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Interactive Comment

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.

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The manuscript shows simulations of black carbon (BC) and organic aerosols (OA) for the Paris metropolitan area for summer and winter periods during the MEGAPOLI field experiment. The main points are: 1) Primary organic aerosol BC are generally well modeled, 2) OA emissions from cooking are developed based on observed data and improve model performance, 3) Secondary OA in summer is well modeled, and 4) Secondary OA in winter is completely underestimated with the current SOA mechanism. The manuscript is well written, is of good quality and has good potential to be published after major changes. Please see my comments below.

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Interactive Discussion

Discussion Paper

General comments

(1) *The explanation of why OOA in winter is underpredicted is not clear, too convoluted, and leaves more questions than answers. I think the authors should work more on it to make this article publishable. They should at least identify where these air masses come from when the extreme underestimation is found. Is this a problem with background concentrations? Maybe boundary conditions are to blame? Does it have to do with residence time over continental regions before reaching Paris? There are still some times where the model performs well, so the authors could also identify when and why this happens to provide better insight into the issue. They also propose a mechanism which could solve this problem, why not test it? This should be relatively simple given the expertise of the authors.*

We have followed the suggestion of the reviewer and performed additional analysis of the observed and predicted wintertime OOA concentrations. This analysis shows that the OOA underprediction is persistent throughout the simulation period (new Figure S5). However, there are certain days (24 and 27 January and 4 and 7 February) during which the analysis of the AMS data suggests very high (more than 6 μg m^{-3}) OOA levels while PMCAMx predicts moderate levels (around 2 μg m^{-3}). Back-trajectory analysis (also added in the supplement as Figure S6) indicates that during these days the air masses arriving in Paris have all continental origin but are coming from a variety of areas (central France, Germany, Belgium, etc.). On the other hand, during the days with reasonable model performance the air masses were mostly clean coming from the Atlantic, the United Kingdom, Ireland, etc.). This further supports our hypothesis in the manuscript of rapid conversion of anthropogenic emissions to OOA during winter.

We have performed a number of sensitivity tests (including changes in boundary conditions) but we could not reproduce these high observed OOA levels in the Paris area without increasing dramatically at the same time the OOA over the rest of Europe. It should be noted that the same model did not show any serious underprediction of OOA over Europe in other sites (Fountoukis et al. 2014b). For example it did not show any

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bias in Cabauw in the Netherlands. Even more, the more than 5 μg m^{-3} of OOA observed during several days is a lot higher than the total OA measured in most sites in Europe. None of the known mechanisms that have been tested in previous applications of PMCAMx (Tsimpidi et al., 2010, Fountoukis et al., 2011, 2014b) explain these very high levels. So at this stage we think that it is important to report this significant discrepancy between observations and predictions thus encouraging future work in this direction. We have added discussion of these issues and two new figures in the supplementary information to address this important issue.

(2) *The analysis performed is mainly for the model representation of diurnal cycles and average concentrations. I think what's missing is how well the model represents the day to day variability. Are the biases found persistent throughout the periods or occur only for exception events? If time series for the whole period are too saturated with data, the authors could plot the time series of daily means or daily distributions (with box and whisker plots). Try to include these plots as additional panels in figures already existent when possible. Please add this analysis for all species and seasons, especially for SOA (OOA) as it would be instructive to see the model representation of these regional events.*

To address this point we have added new figure and revised existing ones in both the main paper and the supplementary information to show the corresponding day-to-day variability. During summer, the POA underprediction seems to be mostly systematic and persistent throughout the simulation period, while the performance for OOA is encouraging for almost all days (with the exception of 21 July). In winter OOA is underpredicted systematically during the majority of the days with some days showing much larger biases than others as discussed in our reply to Comment 1 above. The discrepancies for the POA concentrations during winter show up partly as scatter rather than bias as explained in the text due to discrepancies in the different POA components (i.e. BBOA, COA, HOA, etc.). The comparison for the daily mean concentrations of BC shows encouraging model performance during the summer and an overprediction during several days in winter. This is mostly due to an overprediction of the morning rush hour peak that is illustrated through the average diurnal profiles shown in Fig. 6. We would like to point out that the reproduction of the observed average diurnal variation is an important test of the ability of the model to reproduce observations for the right reasons and unfortunately few model evaluation exercises use this approach. We have also added discussion of the ability of the model to reproduce the day to day variation of the corresponding concentrations.

(3) *The diurnal profile plots (Figs 6,7, S1) provide information only on the mean. The authors could redo these plots as box and whiskers plots, so besides the mean, it could show the spread of the distributions to see how well the model is able to capture it. This could be helpful when trying to explain observation and model discrepancies on the mean throughout the text.*

We have revised these figures following the reviewer's suggestion. The revised figures now show the median as well as the spread (25th and 75th percentiles) of the distributions. We would rather exclude the whiskers (min and max values) as the figures are already busy with modeled and observed values on the same plots.

Comments by line. In the following I'm only including the last 2 digits of the page numbers.

(4) *Section 2,3. What did the authors used for boundary conditions for all species? If they used climatological profiles they could try to use boundary conditions from global models (e.g., MACC reanalysis) and making assumptions on the splitting of OA to see if this helps with the biases found later in the text.*

Concentrations of species at the boundaries of the domain are based on measured average background concentrations in sites close to the boundaries of the domain (Zhang et al., 2007; Seinfeld and Pandis 2006). We have used the same boundary conditions as in Fountoukis et al. (2011). We have now added this information in

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the revised manuscript. Most global models have serious problems reproducing the regional OA concentration levels so it is not clear that their use in this study would be helpful or if it would further complicate the analysis by adding one more potential source of bias.

(5) *Page 53, Line 27. What is the WRF configuration? Or reference where this is stated. What global meteorological conditions are used to force WRF?*

WRF was driven by static geographical data and dynamic meteorological data (near real-time and historical data generated by the Global Forecast System (1 $\times$1 degrees)). 27 sigma-p layers up to 0.1 bars were used in the vertical dimension. Each layer of PMCAMx is aligned with the layers used in WRF. The WRF runs were periodically (every 3 days) reinitialized to ensure accuracy in the corresponding fields that are used as inputs in PMCAMx. We have added this information in the revised manuscript.

(6) *Page 56, lines 20-26. This is confusing; maybe it would be better presented in a table with the components by site and season.*

This information is now included in the new Table 2.

(7) *Page 57, lines 18-20. Did you conclude this just by looking at the emissions or by also looking at the modeled concentrations?*

This is derived from both the emissions and the results of the source apportionment model (PSAT). This is now explained in the revised manuscript.

(8) *Page 57, lines 23. Why there is a west to east gradient predicted during the summer? Identify source regions.*

A west to east gradient is predicted during summer due to the regional source distribution and the corresponding evolution of photochemistry. We have added this explana-

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tion in the revised manuscript.

(9) *Page 57, lines 24-25. By looking at Fig 2, it looks the other way around for winter, POA seems to dominate for this season.*

We thank the reviewer for pointing this out. We have now corrected this into: "OOA is predicted to account for approximately 90 percent of PM_1 OA at ground level over the Paris greater area (domain-average) during summer and 50 percent during winter." The original sentence referred to the whole European domain.

(10) *Page 58, Line 19. State that you will tackle this problem later in the text, as it reads like you found the problem but did nothing to correct it, which is not the case.*

We added the corresponding statement.

(11) *Page 58 Line 27-54. This paragraph could be improved by adding more analysis, not by just listing possible reasons for the discrepancy. For instance, you mention wind speed as a possible reason, so you could evaluate the model wind speed against observations specifically for the morning and for this site (only overall evaluation is done). Another reason could be that the diurnal cycle of traffic emissions is too sharp, as you also see overestimation in morning BC concentrations. Also, could other sources of HOA that you are not considering in your model exist?*

The evaluation of the WRF predictions (including the wind speed) along with the relevant discussion is in the following paragraph as well as in Section 5.4 (mixing height). As stated in the text, no systematic errors were found for the wind velocity. To avoid confusing the reader here we have deleted this sentence listing the wind speed as a possible source of error since the relevant analysis is presented in the next paragraph. Errors related to diurnal cycle of emissions or other sources of HOA, were meant to be inherent in the phrase "emission rate errors". However, to avoid misunderstandings we have expanded the text to include these as well.

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(12) *Fig S1. What about the 6 am peak in winter not represented by the model? Is this persistent throughout the days or episodic? What about the nocturnal biases?*

Fig. S1 shows only summertime results. The time series analysis indicates that both the 6 am peak and the nocturnal bias in SIRTA during summer are episodic. There are two days (4 and 11 July) with a large vehicular-POA underprediction (by more than a factor of 5) at 10 pm and two other days (21 and 28 July) with a similar (a factor of 3-4) underprediction at 6 am. This shows up as an average bias in Figure S1 during these two times which, in fact, is not systematic. Furthermore in the revised Figure S1 (Fig. S2 now) which shows the median rather than the average, the 6 am peak is well represented by the model. We have added text in the revised paper discussing the above issues.

(13) *Page 59, Line 8-13. Do you find any bias in POA or OOA for the days that the model fails to predict the temperature? You could include this discussion if you add time series of OA components.*

For the specific times of the day and specific days during which WRF has the highest temperature errors we do not see any correlation with the POA or OOA bias. We have added this information in the revised text.

(14) *Page 59, Line 28. Authors argue a problem in the spatial distribution of BB emissions. How were these emissions distributed? By population only? It is expected that sub-urban or rural homes use more wood-burning for heating than urban homes. Was this taken into account when distributing? If not, can you re-distribute the emissions using this criteria and see if you get an improvement?*

In the MEGAPOLI emission inventory used in this work, BBOA emissions are distributed not only by population but also by taking into account the rural/urban areas as mentioned by the reviewer. In this paragraph, however, we argue that this approach might still include errors.

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(15) *Page 61, Lines 14-5. This paragraph is hard to follow. First you blame remote sources, but then you say that this shouldn't be the reason as you found in your previous study. But then at the end of the paragraph you go back to point to remote sources (BBOA). Please make it clearer.*

We have added text and rephrased this paragraph to make it clearer to the reader. The main point here is that errors in remote sources (upwind of Paris) could only partly explain the OOA underprediction. There seems to be another reason as well.

(16) *Page 62, Line 19. The minima of the average diurnal cycle are not the background values. Background values cannot be extracted from means as polluted and background conditions are averaged. You can use box and whisker plots and compare the lower end of the modeled and observed distributions to get at how well the model represents background values.*

We have rephrased that part and also added the spread (25th and 75th percentiles) of the modeled and measured concentration distributions.

(17) *Page 62, Lines 27-3. I think you should focus this analysis to the morning rise of the boundary layer rather than to the daily peaks, as is in the morning when you have the model misrepresentation. Compared to the observations, is the model able to capture the timing of the rise of the BL? If it's too slow then this would be a good explanation of what's happening. Maybe a plot of the derivative in time of the BL (maybe the diurnal cycle of it) could help. This is an important issue for primary aerosols representation which seems to be consistent across species, so you should dedicate a figure to it, at least in the supplement.*

We thank the reviewer for pointing this out. We have added a figure in the supplement (Figure S7) with the diurnal cycle of the PBL height for both summer and winter campaigns. It seems that there is an underprediction of the morning rise of the boundary

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layer in SIRTA that could explain part of the BC underprediction. We have added this explanation in the revised manuscript.

(18) *Page 63, Line 13. Why cooking emissions in summer are x2 in winter? Barbecues? Do you see variations between weekdays and weekends? Please elaborate.*

This issue needs additional clarification. The primary OA emissions during winter were increased by a factor of 1.5 (compared to a factor of 3 in summer) because the original primary OA wintertime emissions were higher (in absolute values) than the summertime ones. As stated in the text, the total (absolute) OC emissions that were added to account for the missing cooking OA were 5.3 t/d for the summer and 5.1 t/d for the winter period. This rather small seasonal difference could indeed be due to summertime barbecues.

We did observe a variation of COA emissions during weekdays/weekends. Based on observed COA concentrations, the added weekend COA emissions were higher compared to the weekday emissions. Approximately 18.5 percent of total weekly COA was emitted during each weekend day and 12.5 percent on each weekday. We have added this in the revised text.

(19) *Page 64, Line 2. Explain why this happens.*

This is due to a well-mixed layer and strong vertical mixing during the day. We have added this in the text.

(20) *Section 5.5. Show and discuss scatter-plot for POA in after adding cooking emissions for both seasons.*

We have added the scatter-plot for POA including now the COA emissions (new Figure 7) as well as a new table (Table 4) showing the statistics of this comparison for both seasons and for both POA and OOA. Text has been added to discuss this.

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Technical Corrections

(21) *Page 52, line 13. "fine" grid resolution.*

Corrected.

(22) *Page 53, line 21. Replace by advection and dispersion by transport.*

Replaced.

(23) *Page 62, Lines 6-7. This is statement cannot be deduced from Table 2. This probably should be Fig 5.*

Corrected.

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

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