Review of: Simulating organic aerosol in Delhi with WRF-Chem using the VBS approach: Exploring model uncertainty with a Gaussian Process emulator.

Reyes-Villegas et al., presents a modelling study with the WRF-Chem regional chemical transport model aiming at exploring the uncertainties of several parameters of the volatility basis set (VBS).
Up to 111 different model runs are performed with different SVOC and IVOC scaling factors, SVOC volatility distributions, as well as oxidation and aging rates using a 15km domain centred over New Delhi. Model results are compared against aerosol mass spectrometer (AMS) measurements performed during May 2018 in New Delhi. Additionally, the authors used the WRF-Chem model output as an input for Gaussian process emulators to explore the sources of model uncertainties.

The results indicated that the anthropogenic SVOC scaling factors, applied on top of the EDGAR-HTAP emission inventory (among others), had the highest contribution to the variance between the different model runs, with better agreements for low VBS ageing rate, low SVOC volatility distribution and low SVOC scaling factors combined with either high SVOC oxidation rate or high IVOC scaling factors.

The analysis of the model diurnal profiles against AMS measurements reveal several structural errors, with OA concentrations overestimated up to about a factor of 3 during morning peak hours. The authors conclude that deficiency in the model capability of correctly reproducing meteorological parameters, such as the dynamics of the planetary boundary layer, are likely the cause of these structural errors.

I find the study to be innovative, well designed and potentially useful for the atmospheric modelling community. The large amount of chemical transport model (CTM) runs (even though at coarse resolution) allow exploring the wide range of several physical and chemical parameters that are usually probed by the means of box-model simulations applied in more confined and constrained environments. Therefore, the comparison of such large amount of perturbed model runs with ambient AMS data, especially at hourly resolution, is novel and add significant insights on modelling organic aerosol. Finally, the statistics used for the model evaluation section is solid.

I have several minor suggestions, mainly on the terminology and the analysis used throughout the paper, and two major comments, before I can recommend final publication to ACP:

Major comments:

1) The large amount of CTM simulations are very helpful to explore the influence of the perturbed parameters on the modelled OA fraction, and I congratulate the authors for such analysis. However, I believe that it is important to mention (i.e. in the introduction, methodology or conclusion sections) that the perturbed space explored here is embedded in the parent VBS scheme that has been adopted. Therefore, the results should be interpreted in the light of the original VBS blueprint used by the author (e.g. 1D VBS, among other details). For instance: what will happen to the whole perturbed parameter space, if the original VBS will account for processes such as Fragmentation/Formation of HOMs/Autoxidation or for different kernel distributions of the oxidation products (e.g. different number of oxygen additions during the first oxidation steps, and therefore different allocation of the oxidation products across the volatility bins)?
   I am not asking for more runs, but I believe this is an important point that needs to be considered.

2) The analysis of the diurnals is suggesting a strong influence of the meteorological parameters on the OA concentrations, likely of the boundary layer height diagnosed by the model. Even for the model

runs with the best overall statistics and optimized scaling factors, there is a positive bias during the early morning hours. This is an important result. Uncertainties in the diurnal emissions factors are sound, but I think the work will benefit from more analysis on the meteorological side. Are there any radiosondes measurements available to better understand/evaluate the model vertical output (see my additional comments below for the Results section)? In addition, how are the synoptic conditions of the pre-monsoon season reproduced by the model?

Minor comments:

Abstract

Abstract, line 28: Suggestion: I would replace "POA production and aging" with: "emissions of POA and its chemical evolution".

Abstract, line 29: Suggestion: "The main disadvantage is its complexity". I would not consider the complexity of the VBS a disadvantage per se, and I would suggest removing that sentence (I think it is already clear stated that the high numbers of parameters are difficult to probe).

Abstract, line 40: Suggestion: "in two periods: the full period (1st -29th May) and the period 14:00- 16:00 hrs local time, 1st-29th May". One period and one sub-period?

Introduction

Introduction, Line 50: Suggestion: I would move the sentence beginning at this line in the following paragraph. It is a bit detached from its current paragraph.

Introduction, Line 58: Please consider including Ghosh et al., 2021 who has recently applied the online version of the WRF-CHIMERE model over the same domain, i.e. the Indo-Gangetic Plain.

Introduction, Line 64: Please consider replacing "POA processes" with "OA processes".

Introduction, Line 70: Suggest using a more updated and theoretically oriented reference for the definition of the VBS domain, e.g. Bianchi et al., 2019.

Introduction, Line 73: Please consider rephrasing "The resulting low volatility oxidized organic vapors can condense to produce oxidized primary organic aerosols (oPOA) (Shrivastava et al., 2008)" with ". The resulting low volatility oxidized organic vapors can condense to produce secondary organic aerosol (SOA) (Shrivastava et al., 2008)". I think the model assigns those products to SOA (?). (oPOA is considered secondary, and there is no heterogeneous chemistry accounted for, or?).

Introduction, Line 92: "the partitioning of matter between gaseous and particulate phases, and the chemical aging of POA". Suggest rephrasing the sentence as: "The partitioning of organic matter and the evolution of POA".

Methodology

Methodology: How long was the spin-up period? In addition, was WRF nudged towards some reanalysis datasets (ERA5 or GFS)?

Methodology: What is the vertical resolution of the model and the first model layer height?

Methodology: Which scheme was used for the PBLH in the model? Please consider adding this information (see also my additional comment below).

Methodology, Line 121: “SOA components, each covering 4 volatility bins”. At which C* (and T)? Please add such information. Do they cover the SVOC range, or also the LVOC/ELVOC range? In addition, it would be nice the report the IVOC yields with their oxidation pathways in the Methodology section.

Methodology, Line 130: “To investigate the impact of these assumptions on the model predictions, we have modified the model code so that the VBS emissions, the oxygenation rates and VBS reaction rates, can be directly controlled via namelist options”. This is very nice.

Methodology, Line 134: “The volatility distribution of biomass burning”. Suggest rephrasing with “The volatility distribution of open biomass burning” so to differentiate from the residential biomass burning sector contained in EDGAR-HTAP, which has different patterns and estimation methods (this is already specified in Table 1, and I suggest correcting it also in Table 2 accordingly).

Methodology, Line 137: “The volatility distribution for anthropogenic emissions is also multiplied by a scaling factor of 3”. For anthropogenic emissions, I suggest adding also Denier van der Gon et al., 2015 which proposed a revisited emission inventory for the residential wood combustion sector based on a bottom-up approach. The revisited emission inventory, which try to account for the semi volatile components of the primary organic matter fraction, was a factor 2-3 higher compare to previously used emission inventories.

Methodology, Line 140 “Anthropogenic emissions are derived from the EDGAR-HTAP”. Is there any spatial interpolation performed by the model on top of the original EDGAR-HTAP emission datasets? Please add. What was the projection used in the model?

Methodology, Line 163 ‘’PBLH data were sourced from ECMWF ERA5 with 0.25 deg”. Why not directly from WRF at 15km? After further reading, I have realized that the aim is to compare WRF and ERA5. Are there any direct radiosondes measurements available?

Methodology, Figure 1: Is this an average over the whole period? Please add. I would actually consider adding the model topography instead of the PM_1 concentrations.

Methodology, Line 202: I am not sure I have understood this passage: The IVOCs are included by multiply by a factor of 1.5 the non-volatile OA mass (0.2+0.5+0.8). Several models have implemented such scaling factor (Koo et al., 2014; Tsimpidi et al., 2010) on top of the already increased emission inventory (i.e. by a factor of 3 to include SVOC based on partitioning theory calculation), so that the additional organic mass added to the model runs is 7.5 (i.e. 3 + 3*1.5). Is this the case also for this application? In addition, are the IVOCs factors, when increased or decreased during the sensitivity tests, applied to all the EDGAR sectors (i.e. one factor for all the sectors)? Please specify.

Methodology, Line 209: Are those factors, i.e. the parameter number 5 (i.e. 0.1 - 4), applied on top of the emissions inventory as described in the paragraph at line 137? I.e. a value of one would correspond to the original emissions inventory (i.e. increased by a factor of 3).

Methodology, Line 230, Table2. What are the units of the anthropogenic VBS ageing rate and of the open biomass burning VBS ageing rate? Please add.

Results and discussions

Results and discussions, Line 255: “the VBS setup will affect OA concentrations and PM, with no implications to inorganic aerosols or gaseous species”. Would not the different oxidation schemes, and amount of organic precursors available such as SVOCs, IVOCs and VOC, alter the overall radical budget (e.g. HO_x concentrations), and therefore also the inorganic and gas-phase chemistry?

Results and discussions, Line 267: “Total_OM”. I guess it refers only to the modelled particle phase?

Results and discussions, Line 289: "we identified two potential structural errors in the WRF-Chem outputs, the early morning peak and the late evening low concentrations". This is an interesting part of the model evaluation section:

- I would suggest plotting also the diurnal of CO, which is probably a better candidate then OA for identifying the performance of the PBLH in the model. I think a diurnal of CO is already included in Figure S5 but I am not sure I have understood it properly (I would suggest improving the readability of Figure S5, mainly the choice of the colors. I think Figure S5 is also not referenced in the manuscript). It seems model data are reported every three-hours (?) and have a more flat profile compared to the observations (?). How this relate to the diurnal of OA? Along this line, why do the diurnals of O:C decrease in the model during daytime considering that daytime O_3 concentrations are over predicted ?

- For the analyzed period, it would be nice to know which one of the emissions sector in the EDGAR-HTAP dataset is the dominant one in that area, and what is the relative contribution of each of the sources.

- Is there any factor analysis on the AMS data to compare with? How are the modelled and the observational-based OA components (i.e. HOA and OOA) reproduced? Is it an overestimation related to the primary or to the secondary fraction? In addition, how will the model resolution (i.e. 15km) affect the transport and therefore the equilibration time between the particle and gas-phase organic material?

- I am not sure how much the comparison between ERA5-PBLH and WRF-PBLH could help here. Following one of my previous comment, I think it would be beneficial to clarify which parameterization has been used in WRF for the PBLH. ERA5 is based on the Richardson number method. How about those WRF runs? In addition, why the wind direction is not reported in Figure S4?

Model evaluation, line 340: "these model runs have low SVOC volatility distribution". Does this mean low factors (e.g. towards 0.05) and therefore a more volatile distribution (e.g. first panel in Figure S2)? Please add more details in the manuscript.

Model evaluation, Figure 5: This is a very nice plot, and clearly elucidates the various performance of the model runs. If possible, I would consider adding short acronyms for each of the 5 parameters at the pentagon's vertices (at least for one pentagon). It would highly facilitate the comprehension of the panel. In addition, even though provided on the github link, I would suggest to include a supplementary table were the 5 model's parameters are reported for the TRAIN 127, 121 126, 036, 117, 104, 115, 119 and 058. This will facilitate the use of such values by the modelling community. Finally, at least for TRAIN127 and TRAIN120, I would suggest producing in the manuscript a plot similar to Figure S2 where all the parameters are reported as insets on top of the volatility distribution plot associated to the final scaling factors. Again, this will highly facilitate the use of such parameters.

Conclusions

Conclusion, line 493: "low SVOC scaling". A more volatile distribution?

References

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