Responses to the comments of anonymous referee #2

We thank the referee for the valuable comments that have greatly helped us to improve the manuscript. Please find below our responses (in black) after the referee comments (in blue). The changes in the revised manuscript are written in *italic*.

General Comments Jiang et al. report a modeling study that: (1) evaluates the impact of vapor wall losses during chamber studies on parameters for SOA formation from residential biomass burning emissions, and (2) simulates the increases in OA and SOA concentrations once a CTM is updated with new SOA parameters that take into account these wall losses. Overall, the manuscript is well-written and addresses an important topic in the field of atmospheric aerosol modeling, since implementing accurate parameters for SOA formation in CTMs is challenging. I support publication in GMD once my comments below have been taken into consideration.

Line-by-line comments

Page 1, Line 27 – 29: What is the difference between the "standard VBS" and the "reference scenario"? Does the reference scenario refer to the traditional two-product approach? Please clarify as this is an interesting finding.

The "standard VBS (VBS_BASE)" refers to the default VBS parameterization in CAMx v6.5, while in the "reference scenario (VBS_noWLS)" and "wall loss corrected scenario (VBS_WLS)" we adopted the optimized parameterizations based on the chamber experiments with different assumptions: there is no loss of condensable gases to the chamber wall for VBS_noWLS, and there is for VBS_WLS. We updated the abstract to clarify it. A paragraph as well as a Table 1 were added to Section 3.2.1 to describe the difference of each OA schemes.

"The modeled results from the VBS schemes with standard (VBS_BASE) and vapor wall loss corrected parameters (VBS_WLS), as well as the traditional two-product approach were compared and evaluated by OA measurements from five Aerodyne aerosol chemical speciation monitor (ACSM)/aerosol mass spectrometer (AMS) stations in the winter of 2011. An additional reference scenario VBS_noWLS was also developed using the same parameterization as VBS_WLS except for the SOA yields which was optimized assuming there is no vapor wall loss. The VBS_WLS generally shows the best performance for predicting OA among all OA schemes, and reduces the mean fractional bias from -72.9% (VBS_BASE) to -1.6% for the winter OA. In Europe, the VBS_WLS produces the highest domain average OA in winter (2.3 μg m^{-3}), which is 106.6% and 26.2% higher than VBS_BASE and VBS_noWLS, respectively." (P1, L21–29)

"*To investigate the effects of vapor wall loss corrected yields, as well as to compare to other modifications/parameterizations that are currently strongly debated in the community, five simulations with different OA schemes were conducted in this study (Table 1). Besides VBS_WLS which uses the optimized parameterization with vapor wall loss correction for the biomass burning sector, SOAP and VBS_BASE represent the two standard parameterization in CAMx; VBS_3POA represents a common approach to offset the missing SVOC emissions in recent modelling studies without vapor wall loss; VBS_noWLS is another reference case for that without vapor wall loss, which uses exactly the same parameters as VBS_WLS except for the SOA yields from IVOCs. Details about each OA schemes are introduced below:…" (P6, L173–179)*

Table 1: Description about the different OA schemes.

OA scheme	IVOB[a] emissions	k_{OH} for IVOB (cm^3 $molec^{-1}$ s^{-1})	SOA yields for IVOB (ppm/ppm)[b]
SOAP	= 4.5*POA_BB	1.34	/[c]
VBS_BASE		4.0	[0.081, 0.135, 0.800, 0.604, 0.0]
VBS_3POA		4.0	[0.081, 0.135, 0.800, 0.604, 0.0]
VBS_noWLS	= 12*POA_BB	1.5	[0.014, 0.036, 0.076, 0.136, 0.44]
VBS_WLS		1.5	[0.078, 0.118, 0.157, 0.177, 0.312]

[a] IVOB is the abbreviation of "IVOC from Biomass Burning" in CAMx

[b] The yield values are corresponding to volatility bins with saturation concentrations of 10^{-1}, 10^{0}, 10^{1}, 10^{2} and 10^{3} μg m^{-3}.

[c] SOAP does not separate IVOC from biomass burning and other anthropogenic sectors, and therefore is not comparable with the SOA yields for IVOBs.

Less importantly, I also don't understand why the authors have specifically highlighted the result from Romania.

We rephrased the sentence as *"VBS_WLS leads to an increase in SOA by up to ~80% (in Balkans)" (P1 L30)*

Page 2, Line 37 – 39: The authors imply that residential biomass burning emission is "the dominant source for. . . secondary organic aerosols in winter". However, this statement is supported exclusively by three European studies that are referenced on line 39. Therefore, the authors need to clarify that this conclusion is specific to the European domain.

We rephrased the sentence (in P2, L39) to clarify it refers to the European domain.

"…residential biomass burning emissions have been recognized as the dominant source for both primary (POA) and secondary (SOA) organic aerosols in Europe during winter time"

Page 2, Line 47: I would kindly suggest that the authors specify that the POA emissions are treated as semi-volatile when this "scaling-up" is performed, since some models still assume that POA is nonvolatile.

Done. We updated the sentence as follows:

"In order to compensate the effects from missing precursors, various modeling studies treated the POA as semi-volatile and increased the POA emissions by a factor based on findings from chamber experiments…" (P2, L49)

Page 2, Line 57: The work of Hayes et al. 2015 concerning vapor wall losses used a box model and not a CTM.

We apologize for the mistake. The statements are now corrected in P2, L60.

"This factor was also implemented in a box model with volatility basis set (VBS) scheme (Hayes et al., 2015),…"

Section 2.1: How does the utilization of beech wood as the only fuel potentially bias the results? Is this a fuel commonly used in residential biomass burning in Europe? Basing the parameterization of the VBS scheme on a single fuel is not necessarily a flaw in the study, but some contextualization is needed here to understand how this limitation might influence the model results.

Beech is one of the major forest trees in Europe, and beech wood is widely used for combustion as a heating fuel in European households. That's why beech wood is selected in our chamber experiments.

Different biomass fuel types may influence the vapor composition. Nevertheless, a recent study showed that despite large differences in fuel type and burning conditions, SOA formed is consistent for similar total NMOG load and OH exposure (Lim et al., 2019). This is also consistent with our results showing no dependence of the closure on the combustion regime (flaming vs. smoldering). Therefore, while we think that the fuel type may have an influence on the results this influence is minor compared to other sources of uncertainties (e.g. vapor wall losses or not considering the influence of SVOC on SOA formation). We added the explanation in P3, L83–L87.

"Beech wood is selected as it is one of the major forest types in Europe, and is widely used for residential heating and cooking in Europe. Although different biomass fuel types may largely affect the emitted organic gas species and affect the SOA formation, a recent study showed that the effect of biomass fuel type on SOA formation is much smaller than the effects of initial OM load and OH exposure (Lim et al., 2019)"

Line 105: The phrase "gas-phase equilibrium concentrations in particle phase" is not coherent. Please clarify. Furthermore, I don't think equation (3) can be correct. A partitioning coefficient of 1 would indicate complete partitioning to the particle phase, but this would give a Ceq (i,p) value of zero. This comment also applies to equation (4).

Here is an ambiguity in the term we have used. The C_{eq} refers to gas-phase concentrations at equilibrium with respect to the particle phase ($C_{eq,p}$) and to the chamber walls ($C_{eq,w}$). Therefore, a partitioning coefficient of 1 would indicate complete partitioning to the particle phase, which is consistent with a gas-phase concentration Ceq (i,p) of zero. We updated the explanation of C_{eq} in P4, L122–L123.

"the gas-phase concentrations at equilibrium with respect to the particle phase ($Ceq_{i,p}$) and to the chamber wall ($Ceq_{i,w}$)"

Line 201: The reasoning why the OM loading would have an effect on the box model's accuracy is not clear. Please elaborate. Also, there are some runs when the are low when the model accuracy is very reasonable, for example a11, so the OM loading does not seem to explain by itself the poor accuracy of the model observed for experiments 9 and 14.

Since we optimized the parameters by the sum of mean bias (MB) and RMSE between modelled and measured OA for all 14 experiments, experiments with higher OM loads and SOA productions (which normally have larger MB and RMSE), have higher impact during the model optimization, leading to a better model performance for experiments with higher OM loads after optimization. That is why we mentioned that experiments with lower OM loads may have lower accuracy. We clarify this in the new version of the manuscript.

"The model reproduces the process of OA formation for most of the experiments well, except for experiment #9 and #14 which have relatively lower OM loads (26 and 48 μg m^{-3} for Exp9 and Exp14, respectively). It can be partially explained by different weighting impact for experiments with high or low OM loads. The experiments with higher OM loads normally have larger MB and RMSE in the beginning of optimization, and therefore have higher impact during the model optimization. A direct consequence is the optimized parameters would work better for those experiments with higher OM loads. However, the model performance on each experiment could also be influenced by a series of other factors such as temperature and chamber conditions… " (P8, L230–L234)

Figure 1: It would be useful if a table of the experiment conditions was provided.

We did not describe the experimental conditions in detail as they have already been reported in our previous publication (Stefenelli et al., 2019). As suggested by the referee, we added additional information in Table S1.

"The conditions of each chamber experiment are shown in Table S1. More detailed description of the experiments can be found in Stefenelli et al. (2019), Bertrand et al. (2017) and Bruns et al. (2016)." (P3, L90)

Table S1: Experimental conditions for the 14 chamber experiments used in this study.

#Exp	References	Date	Experimental temperature (°C)	Stove type [a]	OM loads (μg m^{-3})
1	Bertrand et al. (2017)	29.10.2015	2	stove 1	198
2	Bertrand et al. (2017)	30.10.2015	2	stove 1	285
3	Bertrand et al. (2017)	04.11.2015	2	stove 1	123
4	Bertrand et al. (2017)	05.11.2015	2	stove 1	46
5	Bertrand et al. (2017)	06.11.2015	2	stove 2	75
6	Bertrand et al. (2017)	07.11.2015	2	stove 2	134
7	Bertrand et al. (2017)	09.11.2015	2	stove 2	81
8	Bruns et al. (2016)	02.04.2014	-10	stove 3	19
9	Bruns et al. (2016)	17.03.2014	-10	stove 3	26
10	Bruns et al. (2016)	25.03.2014	15	stove 3	62
11	Bruns et al. (2016)	27.03.2014	15	stove 3	45
12	Bruns et al. (2016)	28.03.2014	15	stove 3	42
13	Bruns et al. (2016)	29.03.2014	15	stove 3	48
14	Bruns et al. (2016)	30.03.2014	15	stove 3	48

[a] Stove 1 manufactured before 2002 (Cheminées Gaudin Ecochauff 625), stove 2 fabricated in 2010 (Invicta Remilly) and stove 3 (Avant, 2009, Attika).

Line 203 – 204: I think the text contains an error here. If anything there is an underestimation at short times and an overestimation at long times. More generally, it seems like the comparison between the model and the measurement varies a lot between experiments, so it is difficult to make conclusions regarding whether the box model is overestimating or underestimating.

We believe there is some misunderstanding here. We do not mean to conclude whether the model is overestimating or underestimating here, but to show the agreement between the shapes of measured and modeled curves is improved for most of the experiments when the vapor wall loss is considered. We clarify the point in P8, L235–L236

"… the agreement between the modeled and measured trends was improved when the vapor wall loss is taken into account."

Lines 205 – 207: I think using percentages here to compare the two box model versions (with or without vapor wall losses) overstates the difference between the models. In the end, the differences in the MB and RMSE are only about 6 ug/m3, which is not very much when most of the experiments are run at OM concentrations near or above 100 ug/m3.

The average mean bias was improved from -12.8 to 6.7 μg m^{-3}. The change from underestimation to overestimation somehow indicates we find a potential reason for the long-existing problem – underestimated OA in modeling studies, which means more than the absolute change of 6 μg m^{-3}. In addition, the model performance is not only about the bias at the end point, but also the capability to reproduce the chemical processes of OA formation. As we mentioned before, the shape of the

modeled curves for OA formation with vapor wall loss corrections shows higher agreement with the measurements.

Figure 3: I very strongly suggest that these data also be given in a table so that the quantitative results can be used by other researchers.
We will upload the data for each figure after the acceptance of the manuscript.

Lines 284 – 287: This sentence is confusing. Which model cases are specifically being compared? In addition, in Figure 7, only the schemes VBS_WLS and VBS_noWLS are compared, but then in the text the 3POA scheme is mentioned as well.
We updated the sentence in L284 (current L316) as follows to make it clear.

"*The overall relative differences between VBS_WLS and VBS_noWLS are more than 80% and the highest grid-scale increment reaches 5.6 μg m^{-3} in the region of Balkans.*"

We are sorry for the confusion for Fig. 7. The "Fig. 7" here is a typo, and we have corrected it to "Fig. 6" (P11, L320).

Figure 8: It would be helpful to specify in the figure caption that these plots are annual averages. In addition, why are annual averages used and discussed rather than wintertime measurements, as is done in the other sections of the manuscript?
We specified it is the annual average in the caption of Fig. 8 as the referee suggested. We compared the modeled SOA/OA fraction (fSOA) with the measurements from literature covering different seasons in Section 4.3.2. The number of reported measurements is limited if only winter data were included. That's why we discussed the whole year in this section.

*"**Figure 8:** Modeled fractions of annual mean SOA to total OA (fSOA) using different OA schemes. Modeled results of VBS_3POA are very similar to VBS_noWLS, and therefore are not shown here."*

Lines 318 – 322: The comparisons summarized here are rather haphazard. First, for OA, the VBS_WLS scheme is compared to the VBS_BASE scheme. Then next, for SOA, the VBS_WLS scheme is compared to the SOAP scheme. The authors should be consistent in what schemes they are comparing to as "base cases".
Here we intend to show the ranges of improvement. We mentioned the VBS_BASE and SOAP specifically because they had the largest absolute MFB for OA and SOA, respectively. We updated the sentence (P12, L354) to specify they refer to the largest mean fractional bias.

"Comparison of the modeled results with different OA schemes with the field measurements from five ACSM/AMS stations in Europe in winter, suggests that VBS_WLS generally has the best performance to predict OA, which lowers the highest mean fractional bias from -72.9% (VBS_BASE) to -1.6% for OA, and -77.8% (SOAP) to 20.0% for SOA."

References

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