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

The authors appreciate the reviewer comments, the response is in red

The authors have successfully addressed most of the reviewer's questions. I do not have any major comments. However, there are a few minor comments, primarily concerning some clarifications and discussions that were provided in the response to reviewers but not included in the manuscript. Including these points in the manuscript would be useful to the community.

1) Line 95-97: The manuscript now clearly mentions that this study does not perturb SOA parameters. However, it is still missing a statement acknowledging that this study also assumes that dry and wet deposition simulation uncertainties are not important in Delhi (or assumes that the model accurately represents these processes).

We have edited the paragraph as follows: In this parameterisation we explore the perturbation to the chosen anthropogenic POA and biomass burning POA parameters that would be needed to give the best fit to the observed OA. We are not perturbing the SOA parameters from the base case nor the dry and wet deposition simulation uncertainties, analysis that is out of scope of this work. We also appreciate that there will be sensitivity to the deposition rate of OA components. We have focused our study on the sensitivity of the OA production processes at a constant deposition rate within WRF-Chem allowing reasonable conclusions about the plausible range of the other parameters to be drawn notwithstanding this limitation.

2) Line 145-149: While this information was provided in the response to reviewers, it would be beneficial for readers and the community to include the description of OM/OC ratio, non-oxygen mass, and oxygen mass treatments in emissions before applying the SVOC scaling factor, in the main text.

As suggested, we added the following text to the manuscript: Before applying the scaling factor we assumed a ratio of matter mass to carbon mass of 1.4, dividing the emission inventory matter mass by this to obtain the carbon mass. Within the model each VBS compound is stored as two variables, the oxygen part and the 'non-oxygen' part. When adding the emissions we multiply the carbon mass by 1.17 to get the 'non-oxygen' mass (carbon, plus other atoms), and by 0.08 to get the oxygen mass. These scaling factors were taken from Shrivastava et al. (2011). We then apply the SVOC scaling factor, and volatility distribution, to give the final SVOC emission profile. The IVOC scaling factor is applied to the same base emissions to get the IVOC emission profile.

3) Line 201-213: I found that the author's response to reviewer #2 concerning this scalar variable would be valuable to the community who will use the method presented in this study. Please consider adding more discussion on why creating this variable is essential for the emulator to function correctly for future studies.

The following paragraph, taken from the response, has been added to the manuscript:

The scalar variable represents a sensible range of possible emitted volatility distributions. A method was needed, by which we could represent the variation of possible volatility distributions within the process emulator. The direct approach would be to include a scaling factor for each volatility bin as separate parameters. However, this would have greatly increased the complexity and size of our parameter space, and these parameters would not be independent of each other, leading to a lot of wasted parameter space, waste in the use of our limited computer resources available for the PPE simulations and the assumptions for our variance-based sensitivity analysis becoming invalid.

Instead, we used a simple reaction model, where each step in a fraction of each volatility bin would be 'aged' and moved to the next volatility bin. This approach also allowed us to include some simple partitioning, with aging process stopped for any condensed matter; replicating the behaviour of the model these distributions will later be injected into. Given that we used a simple, fractional, aging process, it would not be appropriate for us to try to relate it to a physical variable. We have included Figure S2 instead, which gives example volatility distributions through the range of this scalar value used in our study.

4) 276-277: I think this line should be removed from the revised manuscript.

The line has been removed as suggested.

5) Line 296-297: It would be helpful for the authors to incorporate the explanation for this model trend in the manuscript since it has already been provided in their response to the reviewers.

We have added the requested explanation: We explored a range of emission multipliers (both IVOC and SVOC scaling). These upper limits, which have been of an appropriate magnitude for previous studies in other locations using different emission datasets (e.g. Shrivastava et al. (2011)), turned out to be too high for our emission dataset, and these are the model runs which produced the very high OM mass loadings (rather than these being predominately caused by high oxidation rates). When the OM mass loading is high, more of the higher volatility (and, here, less aged) compounds condense into the condensed-phase. The VBS scheme we have used has only gas-phase reactions, and so once in the condensed-phase these compounds do not age further. This process leads to the lower mean O:C ratios that are observed here.

6) Line 496-499: Please include the limitations of this study that arise from considering only semi-volatile POA processes. This study does not account for perturbations in SOA parameters (enthalpy of vaporization, SOA yield, photolytic loss) and deposition processes. I believe these processes are more important in Delhi than aqueous chemistry, as IEPOX-SOA is significant for biogenic-influenced regions (Shrivastava et al., 2022) rather than urban regions.

The paragraph has been edited as follows: It is worth mentioning that these structural errors might also be related to representation of other organic aerosol processes not represented by the VBS approach. As mentioned early in the introduction, this study only considers semi-volatile POA processes, without accounting for perturbations in SOA parameters and deposition processes. Recent studies, for example, have examined particle-phase and multiphase chemistry in aqueous aerosols and clouds (Shrivastava et al., 2022), and reactions of SOA precursors with other radicals like chlorine relevant to Indian conditions (Gunthe et al., 2021). Future studies could be focused on studying other parameters such as deposition processes and the perturbations in SOA parameters.