Implementation and Evaluation of an Observation-Constrained Secondary Organic Aerosol Parameterization in MOZART–GOCART Chemistry in WRF-Chem

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Replies to the reviewer 1

Dear Reviewer,

We are thankful to you for providing your suggestions. Your comments and their responses are given below such that comments are in plain text and responses are in blue text. We will incorporate all necessary additions and revisions into the manuscript and submit the revised version during the resubmission stage.

General comments

This is a very narrow-focused study which only looks over one city in India, and only for one month of one particular year. This is very limited both for a comprehensive model evaluation (39 stations only in Delhi, while the whole Indian subcontinent was simulated) and the relevance to the journal itself, which clearly states in its aims and scope statement "Articles should have important and clearly argued implications for our understanding of the state and behaviour of the atmosphere and climate or present substantial new insights into the atmosphere's role in other parts of the Earth system. Articles with a local focus must clearly explain how the results extend and compare with current knowledge" (https://www.atmospheric-chemistry-and-physics.net/about/aims_and_scope.html).

Beyond relevance, the manuscript is weak both in terms of model development not being novel and ignoring important SOA-related processes, and by a weak model evaluation. My recommendation is to reject the manuscript.

We thank the referee for examining the manuscript. The novelty of our study lies in the fact that we identified source sector plumes using detailed VOC observations in Delhi, and using these plume-wise observations, the emission ratios (VOCs/CO) corresponding to each source sector were estimated, which has not been carried out in any previous study. In complex emission environments, such novel VOC fingerprinting provides useful novelty and this is a major postulate and demonstration from our study that has not been demonstrated previously in the literature. Secondly, the comparison of the developed SOA parametrization with other complex gas–aerosol chemistry schemes highlights another very important aspect of our study, because it shows that the developed parametrization is nearly 5 times faster than the complex MOZART-MOSAIC scheme and shows comparable performance (MOZART-MOSAIC NMB: -33%, MOZCART-with-SOA NMB: -45.8%), which makes it very useful for operational air quality forecasting systems where both accuracy as well as computational efficiency are essential.

The present study focuses on Delhi as it serves as the primary testbed for further improving our existing high-resolution air quality forecasting system developed for Delhi (Ghude et al., 2024). The complex gas–aerosol composition, geographical location, and socio-economic and national importance of Delhi necessitate accurate air quality forecasting so that exposure of the public to acute air pollution levels can be minimized. In this study, the SOA parameterization is constrained by detailed, high temporal resolution VOC observations obtained in Delhi. In contrast, such high-resolution observational datasets for SOA and OA are not available for other regions of India, which limits the ability to rigorously evaluate model performance beyond Delhi. Although VOC measurements may not be strictly required in other regions (rural and urban areas) where similar fuel-use profiles are present in emissions, the absence of direct observations of SOA and OA restricts the scientific robustness of model evaluation. In the absence of our observations in Delhi, we did not have any option other than relying on the parameters introduced by Hodzic and Jimenez (2011), which were based on measurements in Mexico City. We show that the same parameters are not valid for Delhi. Therefore, while simulations were conducted over the entire Indian subcontinent to ensure we are able to capture regional transport patterns, the analysis and interpretation of SOA, OA, and $PM_{2.5}$ are primarily confined to Delhi.

However, for use by the wider scientific community, we have made provision to apply the plume-wise emission ratios through the run-time configuration file in the WRF-Chem model, so that if one has derived these emission ratios based on similar observations in other cities, they can be easily incorporated into the model, making this parameterization more generally applicable.

Specific comments

Lines 67-74: Both the two-product model and VBS scheme follow the equilibrium assumptions of Pankow (1994), the difference is that in the two-product model there are effectively two volatility bins and OA mass is not transported from one to the other, while in the VBS there are more than two volatility bins, they are equally-spaced in the volatility space, and there is mass transport from one volatility bin to the other(s) via chemical processing.

Reply: We will revise the statement as follows in the revised manuscript:

"The VBS scheme is an advanced SOA parameterization scheme that also follows Pankow (1994) theory for gas–aerosol partitioning but resolves SOA formation using multiple volatility-resolved bins of condensable compounds (Donahue et al., 2006). These bins span one order of magnitude in effective saturation concentrations and allow dynamic mass transfer between them. The VBS scheme substantially improves simulated SOA concentrations, providing better agreement with observations (Hodzic et al., 2010; Shrivastava et al., 2011)."

Lines 128-130 and line 200: It is my understanding that SOA is implicitly represented in GOCART, even early versions of it, by simply assigning some OA source from biogenic volatile organic compounds (terpenes and maybe even isoprene). Yes, this is not an explicit SOA formation via equilibrium partitioning, but it is SOA formation nevertheless. Is this a capability that has been removed from MOZCART as used in AIRWISE? This is also relevant when presenting results in e.g. figure 3, do you literally mean "without SOA"? Because if indeed there is no SOA at all, then having lower results is only natural.

Reply: Thank you for the comment. We developed the SOA parameterization for anthropogenic emissions and open biomass burning emissions, and we have not altered any part of the biogenic aerosol module(s) in WRF-Chem. Additionally, we carefully looked through the biogenic modules present in WRF-Chem, and as per our understanding, there is no OA source from biogenic volatile organic compounds (terpenes and isoprene) in the MOZART-GOCART chemical mechanism in WRF-Chem.

SOA parameterization: The approach of linking SOA to VOC and CO is over a decade old, and is very sensitive to the source terms used. It is not shown in the manuscript, but the simulations hold the data that can prove or disprove whether this parameterization is applicable elsewhere, or even in Delhi at other seasons and/or years. The authors should had made comparisons across India, in more urban areas as well as rural and remote areas. As a matter of fact, since the Hodzic and Jimenez (2011) work and its derivatives several new processes have been identified as critical to SOA, like photolysis (also studied by Hodzic), oligomerization, and phase state, which make the presence of a complicated mechanism rather than a simple one like the one presented here necessary. On top of that, such an approach completely ignores the temperature dependence of partitioning which forms SOA higher in the atmosphere, as demonstrated by measurements in the Amazon following deep convection.

Reply: Although this has been suggested and applied by Hodzic and Jimenez (2011) in 2011 in CHIMERE regional chemistry model, It has been adopted and presently being used in models like GOCART-2G (Collow et al., 2024) because of its computational efficiency which is one of the critical parameters for operational air quality forecasting. Further, in the present study, unlike the Hodzic and Jimenez (2011), we identified source sector plumes of different anthropogenic sectors along with open biomass burning by detailed VOC measurements in Delhi and then derived sector specific emission ratios, which has not been done in any previous study.

The simulations cover the entire Indian subcontinent and contain information for other regions and periods to ensure that regional transport patterns are adequately simulated by the model. However, the present study focuses on Delhi as the primary testbed for improving our high-resolution air quality forecasting system (Ghude et al., 2024). This is because the SOA parameterization is constrained using detailed, high temporal resolution VOC observations available only for Delhi. Comparable high-resolution datasets for SOA and OA are not available for other regions of India, which limits rigorous model evaluation beyond Delhi. Although VOC measurements may not be strictly required in regions with similar fuel-use profiles, the absence of direct SOA and OA observations restricts the scientific robustness of such evaluations over other parts of India. Therefore, the analysis and interpretation of SOA, OA, and $PM_{2.5}$ are confined to Delhi despite simulations being performed over the full domain.

Moreover, we acknowledge the importance of additional SOA processes and their explicit treatment in complex mechanisms with higher computational cost. However, the primary objective of this study is to improve operational air quality forecasting without compromising computational efficiency. Notably, comparison with a comprehensive scheme (MOZART-MOSAIC), which includes these processes, shows that our approach achieves comparable performance (NMB: -33% vs. -45.8%) while being nearly five times faster. Thus, the developed parameterization provides a practical balance between accuracy and computational efficiency for real-time forecasting applications.

Lines 295-296 and figure 2: The authors state "reasonably captures the observed variability", but the black (measurements) and blue (model) lines differ by a factor of 3 for most of the period. What am I missing here? The discussion that follows simply cherry-picks dates and makes qualitative statements instead of doing a proper statistical analysis. I do acknowledge table 2, which says e.g. -45.8 % normalized mean bias. This is nowhere near “reasonably captures”.

Reply: We thank the reviewer for this observation. We agree that the term “reasonably captures” may not fully reflect the magnitude of the bias over the entire period. The model performance is relatively better during 9–12 November, whereas the substantial underestimation during 13–19 November is primarily attributed to under-detected fire counts in satellite observations due to cloud cover, which affects emission inputs. We also acknowledge that the NMB of −45.8% for SOA is relatively high. However, the inclusion of SOA in MOZART-GOCART leads to substantial improvements in $PM_{2.5}$ and OA predictions over Delhi. Specifically, the NMB improves from −39.4% to −18.3% for $PM_{2.5}$ and from −57.8% to −19.1% for OA, indicating a significant overall benefit despite biases in SOA alone. We hope to achieve additional improvements by incorporating other processes in the model (e.g., nitrate aerosols) and will report those findings in future studies. This study is dedicated to implementation of a simple SOA formation scheme.

References:

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