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Discussions

Interactive comment on "Formation of secondary organic aerosol in the Paris pollution plume and its impact on surrounding regions" *by* Q. J. Zhang et al.

Anonymous Referee #4

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Zhang et al. describe a study that compares airborne measurements taken during the MEGAPOLI campaign against the CHIMERE regional air quality forecasting model. In particular, the authors focus on SOA formation and evaluate the model performance with respect to OA and OA / Ox. Then the model is used to (1) evaluate the evolution of the Paris plume as it is transported from the city, and (2) attribute the total OA mass to different components: POA, biogenic SOA (BSOA), anthropogenic SOA from VOCs (ASOA), and SOA from primary SVOCs and IVOCs (SI-SOA). There are also comparisons for other atmospheric species such as BC and NOx.

This is an interesting analysis given the recent interest in SOA parameterizations for

atmospheric chemistry models, as also noted by the other reviewers, and the airborne measurements represent a valuable opportunity to test these parameterizations. I think the use of OA / Ox is an interesting approach that allows the authors to compare a pseudo-intensive property rather than an extensive property. However, after reading the manuscript, I think that there are four major unresolved points, which should be addressed before publication.

BC Carbon and POA: The model does not accurately reproduce BC concentrations, which is acknowledged by the authors. However, given that BC and POA share sources and are often correlated, it is likely that POA is underestimated in the model as well. It is impossible to be certain if this is the case though, because no comparison of modeled and measured POA is performed. Given that the emissions of the SI-SOA precursors are calculated using the POA emissions, if the POA is underestimated in the model, then the SI-SOA will be underestimated as well. A comparison of modeled and measured POA concentrations is therefore necessary. This would require carrying out PMF analysis of the AMS data, which has its own uncertainties, as mentioned by the authors. However, I would point out that these uncertainties are most likely smaller than the underestimates of 70% and 74% for BC, and thus such a comparison would still be valuable. The authors could also estimate POA using the method of Ng et al. (2011).

SI-SOA: Assuming that POA is underestimated by a similar percentage as BC, the authors should run a sensitivity study in which the concentrations for the precursors of SI-SOA are increased by a factor of $2 - 4$, depending on the outcome of the POA comparison. This sensitivity study would allow the evaluation of how the model predictions depend on the emissions, which are highly uncertain.

Background OA: The article should include a better explanation of why 30% of the maximum concentration was used as the threshold for identifying the background. At the moment there appears to be no explanation for the choice of this percentage, and so the division between background and non-background seems arbitrary.

Model selection: It seems that this version of CHIMERE was run with parameterizations that will favor the production of ASOA relative to SI-SOA. The parameterization used for ASOA includes multi-generation oxidation that will result in quite high SOA yields. At the same time, the Robinson et al. mechanism produces less SI-SOA than the parameterization published by Grieshop et al. (2009). It is not discussed however in the manuscript how the selection of parameterizations used could potentially alter the results. Given the value of this data set, I encourage the authors to expand their analysis to include the Grieshop mechanism as well as the single-generation oxidation mechanism for VOCs, which I believe is already incorporated into CHIMERE (Zhang et al. 2013). At least, the authors should add caveats in the abstract and conclusions that their study is limited to only one specific parameterization, which is a major uncertainty in this study, and thus the relative importance of ASOA versus SI-SOA cannot be determined.

Additional Minor comments:

OOA versus Ox scatter plots: What is the significance of the x-intercept in these plots? Why does the x-intercept change so much, for example in Figure 7c, when comparing the measurements against the model?

P8088/L15: Following my comment above, it seems likely that the average POA concentration of 0.15 ug m-3 for Paris is underestimated.

P8089/L22: "It is likely that these values remain rather similar for a large range of emission and photochemical conditions" This phrase is very vague. What is meant mean by a "large range" or "rather similar"? It would be best if the authors could provide specific numbers. Otherwise, I would simply delete this sentence and note that similar OA to Ox (or SOA to Ox) ratios are observed for many, but not all, cities.

Quality of writing: While the writing in the manuscript is acceptable. The presentation of the science would be clearer if the text was edited for the quality of the English. As an example, the last sentence of the abstract is very confusing.

Grieshop AP, Logue JM, Donahue NM, Robinson AL. Laboratory investigation of photo-chemical oxidation of organic aerosol from wood fires 1: measurement and simulation of organic aerosol evolution. Atmos Chem Phys. 2009;9(4):1263-77.

Ng NL, Canagaratna MR, Jimenez JL, Zhang Q, Ulbrich IM, Worsnop DR. Real-Time Methods for Estimating Organic Component Mass Concentrations from Aerosol Mass Spectrometer Data. Environ Sci Technol. 2011;45(3):910-6.

Zhang QJ, Beekmann M, Drewnick F, Freutel F, Schneider J, Crippa M, et al. Formation of organic aerosol in the Paris region during the MEGAPOLI summer campaign: evaluation of the volatility-basis-set approach within the CHIMERE model. Atmos Chem Phys. 2013;13(11):5767-90.

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