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 #3

The manuscript "Investigation of particle and vapor wall-loss effects on controlled wood-smoke smog-chamber experiments" by Bian et al. studies how different loss processes and assumptions in describing these processes affect estimates of secondary organic aerosol formation in smog-chamber experiments. They combine experiments with model simulations, which describe evolution of an aerosol population in a Teflon smog-chamber. The manuscript is well written and fits in the scope of Atmospheric Chemistry and Physics.

I recommend the paper for publication provided the authors address the following issues:

- My main concern is the determination of particle wall losses. Parameter ke in Equation (1) is a function of turbulent kinetic energy yet the difference in its values for different fuel types range over more than two orders of magnitude (also kw,p0 varies more than two orders of magnitude). What can explain this? It does not seem intuitive that chemical or physical properties of different compounds can affect this parameter so much.

Is there a possibility that the APE model inadvertedly e.g. includes wall losses of evaporating compounds in particle wall losses? This would have implications in determining the relative contribution of gas-wall losses.

Thanks for this comment, which got us thinking critically about the best way to run the APE model. We realized that we were weighting the small and large ends of the size distribution, which were susceptible to poor counting statistics, too heavily in our optimization. We re-ran APE model by reducing the number of fitted moments from the set of diameter moments (-2, -1.5, -1, -0.5, 0, 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4) to (0, 1.5, 3). Note that the 0 moment is total number, 1 is proportional to total diameter, 2 is proportional to total surface area and 3 is proportional to total volume (other moments do not have physical proportionalities). As described in Pierce et al. (2009), the APE model fits the predicted size distributions to the observed size distributions by minimizing the difference between the simulated and observed diameter moments. We therefore narrowed down the fitted moment range, and the new results of k_w and k_e are more consistent between burns as shown in the table and figure below. The two cases (Burn 65 and Burn 67) bound various burns, and we suspect this is related to the loading of small size particles as shown in Table 1.

APE model also constrains the condensation/evaporation of OM in the particle phase as mentioned in Pierce et al., (2008), which actually includes the vapor wall loss. In our cases, APE diagnosed a net evaporation in 16 of the 18 experiments corroborating our results that evaporation of the primary particles shapes the particle size distribution in the chamber.

We have added Figure R1 in the supporting material as Figure S2 and the sentences in line 4, Page 15252"…*D_1* is the Debye function (Aramowitz and Stegun, 1964). We use non-linear least-squares fitting optimization to best estimate the condensation/evaporation and wall-loss parameters in the APE model. The goal of fitting optimization is to minimize X^2 as follows:

$$X^2 = \sum_{a=1}^{3}\left(\frac{(M_{i(a),p} - M_{i(a),o})}{M_{i(a),o}}\right)^2 \qquad \text{Eqn.2}$$

where i(a) is the set of total diameter moments (0, 1.5, 3). The chosen range of moments ensures the model fits both the total number (0^{th} moment) and the total volume (proportional to the 3^{rd} moment). The subscripts p and o indicate the predicted and observed moments, respectively. Derived k_{w0} and k_e for 18 experiments are listed in Table 1. Calculated wall loss rates (k_w, s^{-1}) for the 18 experiments are generally consistent with each other as shown in Figure S2." and we add Figure R1 in the supporting materials.

Table R1 Comparison of original and new fitting kw0 and ke and average number concentration for the particles less than 20 nm during POA characterization for 18 experiments.

Burn ID	Fuel type	Original fitting		New fitting with less moments		Average Num. concentration for particles less than 20 nm
		k_{w0}	k_e	k_{w0}	k_e	
37	lodgepole pine	6.68E-05	4.81	8.03E-05	1.07	232.54
38	lodgepole pine	2.06E-05	6.05	6.27E-05	1.41	163.81
40	ponderosa pine	3.76E-05	3.19	8.67E-05	0.69	210.46
42	wiregrass	6.92E-05	2.46	1.07E-04	0.77	85.93
43	sawgrass	9.84E-05	7.08	1.07E-04	0.52	158.48
45	turkey oak	5.87E-05	2.91	8.11E-05	0.99	142.35
47	gallberry	7.90E-05	0.54	7.37E-05	0.19	271.63
49	sagebrush	4.78E-05	6.12	8.84E-05	0.84	184.86
51	alaskan duff	3.41E-05	0.92	7.00E-05	0.32	115.46
53	sagebrush	2.31E-05	7.57	8.43E-05	0.91	216.59
55	white spruce	1.95E-05	3.31	8.13E-05	0.31	143.68
57	ponderosa pine	5.10E-05	4.08	8.43E-05	0.96	291.67
59	chamise	2.06E-05	5.22	7.58E-05	0.83	279.33
61	lodgepole pine	3.06E-05	3.40	6.30E-05	0.29	215.63
63	pocosin	5.26E-05	1.25	8.46E-05	0.37	316.75

65	gallberry	1.35E-04	0.03	1.43E-04	0.62	61.89
66	black spruce	3.11E-05	1.42	1.02E-04	0.36	157.93
67	wiregrass	4.55E-06	0.49	5.78E-05	0.28	400.13

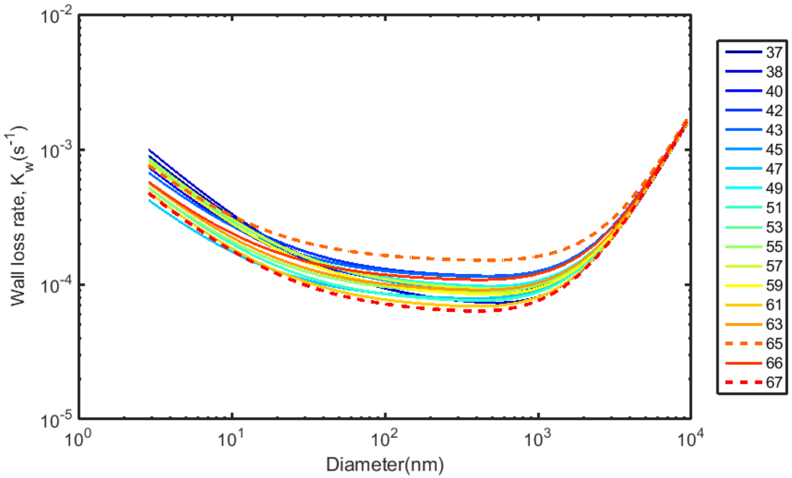

Figure R1 Size resolved wall loss rate, k_w (s^{-1}) for 18 experiments. The number represents the Burn ID for each experiment.

- The model is initialized assuming equilibrium between the gas and particle phase. Are the walls assumed to be initially “empty” from SOA? If so, would this assumption cause overestimation of gas-wall losses? In addition, instant 25:1 dilution seems like an unreasonable assumption.

To answer the first question: yes, in each experimental simulation, the walls assumed to be initially “empty” of OA. To test the possible influence of this assumption and address the second question, we ran additional “10-day” simulations. In these simulations, we kept the deposited particles and vapor on the wall in the model in the first hour and “flushed” the bag overnight (for 12 hrs) by assuming concentrations in the bag to be exactly 0 (perfect flushing) and simulating the evaporation of the vapors and particles in/on the wall. We then repeated the same experiment the next day using the particles/gasses on the walls after flushing. We repeated this cycle 10 times to simulate the effects after 10 days of experiments. In this test, the partitioning between vapor and particles on the wall was included in the model. The figure below shows the 13-hr evolution of organic mass (OM) and number concentration for this test with each line representing a different day. We can see that the OM lost to the walls initially increases because the "experiments" occur in the first hour of each line and the vapor and suspended OM decrease to 0 when “flushing” begins in the following 12 hrs. Deposited particle mass decreased slightly while deposited vapor decreases by 1/2 or 1/3 of the concentration for each experiment due to the vapor evaporation from the wall. Deposited particles and vapor on the wall accumulate across the experiments but exert little influence on OM particle and vapor in the chamber for next experiment. It is possible that over >>10 experiments the effect may be more substantial, but we do not know the history of this bag outside of the 18 FLAME experiments.

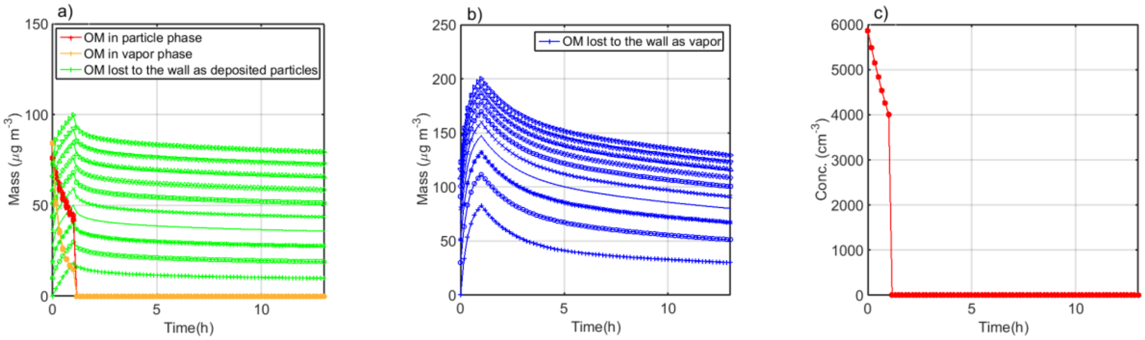

Figure R2 The 13-hr evolution with 1th hr experiments and following 12 hrs “flushing” of a) organic matter (OM, µg m^{-3}) in the particle phase, vapor phase and lost to the wall as deposited particles; b) OM lost to the wall as vapor; c) number concentration (cm^{-3}) taking into account the influence of the deposited particles and vapor on the wall based on the data for Burn 37. The different symbols represent the different times of experiments. The order of symbols from 1 to 10 time simulation is ‘+','o','*','.','x','□','◇ ','△','▽', and '▷ '.

In the manuscript, we have added the Figure R2 in the supporting material as Figure S3 and modified the sentences after line12, page 15254.

“…across the 18 experiments is within 1%.

The buildup of wall-deposited particles and vapor on the wall of the chamber that is retained between experiments might have impacted the observations we used to initialize and compare with our simulations. We therefore modeled this potential influence by retaining the wall-deposited particles and vapors and repeating the same flush / fill experiment 10 times, simulating 12 hours of “flushing” (the particle and vapor concentrations in the volume of the chamber set to 0) between experiments. In these 10-repeat tests, the 10th experiment had an increase of 8.8% in OM in the suspended particle phase and 2.9% in the vapor phase due to the slow and thus incomplete evaporation of the wall-deposited compounds during the flushing process, and the resulting buildup of particles/vapor on the walls slowing vapor deposition to the walls in the subsequent “experiment” (Figure S3). These results suggest that after a number of experiments, the accumulated wall-deposited particles and vapor in the chamber could have some influence on the next set of experiments even after the chamber was flushed overnight. However, since we do not know the history of the bag outside of these FLAME III experiments, we do not attempt to account for these effects in this modeling study but suggest they may represent an important uncertainty in SOA formation studies and should be explored.”

Regarding the third question (25:1 instant dilution), the ratio was roughly estimated by the study of Hennigan et al. (2011) as we mentioned line 1-4, page 15249. We ran the test as a bounding case to the influence of dilution. In the text, we acknowledge the uncertainty associated with this simulation, “It is not clear if this sensitivity study is a better assumption than our base assumption (particles and vapors start in equilibrium in the CMU chamber) as the CMU smog chamber was actually filled over 30 min and thus particles and vapors will move towards equilibrium (and particles and vapors will also be lost to the walls) during these 30 min. However, since we do not know the actual vapor-phase concentrations at the time when the CMU chamber is full and particle-phase measurements start, we are left with these two assumptions for bounding our results."

Minor comments:

- Page 15245, Line 22: What cooling effects?

We have rephrased the sentence as "The net effect of the climate forcing from biomass burning aerosol has been estimated in some studies to be nearly zero or negative due to the dominant cooling direct effect of primary organic aerosol (POA) over the warming from BC, as well as an indirect cooling effect from the particles' interactions with clouds by modifying the cloud albedo (Bond et al., 2013)."

- Page 15251, Lines 15-17: Why wouldn't the lower "effective" accommodation coefficients be appropriate for the POA partitioning?

We are unaware of studies showing mass transfer limitations in fresh POA from combustion. We have modified the text to say, "… because we are unaware of observed mass transfer limitations in fresh POA."

- Page 15260, Line 21: Fig 8. should be Fig. 9

Corrected.