Atmospheric
Chemistry
and Physics
Discussions

Open Access

ACPD

Interactive
comment

Interactive comment on "Impacts of water partitioning and polarity of organic compounds on secondary organic aerosol over Eastern China" *by* Jingyi Li et al.

Anonymous Referee #2

Received and published: 13 March 2020

In this paper, Li et al. have modified the CMAQ model to take into account the impacts of water partitioning and polarity of organic compounds on SOA formation. The model was applied over Eastern China to estimate the regional and seasonal impacts of these modifications on SOA and the aerosol water content. This study may have potential to contribute in the organic aerosol modeling field but major revisions needs to be done before publication. In particular, I have several concerns regarding the validity of the scientific methodology used and the presentation of the study. Therefore, I would recommend publication only if these comments will be addressed and fundamental changes will be contacted.

Major comments

Page 4 line 94: The majority of current CTMs have replaced the 2-product model with the VBS approach. Please make this clear and refer to the 2-product model of Odum et al. (1996) for historical reasons.

Page 6, line 162: Please add the appropriate references to support the nonvolatile nature of the products by these oxidation pathways.

Page 6 line 149: There is no discussion in the methodology about the observations and the statistical analysis metrics used to evaluate the model performance. Especially for the OA observations, there is no reference provided or description of the methods used.

Page 7 line 179: More details are needed here. How the model defines the low NOx and high NOx conditions? Which compounds each of the lump species represent? What is the difference between the lumped species of the same precursor (e.g., BNZ1, BNZ2, BNZ3)? Can you include the aerosol yields for each lumped species in tables S1 and S2? Are these aerosol yields NOx-dependent? What does the SVP stands for in Tables S1 and S2?

Page 8, lines 194-198: Does the absorbing phase of equation 1 includes only the water associated with the organics (from eq. 3) or it includes the total water (including the water associated with the inorganic aerosol components)? Under high RH (higher than the organic/inorganic phase separation RH, SRH), the aerosol organic phase is well mixed with the inorganic salts and, therefore, the aerosol water associated with the inorganic constituents can also contribute to the SOA absorbing medium (Pye et al., 2017). Please clarify what you have assumed here and add the relative discussion.

Page 8, Equation 3: This equation gives the volume of water associated with the organic fraction of the aerosol. However, ALW on the left hand side refers to the mass of water. Please correct.

Page 8, lines 203-204: How do you estimate the hygroscopicity?

Page 8, lines 204-205: How do you calculate the ALW? Are you using κorg and the eq3? Do you use the kappa hygroscopicity to calculate the ALW? If so, how you estimate the kappa?

Page 9, lines 212-216: Add a reference to tables S1 and S2. Furthermore, why the values of OM:OC in tables S1/S2 are different than the values provided by your reference (Pye et al., 2017)?

Page 9 lines 217-218: Can you add the total size of the model domain?

Page 9 lines 233: What boundary conditions are used? Please make a comment on how these can affect the simulation results. I would recommend adding spatial maps of primary organic aerosol emissions and SOA precursor emissions and summarizing in a Table the domain average emission rates of POA and each SOA precursors.

Pages 9-10 lines 234-238: This paragraph needs to be expanded and written in a separate section. In this section the authors should describe in more detail the following: i) Basecase simulation. Please explain how the default CMAQ is simulating POA and SOA and how different is this modelling configuration with the one the authors are testing, ii) Sensitivity simulations. Please explain in much more detail the sensitivity simulations conducted in this work. In addition the authors say that they have conducted three sensitivity scenarios named S1, S2 and S3. In their manuscript they only show results from S3 and they never discuss the results of S1 and S2.

Page 10 line 240: The section "Model evaluation" is extremely problematic and raises questions on the validity of the modelling results given that the model evaluation is insufficient. More specifically: 1) Given that the CMAQ default configuration has been modified to consider the importance of water and organic compound polarity on SOA formation, an accurate evaluation of the updated model performance is needed. 2) You should compare the model results for organic compounds during both July and January 2013. Currently, the evaluation includes a comparison with OC observations only during January over only three locations of the relatively large model domain. Fur-

C3

thermore, please mention in the text what factor have you used to convert the modeled OA to OC. 3) The total PM2.5 measurements have been used to evaluate the model performance during July without explaining the rationale of this choice since the focus of this study is solely the organic fraction of the aerosols. I suggest removing the PM2.5 evaluation or at least moving it to the supplement. 4) Can you include more OC/OA observations over other areas of their domain in your evaluation? 5) It is also important to compare the simulated POA and SOA against observations (e.g., from AMS). Furthermore, it would be helpful to show how the model performance against SOA measurements changes between the BC and the S1, S2, S3 cases.

Page 10 lines 251-253: The authors state here that the impacts of water-co-condensation and polarity of organic condensed species on SOA formation are not significant during winter. This highlights the need to evaluate their model results during July where they have found significant changes with the basecase simulation. Furthermore, the results from the three sensitivity simulations should be evaluated individually.

Page 10 lines 254-256: These are indeed possible factors. Can the authors comment, based on their analysis, which of these two possible factors is more important and try to be more specific? A comparison with AMS observations would be helpful here.

Page 11 line 263: The aging of POA, under specific conditions can enhance the SOA formation, especially over polluted areas. Can the authors comment how this important omission of their model configuration can affect their result? Once again, a comparison against POA and SOA from AMS observations will be helpful to identify the limitations of their model due to the treatment of POA as non-volatile and non-reactive.

Page 11 line 264: Please provide two spatial maps of the fraction SOA/Total OA during January and July 2013 so as to show the contribution of POA and SOA to total OA during each simulation period.

Page 13 line 317: You need a zonal map to show how the water partition changes with altitude and not the total column.

Page 13, lines 322-336: It is not clear how you calculate the κ_{org} in your model. This is very important for this section.

Figure 1: The quality of the figure is poor. It is extremely difficult to see all the plotting data and the changes due to the use of different scenarios (especially in figure 1b).

Figure 2: I found the use of daily maximum concentration in the "difference" maps misleading. Given that you have the monthly average SOA from the basecase simulation, I would prefer to see the absolute (and relative) change of the monthly average SOA due to the use of S3 as well, and not the daily maximum. Furthermore, please add (a), (b), etc. to each subplot of the figure and add this information to the figure caption (apply this change in the rest of the figures as well).

Figure 4: All the fitted correlations listed here suggest that compounds with very low or zero O:C have negative hygroscopicity. Can you comment on this limitation and include a discussion in the text?

Table S3. Please explain in the table what the fraction in the last column stands for. How have you estimated the molecular weight and fraction of the POA from unknown compounds?

Minor Comments

The language and structure of the sentences can be substantially improved in many parts of the manuscript. Just a few examples are listed here, but I suggest revising thoroughly the wording in the whole text.

Page 5, lines109-11: Please rephrase. The sentence sounds wrong.

Page 5, lines117-118: Please rephrase.

Page 6 line 144: OC and OA abbreviations have not been used before in the main text.

Page 8, line 204: Please correct the "can estimated" to "can be estimated".

Page 8 line 206: I would use the word "correlate" instead of "dependent"

Page 9 line 229: Change "in" with "on"

Page 10 line 254: Change the sentence to: "In Beijing and Guangzhou, these impacts are not significant during winter"

Page 11 line 274-275: Please rephrase.

Page 11 line 282: Which two areas? You have mentioned several areas in the previous sentence.

Page 13 line 317: "column water". Please rephrase