

We thank reviewer 2 for their thoughtful and helpful review. Our responses are below.

The authors found that accounting for vapor wall losses leads to 2-3 times increases in the total SOA production in chamber experiments. This conclusion, however, depends on how the adjustable parameters in the model are tuned against chamber observations, and as a result the enhancement in SOA production in the absence of vapor wall loss could vary with different model parameterizations. The authors mentioned that the OA concentrations from FLAME-III experiments are used to constrain the model performance, yet the comparison of simulations with experimental observations are not given in details throughout of the paper. Ideally, the organic aerosol temporal profile by AMS/SMPS during one representative experiment should be given together with corresponding simulations (e.g., Figure 3) to better visualize the model performance.

Response: We do not compare each individual simulation with each corresponding observation because May et al. (2015) was only able to derive a single volatility distribution across the FLAME III 18 experiments. Additionally the IVOC volatility distribution that we use was from FLAME IV experiments that do not directly correspond to the specific FLAME III experiments. Thus, we do expect error in individual experiments due to these assumptions, but we seek to capture the mean behavior across all of the experiments.

We added the following text near Line 408: "...are in very good agreement with those observations. May et al. (2015) was only able to derive a single volatility distribution across the FLAME III 18 experiments and the IVOC volatility distribution from FLAME IV experiments do not directly correspond to the specific FLAME III experiments. Thus, we expect the error in individual experiments due to the single volatility distributions across all simulations. We thus seek to capture the mean behavior across all of the experiments rather than comparing individual simulations to their corresponding experiments. Our simulations also show that..."

Another question related, have the authors conducted optimal fitting of simulations to chamber measured quantities such as organic aerosol mass, O:C and H:C ratios?

We do not simulate O:C and H:C. We do compare organic aerosol mass, at least implicitly; this is what is being evaluated in Figures 4 and 5. The initial organic aerosol masses in the simulations are identical to the measurements, $OAER_{inert}$ effectively evaluates OA mass as the inert-tracer wall losses for these experiments have been evaluated in Bian et al., 2015.

We added the following text around Line 406: "...Since the initial organic aerosol masses in the simulations are identical to the measurements, we use $OAER_{inert}$ to

evaluate simulated OA mass against measurements in Figs. 4 and 5 as the inert-tracer wall losses for these experiments have been evaluated in Bian et al. (2015).”

Is there more than one set of parameters that could well represent the observations? What is the physical meaning of each best-fit parameter that is chosen to describe the BBOA evolution?

Yes, more than one set of parameters could well represent the observations because we do not have enough experimental data to determine which combination of parameters for the study of FLAME III. This is shown in Figures 4 and 5, and we discuss the conclusion that multiple sets of assumptions can describe the measurements from line 415 to 456. We neither evaluate nor declare “best fit parameters” for the study of the influence of wall loss on secondary-organics evolution.

To reinforce this point, we modified the text from line 453, “...can better represent the FLAME-III experiments; however, we are unable to provide the “best-fit parameters” for the simulations as we cannot determine which set of α_w , $C_w/M_w\gamma_w$, and chemistry assumptions best represent the actual processes occurring in the chamber...”

For the ambient plume simulations, the authors are suggested to add discussions on how the values of key parameters, such as fire sizes and atmospheric stability classes, are assigned. Are they representative of the fire plume transportation in the air? A thorough search on the ambient fire plume properties in literatures might be useful to rationalize the sensitivity tests conducted in this study.

Cochrane et al. (2012) reported 14 wildfires with fire size from 5.28 to 1868.78 km². The burning areas for prescribed fires observed in Akagi et al. (2013) ranged from 0.162 to 1.47 km². The fire size for agricultural and pile burns can be as small as 7×10^{-5} km² (Springsteen, et al. 2015). The fire sizes in our sensitive test are 10^{-4} , 0.01, 1, 100 km², covering most of this observed range. Atmospheric stability includes six classes from A (unstable) to F(stable), which represent all the possible atmospheric stability conditions that range from clear sunny days (very unstable) to calm clear nights (very stable). We do not try to simulate any specific fire, just a range of possible conditions. We revised the manuscript from lines 477 to 479 accordingly:

“...The initial plume width is associated with fire size, which means that the fire size could largely influence the plume evolution (Sakamoto et al., 2016). Cochrane et al. (2012) reported 14 wildfires with fire size from 5 to over 1000 km². Akagi et al. (2013) also recorded the burn areas for the observed prescribed fire range from 0.162 to 1.47 km². The burning area for Williams fire was 0.81 km² (Akagi et al., 2012). The fire size for agricultural and pile burns can be as small as 7×10^{-5} km² (Springsteen, et al. 2015). We therefore perform simulations on the evolution of ambient OA concentrations over 4

hours of simulated transport, for four different fire areas of 1×10^{-4} , 1×10^{-2} , 1×10^0 and 1×10^2 km² (with the fire width assumed to be the square root of these areas), which largely cover the reported burned areas above...”

Recent two-dimensional VBS frameworks have incorporated gas-phase fragmentation processes as a function of the O:C ratio of individual volatility bins (e.g., Jimenez et al. 2009). The original distribution of volatility bins upon one generation of oxidation (drops in volatility per reactions) would correspondingly change by adding this branch of mechanism into the model framework. Upon OH-exposure in the order of $\sim 10^{10}$ molecules cm⁻³ s (typically several hours of reactions in the atmosphere), fragmentation should have occurred to some extent, depending on the OH reactivity of the parent precursors. The authors are suggested to discuss uncertainties caused by the assumption of zero fragmentation in the reaction mechanisms.

We agree that lack of simulating fragmentation is a limitation of our study. We have added text to emphasize this after line 341: “...and the alkane k_{OH} set with the two-volatility-bin drop per reaction as a lower bound for SOA formation. Jimenez et al. (2009) showed that fragmentation would produce more-volatile species compared with parent species. The assumption of zero fragmentation and unity SOA mass yield may cause overestimation of SOA production in our study.”

We also changed the text from Line 581: “...Uncertainties in parameters that control vapor wall losses, such as the wall saturation concentration and wall accommodation coefficient, as well as uncertainties in gas-phase chemistry with the assumption of zero fragmentation and unity SOA mass yield, lead to uncertainties in our simulations.”

Minor:

Page 5, Line 168: How are the black carbon and organic contents treated in each particle size bin in the model? Are they well mixed?

We assume that all species are internally mixed within each size section, meaning the black and OA exists in the all particles at the same ratio within each size bin. However, for purposes for calculating OA partitioning, we assume that OA and black carbon exist in separate phases within each particle, and thus this presence of black carbon does not enhance partitioning of OA to the particle phase. We have added text after Line 168: “... and water with 36 logarithmically spaced size sections from 3 nm to 10 μ m. We assume that all species are internally mixed within each size section, meaning that the ratio of BC and OA are the same for all particles within each size bin. When calculating OA partitioning, we assume that OA and BC exist in separate phases, and thus the presence of BC does not influence OA partitioning to the particle phase in the model. In our previous study examining the influence of wall loss...”

Page 9, Line 328: Add 'are' before 'shown'.

Corrected.

Page 29, Figure 1: How are the vapor concentrations calculated, based on equilibrium partitioning?

We assume that you are asking how the initial vapor concentrations are calculated. We set the organic aerosol concentration equal to that measured by the AMS, assume the total-organic volatility distributions from May et al. (2015) and Hatch et al. (2016), and estimate the vapor concentration necessary to sustain the AMS-measured OA mass based on aerosol partitioning theory (Pankow, 1994) on the assumption of gas and particle equilibrium partitioning.

We revised lines 175 to 179 : “In this current study, we expand the simulated organics from eight to fifteen “species” including more volatile organics between 10^6 to 10^{11} $\mu\text{g m}^{-3}$, based on the FLAME-4 study of Hatch et al. (2016), to account for chemical transformations from both volatile and semivolatile organic species and estimate the initial organic vapor concentration based on aerosol partitioning theory (Pankow, 1994) on the assumption of gas and particle equilibrium partitioning (Fig. 1a). The evolution of the organic vapors is calculated based on partitioning theory (to get equilibrium vapor pressures above the particle), wall-equilibrium vapor pressures, and kinetic mass transfer to/from the particles and the walls. “

References:

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