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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling winter organic aerosol at the European scale with CAMx: evaluation and source apportionment with a VBS parameterization based on novel wood burning smog chamber experiments" *by* Giancarlo Ciarelli et al.

Anonymous Referee #2

Received and published: 16 December 2016

General Comments:

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

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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 PM2.5 mass to PM1.0 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.

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.

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?

4. On page 5, lines 150-151, the authors point out that CAMx is predicting PM2.5. But the evaluation is against AMS observations which I presume are primarily PM1.0. Doesn't this fact make the frequent underprediction in SOA even more troubling? Is anything more specific known about the diameter of PM2.5 particles to allow the authors to estimate the fraction that would be PM1.0 and thus more applicable to the measurements?

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.

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.

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.

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?

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%?

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.

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.

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„,".

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

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.

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 cites (Crippa et al., 2014)."

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.

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

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

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

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

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?

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

15. Page 10, line 308-315: The authors can also add here the potential double-counting of SVOC emissions and the application of PM2.5 prediction to a (nominal) PM1.0 measurement.

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.

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

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.

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.

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?

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

22. Figure 11: Is BBOA actually just primary BBOA? Please make this clear in this

figure and throughout the text as it gets confusing.

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