

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: *“Line 278–282: What does the theory for charging effects assume about charge on the chamber walls? This can vary due to contact when conducting experiments and so may not be reproducible.”*

**Author response:** In the manuscript, we assume that the chamber walls are not charged. This assumption is reasonable since the FEP Teflon chamber used is housed in an enclosure (i.e., GTEC facility) to minimize contact with the chamber walls. The chamber walls were never exposed to contact in this series of experiments. Furthermore, the uncorrected particle wall-deposition coefficients for the low-SA-seed-only (experiments 1 through 4) agree with previously measured particle wall-deposition coefficients. This suggests that charging effects (if any) are reproducible as long as the chamber walls are not exposed to contact.

2. Referee comment: *“Lines 514–518: How is it known that SOA growth should stop after the  $\alpha$ -pinene reacted? Why can't there be continued growth due to condensation driven by oligomer formation?”*

**Author response:** Previous studies have shown that even though  $\alpha$ -pinene ozonolysis forms oligomers from particle phase reactions, SOA growth still essentially stops once all the  $\alpha$ -pinene has reacted (Gao et al., 2004a and 2004b; Ng et al., 2006). These references have been added to the revised manuscript:

**Page 13 line 378: “SOA growth basically ceases once all the  $\alpha$ -pinene has reacted, indicating that the first step of  $\alpha$ -pinene ozonolysis is rate-limiting and the first-generation products are condensable (Gao et al., 2004a and 2004b; Ng et al., 2006; Chan et al., 2007).”**

**Page 13 line 386: “In cases where the coagulation-corrected size-dependent particle wall-deposition coefficients are used to correct for particle wall-deposition (Figs. 3a-d), the SOA growth profile is similar to that of the low-SA experiments; SOA growth essentially stops once all the  $\alpha$ -pinene has reacted, as expected (Gao et al., 2004a and 2004b; Ng et al. 2006; Chan et al., 2007).”**

#### **References:**

**Chan, A. W. H., Kroll, J. H., Ng, N. L., and Seinfeld, J. H.: Kinetic modeling of secondary organic aerosol formation: effects of particle- and gas-phase reactions of semivolatile products, Atmospheric Chemistry and Physics, 7, 4135-4147, 2007.**

**Gao, S., Ng, N. L., Keywood, M., Varutbangkul, V., Bahreini, R., Nenes, A., He, J. W., Yoo, K. Y., Beauchamp, J. L., Hodyss, R. P., Flagan, R. C., and Seinfeld, J. H.: Particle phase acidity and oligomer formation in secondary organic aerosol, Environmental Science & Technology, 38, 6582-6589, 10.1021/es049125k, 2004a.**

Gao, S., Keywood, M., Ng, N. L., Surratt, J., Varutbangkul, V., Bahreini, R., Flagan, R. C., and Seinfeld, J. H.: Low-molecular-weight and oligomeric components in secondary organic aerosol from the ozonolysis of cycloalkenes and alpha-pinene, *J. Phys. Chem. A*, **108**, 10147-10164, 10.1021/jp047466e, 2004b.

Ng, N. L., Kroll, J. H., Keywood, M. D., Bahreini, R., Varutbangkul, V., Flagan, R. C., Seinfeld, J. H., Lee, A., and Goldstein, A. H.: Contribution of first- versus second-generation products to secondary organic aerosols formed in the oxidation of biogenic hydrocarbons, *Environmental Science & Technology*, **40**, 2283-2297, 10.1021/es052269u, 2006.

3. Referee comment: *“Lines 514–518: How can all three methods be effective for correcting for wall loss when they give such different results?”*

**Author response:** In our experiments that use low seed aerosol surface area concentrations ( $< 3000 \mu\text{m}^2 \text{cm}^{-3}$ ), SOA mass yields at peak SOA growth obtained from the “size-dependent”, “number averaged” and “volume averaged” methods agree within 14% (Figs. 5 and 6). Hence, our results indicate that these three methods are effective in correcting for particle wall loss in experiments that use low seed aerosol surface area concentrations.

In our experiments that use high seed aerosol surface area concentrations ( $\geq 8000 \mu\text{m}^2 \text{cm}^{-3}$ ), SOA mass yields at peak SOA growth obtained from the “size-dependent”, “number averaged” and “volume averaged” methods differ substantially (Figs. 5 and 6). As explained in the manuscript, these differences arise from assumptions made in the particle wall-deposition correction method regarding the influence of coagulation on the first-order particle wall-loss rate. We find that coagulation needs to be accounted for when the “size-dependent” and “number averaged” particle wall-deposition correction methods are used in chamber studies that utilize high seed aerosol surface area concentrations.

To remove any confusion regarding the effectiveness of the “size-dependent”, “number averaged” and “volume averaged” methods in correcting for particle wall deposition at different seed aerosol surface area concentrations, the following changes have been made to the revised manuscript:

**Page 19 line 567: “In the experiments with low seed aerosol surface area concentrations ( $< 3000 \mu\text{m}^2 \text{cm}^{-3}$ ), the SOA mass yields obtained by the different particle wall-deposition correction methods (i.e., the “size-dependent”, “number averaged” and “volume averaged” methods) are generally consistent with one another. This indicates that these three methods are effective in correcting for particle wall-deposition in experiments that use low seed aerosol surface area concentrations. However, in the experiments with high seed aerosol surface area concentrations ( $\geq 8000 \mu\text{m}^2 \text{cm}^{-3}$ ), the calculated SOA mass yields differ substantially. These differences arise from assumptions made in the particle wall-deposition correction method regarding the influence of coagulation on the first-order particle wall-loss rate. Specifically, we find that coagulation needs to be accounted for in the “size-dependent” and “number averaged” methods in order for them to be effective in chamber studies that use high seed aerosol surface area concentrations. Coagulation does not need to be accounted for in the “volume averaged” method since coagulation does not affect aerosol volume concentrations.”**

4. Referee comment: *“Lines 584–593: Similar to Comment 3. The recommendations are a bit*

*unclear, considering that the methods can give significantly different corrections.”*

**Author response:** Please refer to our response to comment 3. In addition, the original manuscript provides explicit recommendations regarding the use of different particle wall-deposition methods in chamber studies (regardless of VOC system) that use high seed aerosol surface area concentrations:

**Page 20 line 591: “Here we showed that the condensation of SOA-forming vapors in the  $\alpha$ -pinene ozonolysis system can be erroneously concluded as kinetically limited if coagulation is not accounted for in the “size-dependent” and “number averaged” particle wall-deposition correction methods. Similar flawed conclusions in other VOC systems may be drawn in chamber studies that use high seed aerosol surface area concentrations to promote SOA formation but do not account for coagulation. Therefore, we recommend accounting for coagulation when the “size-dependent” and “number averaged” particle wall-deposition correction methods are used in chamber studies that use high seed aerosol surface area concentrations (e.g.,  $\geq 3000 \mu\text{m}^2 \text{cm}^{-3}$ ) to promote the condensation of SOA-forming vapors onto seed aerosol regardless of VOC system. Alternatively, the “volume averaged” and “inert tracer” methods can be used in chamber studies that use high seed aerosol surface area concentrations. In addition, we suggest using multiple techniques (i.e., at least 2) to correct for particle wall-loss in order to determine the effect of SOA mass yield uncertainties introduced by particle wall-deposition correction.”**

5. Referee comment: *“Conclusions. It would be worth noting that it is also possible to use a slightly different inert tracer method that does not assume that the particles on the wall remain in equilibrium with suspended particles. One can use the decay of the AMS seed signal after peak SOA to estimate a wall-loss rate coefficient and then apply this to the rest of the experiment. It is similar then to the volume average method, except that the measured wall-loss rate coefficient is not affected by changes in particle volume due to evaporation after peak SOA is reached. Measuring time-profiles of different AMS masses relative to the seed also gives information on whether the particle composition is changing due to condensation or evaporation.”*

**Author response:** One of the main objectives of this manuscript to evaluate the effectiveness of commonly used particle wall-deposition correction methods. While the referee is correct that the decay of the seed aerosol signal measured by an aerosol mass spectrometer can be used to estimate a first-order particle wall-deposition rate, this particle wall-deposition correction method is not commonly used. Hence, we did not include this suggestion in the revised manuscript.

6. Referee comment: *“In this analysis it is assumed that the results are unaffected by vapor loss to the walls. Are there situations where particle and vapor wall loss are closely coupled, and so cannot be interpreted as separate processes as was done here?”*

**Author response:** We did not assume that our results are unaffected by vapor wall deposition in our analysis. Instead, we showed explicitly in a previous paper that SOA formation in the  $\alpha$ -pinene ozonolysis system is governed by quasi-equilibrium growth (i.e., the timescale for gas-particle equilibrium is less than those for reaction and vapor wall-loss), which results in SOA growth being independent of the seed aerosol surface area concentration (Nah et al., 2016). To remove any confusion, the following changes have been made to the revised manuscript:

**Page 5 line 142: “This work builds on our previous study on the influence of seed aerosol surface area concentration and hydrocarbon oxidation rate on vapor wall deposition and SOA mass yields in the  $\alpha$ -pinene ozonolysis system (Nah et al., 2016). In our previous study, we used a coupled vapor-particle dynamics model to show that the condensation of SOA-forming vapors onto seed aerosol in the  $\alpha$ -pinene ozonolysis system is dominated by quasi-equilibrium growth. This present work is aimed at understanding the uncertainties in the SOA mass yields due to the application of different particle wall-deposition correction methods.”**

**Reference:**

**Nah, T., McVay, R. C., Zhang, X., Boyd, C. M., Seinfeld, J. H., and Ng, N. L.: Influence of seed aerosol surface area and oxidation rate on vapor wall deposition and SOA mass yields: a case study with  $\alpha$ -pinene ozonolysis, *Atmos. Chem. Phys.*, **16**, 9361-9379, 10.5194/acp-16-9361-2016, 2016.**

It is possible that particle and vapor wall deposition processes may be closely coupled in VOC systems where the condensation of SOA-forming vapors onto seed aerosol is kinetically limited (i.e., the timescale for gas-particle equilibrium is competitive with or greater than those for reaction and vapor wall-loss). In these systems, large concentrations of seed aerosol can be added to promote gas-particle partitioning, and consequently increase SOA mass yields. Hence, it is possible that faster particle wall deposition rates may result in a larger effect of vapor wall deposition on SOA formation (and vice versa) in systems where SOA formation is governed by kinetically limited growth.

7. Referee comment: *“It would also be useful to provide some comments on at least one approach that could be used to avoid complications from wall loss of particles and vapor, which is to conduct short experiments.”*

**Author response:** We thank the referee for this suggestion. This is included in the revised manuscript:

**Page 20 line 605: “Complications arising from particle and vapor wall deposition may also be potentially minimized by conducting shorter chamber experiments. This can be achieved by using excess oxidant concentrations, which will increase the oxidation rate, and consequently reduce the time at which peak SOA growth is achieved (Nah et al., 2016).”**

8. Referee comment: *“Technical Comments. Throughout the paper the authors jump between the term “volume averaged” and “volume dependent” method. I suggest sticking with one or the other.”*

**Author response:** In the revised manuscript, we use the term “volume averaged”.

**Response to Referee 2 (Referees’ comments are italicized)**

We thank the referee for the positive comments.