

Responses to the comments of anonymous referee #2

Thank you for your comments that helped to improve our manuscript. Please find below your comments in blue, our responses in black and modifications in the revised manuscript in *italic*.

Ciarelli et al. follow up two other recent publications by augmenting the CAMx VBS implementation with their new parameterization for emission and aging of BBOA emissions. The study itself is a useful application and soundly conceived. The authors find better model-measurement agreement than their previous implementation, but I am troubled by some aspects of their methods and analysis, as described below. Their inclusion of the factor of 3 multiplier to account for missing SVOCs was an approach originally recommended for Mexico City but has not been used for Europe by previous EUCAARI model studies (e.g. Fountoukis et al., 2014). I am open to the authors' interpretation/justification for this choice (especially if I've misinterpreted the situation), but on its face this is a rather critical assumption that could put major aspects of the paper's conclusions in jeopardy. Moreover, the application of modeled PM_{2.5} mass to PM_{1.0} measurements raises questions about how much of the model agreement is spurious. Considering both of these potential biases together, it is concerning that the model predictions for SOA and POA are still lower in many cases than the VBS predictions published by Fountoukis et al. (2014) for the same model scenario. I could recommend this paper for publication after these issues are resolved.

Specific comments:

1. Page 4, line 108-113: The ratio of semivolatile to nonvolatile material is, as the authors know, a function of the emission source, fuel, and operating conditions – I think it is overly simplistic and actually unhelpful to state that the ratio is predicted to be “roughly 3.” The Shrivastava et al. (2011) and Tsimpidi et al. (2010) studies argued that those SVOCs at Mexico City were missing from the inventories because the emissions were parameterized using ambient observations of OA, which would have already equilibrated to atmospheric conditions. On the other hand, the emission factors used to inform the gridded inventories of Europe and the US are, to my knowledge, derived from laboratory scale tests, where much of those SVOCs are notoriously condensed in the particle phase in undiluted exhaust. My reading of Fountoukis et al. (2014) does not lead me to believe that they enhanced their SVOC emissions by a factor of 3 over POA. Rather, I believe they simply repartitioned the existing POA, and they added an additional 1.5*POA for the IVOCs as the authors state. Ciarelli et al. (2016a) shows that the extra SVOCs are needed to improve the model performance (i.e. VBS_BC did much better than VBS_ROB), but I disagree that there is evidence that SVOCs are underestimated in European inventories by so much. Instead, I would argue the real source of this mass is still unknown and is probably a combination of underestimated SOA yields, aqueous processing, aging of anthropogenic and biogenic SOA and some missing SVOCs as well.

At minimum, a considerable amount of rewriting in the methods, conclusions and abstract is necessary so that the authors communicate explicitly that an unknown fraction of these SVOCs are very likely double-counted and that this parameter needs to be refined and probably lowered in the future as more explicit pathways are added to the model.

It is true that our previous studies indicated a deterioration of the model performance for OA when evaporation of primary organic particles was allowed while using the approach proposed for the Mexico city study (Shrivastava et al. (2011); Tsimpidi et al. (2010)) led to a better performance (VBS_BC in Ciarelli et al., 2016a). We agree with the referee that other factors such as underestimation of SOA yields, aqueous processing and aging of anthropogenic and biogenic SOA might also play a crucial role in addition to missing SVOCs and we also agree that the factor of 3 used in this study for the inclusion of semi-volatile organic compounds might have high uncertainties.

This choice however, was based on the recent European study by Denier van der Gon et al. (2015) rather than the Mexico City studies. The OA emissions in Europe have often been claimed to be under-

predicted in current inventories (Bergstrom et al., 2012; Fountoukis et al., 2014) but only recent studies are starting to better elucidate the range of uncertainties related to them, in particular because of the semi-volatile nature of such material. In the work of Denier van der Gon et al. (2015) a revised residential wood combustion (named TNO-newRWC) emission inventory was compiled for Europe using a bottom-up approach. The authors underlined the importance of various sampling methods and measurement protocols or techniques influencing particle emission factors using data from the survey of Nussbaumer et al. (2008a,b). The most important sampling methods used by the countries participating in the survey, were filter measurements of only solid particles (SP) and dilution tunnel (DT) measurements of solid particles and condensable organics (or semivolatile organics). For conventional woodstoves, the authors found a difference in PM emission factors by a factor of up to 5 between the two techniques. The revised emission inventory (TNO-newRWC) was later compiled using the average DT emission factor from different type of appliance (Table 2 in Denier van der Gon et al., 2015) and compared with previously used emission inventory EUCAARI. The authors concluded that the revised emissions were higher than those in the EUCAARI inventory by a factor of 2-3 which is similar to the correction factor used in our study and in Shrivastava et al. (2011) and Tsimpidi et al. (2010). However, it should be noted that a substantial inter-country variation was reported within the gridded emission inventories which might lead to over or underestimation of emissions depending on the country (for example the ratio between the TNO-newRWC and EUCAARI emission inventory was around 1-3 in France and up to a factor of 5-10 in Sweden and Finland).

Denier van der Gon et al. (2015) also used the revised emission inventory in two commonly used chemical transport models carrying the VBS scheme to perform the organic chemistry: PMCAMx and EMEP model. They found that the revised emission inventory substantially improved the agreement between measured and predicted organic aerosol for the same period presented in this study (Feb-Mar 2009) with results in line with the VBS_BC scenario performed in Ciarelli et al. (2016a) and VBS_BC_NEW presented here. Therefore, we think that the correction factor proposed in this study (factor of 3) can be used until detailed emission inventories including semivolatile compounds are available for the modeling community. Moreover, other explicit SOA formation pathways must be included as more experimental data will be available.

We inserted the following statements in the abstract, conclusions and method part as suggested by the referee.

In the abstract as below:

Although the new parameterization leads to a better agreement between model results and observations, it still under-predicts the SOA fraction suggesting that uncertainties in the new scheme and other sources and/or formation mechanisms remain to be elucidated. Moreover, a more detailed characterization of the semivolatile components of the emissions is needed.

In the method at line 171 of the revised manuscript:

In order to include gas-phase organics in the semivolatile range in the absence of more detailed inventory data, we used the approach of increasing the standard emissions by a factor of 3 proposed by previous studies (Shrivastava et al., 2011; Tsimpidi et al., 2010) which is also in line with the recent European study on the revision of the residential wood combustion emissions (Denier van der Gon et al., 2015). This approach of including the semivolatile compounds can be used until detailed emission inventories with more realistic inter-country distribution of the emissions become available (e.g. Denier van der Gon et al., 2015).

In the conclusions, line 417 of the revised manuscript:

On the other hand, the modelled BBPOA was higher than the measurements at several stations indicating the need for further studies on residential heating emissions, their volatility distribution and oxidation pathway of the semivolatile organic gases. In addition, more detailed emission inventories are needed to characterize the semivolatile components better, as proposed by Denier van der Gon et al. (2015).

2. I agree with the first reviewer that there needs to be significant more description of the VBS framework used here. The diagrams in Ciarelli et al. (2016b) are helpful and there should be a table or diagram in this manuscript that summarize that information for the entire VBS picture including emissions and aging.

We agree and revised the part about the VBS scheme as suggested also by Referee #1 in Section 2.2 as below. We added Table 1 to summarize the description of the VBS spaces.

2.2 Organic aerosol scheme

The biomass burning organic aerosol scheme was constrained using recently available wood burning smog chamber data (Bruns et al., 2016) as described in Ciarelli et al. (2016b). The model deploys three different basis sets (Donahue et al., 2011) to simulate the emissions of organics from biomass burning and their evolution in the atmosphere. The first set allocates fresh emissions into five volatility bins with saturation concentrations ranging between 10^{-1} and $10^3 \mu\text{g m}^{-3}$ following the volatility distribution and enthalpy of vaporization proposed by May et al. (2013). In order to include gas-phase organics in the semivolatile range in the absence of more detailed inventory data, we used the approach of increasing the standard emissions by a factor of 3 proposed by previous studies (Shrivastava et al., 2011; Tsimpidi et al., 2010) which is also in line with the recent European study on the revision of the residential wood combustion emissions (Denier van der Gon et al., 2015). This approach of including the semivolatile compounds can be used until detailed emission inventories with more realistic inter-country distribution of the emissions become available (e.g. Denier van der Gon et al., 2015). The second set allocates oxidation products from SVOCs after shifting the volatility by one order of magnitude. The third set allocates oxidation products from the traditional VOCs and biogenic precursors (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 semivolatile range, Ciarelli et al., 2016b). Primary and secondary semivolatile compounds react with OH in the gas-phase with a rate constant of $4 \times 10^{-11} \text{ cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$ (Donahue et al., 2013), which decreases their saturation concentration by one order of magnitude. This implies that also aging of biogenic products is implicitly taken into account. A reaction rate of $4 \times 10^{-11} \text{ cm}^3 \text{ molec}^{-1} \text{ s}^{-1}$ was also applied to the rest of the anthropogenic sources (referred to as HOA) in order to be consistent among all the other anthropogenic sources as already proposed by more recent studies for the range of saturation concentrations used here (Donahue et al., 2013). No heterogeneous oxidation of organic particles or oligomerization processes is included in the model. The new model parameterization described in this study is referred to as VBS_BC_NEW throughout the paper to distinguish it from the previous base case called VBS_BC as given in Ciarelli et al. (2016a). All the VBS sets are listed in Table 1. More details on the VBS scheme can be found in Ciarelli et al. (2016b) and Koo et al. (2014).

Table 1. Properties of the VBS space. Oxygen numbers for each volatility bin were calculated using the group-contribution of Donahue et al. (2011). Hydrogen numbers were calculated from the van Krevelen relation (Heald et al., 2010).

	log (C*)	Oxygen number	Carbon number	Hydrogen number	Molecular weight
	-1	4.11	11.00	17.89	216
POA set1* (BBOA-like)	0	3.43	11.75	20.07	216
Primary biomass burning (BBPOA)	1	2.73	12.50	22.27	216
	2	2.01	13.25	24.49	216
	3	1.27	14.00	26.73	215
SOA set2* (BBOA-like)	-1	4.53	9.00	13.47	194
	0	4.00	9.25	14.50	189
SOA from SVOCs biomass burning	1	3.40	9.50	15.60	184
	2	2.83	9.75	16.67	179
SOA set3* (BBOA-like)	-1	5.25	5.00	4.75	149
	0	4.70	5.25	5.80	144
SOA from VOC/IVOCs biomass burning and biogenics	1	4.20	5.50	6.80	140
	2	3.65	5.75	7.85	135
	3	3.15	6.00	8.85	131
POA set1** (HOA-like)	-1	2.69	17.00	31.3	278
	0	2.02	17.50	33.0	275
Rest of primary anthropogenic sources	1	1.34	18.00	34.7	272
	2	0.63	18.50	36.4	268
	3	0.0	19.00	38.0	266
SOA set1** (HOA-like)	-1	4.90	7.00	9.10	172
	0	4.38	7.25	10.1	167
SOA from rest of all anthropogenic in all volatility range (SVOCs,IVOCs,VOCs)	1	3.84	7.50	11.2	163
	2	3.30	7.75	12.2	158
	3	2.74	8.00	13.3	153

*Based on Ciarelli et al. (2016b).

**Molecular structure as in Koo et al. (2014) and Ciarelli et al. (2016a).

3. What is being done about wildfires in the model? Were there any during the EUCAARI scenario? Are they represented well in the emissions inputs? If so, how do they effect the source apportionment analysis that is presented?

Emissions from wildfires were not considered for this application since they were not delivered in the EURODELTA3 exercise for the year 2009 (Bessagnet et al., 2016). We analyzed the fire emission data (non-domestic fires) as available in IS4FIRES data (Sofiev et al., 2009) obtained by re-analysis of fire radiative power data from the MODIS instrument. Figure 1 below shows the cumulative emissions from wildfires in kg/s in March 2009. Significant fires occurred mainly in the north of Portugal. We think that the effect on the simulated OA concentrations might be quite limited since all the investigated stations are located quite far from that area.

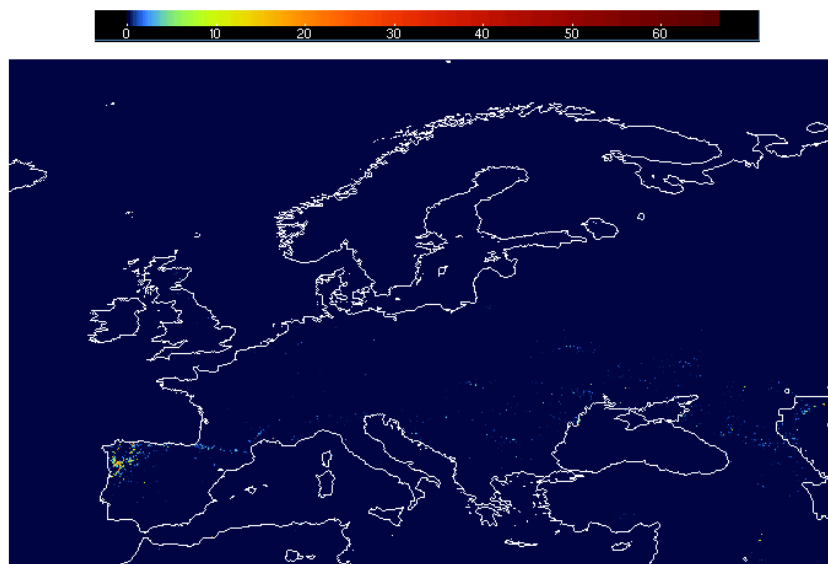


Figure 1. Cumulative emissions from wildfires in kg/s during the month of March 2009 as in IS4FIRES (Sofiev et al., 2009).

4. On page 5, lines 150-151, the authors point out that CAMx is predicting PM_{2.5}. But the evaluation is against AMS observations which I presume are primarily PM_{1.0}. Doesn't this fact make the frequent underprediction in SOA even more troubling? Is anything more specific known about the diameter of PM_{2.5} particles to allow the authors to estimate the fraction that would be PM_{1.0} and thus more applicable to the measurements?

This issue was discussed in Aksoyoglu et al. (2011) where PM₁ and PM_{2.5} measurements in Payerne during both winter (January 2007) and summer (June 2006) periods were compared. The authors concluded that the differences between the two fractions were usually rather small (Figure 14 in Aksoyoglu et al., 2011). These results are also supported by a recent study where comparisons between the organic matter concentrations in PM₁ and PM_{2.5} fractions in winter were found to be in the same range (Bozzetti et al., 2016).

5. Given that points 1 and 4 would lead one to expect substantial overprediction by the model, please also explain why the current predictions are lower than those in Fountoukis et al. (2014) at many sites.

We don't expect substantial overprediction due to particle size as we explained above at point 4. Even though both model simulations (this study and Fountoukis et al., 2014) mostly cover the same domain and time period, some differences are expected due to model resolution, different input data used in simulations as well as the differences in chemical mechanisms. Fountoukis et al. (2014) account for marine OA emissions which are not included in our simulations. In Fountoukis et al. (2014), the emissions were calculated based on the scheme of O'Dowd et al. (2008) and the organic aerosol fractions allocated in both fine and coarse mode. Gantt et al., (2015) showed that primary marine organic aerosol have a weaker coastal-to-inland concentrations gradient than sea-salt aerosol with some inland European cities having more than 10% of the submicron organic aerosol mass as a marine source. Differences in fire emissions, even if present, are likely to be less important for this comparison since there were few fires activity data during the considered periods (as addressed in comment 3).

Another difference is the way the boundary conditions for OA are taken into account. Fountoukis et al. (2014) used fixed boundary conditions based on measured average background concentrations in sites close to the boundaries of the domain whereas we derived OA boundary fields from MACC reanalysis data (Inness et al., 2013; Benedetti et al., 2009). In our study, OA fields at the domain boundaries are distributed as half-half between POA and SOA, as prescribed by the EURODELTA3 exercise, whereas in Fountoukis et al. (2014) OA at the boundaries are assumed to be all oxidized (SOA).

In addition to the input data, different gas-phase mechanisms used in both studies (CB05 in this study,

SAPRC99 in Fountakis et al., 2014) might lead to different results. One has also to keep in mind the different grid resolution ($0.25^{\circ} \times 0.25^{\circ}$ in this study, $36\text{km} \times 36\text{km}$ in Fountoukis et al., 2014) while comparing the two studies.

6. Page 9, lines 269-272: This discussion of Fig. 5 is very light. If there is not more to discuss, I recommend removing the figure and just stating the improvement in MB and r

We prefer to keep Fig. 5 since the figure allows the readers to see the differences for POA and SOA. We combined the two paragraphs where the discussion involves Fig. 5 as follows in line 284-294 of the revised manuscript:

Comparison of results from this study (VBS_BC_NEW) with the earlier one (VBS_BC, Ciarelli et al., 2016a) suggests that the new VBS scheme predicts higher SOA concentrations by about a factor of 3 (Fig. 5) and improves the model performance when comparing assessed OOA from measurements with modelled SOA (Table 4). POA concentrations, on the other hand, are clustered below $1 \mu\text{g m}^{-3}$ except in Barcelona (Fig. 5), showing an $R^2=0.36$ (Table 3). Although predicted POA concentrations at Barcelona were lower than the measurements, $MFB=-47\%$ and $MFE=69\%$ were still in the range for acceptable performance criteria ($MFE \leq +75\%$ and $-60 < MFB < +60\%$, Boylan and Russell, 2006). On the other hand, the model over-predicted the POA concentrations at Hyytiälä ($MFB=131\%$ and $MFE=131\%$), Helsinki ($MFB=95\%$ and $MFE=100\%$) and Cabauw ($MFB=76\%$ and $MFE=86\%$) mainly due to the overestimated BBPOA fraction as seen in Fig. 6.

7. How does the BBOA doubling sensitivity case fit in the context of the VBS_BC_NEW case which is multiplied by 3 and then by 1.5 again? What fraction of that total added vapor mass makes it into the particle phase? This is related to point 8

The BBOA doubling sensitivity case performed in Ciarelli et al. (2016a) with the original VBS scheme (Koo et al., 2014) was reported in order to show that although the model performance for total OA concentration improved significantly, that was not the case for the OA components, with POA being over-predicted at almost all the sites and no significant effect was observed on the modelled SOA concentrations (Figure 10). The rest of the comment is addressed in comment Nr. 8 (below).

8. The description and discussion of BBOA aging should be expanded. Please summarize the aging process as described in Ciarelli et al. (2016b). How is this similar/different to the aging of the traditional biogenic SOA? I assume the authors are not using the Koo et al. (2014) approach where the BBOA ages once and then stops? What is the fractional contribution of the various volatility bins to the total in time and space? Do they actually need 4 VBS bins to represent the aging, or would just using one bin and an IVOC precursor also work reasonably well? Why did they not use the O:C obtained from these AMS data to constrain the aging of the BBOA or the SOA?

A total number of 3 sets were used to describe the evolution of organic material. The first set was used to distribute the primary emissions (set1). Two other sets were used to model the formation and evolution of SOA. Oxidation products of SVOC material arising from primary emissions were allocated to set2, whereas oxidation products from NTVOCs (non-traditional VOCs) were allocated to set3. The specific molecular structures for each of the sets and bins were retrieved using the group contribution approach and the Van Krevelen relation (Donahue et al., 2011; Heald et al., 2010). The oxidation of semi-volatile material would tend to increase the compounds' oxygen number and decrease their volatility and carbon number, due to functionalization and fragmentation. We assume that the oxidation of the primary semi-volatile compounds with C11-C14 decreases their volatility by one order of magnitude and yields C9-C10 surrogates, placed in set2, based on the work of Donahue et al. (2011, 2012). Based on these assumptions and using the group contribution approach, the oxygen numbers for set2 is predicted to vary between 2.26 and 4.56. Thus, the model implicitly accounts for the addition of 1.1 to 1.5 oxygen atoms and the loss of 2.75 to 4.25 carbon atoms, with one oxidation step. Set3 was constrained based on the PTR-MS data. The measurements suggested an average NTVOC carbon and oxygen number of about 7 and 1, respectively. Based on reported molecular speciation data (e.g. Kleindienst et al., 2007), we expect the products of C7 compounds to have a C5-C6 carbon backbone. These products were placed in set3 following a kernel function based on the distribution of

naphthalene oxidation products. At least two oxygen atoms were added to the NTVOC mixture upon their oxidation. The overall O:C ratio in the whole space roughly spans the range from 0.1 to 1.0. Multigeneration chemistry (aging) is also accounted for by the model. Unlike the 2D-VBS, the 1.5D-VBS does not use different kernel functions, to discretize the distribution of the oxidation products according to their $\log(C^*)$ and O:C ratios, when functionalization and fragmentation occur. Instead, to reduce the computational burden of the simulations, the model assumes that the oxidation of a given surrogate yields one other surrogate with lower volatility, higher oxygen number and lower carbon number. These properties should be considered as a weighted average of those relative to the complex mixture of compounds arising from functionalization and fragmentation processes. Accordingly, the 1.5D-VBS approach represents the functionalization and fragmentation processes effectively while reducing the parameter space and the computational burden. Gas-phase products in the semi-volatile range in set2 and set3, once formed, can further react with a rate constant of $4 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ as proposed by previous studies (Donahue et al., 2013; Grieshop et al., 2009; Robinson et al., 2007), further lowering the volatility of the products by one order of magnitude. This implies that for every additional oxidation step, the organic material receives around 0.5 oxygen atoms. However, we don't have a separate set to allocate oxidation products from biogenic precursors, and they follow the same oxidation pathways of biomass burning like aerosol as in the previous case (Ciarelli et al., 2016a), including aging. We are currently working on an updated version of CAMx that includes the separation of biogenic sources.

Minor Issues/Typos

1. Page 2, line 53: What do the authors mean by “higher volatility?” Are these IVOCs or VOCs? And do they mean that the products of these and the semivolatile precursors contributed 15 to 38%?

Higher volatility refers to IVOCs and VOCs. Only the products of IVOCs and VOCs contributed to 15 to 38%. We rephrase for clarification as below:

On the other hand, the oxidation products of higher volatility precursors (the sum of IVOCs and VOCs) contribute from 15 to 38% with no specific gradient among the stations.

2. Page 3, line 62: Consider replacing “qualitatively” with “nominally.” They are very similar for sure but while qualitatively to me suggests one knows a lot about the relative importance of each source (just not the actual numbers), nominal suggests you just know that the sources are there and you can name them. The latter to me is more representative of our knowledge of sources for SOA.

Done.

3. Page 3, lines 65-71: Please also mention aqueous-phase formation and the importance of solubility in water somewhere here to make the picture more complete.

We added the following sentence at line 70-75 of the revisited manuscript to mention the importance of aqueous-phase formation as below:

The physical and chemical processes leading to the formation of SOA are numerous, e.g. oxidation and condensation, oligomerization or aqueous-phase formation, and they are very uncertain and currently under debate (Hallquist et al., 2009; Tsigaridis et al., 2014; Fuzzi et al., 2015; Woody et al., 2016). Moreover, the solubility of organic compounds in water is also a crucial parameter affecting the life time of organic particles and gases in the atmosphere (Hodzic et al., 2016).

4. Page 3, line 82: Consider removing the word “common.” And refer to SOA explicitly here. For example: “Most CTMs today account for SOA formation from biogenic and anthropogenic...A few models also include SOA formation from intermediate volatility,,”.

Done.

5. I don't think you need a hyphen in "semi-volatile" anywhere in the text, but this is your preference.

Done.

6. Page 4, line 114-115: The higher volatility emission parameters were also constrained using monitoring network measurements in the previous modeling studies. Several studies have played with 1.5 factor for instance and it has remained as the parameter of choice despite uncertainties.

We thank the referee for this comment. We reformulated and shortened the sentence from line 116-119 of the revised manuscript as below:

*This implies that, in these applications, the new emitted organic mass (POA + SVOCs + IVOCs) is 7.5 times higher than in original emissions (i.e., $OM = (3*POA) + (1.5*(3*POA))$) which could be used as an indirect method to account for missing organic material in the absence of more detailed gridded emission inventories.*

7. Page 7, lines 193-199: I was confused by this group of sentences. Consider rewriting for clarity. Maybe something like, "We assumed OA emissions from SNAP2 (emissions from non-industrial combustion plants in the Selected Nomenclature for Air Pollution) and SNAP10 (emissions from agriculture, about 6% of POA in SNAP2), to be representative of biomass burning emissions and thus comparable to the BBOA PMF factor. OA from all other SNAP categories were compared against HOA-like PMF factors. Unfortunately, gridded emissions for SNAP2 include other emission sources (i.e., coal burning which might be important in eastern European countries like Poland). We could not resolve our emission inventory with sufficient detail to separate the contribution of coal for these European sites (Crippa et al., 2014)."

We agree and modified the sentence as suggested by the referee (line 205 in the revised manuscript).

8. Page 8, line 219: Please do not call it deposition "capacity" as this suggests something about the ability of the sea to hold pollution. Please reword. "Efficiency" might make more sense. Or just say "reduced deposition". Also change on page 9, line 267

Done.

9. Page 8, line 236: Please provide some statistic for this statement.

We added the statistics: ($R^2=0.72$).

10. Fig. 3: Consider adding error bars to this plot showing variability to make this figure more useful.

Done.

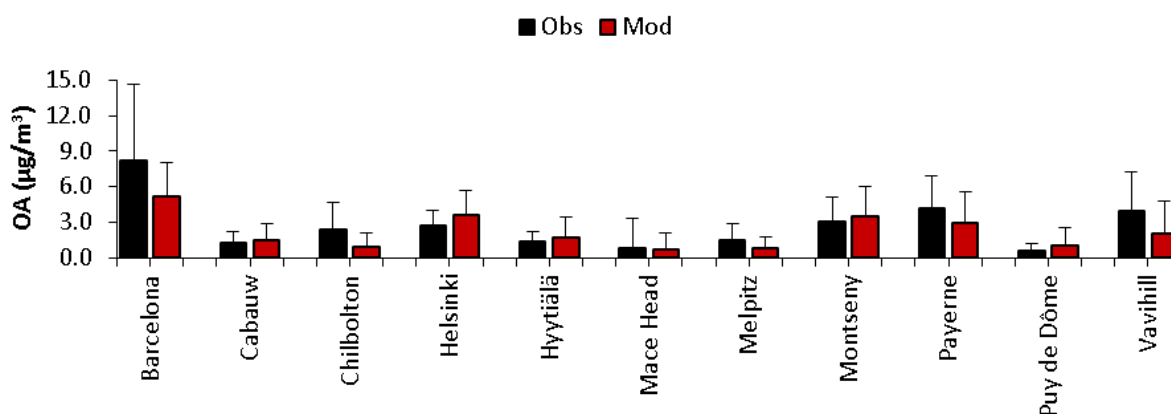


Figure 3. Observed (black) and modelled (VBS_BC_NEW) (red) average OA mass at AMS sites for the period between 25 February and 26 March 2009.

11. Page 9, lines 258-262: This sentence needs to be split into two sentences and reworded for clarity.

We reworded the sentences at line 274-278 of the revised manuscript as below:

Bergström et al. (2012) reported that emissions of organic carbon (OC) from the residential heating sector in Sweden were lower than those in Norway by a factor of 14 in spite of its higher wood usage by 60%. This indicates an underestimation of emissions from residential heating in the emission inventory.

12. Page 10, line 288-290: Do you have evidence from other PM species or pollutants to back up this claim?

We added a comparison for the modelled PM₂₅ components for Puy de Dome and Montseny. At both sites all the components were over-predicted (Fig. S3). We added the following sentence at line 304 of the revised manuscript.

as confirmed by the over-prediction of other PM species at these sites (Fig. S3).

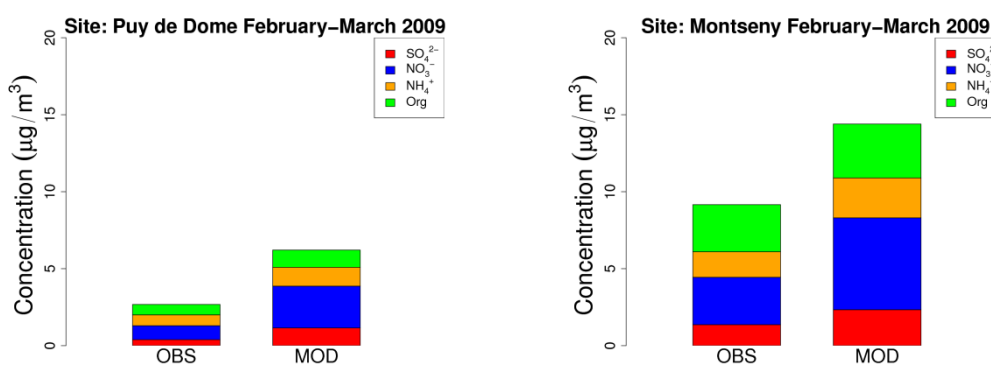


Figure S3. Comparison of modelled non-refractory PM₂₅ components at Puy de Dome and Montseny with the AMS measurements in February-March 2009.

13. Page 10, line 291-305: This sentence should be revised for clarity. The authors have blamed the meteorology and the host model configuration itself but why not the emissions? The activity data for the emissions could be wrong, or the emission factors could be wrong, no? Ok, CAMx has issues like any other CTM, but what makes the authors so sure that most of the problem is not in the emissions data?

We agree with the referee. Emissions might also represent a large source of uncertainties, recently, even more than previously thought. We added more emphasis on this point at line 311-315 of the revised manuscript as below:

In addition, the gridded emission inventories still represent a large source of uncertainties for CTM applications. The majority of the NO_x (NO+NO₂) emissions in Europe arises from the transportation sector (SNAP7), which might have much larger uncertainties than previously thought (Vaughan et al., 2016).

14. Page 10, line 296: course should be spelled coarse

We corrected the typo.

15. Page 10, line 308-315: The authors can also add here the potential double counting of SVOC emissions and the application of PM_{2.5} prediction to a (nominal) PM_{1.0} measurement

We modified the sentence from line 330 of the revised manuscript in order to include other reasons for the over prediction of the BBOA fraction as below:

- 4) The simplistic way of accounting for the semi-volatile part of primary emissions might lead, in some areas, to the double counting of such compounds.
 5) Uncertainties in the retrieved BBOA fraction from PMF analysis.

16. Page 11, line 316-318: How many of the peaks were captured well? What statistic determines how well they were captured? Unless this statement can be quantified, please remove it.

We removed the sentence.

17. Page 11, line 322: Please consider changing “likely” to “possibly.”

Done.

18. Figure 10. Please consider using median values in these plots rather than averages. 1) It will more effectively reduce the influence of extreme pollution days. 2) It will be more consistent with your use of percentiles. Consider also adding percentiles for the model run data.

Done.

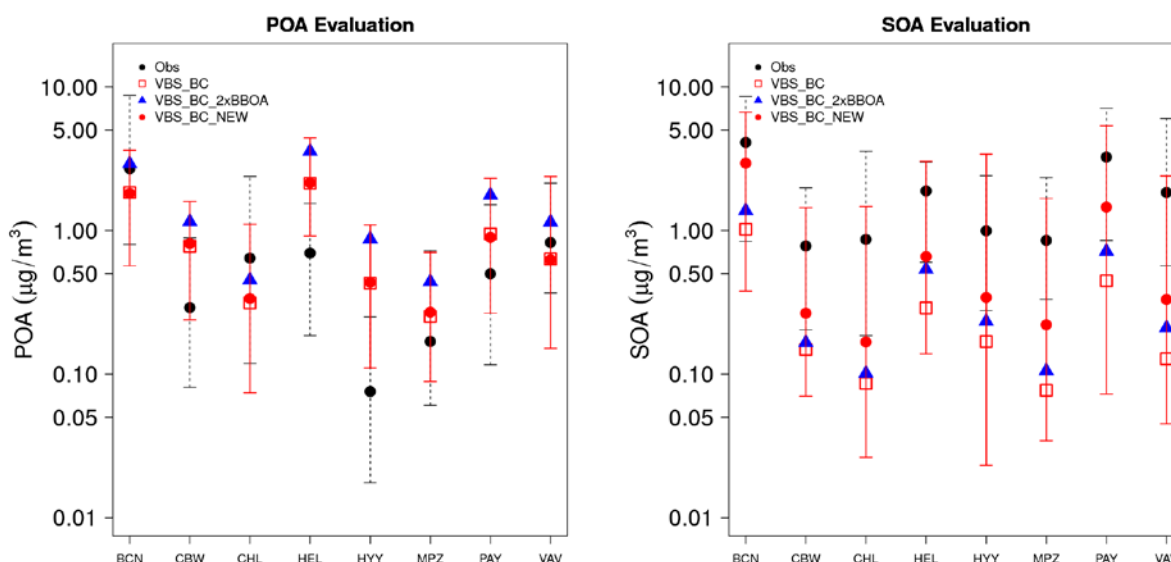


Figure 10. POA (left) and SOA (right) median concentrations at 8 AMS sites for February-March 2009 in the VBS_BC, VBS_BC_2xBBOA and VBS_BC_NEW cases. Dotted lines indicate the 10th and 90th quartile range (also reported in red for the VBS_BC_NEW case). Data for the Puy de Dôme and Montseny sites at higher layers are not available for the VBS_BC_2xBBOA scenario.

19. Figure 11: This data would be better represented as a bar plot since the x-axis is not really a continuum, even though you are trying to approximate one by ordering them south-north.

We agreed and replaced the Figure 11 with a bar plot.

20. Tables: please add one more significant figure to all data. I can't figure out why the mean biases are different than the differences in the mean model and mean obs. Is it a rounding issue?

The observed differences are indeed due to the rounding. However, we prefer to keep one significant digit in the tables.

21. Page 13, line 380-388: Please quantify “reasonably good.” Compared to what?

We modify the sentence at line 413 as below:

Predicted HOA concentrations were in the range of those retrieved from the PMF analysis.

22. Figure 11: Is BBOA actually just primary BBOA? Please make this clear in this figure and throughout the text as it gets confusing.

We thank the reviewer for this comment that was also addressed by referee #1. Yes, BBOA refers only to primary BBOA. We changed the legend in Figure 11 as presented in comment 19. We changed BBOA to BBPOA in the whole manuscript.

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