.66

10^2 branching ratio	**0.3**	**0.16**
10 branching ratio	**0.05**	**0.06**
1 branching ratio	**0.05**	**0.06**
0.1 branching ratio	**0**	**0.06**
Percentage error for combination	**21%**	**37%**

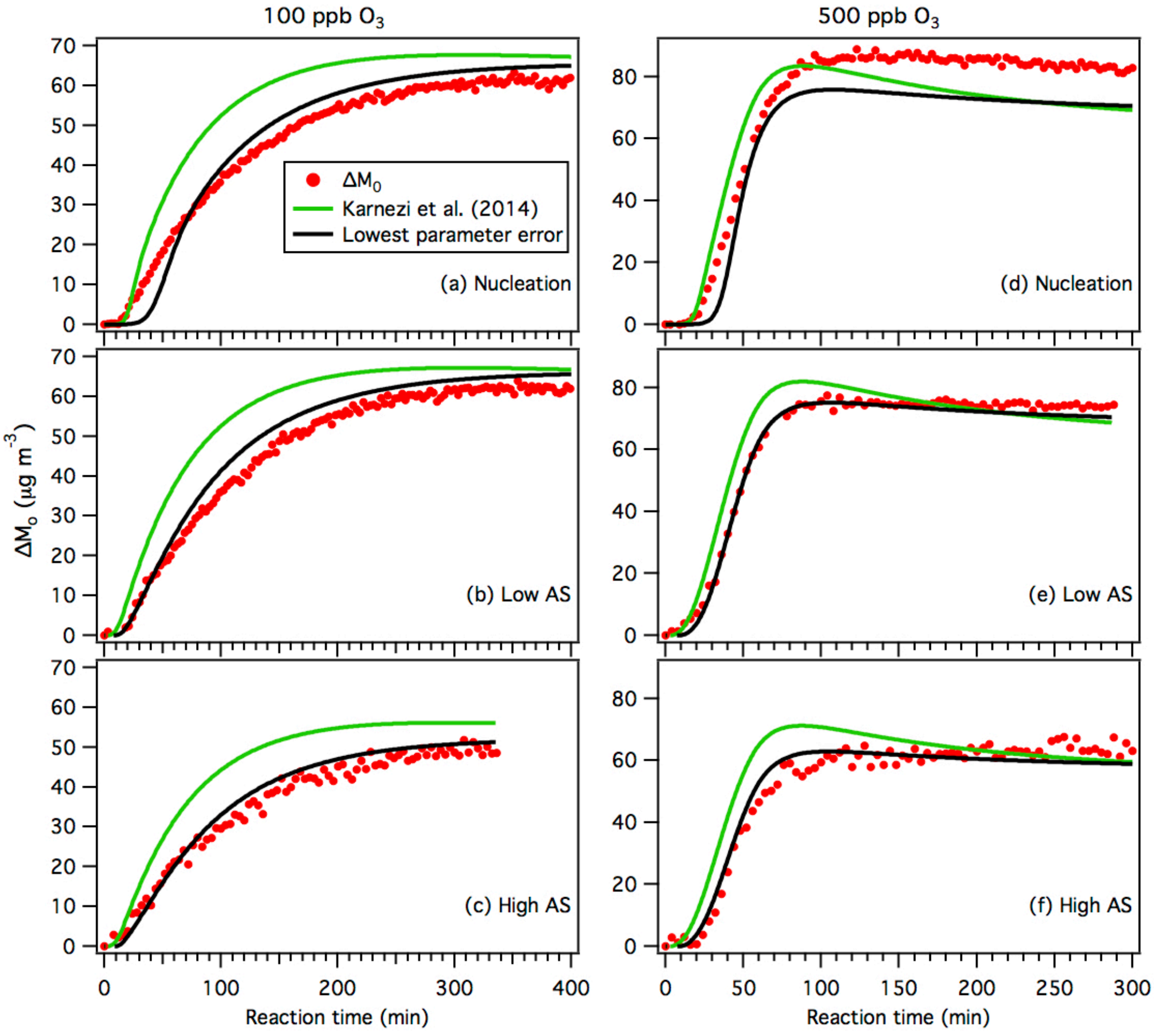


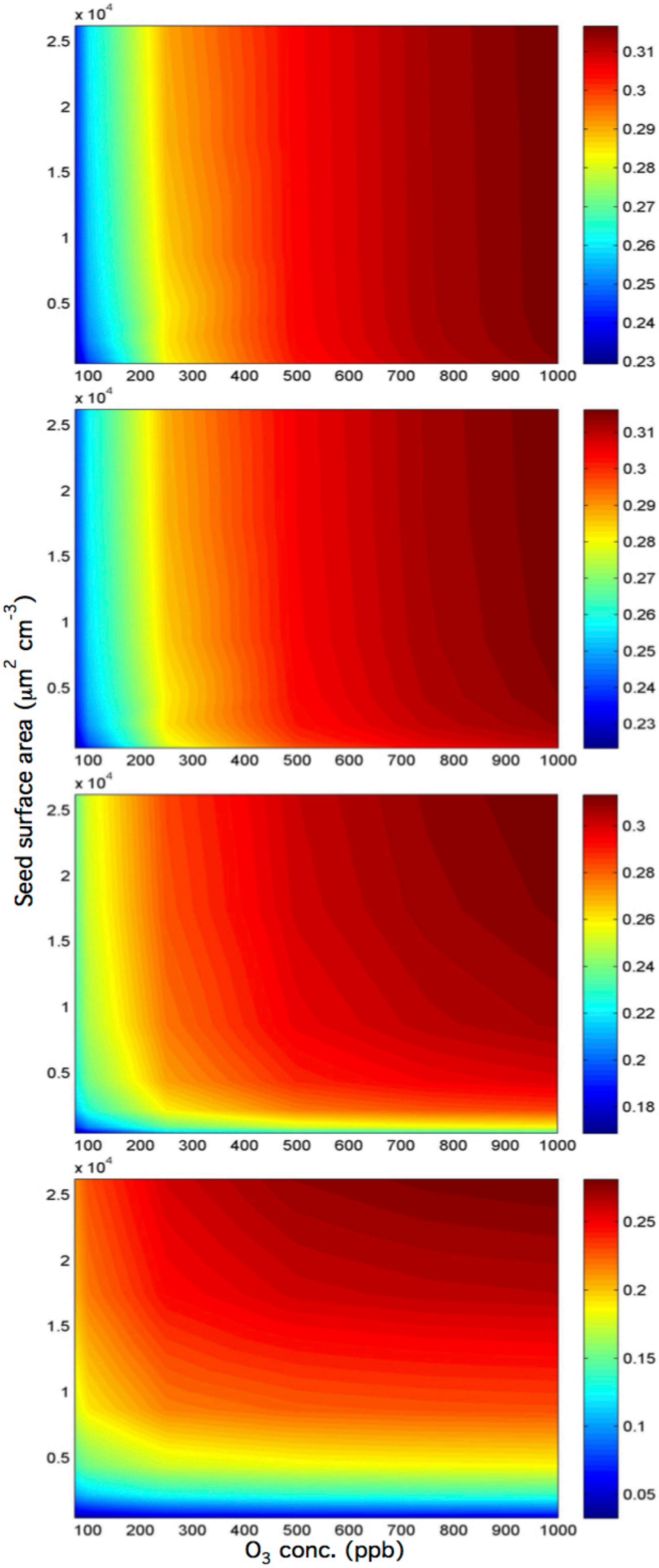
Figure S9: Reaction profiles for measured and modeled SOA concentration, using both the Karnezi et al. (2014) parameters and the lowest-error combination of parameters (see Table S4). Panels (a), (b) and (c) show results from the nucleation, low AS and high AS 100 ppb O_3 experiments, respectively. Panels (d), (e) and (f) show results from the nucleation, low AS and high AS 500 ppb O_3 experiments, respectively.

Page 21 line 588: "The best-fit α_p = 0.1 (or α_p = 1, with almost the same percentage error) suggests the absence of significant limitations to vapor-particle mass transfer in the present α-pinene ozonolysis study, and that SOA formation is governed by

quasi-equilibrium growth (Saleh et al., 2013; McVay et al., 2014), which occurs when SOA-forming vapors are produced at a rate that is significantly slower than that required to establish gas-particle equilibrium (Shiraiwa and Seinfeld, 2012; Zhang et al., 2012)."

Page 21 line 598: "The best-fit α_p = 0.1 is within the range of α_p coefficients determined from α-pinene ozonolysis SOA thermodenuder studies (α_p = 0.1) (Saleh et al., 2013; Saha et al., 2016) and α-pinene photooxidation chamber studies (α_p = 0.1 or 1) (McVay et al., 2016)."

Page 24 line 690: "A key aspect of vapor wall deposition is the potential interplay between the seed aerosol surface area effect and the oxidation rate effect. To examine this interplay in the α-pinene ozonolysis system, simulations were carried out by varying the seed aerosol surface area and the O_3 concentration simultaneously, while using the branching ratios, oligomerization rate, and vapor wall deposition rate parameters obtained in the present study. The initial α-pinene concentration was set to 50 ppb, and a fixed O_3 concentration was used in place of a linear injection. α_p was varied at 0.001, 0.01, 0.1, and 1 in these simulations. Figure 6 shows the SOA mass yield at peak SOA growth as a function of both the seed aerosol surface area and O_3 concentration for α_p = 1, 0.1, 0.01, and 0.001. For α_p = 1 or 0.1, the oxidation rate dominates: SOA mass yield increases significantly as O_3 concentration increases while the seed aerosol surface area has a negligible effect. For α_p = 0.01, both effects can be observed in different regions: at low O_3 concentrations and high seed aerosol surface areas, the oxidation rate effect dominates; at low seed aerosol surface areas and high O_3 concentrations, the seed surface area dominates. At low seed aerosol surface areas and low O_3 concentrations, both effects are present. For α_p = 0.001, the seed aerosol surface area effect dominates except at very high seed aerosol surface areas. These observations show that the presence of an oxidation rate effect and/or seed aerosol surface area effect depends on a complex interplay of factors, such as α_p, the rate of hydrocarbon oxidation, and the amount of seed surface area present."



(a) SOA mass yields at $\alpha_p = 1$

(b) SOA mass yields at $\alpha_p = 0.1$

(c) SOA mass yields at $\alpha_p = 0.01$

(d) SOA mass yields at $\alpha_p = 0.001$

Figure 6: SOA mass yields at peak SOA growth as a function of both the seed surface area and O_3 concentration for α_p = 1, 0.1, 0.01, and 0.001. The SOA mass yields at peak SOA growth are indicated by colors and contours. Note that the color bars for panels (a), (b) and (c) have different SOA mass yield ranges. Simulations were carried out using the optimal branching ratios, oligomerization rate, and vapor wall deposition rate parameters obtained in this study. The initial α-pinene concentration was set to 50 ppb, and a fixed O_3 concentration was used in place of a linear injection.

4. Referee comment: "*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.*"

Author response: As requested, we have expanded our discussion to include recommendations on how future chamber experiments should be conducted:

Page 30 line 850: "More importantly, the impact of vapor-wall deposition on SOA formation and evolution in various VOC systems conducted under different reaction conditions (regardless of atmospheric relevance) needs to be quantified through a combination of experimental and modeling efforts. Similar to this study, experiments should be performed using different seed aerosol surface area and oxidant concentrations to study their influence on vapor-wall deposition and SOA mass yields. If the effects of vapor-wall loss are found to be strongly dependent on seed aerosol surface area and/or oxidant concentrations (e.g. toluene photooxidation, where SOA formation may be underestimated by factors as much as four (Zhang et al., 2014)), further experiments aimed at measuring the wall deposition rates of the oxidation products should be performed. These wall deposition rates can then be used in predictive models to determine the vapor-wall and vapor-particle mass accommodation coefficients of these oxidation products. Consequently, this will allow us to determine the fraction of SOA-forming vapors partitioning to the particle phase vs. lost to the chamber walls (Zhang et al., 2015; Krechmer et al., 2016)."

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

Author response: Based on the concentrations of cyclohexane and α-pinene injected into the chamber, we expect the reaction rate of OH with cyclohexane to be ~60 times greater than α-pinene. This information is added to the revised manuscript:

Page 5 line 137: "Based on the concentrations of cyclohexane and α-pinene injected into the chamber, the reaction rate of OH with cyclohexane is ~60 times greater than that with α-pinene"

6. Referee comment: "*lines 149-150: I wonder whether it is appropriate to call this an "initial ratio" since the a-pinene is reacting away while the ozone is injected. Please address how much a-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 a-pinene initially reacting with ozone "hot spots"*."

Author response: We agree with the reviewer that it is not appropriate to call this an "initial ratio" since α-pinene was reacting away while O_3 was being injected. Based on our GC measurements, we estimate that ~11 % and ~98 % of the α-pinene had reacted when O_3 injection was completed in the 100 and 500 ppb O_3 experiments, respectively. This information is added to the revised manuscript. Since α-pinene was reacting away while O_3 was being injected and the chamber was operated in batch mode in this study, the α-pinene:O_3 ratio cannot be controlled exactly in these experiments. Therefore, to remove any confusion, we have made the following changes in the revised manuscript:

Page 5 line 154: "The injected α-pinene:O_3 molar ratio were approximately 1:2 and 1:10 in the 100 and 500 ppb O_3 experiments, respectively."

Page 6 line 158: "Approximately 11 % and 98 % of the initial α-pinene had reacted when O_3 injection was completed in the 100 and 500 ppb O_3 experiments, respectively."

Page 16 line 445: "At either O_3 concentration, the molar ratio of O_3 reacted to α-pinene reacted is approximately 1:1 (i.e., 50 ppb α-pinene reacted with 50 ppb O_3), which indicates that O_3 reacts only with α-pinene and not its oxidation products."

Page 21 line 581: "Figure 4 shows that for both O_3 mixing ratios used, the time-dependent SOA mass yield is similar at any given AS seed surface area (see also Table 1)."

We estimate the O_3 mixing timescale for all experiments to be ~12 min based on the O_3 time series traces in the O_3-only experiments. This information is added to the revised manuscript. Due to the design of the GTEC facility, α-pinene closest to the O_3 injection port of the chamber likely reacted first in the α-pinene ozonolysis experiments. However, we do not anticipate this experimental limitation to affect our conclusions (which are derived from SOA mass yields at peak SOA growth) since the O_3 mixing timescale is significantly shorter than the times at which SOA peak growth is achieved. As noted in the manuscript, SOA peak growth is achieved at reaction times ~300 to 350 min and ~100 min for the 100 and 500 ppb O_3 experiments, respectively (the start of O_3 injection into the chamber is reaction time 0 min).

We have made the following changes in the revised manuscript:

Page 6 line 159: "In the GTEC chamber, α-pinene closest to the O_3 injection port likely reacted first in the α-pinene ozonolysis experiments. The O_3 injection times were established in separate experiments in which only O_3 was injected into the chamber. Based on the O_3 time series traces in the O_3-only experiments, the O_3 mixing timescale was estimated to be ~12 min for all experiments."

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

Author response: The units for the particle number concentration are added in the revised manuscript.

Response to Referee 2 (Referees' comments are italicized)

1. Referee comment: "*Lines 674-677. This sentence does not include any mention of vapor loss to Teflon walls, only to organic matter deposited on the walls. Studies indicate that the loss to Teflon is more important.*"

Author response: The referee is correct in pointing out that the loss of SOA-forming vapors to Teflon walls is more important. Therefore, we have made the following changes to the revised manuscript to better reflect this point:

Page 28 line 806: "One possible explanation for the higher SOA mass yields in the continuous-flow, steady state, mode is that the SOA-forming vapors are in equilibrium with the chamber walls and seed aerosol, hence minimizing the irreversible loss of SOA-forming vapors to the chamber walls (Shilling et al., 2008)."

2. Referee comment: "*The discussion about kinetically-limited vs. quasi-equilibrium controlled SOA formation does not mention the effect of the time constant for oligomer formation. I would think that this has a significant effect on the growth regime, and so should be discussed.*"

Author response: The referee is correct in pointing out that the timescale of oligomerization may play an important role in the SOA growth regime (i.e., kinetically-limited vs. quasi-equilibrium). Our model results suggest that the timescale of oligomerization for the α-pinene ozonolysis system (where SOA formation is governed by quasi-equilibrium) is ~4 hours based on best-fit model predictions with the experimental data (Fig. S9). Similar modeling work (in addition to experimental work) will need to be performed on a VOC system where the SOA formation is governed by kinetically-limited growth (e.g. toluene photooxidation (Zhang et al., 2014)), in order to determine how the timescale of oligomerization may differ between the two SOA growth regimes. This is currently outside the scope of this paper, which is focused primarily on the analysis of SOA formation from the α-pinene ozonolysis system. However, we have made the following changes to the revised manuscript to raise the possibility that the timescale of oligomerization may play an important role in the different SOA growth regimes and needs to be investigated in future studies:

Page 27 line 784: "While not investigated in detail in this study, the timescale of oligomerization may play an important role depending on the SOA growth regime (i.e., kinetically-limited vs. quasi-equilibrium). Currently, it is unclear how the timescale of oligomerization in a VOC system where SOA formation is dominated by quasi-equilibrium growth (e.g. α-pinene ozonolysis) may differ from one that is dominated by kinetically-limited growth (e.g. toluene photooxidation (Zhang et al.,

2014)). This requires further investigation through a combination of experimental and modeling efforts to improve our understanding of how particle-phase processes (e.g. oligomerization) affect gas-particle partitioning, and consequently influence the magnitude by which vapor-wall deposition affects SOA mass yields."

3. Referee comment: "*It appears that the time constant for wall loss is much smaller that the values inferred from previous measurements of wall loss for products of a-pinene photooxidation by Zhang et al. in the Caltech chamber. Do the authors have any comments on why?*"

Author response: A wide range of vapor wall loss rates have been reported in different studies, and the reason for this wide range has not yet been determined. We address this briefly as follows:

Page 19, line 539: "The best-fit $\alpha_w = 10^{-6}$ corresponds to a first-order vapor-wall deposition rate constant ($k_{wall,on}$) of 10^{-4} s^{-1}. A wide range of vapor wall loss rates has been reported (Figure 3 of Krechmer et al. (2016)). This $k_{wall,on}$ value is comparable to that reported by Matsunaga and Ziemann (2010) for a 8.2 m^3 chamber but significantly faster than wall loss rates that have been measured in the Caltech chamber (Zhang et al., 2015). The reason for this wide range of reported vapor wall loss rates is currently uncertain and outside the scope of this study."

4. Referee comment: "*For reactions conducted under low NOx conditions the oxidation rate will affect the chemistry of RO2 radicals. In the O3 reaction, where it has been previously observed that products of both RO2 + RO2 (such as pinic acid) and RO2 + HO2 (organic peroxides) are formed, it is to be expected that at higher oxidation rates the system will shift more towards RO2 + RO2 reactions. This can have a significant effect on SOA yields. This is also true for the referenced studies on monoterpene + NO3 reactions. The authors should discuss this effect and how it might alter the interpretation of their results.*"

Author response: We acknowledge that it is possible that the RO_2 radical chemistry may be different at low (100 ppb O_3) and high (500 ppb O_3) oxidation rates, which in turn, may affect the SOA mass yields. We also agree with the referee that differing RO_2 radical chemistry may also play a role in the different SOA mass yields measured in the reference NO_3+monoterpene studies. Therefore, we have made the following changes in the revised manuscript:

Page 24 line 677: "It should be noted that while we showed that the observed oxidation rate effect (i.e., higher SOA mass yields as a result of faster hydrocarbon oxidation rates) is a consequence of vapor-wall deposition, the possibility that differing peroxy radical (RO_2) chemistry in the 100 and 500 ppb O_3 experiments may play some role in influencing the SOA mass yields cannot be discounted. RO_2 radicals, which are formed from the decomposition of excited Criegee intermediates (Docherty et al., 2005), may be produced at faster rates in the 500 ppb O_3 experiments. This may lead to the higher production of condensable oxidation products from the RO_2+RO_2 reaction pathway in the 500 ppb O_3 experiments

(relative to those formed in the 100 ppb O_3 experiments), which may result in higher SOA mass yields."

Page 29 line 825: "In addition to differences in the experimental conditions of the two studies (which may lead to differing RO_2 chemistry), Boyd et al. (2015) hypothesized that the higher SOA mass yields could also be a result of the higher NO_3 concentrations used in their study (which led to faster β-pinene oxidation rates) compared to those used by Fry et al. (2009, 2014)."

5. Referee comment: "*Similar to the comments made in #4, when comparing studies the authors should consider the fact that oxidation rate is unlikely to affect RO2 chemistry under high NOx conditions, such as in aromatic VOC experiments.*"

Author response: We thank the referee for the comment. We have made the following changes to the revised manuscript:

Page 29 line 829: "The oxidation rate effect was also observed in the *m*-xylene photooxidation system, where Ng et al. (2007) showed that the SOA mass yields were dependent on the *m*-xylene oxidation rate, with higher OH concentrations (and hence faster oxidation rates) resulting in higher SOA mass yields. The authors dismissed the possibility of the different SOA mass yields being a result of different RO_2 chemistry since all their *m*-xylene photooxidation experiments were performed under high-NO_x conditions and the RO_2 reacted virtually entirely with NO."

6. Referee comment: "*Line 592: Should add "of" after "regardless".*"

Author response: This is corrected in the revised manuscript.

The following are additional minor changes the authors have made to the manuscript:

1. This sentence was added into the revised manuscript:

Page 3 line 68: "Determination of particle wall loss coefficients may be complicated if coagulation is significant. Particle dynamics models can be used to correct particle wall loss coefficients for coagulation."

2. We corrected the grammatical errors in the sentence:

Page 5 line 128: "Before each experiment, the chamber was flushed with dried, purified air for at least 36 h until the aerosol number concentration was < 30 cm^{-3}. All experiments were conducted under dry conditions (< 5 % RH) at room temperature (25 °C). All experiments were conducted under dry conditions (< 5 % RH) at room temperature (25 °C). NO_x mixing ratios in these experiments were < 1 ppb. Experimental conditions are summarized in Table 1."

3. This sentence was added into the revised manuscript:

Page 7 line 212: "As we describe subsequently, β_i may be measured directly during seed-only experiments or may be corrected for the influence of coagulation using a particle dynamics model."

4. This sentence was added into the revised manuscript:

Page 9 line 248: "Modeled O_3 and α-pinene concentrations are compared with observed concentrations in Fig. S5. The good fit of modeled and observed O_3 and α-pinene concentrations indicates that our representation of O_3 is appropriate."

5. This sentence was added into the revised manuscript:

Page 9 line 266: "Coagulation is not considered in the present model; we address the potential impact of coagulation later in the paper."

6.We added the word "measured" to the sentence:

Page 17 line 479: "It is important to note that these conclusions are robust even when the average of the measured low AS-seed only and high AS-seed only particle wall loss coefficients are used to account for particle wall loss in all the experiments (Fig. S6)."

7. Figures S6, S11, S12 and S13 in the original manuscript are changed to Figures S5, S10, S11 and S12, respectively, in the revised manuscript. Table S2 in the original manuscript is changed to Table S3 in the revised manuscript. Equations 13 and 14 in original manuscript are changed to equations 15 and 16, respectively in the revised manuscript.

8. We changed "ΔM_0" to "ΔM_o" in Table 1 headers.

9. We added a period after Shilling et al in Figure 5 caption:

"Figure 5: Comparison of SOA mass yields obtained in this study to those of previous dark α-pinene ozonolysis studies (Table S2). The SOA mass yields and concentrations of majority of these previous studies (Hoffmann et al., 1997; Griffin et al., 1999; Cocker et al., 2001b; Gao et al., 2004; Presto et al., 2005; Presto et al. 2006; Pathak et al., 2007b; Song et al., 2007) were previously compiled by Shilling et al. (2008). Similar to Shilling et al. (2008), all the data shown here (including those reported in this study) have been adjusted using an organic density of 1.0 g cm^{-3}, and to 298 K using a temperature correction of 1.6 % per K, as recommended by Pathak et al. (2007b) to facilitate easier comparison among the different studies."

10. We corrected Figure S4 caption:

"Figure S4: Raw and particle wall loss (PWL) corrected number and volume concentration data for the 500 ppb O_3 experiments. All the raw data are particle wall loss corrected using the average particle wall loss rates (i.e. average of the particle wall loss rates obtained from low AS-seed only and high-AS seed only experiments)."

11. We revised Figure S6 caption:

Figure S6: 10 min-averaged SOA mass yields over the course of an α-pinene ozonolysis experiment as a function of initial total AS seed surface area concentration for the (a) 100 ppb O_3 experiments, and (b) 500 ppb O_3 experiments. Here, all the data have been particle wall loss corrected using the average particle wall loss rates (i.e. average of the particle wall loss rates measured from low AS-seed only and high-AS seed only experiments). Symbol color indicates the SOA mass concentration and symbol size indicates the time after O_3 is injected into the chamber. The × symbols are the SOA mass yields at peak SOA growth. The y-axis error bars represent the uncertainty in the peak SOA mass yield, which originates from the α-pinene injection and the aerosol volume concentration measured by the SMPS at peak SOA growth (one standard deviation). As discussed in the main text, the use of average measured particle wall loss rates for particle wall loss correction does not change the conclusions of this work: 1) SOA mass yields are enhanced at higher O_3 concentrations, and 2) there is a lack of a SOA mass yield dependence on the seed surface area within the range of AS seed surface area concentration used in this study.

12. We corrected the grammatical errors in Figure S12 caption:

"Figure S12: Results from the coupled vapor-particle dynamics model showing how SOA mass concentration (ΔM_o) changes as a function of reacted α-pinene at different O_3 concentrations. In these model simulation runs, the initial α-pinene concentration is fixed at 48 ppb, while the O_3 concentration is varied from 75 to 1000 ppb. Here, the O_3 injection rate is 5 times faster than the base rate used in the model. The base rate is 500/54.25 ppb min^{-1}, the same rate used to analyze results from the 500 ppb O_3 experiments. As discussed in the main text, the oxidation rate effect persists at a higher O_3 concentration when a faster O_3 injection rate is used. It is important to note that SOA evaporation is predicted at high O_3 concentrations in the coupled vapor-particle dynamics model, but not observed in chamber experiments."

13. We corrected Table S3 caption:

"Table S3: Comparison of experimental conditions used in this work with those of previous dark α-pinene ozonolysis studies. The SOA mass yields and concentrations of these studies are shown Fig. 5."

14. In the original manuscript, the citation for Zaveri et al. (2014) was left out in the Reference section. This citation is added to the revised manuscript:

Zaveri, R. A., Easter, R. C., Shilling, J. E., and Seinfeld, J. H.: Modeling kinetic partitioning of secondary organic aerosol and size distribution dynamics: representing effects of volatility, phase state, and particle-phase reaction, Atmospheric Chemistry and Physics, 14, 5153-5181, 10.5194/acp-14-5153-2014, 2014.

15. The following references have also been added to the revised manuscript:

Docherty, K. S., Wu, W., Lim, Y. B., and Ziemann, P. J.: Contributions of organic peroxides to secondary aerosol formed from reactions of monoterpenes with O-3, Environmental Science & Technology, 39, 4049-4059, 10.1021/es050228s, 2005.

Krechmer, J. E., Pagonis, D., Ziemann, P. J., and Jimenez, J. L.: Quantification of Gas-Wall Partitioning in Teflon Environmental Chambers Using Rapid Bursts of Low-Volatility Oxidized Species Generated in Situ, Environmental Science & Technology, 50, 5757-5765, 10.1021/acs.est.6b00606, 2016.

Saha, P. K., and Grieshop, A. P.: Exploring Divergent Volatility Properties from Yield and Thermodenuder Measurements of Secondary Organic Aerosol from α-Pinene Ozonolysis, Environmental Science & Technology, 50, 5740-5749, 10.1021/acs.est.6b00303, 2016.

Ye, P., Ding, X., Hakala, J., Hofbauer, V., Robinson, E. S., and Donahue, N. M.: Vapor wall loss of semi-volatile organic compounds in a Teflon chamber, Aerosol Sci. Technol., 50, 822-834, 10.1080/02786826.2016.1195905, 2016.