In the manuscript by Ciarelli et al. a modified VBS scheme for biomass burning-like OA is implemented in the chemistry transport models CAMx. The new VBS scheme was developed by the authors and is described in a paper under review in the GMD (Ciarelli et al., 2016b). CAMx is then used to simulate the wintertime OA mass concentration in Europe in Feb-Mar 2009. The model results are compared with model results from simulations using a different VBS scheme for biomass bring-like OA (Ciarelli et al., 2016a) and with AMS measurements from 11 stations around Europe. The manuscript addresses a very relevant and important topic: the contribution of small-scale residential (mainly wood) combustion to the OA mass loading in Europe during wintertime. My review mainly focuses on the method section, which I partly found quite hard to understand. I have a few critical questions conserving how the different VBS methods was applied which would like to get answered and also explained clearly in the manuscript. If this is done properly and the new VBS parameterization is considered to be scientifically sound by the reviewers of the GMD manuscript Ciarelli et al., 2016b, then I think the manuscript can be suitable for publication in Atmospheric Chemistry and Physics.

More general comments mainly concerning the method:
Is it correct that the only difference between the base case model runs from Ciarelli et al., 2016a and this new study is that you use the new VBS sets (called VBS_BC_NEW) instead of VBS_BC to describe the OA formation from biomass burning emissions? If the answer on this question is Yes, which I hope is the case, then please state this clearly in the manuscript. If the answer is No, you have to clearly explain all differences between the two different model runs.

Is it correct that you in total use 3 VBS sets to describe OA formation from biomass burning, 1 set for fresh HOA from fossil fuel combustion, 1 set for aged oxidized HOA, 1 VBS set for BVOC oxidation products (no ageing considered). Thus in total 6 VBS sets? I think you need to describe this more clearly in the manuscript and how this compares to the VBS sets used in Ciarelli et al., 2016a.

In Ciarelli et al. (2016a) for further aging of ASOA and POA vapors from HOA-like emissions you use a reaction rate with OH of 2×10^{-11} cm^3 $molec^{-1}$ s^{-1}. How about this new study? Was it 4×10^{-11} cm^3 $molec^{-1}$ s^{-1} instead or is this only the reaction rate used for BBOA precursors?

I suggest that you create a table where you list all VBS sets used in the two different model simulations and if they represent SOA or POA, the sources (i.e. BVOCs, biomass burning or fossil fuel burning) and if they represent HOA, BBOA or Biogenic OA. As the manuscript is written now I get very confused about which organic compounds that are POA and SOA, their origin and if they are classified as HOA, BBOA or Biogenic OA. If I understand it correctly the BBOA is only the not atmospheric processed (oxidized) POA emission from biomass burning, and the POA from biomass burning sources that evaporates and then form more oxidized OA is treated as SOA (VBS_BC_NEW set 2) and the VBS_BC_NEW set 3 OA is always treated as SOA. I think you need to more clearly state that BBOA is only referring to the POA from biomass burning but not the SOA formed from biomass burning. I suggest that you change from BBOA to BBPOA. It is only in the abstract L40-41 that you mention that BBOA is referring to <u>primary</u> biomass burning-like OA and HOA <u>primary</u> hydrocarbon-like OA. I missed this and was confused about this when I was

reading the manuscript the first time.

The new version of the model underestimates the OA to a less extent than the previous version. The only difference between the model runs is how the BBOA formation is treated. On L306-307 you write: *"The modelled BBOA fraction on the other hand was generally higher than the measurements, with an average MFB of 50% (Table S3, Figs. 6-7)"*.
I interpret this as that the model improves the modeled total OA but at least partly for the wrong reason because it gives too much BBOA.

In Ciarelli et al. (2016a) where you use the VBS_BC method you write: *"Further aging of BSOA is not considered in this study, based on previous modelling results showing overprediction of OA when such process is taken into account (Lane et al., 2008; Murphy and Pandis, 2009). This implies that also further aging of POA vapors from BBOA-like emissions was not considered since it is performed in the same basis set."*
This is a large assumption which I think might be one of the main reason why you get much less OA (especially SOA from biomass burning sources) when you run VBS_BC instead of VBS_BC_NEW. This needs to be discussed and explained in the manuscript. I also think that you should run a model simulation using VBS_BC but separating the POA vapors from BBOA and allow them to be further oxidized in the same way as the HOA vapors. Then you compare the model results from this method with the model simulations with your new VBS sets (VBS_BC_NEW). To me this is a more fair evaluation of your new biomass burning-like OA VBS parameterizations compare to the old parameterization from Koo et al. (2014) that to my understanding allowed oxidation of evaporated POA from biomass burning sources. If your new biomass burning VBS parameterization still gives substantially better agreement with the observations than the Koo et al. (2014) VBS parameterization, then your contribution to the field can be considered substantial and important.

On L49-54 you write: *"Contributions to OA from residential combustion precursors in different volatility ranges were also assessed: our results indicate that residential combustion gas-phase precursors in the semi-volatile range contributed from 6 to 30%, with higher contributions predicted at stations located in the southern part of the domain. On the other hand, higher volatility residential combustion precursors contributed from 15 to 38% with no specific gradient among the stations."*
I don't understand how you can separate the SVOC molecules in the gas-phase from the SVOC molecules in the particle phase. If we assume equilibrium partitioning between a condensed liquid phase and the air, then the same molecule species are cycled back and forth between the gas- and particle phase because of evaporation and condensation. Do you mean: additional OA formed because of SVOC oxidation in the gas-phase as compared to additional OA formed as a consequence of IVOC oxidation in the gas-phase?
Related to this I also wonder if all POA species (which are SVOCs) are allowed to evaporate (assuming equilibrate with the gas-phase at all times) and form more oxidized organic compounds that become SOA in the model when they re-condense. Thus, is it correct that all POA species eventually end up as more oxidized SOA species in the model? Or is only a fraction of the POA species allocated to the gas-phase and the rest is fixed in the particles based on the initial fresh POA VBS distribution (Fig S1)?

On L156-159 you write: *"Hourly emissions of biogenic VOCs, such as monoterpenes, isoprene, sesquiterpenes, xylene and toluene, were calculated using the Model of Emissions of Gases and Aerosols from Nature MEGANv2.1 (Guenther et al., 2012) for each grid cell in the model domain."*

But into what VBS scheme are the BVOC oxidation products added? In Koo et al., (2014) and Ciarelli et al. (2016a) you have a 4th VBS set for BVOC oxidation products which are not allowed to age because then you get too much SOA.

According to Section 3.3 all of the modeled SOA can either be attributed to residential or non-residential combustion. What about the SOA from BVOCs? Did you not consider BVOCs when you calculated the SOA formation in this new study? I thought that the only difference between the base case model run in Ciarelli et al. (2016a) and in this work was the VBS setup for the BBOA and it's transformation to SOA? This needs to be clarified. At least for the southernmost stations I would expect that BSOA formation also is substantial during the wintertime, and transport from south to north could bring this SOA to the northern latitudes too.

On L170-173 you write: *"The third set allocates oxidation products from traditional VOCs (xylene, toluene, isoprene, monoterpenes and sesquiterpenes) and from non-traditional SOA precursors retrieved from chamber data (~4.75 times the amount of organic material in the semi-volatile range, Ciarelli et al., 2016b)."*

So do I understand it correctly that this 3rd VBS set for VOCs originating from biomass burning only considers the traditional VOCs emitted from biomass burning but not the traditional VOCs from other sources? I.e. the same traditional VOC species but from other sources (vegetation and fossil fuel) is added to other separate VBS sets. This, would be desirable since it allows you to distinguish SOA formed from biomass burning, biogenic VOCs and VOCs from fossil fuel sources?

On L232-235 you write: "In spite of the improvements with respect to earlier studies, modelled OA is still lower than measured (mean bias MB from -0.1 μg m^{-3} up to -3.1 μg m^{-3}) at most of the sites, with only a slight overestimation at a few locations (MB from 0.3 μg m-3 up to 0.9 μg m-3)."

Here I think you also should mention that the model might underestimate the OA formation because no gradual BVOC oxidation is considered. Or maybe even more if you did not consider any biogenic SOA formation?

Was the influence of NO considered when you divided the SOA precursors into the different VBS bins as was done by Koo et al. (2014)?

<u>To summarize</u>: The model results looks reasonable and the agreement between the model and observations are as good as you could expect both when using the new VBS set and the old VBS set from Ciarelli et al. (2016a). But to me it still remain to be shown that the new VBS parameterization for biomass burning-like OA substantially improves the model performance as to compared to the VBS parameterization developed by Koo et al. (2014). I.e. you need to compare the model results from the simulations with your new VBS parameterization with a simulation using the Koo et al., (2014) VBS parameterization where you also allow the evaporated BBOA material to be further oxidized in the gas-phase. I also think you need to evaluate if not at least part of the reason why the model underestimates the OA is because it underestimates or maybe not even considers biogenic SOA

formation.

Minor specific comments:

L47, Page 1: Here you use the term "*transportation precursors*". I think you mean precursors from the road transportation sector. I think you should change the formulation a bit to make this clearer.

L78-79 You write: "*Moreover, numerous ambient studies of open burning plumes from aircraft do not show a net increase in OA, despite observing oxidation (Cubison et al., 2011; Jolleys et al., 2012).*"
I suggest that you reformulate this sentence and instead write something like:
Moreover, numerous ambient studies with aircraft of open biomass burning plumes do not show a net increase in OA, despite observed oxidation (Cubison et al., 2011; Jolleys et al., 2012).
When I first read this sentence I thought the open burning plumes came from the aircraft but then I realized that the aircrafts where only used for the measurements of the open biomass burning plumes.

L98-103: The sentence: "*Ciarelli et al. (2016a) showed that allowing for evaporation of primary organic particles as available in European emission inventories degraded OA performance (further under-predicted OA but with POA and SOA components in a better agreement) whereas model performance improved when volatility distributions that implicitly account for missing semi-volatile material (increasing POA emissions by a factor of 3) were deployed.*" is hard to understand. I suggest that you split it into two or three sentences. What do you mean with "*degraded OA performance*"? Do you mean: degraded the model performance concerning the modeled total OA mass?

On L112-115 you write: "*This indirect accounting of missing organic material could be used in the absence of more detailed gridded emission inventories, keeping in mind that the amount of higher volatility compounds was specifically derived from studies conducted with diesel engines (Robinson et al., 2007).*"
In fact I think the Robinson et al., (2007) study was only performed on one single diesel engine (a single-cylinder Yanmar diesel generator), which I expect do not represent modern diesel car engines very well.
I suggest that you instead of "*diesel engines*" at least write: *a singe diesel engine.*

On L284-287 you write: "*On the other hand, the remote station of Mace Head showed a positive bias for SOA (MFB = 30%), even though model and measurement concentrations were very similar (0.54 and 0.35 μg m-3, respectively), which could be attributed to an overestimated contribution from the boundaries.*"
What do you mean by " *overestimated contribution from the boundaries*"? Is it the influence from the model boundary conditions?

On L337-340 you write: "*The model results indicate that non-residential combustion and transportation precursors contribute about 30-40% to SOA formation (with increasing contribution at urban and near-industrialized sites) whereas residential combustion (mainly related to wood burning) contribute to a larger extent, i.e., around 60-70%.*"
I suggest that you change to:

The model results indicate that non-residential combustion and transportation precursors contribute <u>to</u> about 30-40 % <u>of the</u> SOA formation (with increasing contribution at urban and near-industrialized sites) whereas residential combustion (mainly related to wood burning) contribute to a larger extent, i.e., around 60-70%.

On line L349-351 you write: "*In the southern part of the domain, the higher temperature will favour more organic material in the semi-volatile range to reside in the gas-phase, rendering it available for oxidation.*"
I would also expect that the higher UV-light intensity in the south caused more SOA formation because of higher OH concentrations.

On line L351-351 you write: "*On the other hand, no south-to-north gradient was predicted for the higher volatility class of precursors.*"
Do you mean?
On the other hand, no south-to-north gradient was predicted for the SOA formed from the higher volatility class of precursors.

On L 291-294 you write: "*Mostly traffic-related HOA was underestimated at the urban site Barcelona (Table S2, Fig. 6), with the model not able to reproduce the diurnal variation of HOA at this urban site likely due to poorly reproduced meteorological conditions or too much dilution during day time in the model (Fig. S2).*"
Can it not also be because of too weak diurnal variations in the HOA emissions from traffic in the model?
Reflection: But in the case of Helsinki it seem as if the model instead gives substantially more HOA during the morning (6 UTC, 8 am local time, and 15 UTC, 5 pm local time), which is what you would expect if the HOA mainly came from the local traffic. But surprisingly to me the observations do not indicate any increased local HOA contribution during the morning and afternoon rush hours in Helsinki. Could it be related to the vehicle fleet in Helsinki (i.e. is the road traffic very much dominated by gasoline cars which do not emit much primary HOA but precursors for SOA formation)?