We thank the reviewer for the constructive review comments to improve our manuscript. We provide a point-to-point response to reviewer's comments. The manuscript with the tracked changes is submitted together with the response documents.

Reviewer #1

General Comments:
This paper describes a modeling effort aimed at investigating the effects of wall losses of organic particles and gases on the mass and number concentrations of particles measured in a series of experiments in which aerosol was formed by burning various types of biomass and then added to a Teflon film chamber. A state-of-the-art model was used for this purpose and wall loss parameters were calculated from theory or taken from the literature. Although it is standard practice to correct chamber studies for losses of particles, only recently has it been shown that loss of gases can also be important. A few modeling studies of secondary organic aerosol (SOA) formation have investigated the effects of wall loss of gases on SOA yields, and shown them to be important, but to my knowledge this is the first to look at the effects on chamber studies of primary emissions. The results are significant, in that they indicate that both particle and gas losses were important, and roughly equal, thus indicating that these effects should be included in future chamber modeling studies. The study appears to be well done, and the manuscript is clearly and concisely written. I think the paper should be published in ACP, although I have a few comments for authors to consider.

Specific Comments:
1. Page 15250, line 24-28: The volatility distribution was determined on aerosol that had already been added to the chamber. How would losses of vapors and particles affect the validity of this distribution? Are these effects included in the upper and lower bounds to the uncertainties used in Section 3.2.1?

May et al. (2013b) derived the volatility distribution for biomass burning in the smoke chamber using an evaporation model with the thermodenuder data as an input. They characterized the particle number losses in the thermodenuder by measuring the generated ammonium sulfate size distributions using an upstream and downstream SMPS and then apply those losses to correct the measured particulate organic data. However, as stated by the reviewer above, the loss of semi-volatile organics to the walls may lower the average volatility in the particles in the chamber with time. The volatility distribution of biomass burning from May et al. (2013b) may be weighted towards lower volatilities than what entered the chamber. This means the initial volatility distribution could contain more material in the higher volatility bins than what we assume and vapor wall losses could be more important than we calculate.

We have added the following text after line 28, page 15250 to our paper to address this uncertainty, "…across the 18 experiments. In the fitting procedure, May et al. (2013b)

characterized the particle number loss in the thermodenuder by measuring the generated ammonium sulfate size distributions using an upstream and downstream SMPS and then apply those loss to correct the measured particulate organic data. However, the evaporation of semi-volatile organics due to vapor-phase wall losses may have shifted the particle volatility to lower values with time in the chamber. Thus, the volatility distribution of biomass burning adopted from May et al. (2013b) may be weighted towards lower volatilities than those that entered the chamber, which means that vapor wall losses could be more important than calculated in this study. We test the model sensitivity to upper and lower bounds of their derived POA volatility distribution (representing the edges of the shaded region in Figure 6a from May et al. (2013b) and shown here in Figure 1)."

2. Page 15252, lines 18-19: The results of Matsunaga and Ziemann 2010 were consistent with wall loss rates that were independent of the accommodation coefficient, which meant that the value was at least 10ˆ-5, but could have been much larger. So it was not necessarily 10ˆ-5, the value used here. How would the results here be affected if the value was much larger than 10ˆ-5? Also, weren't the values calculated using the approach of Zhang et al. (2015) less than 10ˆ-5?

We have added the three sensitivity tests on the larger accommodation coefficients of vapor with wall using the values of 1×10^{-4}, 1×10^{-2}, and 1. Generally, increasing accommodation coefficients increases vapor wall loss and thus increases the organic aerosol loss in the chamber. But when α_w increases above 10^{-4}, accommodation to the wall no longer limits the rate of vapor wall loss. We find little increase of OA loss applying α_w larger than 1×10^{-4} as shown in the following Figures R1 and R3. The total OA loss increases from 41% to 47% of initial particle-phase organic mass if α_w increase from 10^{-5} to 10^{-4}, causing a lower bias of in the final OA (-14% and -9.5% for high- and low-OF simulations, Table 5). The proportion of OA wall loss via depositional removal and by the transferring from the particle to the vapor phase changes from 65%:35% to 52%:48%, as the increase vapor wall loss rates, suggesting that α_w is an important factor in our calculations though its effect saturates for α_w larger than 10^{-4}.
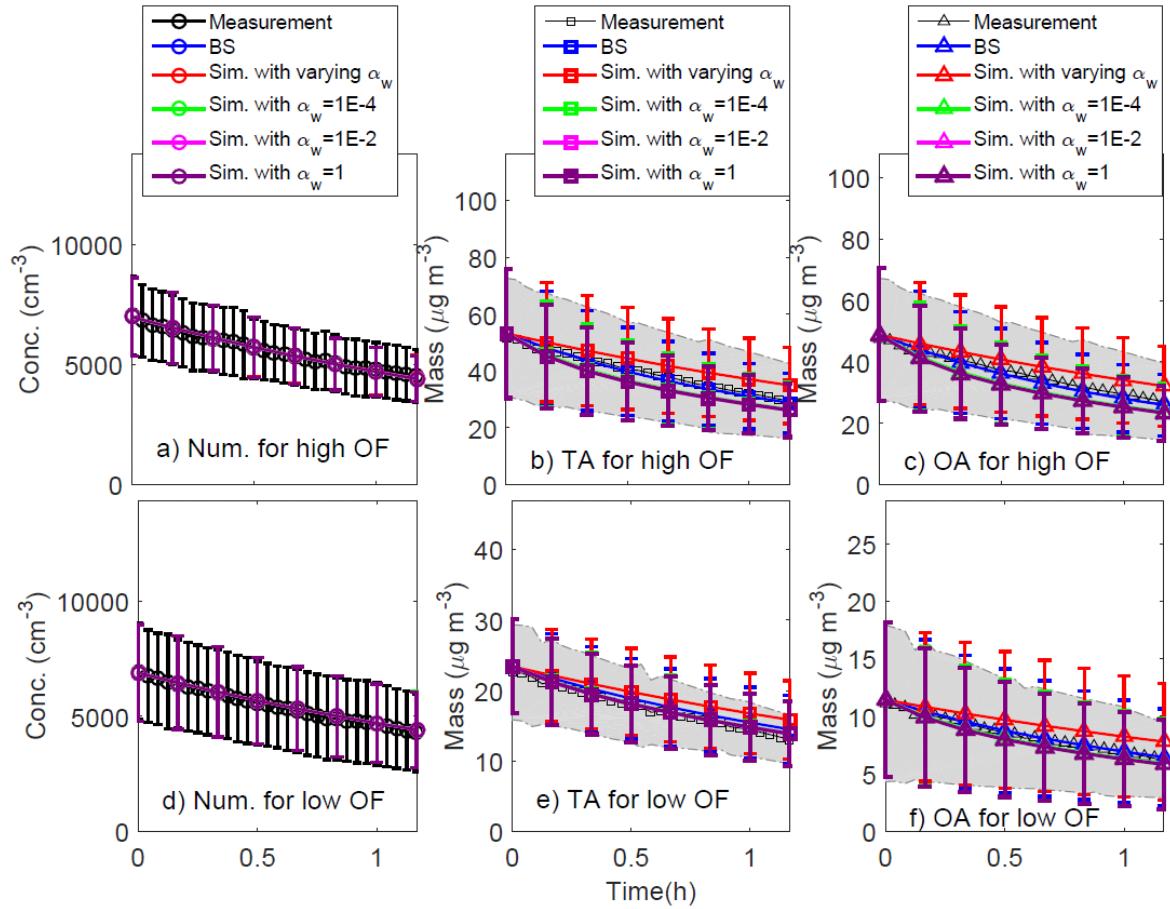
Figure R1 Similar to Figure 2 except for the simulations with different accommodation coefficients of vapor with walls, α_w, computed as a function of C_i^* (Zhang et al., 2015) and taking the values of 1×10^{-4}, 1×10^{-2} and 1.
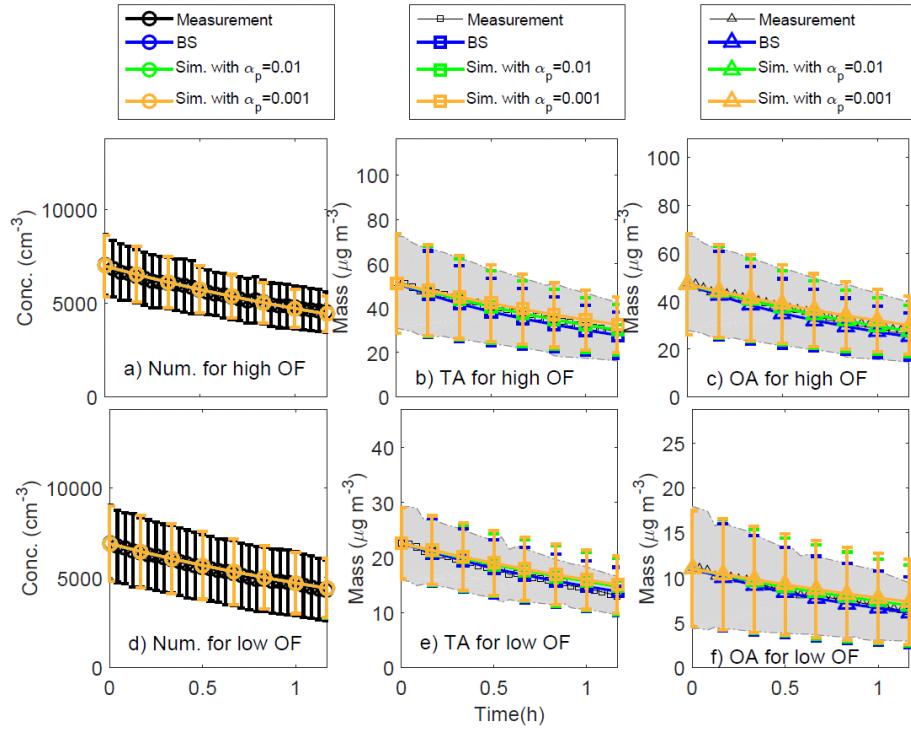
Figure R2 Similar to Figure 2 except for the simulations with different accommodation coefficients of vapor with particles (α_p) of 0.01 and 0.001.
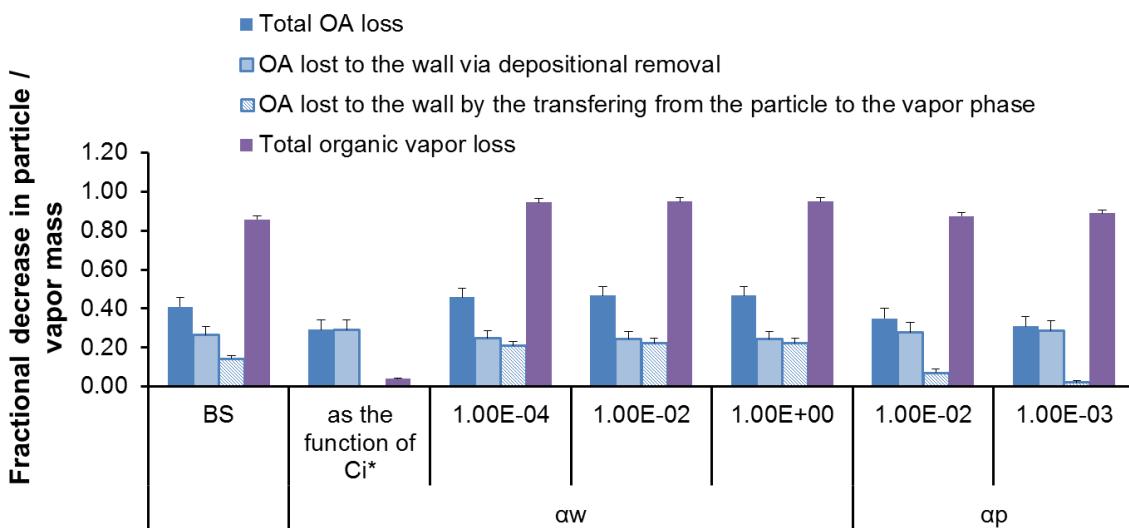
Figure R3 The organic species mass budgets for the base-assumptions simulations (accommodation coefficient of vapor with wall, α_w of 1×10^{-5} and accommodation coefficient of vapor with particle, α_p of 1) and the sensitivity studies on α_w (as the

function of $C_i{}^*$ and of the values 1×10^{-4}, 1×10^{-2}, and 1) and α_p (of 0.01 and 0.001). All other variables as in Figure 3b.

Table R1 Molecular weight (g mol^{-1}) associated with each vapor pressure bin, computed for a temperature of 298K followed the calculation equation in Table 2, varying $C_w/M_w\gamma_w$ as a function of vapor pressure bins, wall loss rates (s^{-1}, $k_{w, on}$ and $k_{w, off}$ for 120, 9 µmole m^{-3} and varying $C_w/M_w\gamma_w$) and accommodation coefficients between gas and wall (α_w) for each volatility bin in the base-assumptions simulation and the sensitivity tests.

C_i^* (µg m^{-3})	10^{-3}	10^{-2}	10^{-1}	10^{0}	10^{1}	10^{2}	10^{3}	10^{4}
Molecular weight (g mol^{-1})	569	524	479	434	389	344	299	254
varying $C_w/M_w\gamma_w$ ^{1}	1.30×10^{-6}	9.16×10^{-5}	6.44×10^{-4}	4.53×10^{-3}	3.18×10^{-2}	2.24×10^{-1}	1.57	11.1
$\alpha_{w, base}$	1.0×10^{-5}	1.0×10^{-5}	1.0×10^{-5}	1.0×10^{-5}	1.0×10^{-5}	1.0×10^{-5}	1.0×10^{-5}	1.0×10^{-5}
$\alpha_{w, sens}$ ^{4}	1.80×10^{-6}	1.16×10^{-6}	7.45×10^{-7}	4.79×10^{-7}	3.08×10^{-7}	1.98×10^{-7}	1.27×10^{-7}	8.17×10^{-8}
$k_{w,on}$(base, s^{-1})	7.33×10^{-4}	7.58×10^{-4}	7.86×10^{-4}	8.18×10^{-4}	8.54×10^{-4}	8.97×10^{-4}	9.47×10^{-4}	1.01×10^{-3}
$k_{w,off}$ (s^{-1},120 µmol m^{-3})^2	1.07×10^{-11}	1.21×10^{-10}	1.37×10^{-9}	1.57×10^{-8}	1.83×10^{-7}	2.17×10^{-6}	2.64×10^{-5}	3.31×10^{-4}
$k_{w,off}$(s^{-1},9 µmol m^{-3})^3	7.01×10^{-11}	8.09×10^{-10}	9.44×10^{-9}	1.11×10^{-7}	1.33×10^{-6}	1.62×10^{-5}	2.02×10^{-4}	2.59×10^{-3}
$k_{w,off}$(s^{-1}, varying $C_w/M_w\gamma_w$)^1	3.95×10^{-4}	6.31×10^{-4}	1.02×10^{-3}	1.67×10^{-3}	2.76×10^{-3}	4.66×10^{-3}	8.04×10^{-3}	1.43×10^{-2}
$k_{w,on}$ (s^{-1},$\alpha_{w,sens}$)^4	1.55×10^{-4}	1.05×10^{-4}	7.15×10^{-5}	4.86×10^{-5}	3.31×10^{-5}	2.27×10^{-5}	1.56×10^{-5}	1.09×10^{-5}
$k_{w,off}$(s^{-1},$\alpha_{w,sens}$)^4	2.27×10^{-12}	1.68×10^{-12}	1.24×10^{-11}	9.32×10^{-10}	7.09×10^{-9}	5.50×10^{-8}	4.36×10^{-7}	3.59×10^{-6}
$k_{w,on}$ (s^{-1},$\alpha_{w,}$ of 1× 10^{-4})	2.77×10^{-3}	2.81×10^{-3}	2.85×10^{-3}	2.89×10^{-3}	2.94×10^{-3}	2.99×10^{-3}	3.05×10^{-3}	3.12×10^{-3}
$k_{w,off}$(s^{-1},α_{w} of 1× 10^{-4},)	4.06×10^{-11}	4.47×10^{-10}	4.96×10^{-9}	5.55×10^{-8}	6.30×10^{-7}	7.25×10^{-6}	8.51×10^{-5}	1.02×10^{-3}
$k_{w,on}$ (s^{-1},$\alpha_{w,}$ of 1× 10^{-2})	3.99×10^{-3}	4.00×10^{-3}	4.01×10^{-3}	4.01×10^{-3}	4.02×10^{-3}	4.03×10^{-3}	4.04×10^{-3}	4.06×10^{-3}
$k_{w,off}$(s^{-1},α_{w} of 1× 10^{-2},)	5.85×10^{-11}	6.36×10^{-10}	6.97×10^{-9}	7.70×10^{-8}	8.61×10^{-7}	9.76×10^{-6}	1.13×10^{-4}	1.33×10^{-3}

$k_{w,on}$ $(s^{-1}, \alpha_{w,}$ of 1)	4.01×10^{-3}	4.02×10^{-3}	4.02×10^{-3}	4.03×10^{-3}	4.03×10^{-3}	4.04×10^{-3}	4.06×10^{-3}	4.07×10^{-3}
$k_{w,off}$ $(s^{-1}, \alpha_w$ of 1)	5.88×10^{-11}	6.39×10^{-10}	7.00×10^{-9}	7.73×10^{-8}	8.64×10^{-7}	9.80×10^{-6}	1.13×10^{-4}	1.34×10^{-3}

Table R2 The percent bias between the mean and the standard deviations of these simulations (including base-assumptions simulation and other sensitivity tests) and measurements after 1 hr of evolution

	High-OF experiments			Low-OF experiments		
	Number Conc.	Total Aerosol Conc.	Organic Aerosol Conc.	Number Conc.	Total Aerosol Conc.	Organic Aerosol Conc.
Base-assumptions simulation	-1.4%±11%	-3.1%±22%	-4.8%±21%	0.94%±4.9%	6.6%±31%	-0.12%±15%
Simulation with lower volatility distribution	-1.4%±11%	3.3%±13%	2.2%±11%	0.90%±4.8%	9.1%±38%	5.1%±23%
Simulation with higher volatility distribution	-1.6%±12%	-13%±28%	-15%±27%	0.94%±4.8%	0.91%±25%	-12%±6.1%
Simulation using Cw/Mwγw of 50 μmole m^{-3}	-1.4%±11%	-3.0%±22%	-4.6%±21%	0.93%±4.9%	6.6%±31%	-0.06%±15%
Simulation using Cw/Mwγw of 20 μmole m^{-3}	-1.4%±11%	-2.6%±21%	-4.2%±20%	0.94%±4.8%	6.7%±32%	-0.09%±15%
Simulation using Cw/Mwγw of 9 μmole m^{-3}	-1.4%±11%	-2.0%±20%	-3.6%±19%	0.94%±4.8%	6.8%±32%	0.34%±15%
Simulation using varying Cw/Mwγw	-1.4%±11%	9.7%±9.5%	9.2%±7.4%	0.91%±4.8%	12%±38%	10.4%±24.5%
Simulation using varying α_w	-1.3%±11%	12%±4.0%	12%±1.9%	0.96%±4.9%	12%±44%	14%±31%
Simulation using α_w of 10^{-4}	-1.7%±12%	-11%±29%	-13%±28%	0.96%±4.9%	2.7%±25%	-8.3%±7.4%
Simulation using α_w of 10^{-2}	-1.7%±12%	-12%±30%	-14%±29%	0.91%±5.0%	2.0%±24%	-9.7%±6.2%
Simulation using α_w of 1	-1.7%±12%	-12%±30%	-14%±29%	0.90%±5.0%	2.0%±24%	-9.7%±6.2%
Simulation using α_p of 0.01	-1.4%±11%	1.3%±16%	0.44%±15%	0.92%±4.9%	8.0%±38%	6.3%±22%
Simulation using α_p of 0.001	-1.4%±11%	8.5%±8.1%	8.3%±6.3%	0.94%±4.9%	10%±41%	12%±28%
Simulation with instantaneous dilution	-2.1%±12%	-24%±40%	-27%±40%	0.69%±5.2%	-4.6%±6.7%	-23%±15%

We have replaced Figures. 9 and 10 with Figures. R1-3, updated wall loss rates ($k_{w,on}$ and $k_{w,\ off}$) for higher α_w of 1×10^{-4}, 1×10^{-2}, and 1 in Table 3 and modified the text from line 25 , page 15259 in our paper, "…in Matsunaga and Ziemann(2010) and May et al. (2013b). To test the effects of uncertainty in α_w, we perform sensitivity studies where (1) α_w is calculated as a function of C^*_i with a range from 10^{-8} to 10^{-6} as proposed by Zhang et al. (2015) and shown in Table 3, and (2) α_w is set to a constant value of 10^{-4}, 10^{-2}, and 1, respectively, and α_p is set to unity. Figure 11 shows that applying the lower α_w as a function of C^*_i, from Zhang et al. (2015) causes only a 3.9% decrease in vapor-organics concentrations, and the vapor evaporation from particles is almost entirely suppressed because $k_{w,on}$ for C^*_i dependent α_w is several orders of magnitude lower than in the base-assumptions simulation (α_w of 10^{-5}, Table 3) and thus leads to negligible vapor wall loss on the 1 hr experimental timescale, compared with more substantial particle wall losses. These results are very similar to our simulations with no vapor loss, and thus they lead to a high bias in final OA (12 and 14% for high- and low-OF simulations, Table 5) relative to the measurements, showing that these lower α_w values may be unrealistic. For the simulations using higher α_w (10^{-4}, 10^{-2}, and 1), we find when α_w increases above 1×10^{-4}, accommodation of vapors to the wall no longer limits the vapor wall-loss rate and exerts limited influence on OA loss, as shown in Figure 11. The total OA loss increases from 41% to 47% of initial particle-phase organic mass if α_w increases from 10^{-5} to 10^{-4}, causing a lower bias in the final OA (-14% and -9.5% for high- and low-OF simulations, Table 5). The proportion of OA wall loss via particle wall loss versus particle evaporation changes from 65%: 35% to 52%:48%, due to increased vapor wall losses. In summary, α_w is an important factor in our calculations although its effect saturates for α_w larger than 10^{-4}.

We also test non-unity values of α_p (0.01 and 0.001)…"

3. Page 15259, line 5: I suggest rewording this to make it clear that it is the values of the CM/g ratio calculated here using the approach of Zhang et al. (2015) that are much smaller, since that study did report values of the ratio that were much larger than those in Matsunaga and Ziemann (2010).

We agree with this suggestion. C_w estimated from Matsunaga and Ziemann (2010) is 2, 4, 10 and 24 mg m^{-3} for alkanes, alkenes, alcohols and ketones. C_w in Zhang et al. (2015) was highly dependent on the species volatility, ranging from 3.83×10^{-4} to 303 mg m^{-3} for C^* bins from ~10^{-1} to 10^6 µg m^{-3}. We only used the sub-range of reported C^* bins from Zhang et al. (2015) from 10^{-3} to 10^4 in the model with corresponding C_w of 3.26×10^{-6} to 2.77 mg m^{-3}.

We have rephrased the sentence as "These findings resulted because the adapted $C_w/M_w\gamma_w$ values (1.30×10^{-5} – 11.1 µmole m^{-3}, Table 3) for C^* bins from 10^{-3} to 10^4 from Zhang et al. (2015) are generally smaller than in the base case (9-120 µmole m^{-3}), which increases k_{off} and thus lowers the net vapor uptake to the wall."

Technical Comments:

1. Page 15248, line 19: Should be "studied".

Corrected.

2. Page 15254, line 10: Something seems wrong with the phrase "…particles are thus in proportional with on the …".

We have rephrased the sentence as "The condensation/evaporation of vapors to/from particles on the wall are treated as identical to suspended particles of the same size (i.e. we assume that the particles on the wall undergo identical gas-particle partitioning and mass transfer as the suspended particles)."