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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 #1

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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.

General comments

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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.

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.

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

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

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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.

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?

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

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

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

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.

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.

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 might exist?

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

13) Page 59, Line 8-13. Do you find any bias in POA or OOA for the days that the

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model fails to predict the temperature? You could include this discussion if you add time series of OA components.

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?

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

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

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.

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.

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

20) Section 5.5. Show and discuss scatterplot for POA in after adding cooking emissions for both seasons

Technical Corrections

21) Page 52, line 13. “fine” grid resolution

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

23) Page 62, Lines 6-7. This is statement cannot be deduced from Table 2. This probably should be Fig5

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

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