

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.

Anonymous Referee #1

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This paper applies the volatility basis set approach for representing organic aerosols, that is becoming more widely used in regional models, and evaluates the model with measurements collected during the MEGAPOLI campaign in the vicinity of Paris. Although the focus is on organic aerosol, the paper also presents the performance of trace gases, inorganic aerosols, and black carbon. The authors run the CHIMERE model with various configurations to examine how predictions of organic aerosol vary based on a previous treatment of SOA and two variations of the VBS approach.

The paper is well written and organized, the discussion of the results is balanced in

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terms of the possible uncertainties that can contribute to model errors, and the figures are easy to read and interpret. However, there are a number of issues described next that need additional clarification from the authors before the paper is suitable for publication.

General comments:

Introduction: The introduction provides a brief review of the volatility basis set approach and its use in models. This material is a bit dated and does not include more recent work in the past 2-3 years that have shown that VBS formulations can also over-predict OA in some instances and have provided lab/field observations (e.g. Virtanen et al., 2010; Vaden et al., 2010, 2011) that question some of the underlying assumptions employed by the VBS. Many of the assumptions employed by VBS are largely unconstrained by data (e.g. assuming SVOC emissions are 3 times POA emissions, each oxidation reaction increases mass by 7.5%). So the work on VBS is a bit more complicated than what is presented in this paper. However, there have not yet been any alternative theories for the entire lifecycle of SOA that can now be used by models so the use of VBS in this paper is still appropriate.

Measurements, Section 2.1: The authors note while other instruments measure aerosols, they only use measurements from the AMS in this study. I can understand perhaps that the measurements may be redundant, but are those other measurements similar to those from the AMS? This information would be useful to know something about measurement uncertainty which will affect interpretation of model results.

Model Configuration, Section 3.1: Does CHIMERE include wet removal of trace gases and aerosols? It is not clear whether this process is important or not for this study, since there is little discussion on whether precipitation is significant or not. In several places the authors introduce features of the model configuration after section 3.1, which is confusing. For example, the authors state that the model does not include an urban canopy parameterization on page 29493, which should also be included in section 3.1. They also state they set a minimum PBL height in the model of 200 m which again

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should be stated up-front in section 3.1.

Section 3.4: Please comment if any attempt was made to include effects of anthropogenically influenced biogenic SOA. There have been recent observations suggesting that anthropogenic precursor emissions enhance biogenic SOA when these two sources mix. Paris is an environment where this is possible. Normally anthropogenic and biogenic SOA are computed separately.

Model Evaluation in General, Section 4: The evaluation of the model relies solely on surface meteorological, trace gas, and aerosol quantities. Many other modeling studies have shown that knowing whether the conditions aloft are simulated well is important since those conditions will affect surface concentrations. Where there no measurements aloft collected during MEGAPOLI that could be used to evaluate model performance? For readers not familiar with the campaign, it would be useful to state somewhere (either in the introduction or Section 2.1) that no observations were made during the campaign.

Meteorological Evaluation, Section 4.1: I suggest including a time series of observed and simulated humidity. Relative humidity is an important quantity for the uptake of water on aerosols. The model underestimates daytime temperatures, as much as 5 deg C on some days which is rather large. What impact will this have on biogenic emissions that are temperature dependent, and subsequently ozone and SOA?

Section 4.2: The model has not been evaluated with VOC or OH measurements. Were these available? Since the VBS approach depends on both, knowing how SAPRC performs in terms of VOCs and OH will provide evidence of whether SOA production would have been higher or lower, had the photochemistry been predicted better.

Inorganic Aerosol Evaluation, Section 4.4: The evaluation of inorganic species seems a bit of a distraction since the main purpose of the paper is to evaluate organic aerosols. I am not recommending removing the section, since it is useful for completeness. However, the rationale for inclusion up-front in the paper and some transition statements

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here would improve the flow of the paper.

Organic Aerosol Evaluation, Section 4.5: The authors need to state how the model predictions of organic aerosols are compared to AMS measurements. The AMS measures aerosols up to about 1 μm , as reported by the manufacturer, but they often could only be measuring up to ~ 0.7 μm if the aerosol concentrations are low. So, which size bins from CHIMERE are used to compare with AMS measurements? Also, do the AMS measurements been compared with independent aerosol size distribution data to derived collection efficiency factors? What are the factors that have been employed? It would be useful to include some sort of uncertainty estimate in the data.

Evaluation using AMS Measurements, Section 4.6: It is good that the paper compares simulated primary and secondary organic aerosol with estimates derived from AMS measurements. But only one site is presented in this study, and the conclusions regarding the performance would have been much stronger if such comparisons could be made at the other sites. The modeling domain is large and only one site is available for evaluation, so the results need to be taken with a grain of salt. I certainly agree that the VBS approach is an improvement over the previous treatment, but much work remains to determine whether the model is getting a better answer for the right reason.

Specific comments:

Page 29480: line 11: Change "(Hodzic et al . . ." to "(e.g. Hodzic et al . . ." There are other papers using the VBS approach for Mexico City.

Page 29481, line 11: Change "of the Paris city" to "of Paris".

Page 29482, line 11: Change "performed by several instruments" to "sampled by several instruments". Change "with AMS" to "from the AMS".

Page 29482, line 12: Suggest starting a new paragraph with the sentence "Source apportionment . . ."

Page 29482, line 22: I am a bit confused by the use of PMF for model evaluation. The

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authors mention using only the 3-factor PMF analysis that includes cooking activities, but cooking activities are not included in the emission inventory so perhaps it would be more appropriate to compare with the other two sites. At LHVP, should we expect the model to be too low when compared to PMF POA or OOA. A bit more clarification from the authors is needed here.

Page 29488, line 28: I assume the author mean that the use of VOC species does not alter the concentration of VOC species. The current phrase is a bit confusing. Is this assumption significant?

Page 29495, line 8: I do not think it has been mentioned what type of instrument is used to obtain BC measurements, but it is mentioned in Table 2. What size range is included for the model evaluation? Please state this somewhere.

Page 29499, lines 16-17: This statement is not quite correct. The model does better for total OA during these periods. While biogenic SOA is high, there are no observations of biogenic SOA to suggest that the peaks were "reproduced" for this reason.

Page 29501, line 6: Is the simulated value 1.21? The phrase with 0.34 and 1.21 is confusing.

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