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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simulating the formation of carbonaceous aerosol in a European Megacity (Paris) during the MEGAPOLI summer and winter campaigns" *by* C. Fountoukis et al.

Anonymous Referee #2

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The manuscript presents simulations of black carbon (BC) and organic aerosol (OA) components (e.g. POA, SOA, and cooking OA) from the PMCAMx model for Paris and compares these results against measurement taken at three ground sites during the MEGAPOLI summer and winter campaigns. It is found that the model provides reasonably good predictions of BC, with some discrepancies during the morning rush hour. In addition, model-measurement agreement is achieved for the summertime SOA concentrations. On the other hand, for the base case, there are significant differences between the model and the measurements for POA and for SOA during the wintertime.

The manuscript concludes that the substantial discrepancy in the POA concentrations

is due to the lack of cooking emissions in the base case. When a cooking emissions inventory based on field observations is implemented in the model, much better model-measurement agreement is found, which supports the importance of including this source category in chemical transport models. For SOA during the wintertime, the reason for the discrepancy is unclear, although it is speculated that missing SOA formation pathways or inaccurate biomass burning emissions may be responsible.

Overall this is an interesting manuscript that is well within the scope of ACP, and the work certainly has the potential to be of high quality. However, there are a number of points in the manuscript where the inclusion of additional data, information, or sensitivity studies is needed and the current discussion lacks sufficient depth. This additional work will need to be included before final publication. The terminology used in the manuscript should be clarified as well, as discussed in the general comment below.

General Comment:

If I understand correctly, the authors are using the term "anthropogenic SOA" to refer to SOA formed from anthropogenic VOCs. This makes the manuscript confusing, since one could have anthropogenic SOA formed from SVOCs and IVOCs as well. This confusion is particularly problematic in the discussion of aging in Section 2 as well as in the conclusions. In Section 2, does the rate constant of 1 x 10ˆ-11 cmˆ3 molecˆ-1 sˆ-1 apply to anthropogenic SOA from only VOCs or to all anthropogenic SOA including SOA-iv and SOA-sv? In the conclusions, the authors state that 13% of summertime SOA "consists of anthopogenic SOA". This is a very dangerous statement as it gives the reader the impression that 87% of SOA is biogenic. I believe the correct conclusion is that 87% of summertime SOA comes from biogenic VOCs or primary SVOCs and IVOCs that are either biogenic or anthropogenic.

There is a similar problem with the alternating use of HOA and POA in the manuscript. Is there a difference between "predicted HOA" and "predicted POA"? This distinction is important because in older work HOA was used as a term to identify a product from component analysis of AMS data that was strongly associated with POA. However, with the improvement of AMS and PMF analysis, HOA has morphed into a quantity that is no longer equivalent to total POA, but instead it is more associated with only the vehicular component of POA. In the specific comments below some instances of this problem are noted. I recommend that the authors use terms such as "predicted total POA" and "predicted vehicular POA" rather than "predicted HOA" to avoid confusion.

Specific Comments:

Page 25551, Lines 3 - 6: I realize this sentence is not based on the authors' own work, but it would be helpful if "larger geographic area" was better defined. Would this larger area be continental-scale versus local/city-scale or something else?

Pages 25554 – 25555, Lines 23 – 7: This paragraph and the discussion of the percentages of OA and BC from various sources should be summarized in a table. Currently, the paragraph is difficult to read and it's hard to compare the different percentages, which would be of interest.

Page 25556, Lines: Lines 26 – 28: Additional information should be provided regarding the instruments used to measure black carbon. For example, what wavelengths were used for the absorption measurement, what are the instrument model numbers, what was the absorption coefficient used to determine the BC concentration, and were possible artefacts such as shadowing corrected? This information is critical for evaluating the model/measurement comparisons with respect to BC and needs to be included in the manuscript directly or via the appropriate references. Similarly, an uncertainty for the BC measurement should be reported in Figure 6.

Page 25557, Line 23: Is there an explanation for why a west to east gradient is predicted?

Page 25557, Line 24: The terminology is confusing here. It seems like "OOA" is being used interchangeably with "SOA" in this paragraph. These aren't exactly the same

thing – OOA is used to identify a component from factor analysis. Practically there is little difference, but only one name should be used, unless the authors are trying to distinguish between two different predicted quantities. This comment applies to the panel labels in Figure 2 as well.

Page 25558, Line 20: Similar to the previous comment, the previous two paragraphs discuss POA concentration predictions by PMCAMx, and starting with this line PMCAMx predictions of HOA are described. Is this really a different quantity in the model? As the authors already mentioned, the baseline emissions inventory used in this work does not include cooking, so that means HOA and POA are the same quantity in the model. For the purpose of clarity, it is critical that the same name is used for the same quantity predicted by the model. Again, phrases such as "the model predicts low concentrations of HOA" are problematic since HOA is a term that is specific to factor analysis, whereas terms such as "vehicular POA" would be more accurate for describing model output.

Page 25559, Lines 25 – 29: The authors should provide the prediction skill metrics of PMCAMx for BBOA in table format, similar to what has already been provided in the supporting information for HOA.

Section 5.3: I agree with the first referee that the discussion of OOA in this section seems incomplete. An important shortcoming in the model predictions has been identified, but then there is no rigorous follow-up such as sensitivity studies. The article is not particularly long, so there seems to be a missed opportunity to explore the origin of this discrepancy. Since it is stated in the manuscript that there are large uncertainties in BBOA emissions, could the authors run a sensitivity study where the emissions of BBOA and the associated SVOCs and IVOCs are increased or modified in some other fashion? Alternatively, could a different parameterization be used for the formation of OBBOA?

Supporting information, S3: All the figures showing model-measurements comparisons are diurnal averages except for this figure. In order to facilitate comparison the comparison of BBOA should be shown as a diurnal average as well.

Page 25561, Lines 19 – 21: Wouldn't the SOA-iv concentrations also be underestimated and not just the SOA-sv concentrations? Based on the model description, it seems that there would be primary IVOCs emitted with the BBOA that has SOA forming potential.

Page 25562 – 25563, Line 27 – 9: The discussion in this paragraph of the possible reasons for the BC model-measurements discrepancy should be expanded; otherwise the conclusions are too weak. Firstly, the variability of the BC and mixing height measurements during the two campaigns needs to be presented in some fashion in the manuscript. (In fact, it seems that mixing height data is not shown anywhere in the manuscript.) For example, time series for the model and measurement results could be given in the supporting information, or the diurnal plots could use a box-and-whiskers format. Presenting only a diurnal average of the BC concentration and then mentioning only in the text the mixing layer heights for three specific days out of the entire campaign period is not sufficient for evaluating why the model has difficulty reproducing the BC concentration during the morning.

In addition, it would be a simple sensitivity study to correct the predicted BC concentration for the underestimated mixing height using the LIDAR observations. I agree that there is a significant uncertainty in the observations, but such a comparison would still be interesting. If the corrected model prediction of BC still does not match the observation, despite a potential positive bias of the LIDAR, then that would strongly indicate that there are other reasons for the model-measurement discrepancy besides an inaccurate representation of the mixing layer height. (In other words a positive LIDAR bias would lead to an over correction of the model, which is currently overestimating the BC measurement.)

Section 5.5: Given that the inclusion of cooking emissions substantially improves the

model predictions, the authors should summarize the prediction skill metrics of PM-CAMx for this sensitivity study in a table. In other words, create a third table that is analogous to Table 2, but for the results with cooking.

Page 25563, Lines 19 – 20: What was the temporal profile of the added cooking emissions during the winter period? Was it the same as during the summer period? If not, why is the temporal profile different?

Page 25563, Lines 24 – 26: The manuscript should also include a comparison of the modeled and measured COA for the SIRTA site. As described in Section 4, a COA factor was identified at the SIRTA site for both summertime and wintertime. So, it is not clear why this comparison is shown currently in the manuscript for only LHVP. This omission is conspicuous.

Figure 7: Similar to a previous comment, showing only the diurnal average of the COA measurement does not give the reader sufficient information to interpret the results. A box-and-whisker plot would be strongly preferable or the corresponding time series should be included in the supporting information.

Page 25564, Lines 11 – 13: This sentence is confusing and its grammar/syntax should be verified. If the cooking OA can undergo aging in the model, does that mean cooking SOA is formed? Is the cooking OA assumed to be semi-volatile? Are IVOCs emitted with the cooking OA similar to other POA sources? More information is needed for a reader to evaluate this sensitivity test. While reading the previous paragraph, one is given the impression that the cooking OA is inert, but now that seems to not be the case.

Pages 25565 – 25566, Lines 25 – 37: How much is the contribution of COA to the total OA during summertime? It seems like the importance of COA for the total OA would be much smaller than the 70% figure given for the fraction of POA contributed by cooking.

Technical Comments:

Introduction: At several points in the text the term “Megacities” is capitalized, but it seems that lowercase should be used as this word is just an ordinary noun (e.g. Cities versus cities).

Page 25551, Line 14: air massES

Page 25553, Line 18: generation reactionS

Page 25556, Line 6: It appears that the acronym “GOLF” is not defined.

Page 25560, Lines 5 – 6: The acronyms SOA-iv and SOA-sv have already been defined.

Page 25560, Line 11: Should this be aSOA-v?

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 25547, 2015.