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Comment on acp-2022-463

Anonymous Referee #2

Referee comment on "Simulating organic aerosol in Delhi with WRF-Chem using the volatility-basis-set approach: exploring model uncertainty with a Gaussian process emulator" by Ernesto Reyes-Villegas et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-463-RC2, 2022

Reyes-Villegas et al. present results from WRF-Chem simulations in Delhi with further analysis using Gaussian process emulators, focusing on organic aerosol mass and O:C ratio. Model results with 10 different perturbed parameters are compared to AMS observation in May 2018, and those model results are used to train Gaussian process emulators. With this approach, the authors could generate 0.5 million sets of model timeseries that can be compared to the AMS observations, from 111 WRF-chem runs. The authors also report the limitation of the model with dynamics related to the planetary boundary layer. This paper suggests that anthropogenic SVOC scaling is the most important factor to improve organic aerosol simulation in Delhi. I find that the method used in this study will be very helpful for the modeling community, not only for organic aerosol modeling but inorganic aerosols and gas-phase chemistry. However, I think a set of parameters the authors have used in this study cannot deliver meaningful results to the organic aerosol modeling community, and the suggestion for organic aerosol modeling would be misreading. I think GMD is more appropriate for this paper, as some comments may not be necessary for GMD. But for publication in ACP, a major revision is needed before it can be acceptable.

Major comments
(1) 10 perturbed parameters: I don't think those are a scientifically meaningful set of parameters to improve OA modeling in Delhi. The authors only include semi-volatile POA only, especially for emission and formation processes, and SOA parameters are not considered in this study. I think biogenic SOA would be minor in May in Delhi, but at least aromatic SOA can not be ignored in the urban atmosphere. Furthermore, loss processes such as wet and dry depositions and photolytic losses (e.g., Zawadowicz et al. 2020) are not considered. If the photolytic loss was not included in the base model, I understand that adding a new reaction would be out of scope in this study. However, the authors

could simply set up a set of parameters to change dry and wet deposition processes in the model, by changing the single parameter - effective Henry's law constant. As there are orders of magnitude variabilities in this constant and it is important for SVOCs (Hodzic et al. 2014), this parameter can give a sufficient range for use in this emulator approach.

(2) Aging with 7.5~45% mass added per reaction: I think those extra masses upon aging reaction can be exaggerated. The studies cited here had been published before the vapor wall loss was reported (Zhange et al. 2014). I guess those additional masses are introduced to compensate for the vapor wall loss (and missing sources like VCPs; McDonald et al. 2018), especially for SVOCs that are sticky and so wall losses are even higher than others. I agree that chemical aging happens in the atmosphere, but this approach only considers functionalization processes that always add mass and shift the volatility towards the lowest saturation vapor pressure bin, and neglects fragmentation processes. Field measurements also showed that there was no mass change with aging (Hodshire et al. 2019). This study is for the collection of field studies from biomass burning plumes, so may not be relevant to the anthropogenic OAs. There are some field studies from urban areas that showed a mass increase with photochemical age, but still I am not sure if the mass increase must be included especially in this study, because of the observed OA profiles shown in Figure 4. Looking at the observed diurnal cycles, O:C ratio increases during the daytime, meaning that there is aging in the atmosphere. However, OA mass decreases during the daytime. PBL evolution couldn't explain this, as observed PBL heights are similar between 12~16 pm. So I guess there were two compensating aging processes (functionalization and fragmentation) at this location. I would suggest that the authors should add the cases with no additional mass (Parameter 3 -> 0) or even no aging process (Parameter 1 -> 0) for meaningful suggestions to the OA modeling community.

(3) Inclusion of 5 biomass burning parameters. I think those 5 parameters related to biomass burning are not necessary, compared to the loss processes and no aging cases suggested above. Those could be excluded for computational efficiency. This can be justified by comparing the magnitude of emissions in this area. For example, if biomass burning emission is 10 while anthropogenic emission is 100, no one expects that the variance of biomass burning scaling will be higher than that of anthropogenic scaling. Unless applying a lot higher maximum scaling factor to biomass burning than anthropogenic, the emulator should give the results that changing anthropogenic parameters is the effective way.

(4) Figure 3: Looking at the top panel of Figure 3, the model runs with low O:C ratios show very high OM concentrations (yellowish). Since all model runs are considering aging reactions with added oxygen rates from 7.5 to 45%, I am not able to understand the results. Since the aging in this model adds oxygen mass only while the carbon mass is fixed, I think O:C ratio should be higher for the higher OM concentrations. Please explain why the model shows this tendency.

Minor comments

1) Line 50 - 51: The authors are talking about the air quality in India in this paragraph, then the last sentence is saying the large fraction of organic aerosols "globally". It would be better to cite other works that measured organic aerosols in India, or the authors can

change the paragraph to present the global air pollution problem instead. But since the next paragraph is presenting OA modeling works in India, the former could be better.

2) Line 59: Spell out "VOC"

3) Line 63-64: Please provide the references to support this statement. I think there are also a lot of uncertainties about SOA mechanism as well, for example, large variabilities of enthalpy of vaporizations, pH-dependency (e.g. IEPOX), aging reaction parameters, photolytic loss, etc.

4) Line 122-123: As far as I know, WACCM doesn't simulate ARO1 and ARO2, instead, benzene, toluene, and lumped xylenes. How did the authors match WACCM aromatics to the aromatics in WRF-Chem? Since the WRF-Chem domain used in this study is small, I think boundary conditions can be also important. A table with species mapping information, especially for OA and OA precursors, would be helpful to understand.

5) Table S1: Please add the corresponding saturation vapor pressure value for each VBS bin. And the unit of the aging rate is missing.

6) Line 140: Emission inventories usually provide OA emissions as carbon mass, not matter mass (e.g., HTAP). How did the authors convert carbon mass to matter mass when those are emitted in the model? Is it implicitly considered when the authors apply the scaling factor of 3? Please clarify that.

7) Line 144: Please specify the version of MEGAN that this study is using. Depending on the version, the authors may need to change the reference to Guenther et al. (2012).

8) Line 189: Can the authors elaborate on the meaning of this scalar variable? Can the authors convert/relate it to some variable with a physical meaning, such as photochemical age?

9) Table 2: Please clarify that "anthropogenic" here includes traditional VOCs (e.g., benzene, toluene) or only for semi-volatile POA.

10) Line 251: Section S2 -> S3

11) Line 255-256: Although the VBS only affects OA and PM, doesn't it affect inorganic aerosols through microphysical processes and gases through changing clouds and aerosol-radiation interactions?

12) Line 258-259: The model also substantially underestimates CO. I understand that this is not easy to fix, but are there any previous studies that reported the same problem in this area? This can be also applied to PM2.5 and NO2, but I think those are not necessary unless there is a previous modeling study at the same location, because those are short-lived species.

13) General: I understand that making a table with all the parameters used for all 111 model runs would be overkill, but it would have been helpful if the authors could provide a table with a set of parameters for some runs, such as model runs used in Figure 4. It is difficult to read parameters from Figures 5 and S7.

14) Line 294-295: Even with the same PBL height, the concentrations can be significantly changed due to the PBL mixing scheme. For example, the early morning peak of OA shown in the model does not exist in the observation, although there are good agreements between the model and the observation before noon. Here or in the methodology section, the authors could discuss the PBL mixing scheme and potential errors from it.

15) Figure 9 and S13: Axis labels are impossible to read.

16) Line 432: These plots -> Figure 10

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