

***Interactive comment on* “Formation of organic aerosol in the Paris region during the MEGAPOLI summer campaign: evaluation of the Volatility-Basis-Set approach within the CHIMERE model” by Q. J. Zhang et al.**

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This paper is an interesting study of the Paris region making good use of measurements and PMF analysis from the MEGAPOLI campaign. The paper is well written, and apart from some suggestions as given below, is suitable for publication in ACP.

We sincerely thank the referee for his comments and suggestions. They certainly improved the paper.

Major comments:

1. On page 29482, the authors point out that they use only the PMF solution from Freutel et al.. Explain why this choice was made, and any possible consequences of other choices.

Reply: The PMF solution from Freutel et al. 2012 was applied for the AMS measurement at all the three measurement sites, while the one from Crippa et al. 2012 was applied only to the high resolution AMS data obtained at the peri-urban SIRTA site at the SW edge of the agglomeration. In addition, the PMF solution from Freutel et al. 2012 was the only available when the paper was written. It yields the three factors HOA (Hydrocarbon like OA), OOA (Oxygenized OA) and COA (OA from cooking activities). The PMF solution from Crippa and al., contains five factors: HOA, LV-OOA (Low Volatile Oxygenated OA), SV-OOA (Semi-Volatile Oxygenated OA), Marine OA and COA. Both solutions are broadly consistent, if one considers that the factors LV-OOA, SV-OOA and MOA in the five factor solution correspond to OOA in the three factor solution.

The sentences on p29482, line13-16 are modified to:

“Source apportionment from positive matrix factorization (PMF) analysis was applied to study the origins and the potential sources of OA (Freutel et al., 2012; Crippa et al., 2012b for the SIRTA site). In this paper, focusing on the urban LHVP site, only the solution from Freutel et al., 2012 is used.”

2. In general, the paper does a good job of considering many of the uncertainties involved in OA modelling. I miss mainly some details of the emissions.

Firstly, a small discussion of the accuracy of the biogenic emissions is needed. The model uses MEGAN. A recent paper by Langner et al. has shown factor 5 differences between BVOC emissions in different European models, with MEGAN giving by far the highest estimates. This is an obvious source of uncertainty in BSOA predictions, and warrants some words. (I also wonder if a lower BVOC emission and higher ageing rate would give similar conclusions.)

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Reply: We agree that uncertainties in BVOC emissions are large. The predicted BVOC concentrations, including isoprene and terpenes were compared with the observed ones during the MEGAPOLI campaign, giving good agreement (see also response to referee 1). Even if these measurements are only locally representative, this gives some more confidence in the BVOC emissions used for our study. In this paper, the chemical aging rates of SVOC's originating from BVOC oxidation in VBS-T2 and VBS-MPOLI simulations are on the higher end of values used in previous VBS studies (Murphy and Pandis, 2009; Fountoukis et al., 2011, etc.) was applied. Putting still higher aging rates would probably not be realistic.

On p29502, line 28, the sentence will be modified as and a figure will be added (Fig. S6):

“This could be related to the uncertainties in BVOC emissions in the MEGAN inventory, which often gives the highest biogenic emissions among European models (Langner et al. 2012) at a continental scale, though the predicted and observed biogenic VOC concentrations give good agreement in the Greater Paris region during the campaign (Fig. S6). “

Reference to be added:

Langner, J. et al., A multi-model study of impacts of climate change on surface ozone in Europe Atmos. Chem. Physics, 2012, 12, 10423-10440

3. In Section 3.2 I wonder about the consistency of the emissions for inorganics versus organic emissions. The emission factors are from different sources, but are the spatial distributions consistent? How does the MEGAPOLI database compare to the standard (LA) one, also even outside Paris? Some maps of the emissions in Paris would be helpful (replacing Fig. 1 say) to clarify such things.

Reply: The spatial distributions are not consistent between the LA and EMEP inventory, as both are independent. The POA emissions from the LA inventory are a factor of

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three higher than those from the MEGAPOLI database in the Paris region, while they are lower by about a factor of three around the Paris region as indicated on p29500, line6 and in a new figure (Fig. S8).

Here in section 3.2, we add a sentence on p29485, line20:

“POA emissions from the LA inventory are about three times higher in the Paris region as those from the MPOLI one (Fig. S8), while they are lower around.”

4. On p29486, last paragraph, the emissions story is also rather confusing. What is the relation between the PROCARB diesel emissions and assumptions, and the factor 1.5 mentioned above. Here the authors mention some other fraction (75%).

Reply: The PROCARB diesel emissions and assumptions referred to the French diesel emissions factor measurement which is similar to the one in the Robinson et al. 2007's study. Thus, the results from Robinson's study are considered to be applicable to the European case (Bergstrom et al., 2012). The factor 1.5 is the ratio of IVOC/SVOC emissions in Robinson's study. The factor of 75% indicates that following Robinson et al. (2007), 75% of SVOC emissions have been measured as POA under the experimental conditions of the PROCARB study (OA loading of about $1000 \mu\text{g m}^{-3}$). Thus, theoretically, one could correct the POA emissions by a factor of 1.33 to obtain the total SVOC emissions. However, this factor is not applied here, as it is uncertain in itself, and probably smaller than the general uncertainty of POA emissions.

On p29486, line21, the sentence is modified to:

“The emission factors for calculating diesel exhaust related POA emissions for both the LA and MPOLI inventories are obtained from laboratory measurements under a low level of dilution with high OA loading (of the order of $1000 \mu\text{g m}^{-3}$ (PROPCARB, 2010)), similar to that described in Robinson et al. (2007). Under such conditions, about 75% of SVOC emissions are expected to occur in the particle phase according to dilution experiments described in Robinson et al. (2007).”

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5. Finally, I wonder how consistent these Junker and Lousse based emissions are with the EMEP emissions used for all else. A common problem is that bottom-up estimates of POA emissions may be larger than estimates of PM_{2.5} emissions. Some words on this for the Paris area would be useful to set the results in the context of other European studies.

Reply: We haven't use EMEP based aerosol emissions for this study. POA emissions (see Fig. S8) in the LA inventory are much higher for Paris by a factor of three than the MEGAPOLI inventory database.

Minor comments:

Abstract, p29477 line 18, rephrase sentence with "model mostly after long range"

Reply: The sentence was rephrased to :

"High concentration events observed mostly after long range transport are well reproduced by the model."

p29479, line 16. POA "used to be" generally considered...

Reply: OK. It was done.

p29479, line 17. One of the Donahue or Robinson references would probably be appropriate alongside Pankow here.

Reply: These references have been added.

p29480, line 3. I don't know why the author say "especially" here, what do the latter papers have to do with the framework. They are rather updates aren't they?

Reply: OK," especially" was deleted.

p29480, line 14. "derived from" instead of "applying"

Reply: OK. It was done.

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p29480, line 18. "Better agreement", compared to what?

Reply: "than for the traditional SOA formation scheme"

p29482, line 10. Say "incoming" rather than "encountered"

Reply: OK. It was done.

p29486. Does Seinfeld and Pandis give much data on Atlantic OA for boundary conditions? What is meant by climatological analysis - whose, where?

Reply: The reference cited in Seinfeld and Pandis is

"Cachier H., Buat-Ménard P., Fontugne M., Chesselet R., Long-range transport of continentally-derived particulate carbon in the marine atmosphere: evidence from stable carbon isotope studies, Tellus Series B-chemical and Physical Meteorology - Tellus B-Chem Phys Meteorol , vol. 38, no. 3-4, pp. 161-177, 1986"

It has been added to the reference list.

p29486, line 1. The 1.5 fraction includes SVOC as well as IVOC.

Reply: According to previous studies (Robinsons et al. (2007), Shrivastava et al. (2008), Murphy and Pandis, 2009; Hodzic et al., 2010; Tsimpidi et al., 2010) to which we make reference in our study, the additional IVOC emissions are 1.5 times to the POA emissions. The sentence will be changed to:

"The additional gas phase IVOC..."

p29486, The PROCARB reference has no address, and the title is given in French. Add address and translation, and some words on what this is for English-only readers.

Pillot D., Darmon A., Besombes A., Cazier JL., Hanoune F., Vidon R., Perret P., Tassel P., 2009. Émissions de polluants de 10 véhicules légers alimentés en biocarburants à différentes teneurs et GNV - Rapport final. Rapport INRETS- LTE N° 0905 INRETS, Bron, France. 89 p, editeurs André M., Joumard R., Clairet JM., 2008. (translation

in English : Pollutant emissions of ten light vehicles working with different fractions of biofuels and natural gas.) <http://www.inrets.fr/ur/lte/publications/publipollu.html>

p29489, line 20. "appropriate"

Reply: OK. It was done.

p29489, line 27. To put the different VBS studies in context, it could be mentioned that other studies (e.g. Lane et al, 2008) use a higher ageing rate.

Reply: OK, A sentence is added on p29489 line 22:

“Other studies (e.g. Lane et al 2008a) use a higher aging rate which didn't improve model performance.”

p29490, item 2. Again, I think SVOC as well as IVOC is included in the factor 1.5 POA.

Reply: Item. The sentence will be changed to:

“The additional gas phase IVOC...”

p29492, lines 13-14. I would have said that the peak temperatures were underestimated, not "well represented".

Reply: OK. The sentence will be changed to

“For the three observed periods with enhanced temperatures (R1, R2 and R3 as described in Sect. 2) over 300 K, simulations show a negative bias.”

p29493, line 5. Say "is reduced" rather than "gets lower".

Reply: OK.

p29494, lines 10-11. No need for 3 significant figures in bias numbers.

Reply: OK, the significant numbers of biases have been reduced to 2.

p29495, lines 12 on. Say just "relative biases", not "biases (relative biases)".

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Reply: In fact, here the related biases are presented within the parentheses, along with biases. The sentence will be changed to:

“The biases (and relative biases) for comparison . . .”

p29495, line 14. Say "larger", not "more important".

Reply: OK.

p29497. What about coarse nitrate?

Reply: No coarse nitrate is taken into account in the simulations as the seasalt and dust aerosol are considered as inert in the model.

p29497. I am not convinced by the arguments concerning sulphate, OH and RO₂.

Firstly, if SO₄ is a result of long-range transport, problems in reproducing radicals at an end-point site inside a mega-city are probably irrelevant. Also, after long range transport I would expect all sulphur to be as sulphate, regardless of moderate uncertainty in chemical formation rates.

Reply: In order to take into account the referee’s concern, we will change “overestimated OH and RO₂ levels” to “the uncertainties related to the formation process”

p29497, last sentence. Why mention just the financial crisis? Surely emission controls, traffic growth, and other factors cause emissions for 2009 to be different to those of 2005.

Reply: OK , instead of specifically pointing to the financial crisis, we make a more general statement about uncertainties in emissions.

p29498, line 6. Say "the CSS method", and remind the reader where to find this explained.

Reply: OK. The sentence will be changed to:

“OA simulated in CSS (see section 3.5) overestimated the morning peaks by about a

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factor of two.“

p29501, lines 20-24. Some of the problems with the morning peaks are probably due to dispersion, as evidenced from the comparisons for NO_x and BC. Some mention of the implications of this is warranted I think.

Reply: We added an argument on dispersion on page 29501 line2,

“...VBS-T1 and VBS-T2 simulations, possibly also due to a problem in dispersion as probable also for for NO_x and BC”

p29504, line 13. Say model evaluation, not control.

Reply: OK. This was done.

Tables:

Table 1. Give full name of sites.

Reply: OK

Table 3. I found this a little confusing. The VBS approach etc should be clearly underlined as being headers. Make it explicit what blanks mean. (e.g. not included, or not available.)

Reply: We made the suggested changes in Table 3.

Figures:

Fig. 2 on, the x-axis for the month-plot is presumably day of month, rather than local time? State which month in the caption. What is white "fond"?

Reply: OK, The x-axis will be indicated by “date (local time)” as for other figures; “background” instead of “fond”

Fig. 3 on, make it clear if the data are hourly or daily averages.

Reply: The data are hourly averages. This is made clear in the text.

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Supplementary:

Table S1. Cite source for mass-distributions used here.

Reply: A reference to the paper Freutel et al. 2012 is added in the manuscript.

Table S3. Make it clear that these are mass-yields.

Reply: This is made clear now in the manuscript.

Table S4. Explain "R" and "RMSE".

Reply: R is the correlation coefficient; RMSE is root mean square error. This is now made clear in the paper.

Table S4, on. Give dates of comparisons, and number of samples.

Reply: It is 31 days of comparison with hourly data (corresponding to the frequency of the model output. This information is added in the Table 4 legend.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 29475, 2012.

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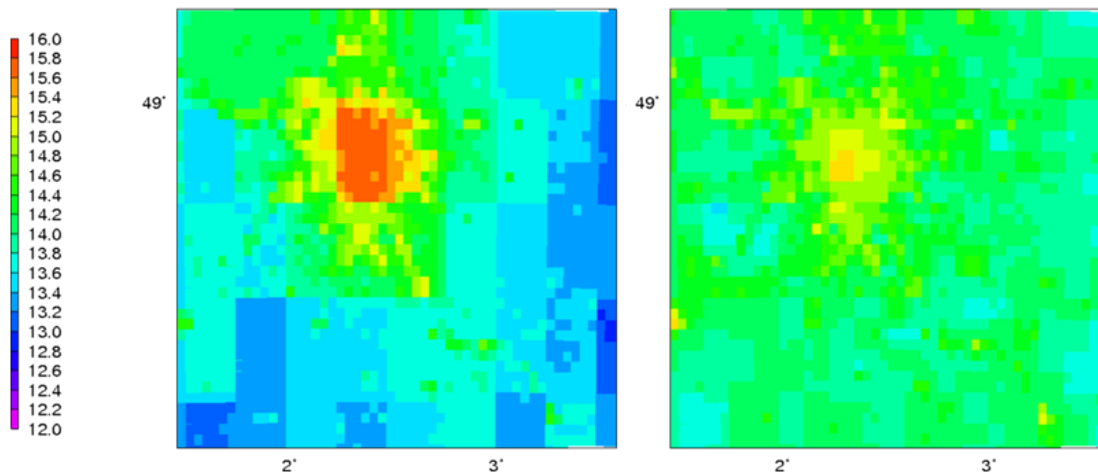


Fig. 1. Fig. S8. $\log_{10}(\text{OM})$ emissions (unit: molecule cm^{-2} of a typical weekday in July) centred for the Paris region from LA inventory (left) and the MEGAPOLI database (right)

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