

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

Rajmal Jat<sup>1</sup>, Akash S. Vispute<sup>1</sup>, Sachin D. Ghude<sup>1</sup>, Rajesh Kumar<sup>2</sup>, Vinayak Sinha<sup>3</sup>, Baerbel Sinha<sup>3</sup>, Gaurav Govardhan<sup>1</sup>, Zhining Tao<sup>4</sup>, Prafull P. Yadav<sup>1,6</sup>, Sandeep Wagh<sup>1</sup>, Sreyashi Debnath<sup>1</sup>, Aditi Rathore<sup>1</sup>, M. Rajeevan<sup>5</sup>

<sup>1</sup>Indian Institute of Tropical Meteorology, Ministry of Earth Sciences, Pune, India

<sup>2</sup>NSF National Center for Atmospheric Research, Boulder, CO, US

<sup>3</sup>Indian Institute of Science Education and Research Mohali, Punjab, India

<sup>4</sup>Morgan State University, Baltimore, MD, USA

<sup>5</sup> National Centre for Earth Science Studies, Thiruvananthapuram, Kerala, India; Atria University, Bengaluru, India

<sup>6</sup>Dept. of Atmospheric and Space Sciences, Savitribai Phule Pune University, Pune, India

**Correspondence:** Rajmal Jat ([rajmal.jat@tropmet.res.in](mailto:rajmal.jat@tropmet.res.in)) and Sachin D. Ghude ([sachinghude@tropmet.res.in](mailto:sachinghude@tropmet.res.in))

## **Replies to the reviewer 3**

Dear Reviewer,

We sincerely thank you for your constructive suggestions, which have improved the manuscript. Your comments and their responses are given below, such that the comments are in plain text and the responses are in blue text. Additions and changes to the manuscript are marked using track changes in the copy of the manuscript with track changes. References to line numbers in the responses correspond to the copy of the manuscript with track changes.

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The manuscript presents a simplified observation-constrained SOA parameterization implemented in MOZCART within WRF-Chem and evaluated over Delhi during November 2024. The work is relevant, however, the parameterization currently relies on several strong assumptions (e.g., 100% SOA yield, OH-only oxidation, constant VOC/CO ratios, simplified lumping) that require more rigorous justification. Additional evaluation, uncertainty analysis, and clarification of the parameterization framework are necessary before the manuscript is suitable for publication. I therefore recommend major revision.

Reply: Thank you for your valuable comments and suggestions, which have helped us substantially improve the manuscript. In response to your major concerns, we have replaced the original parameterization with a bulk volatility basis set (VBS) based scheme that no longer assumes 100% conversion of VOCs to SOA and that explicitly treats gas–particle partitioning, temperature dependence and aging (Major Comments 1 and 2). While addressing your comment on the Aerosol Mass Spectrometer (AMS) measurements (Major Comment 6), we also identified an issue with the high-resolution peak fitting in the original SOA observations. We therefore repeated the observational analysis using the raw data and revised the observed aerosol concentrations accordingly. These changes have led to revisions in the parameterization framework, the observational analysis, the model evaluation and the associated discussions. Our detailed responses to each of your comments and the corresponding revisions are provided below.

## Major Comments

### 1. Physical realism of the 100% SOA mass yield assumption

The parameterization assumes complete conversion of the emitted surrogate VOC mass into SOA with a 100% yield. This is a very strong assumption and appears physically unrealistic. Although the authors cite Hodzic and Jimenez (2011), the implementation used here seems conceptually different from that study. In Hodzic and Jimenez (2011), the emitted surrogate VOC was effectively constrained such that only the SOA-forming fraction of organic vapors was represented, based on the relationship between SOA production and  $\Delta\text{CO}$  (i.e.,  $\text{Emis}(\text{VOC})/\text{Emis}(\text{CO}) = \text{SOA}/\Delta\text{CO}$ ). In the present manuscript, however, the authors derive VOC emissions directly from observed VOC/CO emission ratios obtained from in situ measurements. These measured VOCs represent total VOC emissions and therefore cannot reasonably be assumed to convert entirely into SOA. As currently formulated, the approach appears to imply unrealistically large effective SOA yields. The manuscript therefore requires a much more rigorous justification of this assumption and a clearer explanation of how the surrogate VOC species are defined relative to actual SOA precursors. In addition, sensitivity simulations using lower and more realistic effective SOA yields, constrained by literature or chamber studies, would substantially strengthen the manuscript and help quantify the impacts on SOA, OA, and PM<sub>2.5</sub> predictions.

Reply: Thank you for the suggestion. Following your suggestions, we realized that we had misunderstood the approach of Hodzic and Jimenez (2011). We agree that, in our original

scheme, we were measuring total VOCs and converting 100% of the total VOCs to SOA, which was unrealistic.

In the revision, we have rewritten the scheme, and total VOCs are no longer fully converted to SOA. We have replaced the original scheme with a bulk volatility basis set (VBS) scheme in MOZCART. Instead of using total measured VOCs, we use emissions of seven precursor VOC species—benzene, toluene, xylene, isoprene, monoterpenes, lumped alkenes, and intermediate-volatility organic compounds (IVOCs)—that are known to contribute to SOA formation in the atmosphere. These emissions are obtained from an analysis of each sector’s pollution plume. The emission ratios of each of these species in each sector’s pollution plume are presented in Table S3.

Table S3: The VOC to CO emissions ratio (ERs) (ppbv/ppmv) of different VOC species from Industrial, transportation and biomass burning sectors

| VOC species   | Industrial Sector | Transportation Sector | Biomass Burning Sector |
|---------------|-------------------|-----------------------|------------------------|
| Benzene       | 3.44              | 3.10                  | 3.20                   |
| Toluene       | 8.80              | 9.30                  | 5.90                   |
| Xylene        | 6.50              | 5.80                  | 2.40                   |
| isoprene      | 0.95              | 0.82                  | 1.20                   |
| monoterpene   | 1.16              | 0.79                  | 0.12                   |
| IVOCs         | 0.96              | 0.85                  | 3.30                   |
| Lumped Alkene | 33.00             | 24.00                 | 22.00                  |

Each precursor VOC species undergoes photochemical oxidation by the hydroxyl radical (OH) and forms low-volatility condensable gases. In our VBS approach, we considered four volatility bins (Bins 1, 2, 3, and 4), with respective reference saturation concentrations of 1, 10, 100, and 1000  $\mu\text{g}/\text{m}^3$ . The stoichiometric coefficients ( $\alpha$ ) for the four volatility bins are derived from chamber-based VBS parameterizations (CAMx 1.5-D and CHIMERE for lumped alkenes; Huang et al., 2026). These coefficients differ under high- and low-NO<sub>x</sub> conditions (Table S4). The SOA yield is therefore determined by chamber data and varies with the conditions, rather than being fixed at 100%.

Table S4: SOA formation parameters in the MOZCART VBS scheme

| C*( $\mu\text{g}/\text{m}^3$ )@298K                                               | 1            | 10            | 100          | 1000         |
|-----------------------------------------------------------------------------------|--------------|---------------|--------------|--------------|
| CG/SOA<br>MW(g/mol)                                                               | 167          | 163           | 158          | 153          |
| stoichiometric coefficient (ppm/ppm) under high (low NO <sub>x</sub> ) conditions |              |               |              |              |
| Benzene                                                                           | 0.001(0.035) | 0.079 (0.108) | 0.148(0.185) | 0.222(0.268) |
| Toluene                                                                           | 0.006(0.006) | 0.145(0.145)  | 0.281(0.437) | 0.432(0.281) |
| Xylene                                                                            | 0.001(0.048) | 0.127(0.195)  | 0.201(0.252) | 0.301(0.364) |
| Isoprene                                                                          | 0.0(0.004)   | 0.009(0.013)  | 0.006(0.006) | 0.0(0.0)     |
| Monoterpenes                                                                      | 0.01(0.087)  | 0.101(0.077)  | 0.173(0.309) | 0.451(0.54)  |

|                 |              |              |              |              |
|-----------------|--------------|--------------|--------------|--------------|
| Lumped alkenes* | 0.002(0.015) | 0.015(0.029) | 0.054(0.085) | 0.178(0.247) |
|-----------------|--------------|--------------|--------------|--------------|

\*For lumped alkene, CG/SOA MW of 120 g/mol is considered across the bins as per CHIMERE model VBS parameterization (Huang et al. 2026)

Only for IVOCs was an effective unit SOA yield assumed, consistent with previous modeling studies in which IVOCs undergo OH-driven aging to form lower-volatility products, with an ultimate SOA yield of 100% (Robinson et al., 2007). Similarly, CMAQ AERO7 (Huang et al., 2026) assumes an IVOC SOA yield of 1 g g<sup>-1</sup>, forming effectively nonvolatile products. Following these approaches, IVOCs are represented as forming low-volatility condensable products with an effective unit yield upon oxidation.

We have added these details to Section 2.3, “SOA Parameterization,” of the revised manuscript (Lines 330–360).

## 2. Lack of treatment of semivolatile partitioning

The proposed scheme effectively bypasses gas–particle partitioning physics and volatility evolution, which are central processes in SOA formation. The authors should discuss much more explicitly the limitations associated with omitting SOA thermodynamics and semivolatile partitioning processes. In particular, the manuscript should clarify how the dependence of SOA formation on aerosol mass loading is represented, whether SOA evaporation or repartitioning is possible within the parameterization, and whether any temperature dependence is included.

Reply: Thank you for the suggestion. In our revised VBS-based scheme, we have added gas-to-particle partitioning according to the absorptive partitioning theory of Pankow (1994), which includes the dependence of SOA formation on total aerosol mass loading. For the temperature dependence of SOA formation, we have used the Clausius–Clapeyron equation to adjust the saturation concentrations of the gas-phase condensable gases (CGs) in the different bins according to ambient temperature, which affects SOA formation. The aging of SOA is also accounted for in our parameterization scheme through OH-oxidation based functionalization of gas-phase CGs.

We have added the following information to Section 2.3, “SOA Parameterization,” of the revised manuscript (Lines 345–400).

“To cover the wide range of condensable gases, four volatility bins (Bin 1, Bin 2, Bin 3, and Bin 4), with respective reference saturation concentrations of 1, 10, 100, and 1000 µg/m<sup>3</sup>, are considered. The general form of the chemical equation used is presented in Eq. (1).



Where,  $VOC_{species}$  corresponds to each of the six precursor VOC species,  $\alpha_{species\_i}$  is the stoichiometry coefficient of the particular species for the  $i^{th}$  bin, and  $CG_i$  is the condensable gas in the  $i^{th}$  bin. CG1, CG2, CG3 and CG4 correspond to Bin1, Bin2, Bin3 and Bin4, respectively, with saturation concentrations of 1 , 10 , 100 and 1000  $\mu g/m^3$ , respectively. The oxidation product of IVOCs is converted to SOA (IVSOA) using eq. 2. The rate constant (r) for these reactions is set to  $1.25 \times 10^{-11} \text{ cm}^3 \cdot \text{molecules}^{-1} \cdot \text{sec}^{-1}$ .

The stoichiometric coefficient for each species in each bin is generally derived by fitting the experimental results of chamber-based laboratory studies. Here, we obtained the stoichiometric coefficients for the precursor species under both high and low NO<sub>x</sub> regimes from the CAMx 1.5-D model VBS parameterization, except for lumped alkenes, for which the CHIMERE model VBS parameterization is used (Huang et al., 2026). The species-wise information on the stoichiometric coefficients across different bins under high and low NO<sub>x</sub> conditions used here is compiled from Huang et al. (2026) and presented in Table S4.

These condensable gases (CGs), corresponding to oxidation products of each precursor VOC species, then undergo gas-to-particle partitioning according to the absorptive partitioning theory of Pankow (1994) (Eq. 3).

$$f_{particle}^i = \frac{C_{OA}}{C_{OA} + C_i^*} \dots\dots\dots(\text{Eq.3})$$

where,  $C_{OA}$  is the organic aerosols concentrations,  $C_i^*$  is the saturation concentrations in the  $i^{th}$  bin, and  $f_{particle}^i$  is particle-phase fraction of the total SOA mass (gas-phase + particle-phase) in  $i^{th}$  bin. The  $C_i^*$  values are adjusted according to the ambient temperature using the Clausius Clapeyron equation (Eq.4), to account for changes in saturation concentration due to local temperature and, consequently, their effect on SOA formation.

$$C_i^* = C_{i,ref}^* \frac{T_{ref}}{T} \exp \left[ \frac{\Delta H_{vap}}{R} \left( \frac{1}{T_{ref}} - \frac{1}{T} \right) \right] \dots\dots\dots(\text{Eq.4})$$

where,  $C_{i,ref}^*$  is the reference saturation concentration in the  $i^{th}$  bin at reference temperature ( $T_{ref}$  of 298 K), T is the local ambient temperature, R is the universal gas constant and  $\Delta H_{vap}$  is the enthalpy of vaporization.  $\Delta H_{vap}$  is prescribed as 35 kJ/mol for each volatility bin following the CAMx 1.5-D VBS scheme (Huang et al., 2026).

The aging of SOA is also accounted for in our parametrization scheme through OH-oxidation based functionalization of gas-phase CGs. For SOA aging at each time step, CGs produced in each bin are oxidized by OH $\cdot$  and produce lower volatility products. These oxidized products move to the next lower volatility bin in a step-wise manner. For instance, the oxidized product moves from Bin4 ( $C^* = 1000 \mu g/m^3$ ) to Bin 3 ( $C^* = 100 \mu g/m^3$ ), from Bin 3 ( $C^* = 100 \mu g/m^3$ ) to Bin 2 ( $C^* = 10 \mu g/m^3$ ), and Bin 2 ( $C^* = 10 \mu g/m^3$ ) to Bin1 ( $C^* = 1 \mu g/m^3$ ). The general equation for SOA aging is as follows.

$$CG_i^t = (SOA_i^{t-1} + CG_i^{t-1}) f_{gas}^i \times e^{-k_{OH}[OH]\Delta t} + (SOA_k^{t-1} + CG_k^{t-1}) f_{gas}^k \times (1 - e^{-k_{OH}[OH]\Delta t}) \dots\dots\dots(\text{Eq. 5})$$

$CG_i^t$  is the concentration of CG in the  $i^{\text{th}}$  bin at time  $t$ . The SOA mass at the previous time step ( $SOA_i^{t-1} + CG_i^{t-1}$ ) in the  $i^{\text{th}}$  bin is multiplied by the fraction ( $f_{gas}^i \times e^{-k_{OH}[OH]\Delta t}$ ) that remains in the gas phase after OH oxidation. The  $[OH]$  is the ambient OH concentrations ( $\text{molecule.cm}^{-3}$ ) at time  $t$ .  $SOA_k^{t-1}$  and  $CG_k^{t-1}$  are the particle phase and gas phase SOA mass, respectively, in the  $k^{\text{th}}$  bin, the immediately higher-volatility bin ( $i+1$ ) from which CG mass is transferred to the  $i^{\text{th}}$  bin. Thus, the second term in Eq. (4) represents the transfer of the gas-phase fraction from the higher-volatility bin ( $k$ ) to the next lower-volatility bin in time  $\Delta t$  ( $t - (t - 1)$ ). Please note that there will be no gas-phase mass transfer (i.e., the second term will be zero in eq. 4) to the last bin ( $i=4$ ). Similarly, for the first bin ( $i=1$ ), there will be no loss of CG. Therefore, the exponential term ( $e^{-k_{OH}[OH]\Delta t}$ ) is omitted from the first term for first bin ( $i=1$ ). The rate constant ( $k_{OH}$ ) used is  $2 \times 10^{-11} \text{ cm}^3.\text{molecule}^{-1}.\text{s}^{-1}$ . Furthermore, to understand the contributions from different source sectors to the simulated SOA in the present study, each precursor VOC species, its corresponding CGs, and particle-phase SOA are tagged according to their respective sectors to identify the contributions from different sectors.”

### 3. Broader applicability of the proposed approach is unclear

The parameterization is constrained using VOC/CO ratios measured in Delhi during a single season (post-monsoon/winter). It is therefore unclear whether the proposed parameterization is applicable outside Delhi or under different seasons, especially during summer when photochemistry and biogenic SOA are stronger. The manuscript repeatedly suggests that the approach is suitable for operational applications over “highly polluted urban regions”; however, this broader applicability is not sufficiently demonstrated. The authors should discuss more explicitly the expected limitations of the parameterization to other regions, seasons, and chemical regimes, particularly given the strong dependence of VOC composition and SOA formation pathways on local emissions and meteorological conditions.

Reply: Thank you for this important suggestion. We have kept the focus of the present study on Delhi as the primary testbed because we aim to improve the accuracy of our high-resolution air quality forecasting system (Ghude et al., 2024) for Delhi. Therefore, the SOA parameterization presented here is constrained by detailed, high-temporal-resolution VOC observations available only for Delhi. We agree that the broader applicability of the proposed SOA parameterization beyond Delhi and across different seasons should be tested and requires further evaluation. However, for rigorous model evaluation beyond Delhi, high-resolution datasets of SOA and OA are required for other regions of India. But such datasets are currently unavailable, limiting the detailed evaluation of the broader applicability of the parameterization in other parts of India.

Nevertheless, to provide an initial assessment of broader applicability of the parameterization beyond Delhi, we have evaluated simulated  $\text{PM}_{2.5}$  concentrations with and without the revised SOA parameterization in six additional cities across the Indo-Gangetic Plain (IGP) (Figure S12), using observations from the CPCB monitoring network. This analysis examines the influence of the revised parameterization on simulated  $\text{PM}_{2.5}$  concentrations in other urban areas. Improvements in simulated  $\text{PM}_{2.5}$  may provide an initial indication of the parameterization's applicability beyond Delhi and suggest that the fuel-use profiles and

associated VOC emission characteristics may be broadly similar to those of Delhi. In contrast, the absence of improvement may indicate differences in local emission profiles and the need for region-specific VOC/CO emission ratios derived from high-resolution VOC observations. However, these results remain indicative rather than conclusive, as improvements in total PM<sub>2.5</sub> do not constitute direct validation of SOA formation. The limited availability of comparable high-resolution SOA, OA, and VOC observations for these cities currently restricts a more rigorous assessment.

Although the proposed revised parameterization incorporates several physically based processes that account for the effects of temperature, aerosol mass loading, and chemical aging on SOA formation, allowing it to respond to changes in environmental conditions, we agree that the parameterization should also be tested during other seasons in Delhi, such as summer. However, high-resolution observational data on SOA, VOCs, and OA are currently available only for the study period. We have added a discussion of this limitation to the revised manuscript and suggested future studies to increase confidence in the parameterization's applicability to other seasons.

We have added the following information to the revised manuscript (Lines 595–620).

“To understand the broader applicability of this observation-constrained SOA parameterization, we also evaluated simulated PM<sub>2.5</sub> concentrations at six different cities spanning the IGP (Figure S12) against observed PM<sub>2.5</sub> concentrations. A direct evaluation against SOA observations was not possible for these cities because comparable high-resolution SOA observations from aerosol mass spectrometry (AMS) were unavailable. The evaluations of simulated PM<sub>2.5</sub> without and with SOA are presented in Tables S5 and S6, respectively. A mixed response was observed across these cities. Statistical measures showed improvements in PM<sub>2.5</sub> predictions for Lucknow, Ludhiana, and Chandigarh, while predictions degraded for Agra and Kolkata. The substantial underprediction without SOA was reduced: NMB changed from –36% to +6% in Lucknow, from –36% to –7% in Ludhiana, and from –63% to –45% in Chandigarh. RMSE, MAE, and MFE also improved in Lucknow and Chandigarh. In contrast, adding SOA degraded model predictions in Agra and Kolkata. In Agra, model performance shifted from a nearly unbiased prediction (NMB of –7%) to overprediction (+39%), while in Kolkata, the existing overprediction increased further (NMB of 106% and RMSE of 95 µg/m<sup>3</sup>). The response in Patna was mixed. The NMB changed from –25% to +26%, but RMSE, MAE, and MFE remained almost unchanged, indicating that the reduction in negative bias was accompanied by a shift to positive bias. These results provide an initial assessment of the parameterization's performance beyond Delhi but do not constitute direct validation of SOA formation. The improvements observed in some cities may be consistent with fuel-use profiles and associated VOC emission characteristics similar to those in Delhi, whereas the deterioration in other cities may reflect differences in local emission profiles. At the same time, uncertainties in the emissions inventory and simulated meteorology, as well as the absence of secondary inorganic aerosol (SIA) species in MOZCART chemistry, also contribute to the differences between simulated and observed PM<sub>2.5</sub> concentrations. These findings highlight the limitations of directly transferring the Delhi-constrained parameterization to other regions

and the potential need for region-specific VOC/CO emission ratios derived from high-temporal-resolution VOC observations. Further evaluation using comparable SOA, organic aerosol (OA), and VOC observations is needed to establish its broader applicability.

Table S5: Statistical analysis of PM<sub>2.5</sub> predictions (without SOA) for different cities in IGP during November 2024.

| City       | RMSE (µg/m <sup>3</sup> ) | MB (µg/m <sup>3</sup> ) | MAE (µg/m <sup>3</sup> ) | NMB (%) | MFB (%) | MFE (%) |
|------------|---------------------------|-------------------------|--------------------------|---------|---------|---------|
| Agra       | 36.63                     | -5.04                   | 24.81                    | -7.49   | -0.18   | 36.20   |
| Chandigarh | 101.88                    | -82.14                  | 82.14                    | -63.31  | -90.73  | 90.74   |
| Kolkata    | 58.28                     | 34.13                   | 47.05                    | 46.93   | 39.72   | 53.30   |
| Lucknow    | 54.64                     | -39.68                  | 43.30                    | -35.91  | -39.67  | 45.66   |
| Ludhiana   | 84.48                     | -38.15                  | 49.72                    | -35.73  | -35.46  | 50.70   |
| Patna      | 40.41                     | -26.70                  | 32.53                    | -24.61  | -26.38  | 33.54   |

Table S6: Statistical analysis of PM<sub>2.5</sub> predictions (with SOA) for different cities in IGP during November 2024.

| City       | RMSE (µg/m <sup>3</sup> ) | MB (µg/m <sup>3</sup> ) | MAE (µg/m <sup>3</sup> ) | NMB (%) | MFB (%) | MFE (%) |
|------------|---------------------------|-------------------------|--------------------------|---------|---------|---------|
| Agra       | 52.22                     | 26.35                   | 40.66                    | 39.18   | 36.03   | 49.45   |
| Chandigarh | 86.92                     | -58.28                  | 62.91                    | -44.92  | -60.50  | 64.43   |
| Kolkata    | 95.22                     | 77.20                   | 80.90                    | 106.17  | 69.92   | 72.45   |
| Lucknow    | 46.49                     | 6.78                    | 37.71                    | 6.14    | 7.99    | 34.01   |
| Ludhiana   | 87.85                     | -7.76                   | 59.57                    | -7.26   | -5.22   | 53.11   |
| Patna      | 47.15                     | 28.18                   | 38.40                    | 25.97   | 22.90   | 33.44   |

”

Added the following information to the Abstract of the revised manuscript (Lines 45–50).

“These results demonstrate that the proposed SOA parameterization provides an effective balance between accuracy and computational efficiency and has potential for operational air quality forecasting over highly polluted urban regions, provided that it is constrained by detailed, high-temporal-resolution local VOC observations.”

Added the following information to the section 4 “**Future Outlook and Limitations of the Present Study**” of the revised manuscript (Lines 840–845).

“The applicability of the proposed parameterization across seasons remains to be established. Its performance in Delhi, particularly during summer, has not yet been evaluated because high-

resolution observations of SOA, OA, and VOCs are available only for the study period. Higher temperatures, stronger photochemical activity, and increased biogenic VOC emissions during summer may affect SOA formation. Future studies should therefore evaluate the parameterization using high-resolution summer observations of SOA, OA, and VOCs in Delhi to assess its performance under different seasonal conditions.”

Added the following information to Section 5, “**Conclusions**” of the revised manuscript (Lines 895–900).

“Overall, these findings demonstrate that emission-ratio based SOA parameterizations constrained by detailed, high-temporal-resolution local VOC observations offer a practical approach to improving aerosol predictions in operational air quality forecasting systems over highly polluted urban regions.”

#### 4. **Fire emission underestimation explanation is insufficiently demonstrated**

The manuscript attributes the strong SOA underestimation during 13–19 November to missing fire detections under foggy conditions. While this explanation is plausible, it remains insufficiently demonstrated. The authors should provide additional supporting evidence, such as comparisons with independent fire products, aerosol optical depth (AOD) observations, CO measurements, or sensitivity simulations using scaled fire emissions.

Reply: Thank you for the suggestion. Following your suggestion, we have incorporated additional information from spatial AOD plots, fog/low-cloud distributions from INSAT-3DR, and independent fire activity datasets from MODIS, VIIRS onboard Suomi NPP, NOAA-20, and NOAA-21, along with Fire Radiative Power (FRP) data from CAMS GFAS, to further investigate the potential underestimation of fire emissions during foggy conditions.

The following information has been added to the revised manuscript (Lines 475–495).

“For instance, during 13–18 November 2024, natural-colour images overlaid with the spatial distribution of Aerosol Optical Depth (AOD) from the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the Terra and Aqua satellites (NASA Worldview), together with fog/low-cloud distributions from the Indian National Satellite -3DR (INSAT-3DR) over northern India, clearly show dense fog over the states of Punjab and Haryana (Figures S6 and S7). Likewise, daily fire activity over Punjab and Haryana during November 2024 was analysed using fire count data from MODIS, the Visible Infrared Imaging Radiometer Suite (VIIRS) onboard the Suomi National Polar-orbiting Partnership (S-NPP) satellite, and VIIRS onboard the National Oceanic and Atmospheric Administration (NOAA)-20 and NOAA-21 satellites, together with Fire Radiative Power (FRP) data from the Copernicus Atmosphere Monitoring Service Global Fire Assimilation System (CAMS GFAS) (Figure S8). The daily time series of fire counts shows that the high-resolution (375 m) VIIRS

products indicate a clear decrease in fire counts from 12 to 17 November, and the maximum and cumulative FRP values from CAMS GFAS are also relatively low during this period. Fire emission plumes were probably obscured by the dense fog over these states, so the satellites may have missed those fires (Figure S9). This probably led to underestimated fire counts and biomass-burning emissions in the model, and hence to underestimated SOA. Other factors, such as enhanced aqueous-phase SOA formation under high-humidity and fog conditions, which is not represented in the current scheme, may also have contributed.”

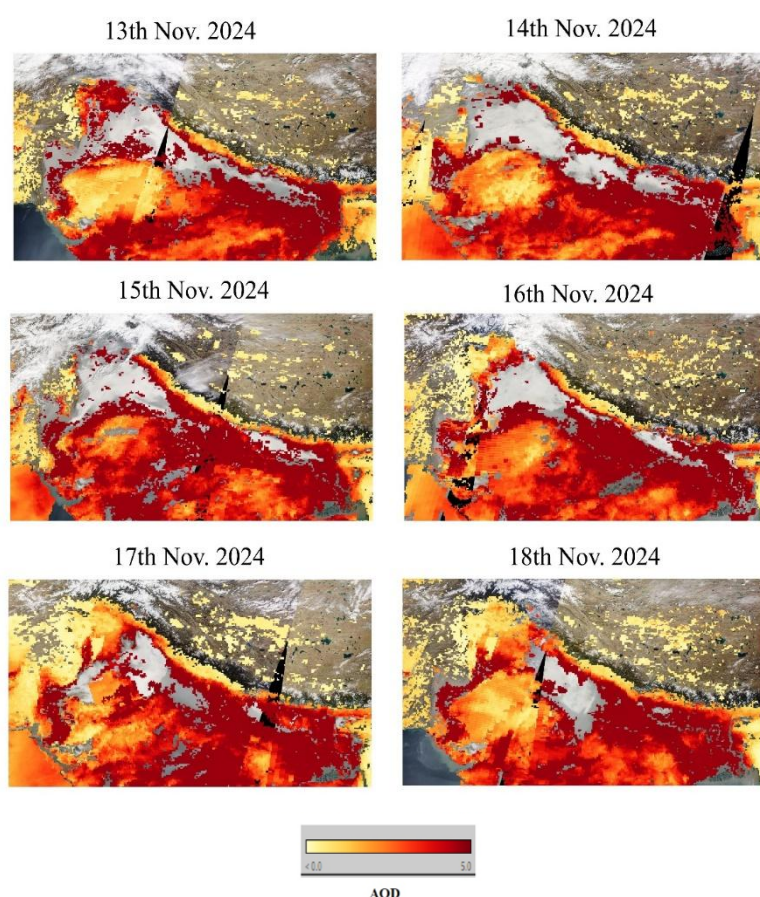


Figure S6: Natural Colour Images with spatial distribution of AOD over Northern India from NASA World View during 13th -18th November 2024.

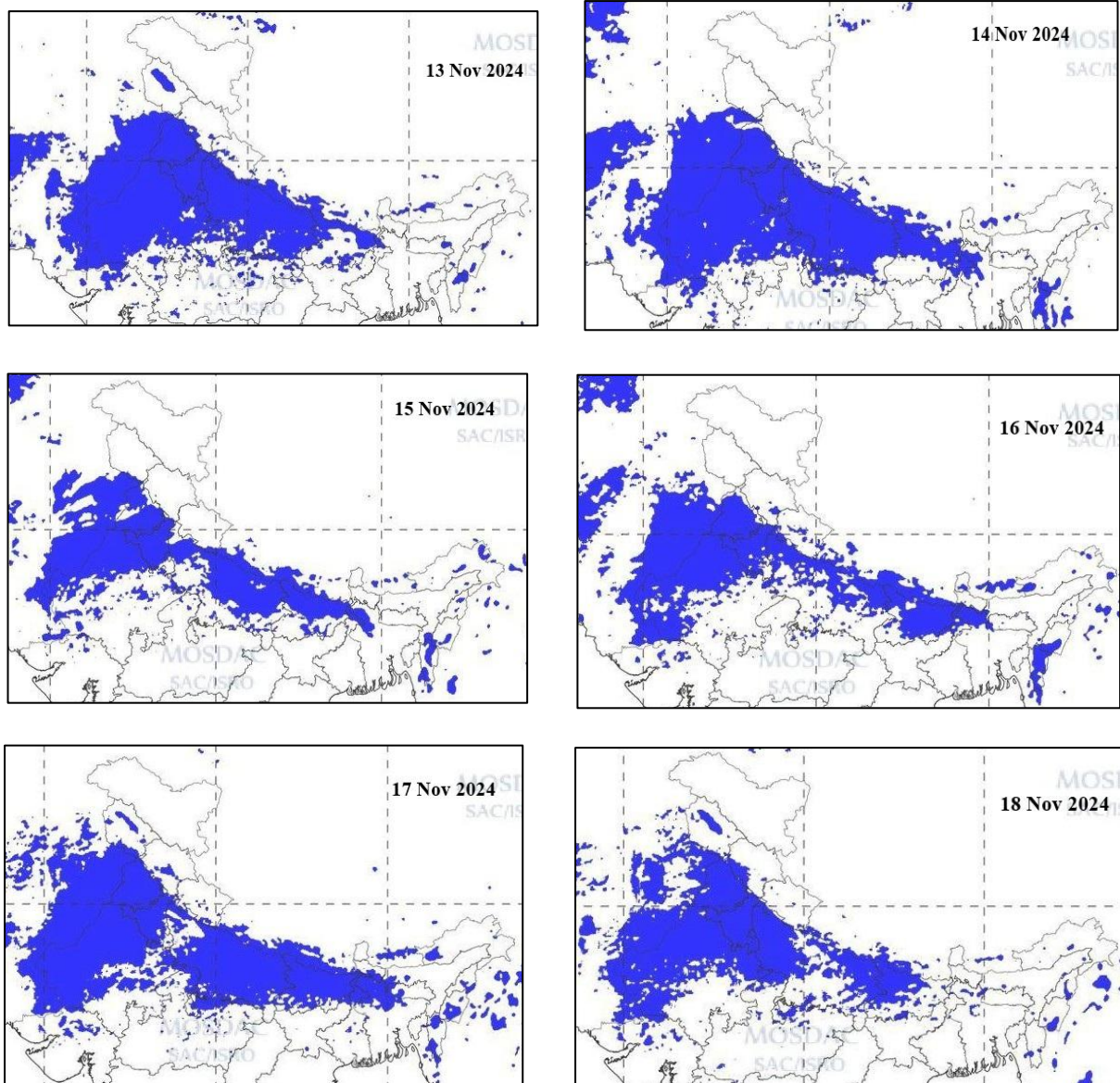


Figure S7. INSAT-3DR satellite-derived fog/low-cloud distribution over northern India from 13 to 18 November 2024.

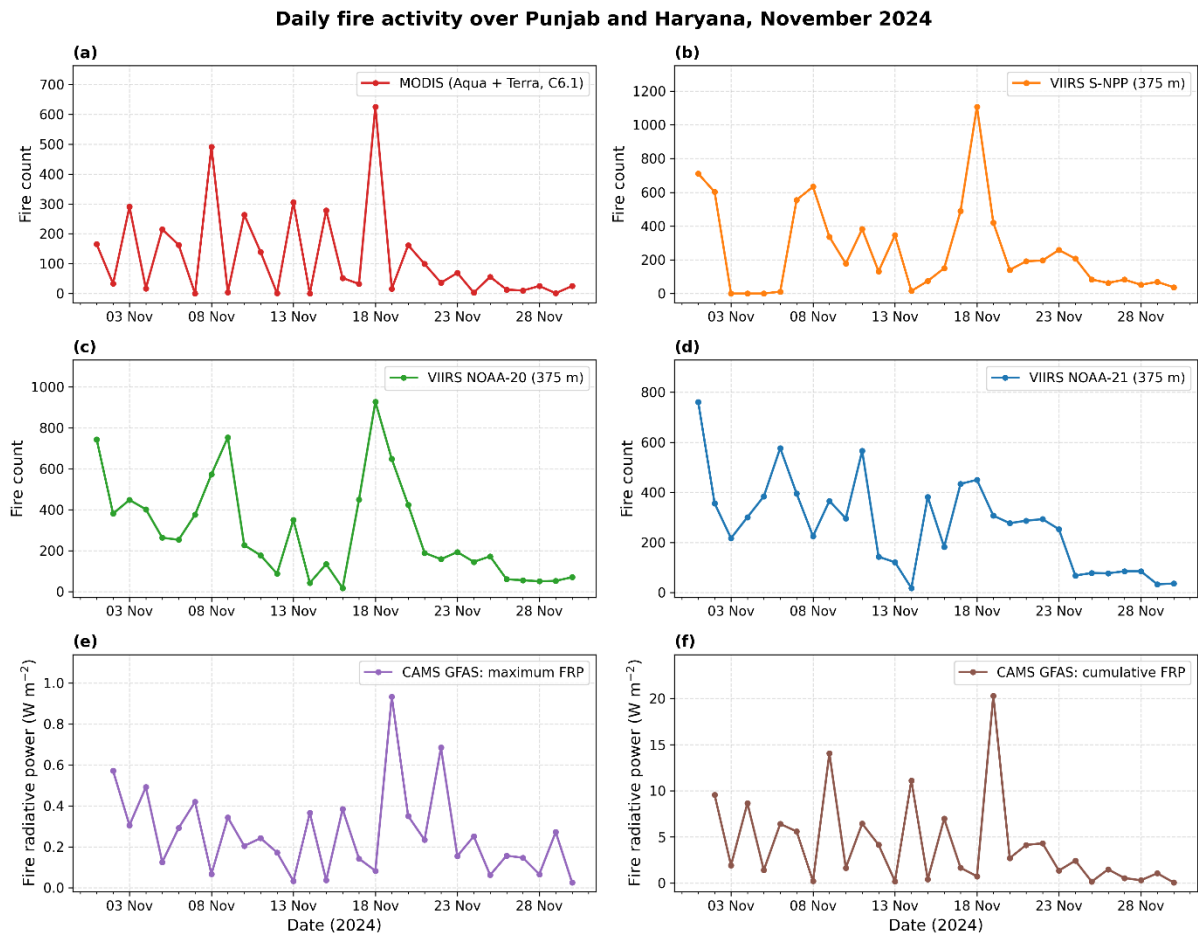


Figure S8: Daily fire counts from MODIS (a), VIIRS S-NPP (b), VIIRS NOAA-20 (c) and VIIRS NOAA-21 (d), and daily maximum (e) and cumulative (f) FRP from CAMS GFAS over Punjab and Haryana during November 2024.



Figure S9: Photograph of crop residue burning plumes recorded on 17.10.2021 near the border of Patiala district and SAS Nagar district in Punjab.

## 5. Comparison with MOSAIC is incomplete

The comparison with MOZART–MOSAIC is interesting but remains somewhat superficial. The authors compare only bulk SOA metrics (e.g., mean concentration, RMSE, and NMB), while important aspects such as diurnal variability, spatial distributions, and OA composition are not discussed. The manuscript would benefit from a more detailed diagnostic comparison to better demonstrate to what extent MOZCART can reproduce the performance of MOZART–MOSAIC despite its much simpler representation of SOA physics and chemistry.

Reply: We agree with your suggestion. In this revision, we have incorporated additional analyses to provide a more detailed comparison of the MOZART–MOSAIC, MOZCART, and RADM2–SORGAM chemical mechanisms used in this study. These include the diurnal variations in SOA concentrations, supported by an analysis of near-surface hydroxyl radical (OH) concentrations, temperature, and planetary boundary layer height (PBLH), as well as the spatial distributions of SOA, primary organic aerosol (POA), and total organic aerosol (OA).

We have added the following information to the revised manuscript (Lines 700–780).

“To further understand this, we have plotted the mean diurnal variation of hydroxyl radical (OH.), near-surface temperature at 2 m (T2), planetary boundary layer height (PBLH) and SOA from the selected chemical schemes (Figure 6). The daytime PBLH magnitude is found to be lower in the MOZART-MOSAIC scheme than in the other two schemes (Figure 6c). Since all the selected schemes share the same meteorological forcing fields, physics parameterizations and anthropogenic emissions, the higher aerosol concentration in MOZART-MOSAIC may be related to its more detailed aerosol chemistry. The resulting aerosol–meteorology feedback could contribute to the shallower boundary layer. The diurnal cycle of T2 (Figure 6b) shows relatively lower values in MOZART-MOSAIC. Lower T2 in MOZART-MOSAIC may also favour the partitioning of semi-volatile material to the particle phase. These factors may partly explain the higher SOA concentrations in MOZART-MOSAIC, in addition to differences in the SOA formation pathways between schemes. The diurnal cycle of T2 shows a consistent rise during the daytime and a reduction at night in all the schemes, as expected. The photochemical production of OH (Figure 6a) is well synchronized with the diurnal T2 pattern across the selected schemes.

The diurnal cycle of SOA (Figure 6d) shows a continuous reduction of SOA concentrations during daytime (06–15 Indian Standard Time (IST)) in MOZART-MOSAIC and RADM2–SORGAM, which then rise and reach stable, higher values during nighttime. In contrast, the observed SOA shows an increase from 06 IST to 09 IST and then a decrease during the daytime. This trend is probably due to the combined effect of photochemical production and PBLH deepening. During the early morning hours (06–09 IST), the PBLH is shallow, so the increase in SOA associated with photochemical production is visible. During the afternoon and late

evening hours, although photochemical production continues, the deepening of the PBLH dominates over the rise due to photochemical production, and hence an overall decrease in concentration is seen in the observed SOA. The decreasing trend in the MOZART-MOSAIC and RADM2-SORGAM simulated SOA during 09–15 IST is also seen in the observed SOA, but the increasing trend during 06–09 IST is completely missed by these two schemes. Similarly, for the updated MOZCART chemistry scheme, a decrease in simulated SOA concentrations is also noticed from 06 to 15 IST, although not as sharp as in the MOZART-MOSAIC and RADM2-SORGAM schemes. To understand this behaviour in MOZCART, we also plotted the diurnal variation of OH concentrations (Figure 6a), which clearly shows broad daytime photochemical OH production in all schemes, accounting for a large part of SOA formation. Interestingly, the daytime OH concentration in the MOZCART scheme, especially during 10–16 IST, is lower than in the other two schemes.

This is probably due to combined effect of differences in the magnitude of precursor VOC species emissions and SOA parametrization approaches between MOZCART, MOZART-MOSAIC and RADM2-SORGAM. Therefore, even though MOZCART and RADM2-SORGAM have nearly similar boundary layer heights during the daytime, MOZCART SOA is closer to the observed daytime values than RADM2-SORGAM. This is further evident from the variation between 06 and 09 IST, when all three schemes have nearly the same PBLH and the OH concentrations in MOZCART and RADM2-SORGAM are nearly the same, which may be associated with differences in SOA production rate in MOZCART than those in MOZART-MOSAIC and RADM2-SORGAM. PBLH may also be overestimated in the models during 06–09 IST, which could contribute to the opposite trend in simulated SOA. An overestimation of wind speed (Sengupta et al., 2022) in the WRF-Chem could further contribute to the underestimation of SOA, particularly during the daytime and early morning hours. The differences may also arise from the complexity of the SOA schemes. MOZART-MOSAIC employs a volatility basis set (VBS) approach that accounts for SOA formation under high and low NO<sub>x</sub> regimes and represents functionalization (ageing) of organic gases through OH oxidation. It is coupled with the MOSAIC sectional aerosol module, which has 4 aerosol size bins, and in each size bin the organic species are tracked in 5 volatility bins. RADM2-SORGAM uses a two-product approach (two volatility bins) to represent SOA formation and tracks organic species in the Aitken and accumulation aerosol modes. In contrast, MOZCART uses a bulk VBS scheme that tracks SOA species in 4 volatility bins without resolving aerosol size bins which makes the VBS in MOZCART computationally cheaper than the two other schemes.

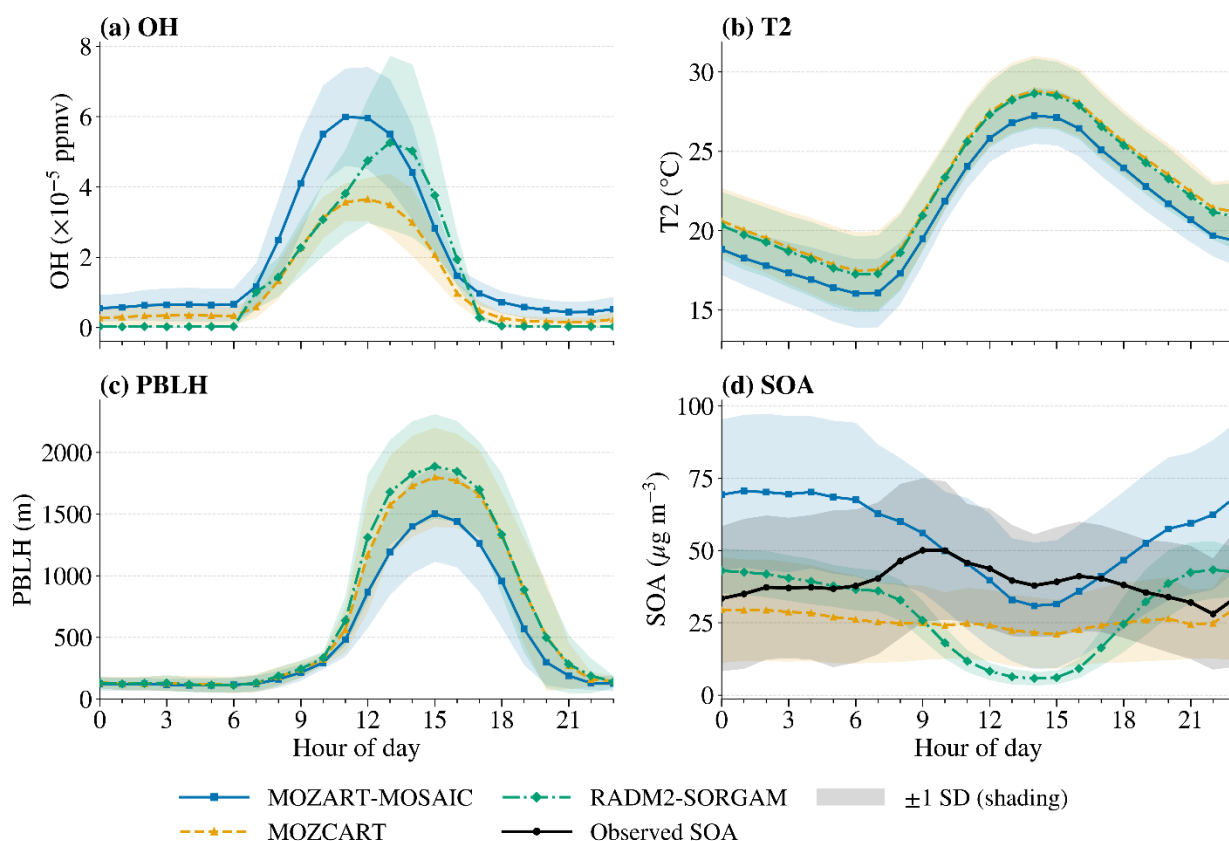


Figure 6. Mean diurnal cycles of (a) OH radical mixing ratio, (b) 2 m air temperature (T2), (c) planetary boundary layer height (PBLH), and (d) secondary organic aerosol (SOA) simulated with the MOZART-MOSAIC, MOZCART, and RADM2-SORGAM chemical schemes at Delhi. Black line shows observed SOA.

The spatial distributions of simulated secondary organic aerosol (SOA), primary organic aerosol (POA) and total organic aerosol (OA) from the MOZART-MOSAIC, RADM2-SORGAM and MOZCART chemistry schemes are presented in Figure 8. Simulated SOA is higher in MOZART-MOSAIC than in the other two schemes, particularly over North India, where concentrations reach  $\sim 60$ – $80 \mu\text{g/m}^3$  over Punjab and Haryana and  $\sim 40$ – $60 \mu\text{g/m}^3$  along the central and lower IGP region. In comparison, MOZCART simulates  $\sim 30$ – $50 \mu\text{g/m}^3$  over the IGP, and RADM2-SORGAM simulates  $\sim 40$ – $50 \mu\text{g/m}^3$  over Punjab and Haryana and only  $\sim 20$ – $30 \mu\text{g/m}^3$  over remaining IGP region. Over Central India and the adjacent ocean, SOA is comparable between MOZART-MOSAIC and MOZCART ( $\sim 20$ – $40 \mu\text{g/m}^3$ ), whereas RADM2-SORGAM shows relatively lower concentrations ( $< 10$ – $20 \mu\text{g/m}^3$ ). POA concentrations over North India are broadly similar in MOZART-MOSAIC ( $\sim 20$ – $40 \mu\text{g/m}^3$ ) and MOZCART ( $\sim 20$ – $30 \mu\text{g/m}^3$ ), and both are higher than in RADM2-SORGAM ( $< 20 \mu\text{g/m}^3$ ). The comparable magnitude with MOZART-MOSAIC and the wider spread of POA in MOZCART could be due to the fact that its POA is the sum of hydrophobic (OC1) and hydrophilic (OC2) organic aerosols. The hydrophobic organic aerosols are assumed to convert into hydrophilic aerosols with an e-folding lifetime of 2.5 days. The sum of SOA and POA, presented as total OA for the three schemes, clearly shows a higher regional OA loading in MOZART-MOSAIC followed by MOZCART and RADM2-SORGAM.

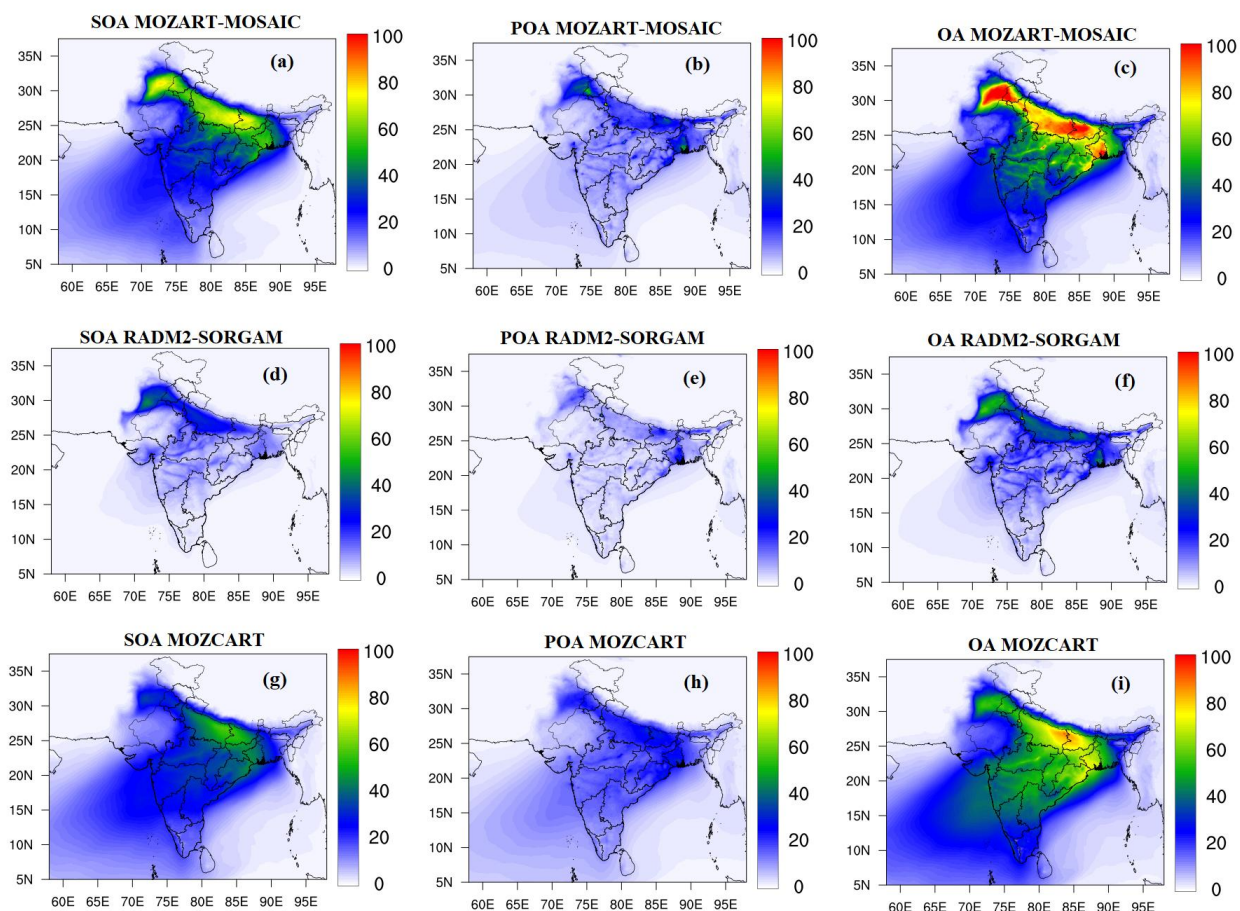


Figure 8: Surface monthly mean concentrations ( $\mu\text{g}/\text{m}^3$ ) of secondary organic aerosol (SOA), primary organic aerosol (POA) and total organic aerosol (OA) over the study domain during November 2024, simulated by WRF-Chem with the MOZART-MOSAIC (a–c), RADM2-SORGAM (d–f) and MOZCART (g–i) chemistry schemes.”

## 6. Clarification needed on AMS size range and comparison with $\text{PM}_{2.5}$

The manuscript states that SOA and OA observations were derived from HR-ToF-AMS measurements of non-refractory  $\text{PM}_{2.5}$ . However, standard Aerodyne AMS/HR-ToF-AMS instruments typically measure non-refractory submicron aerosol ( $\text{NR-PM}_{1.1}$ ). This discrepancy is important because the model evaluation compares modeled SOA/OA in  $\text{PM}_{2.5}$  with AMS-derived OA/SOA. The authors should verify whether their analysis involves a size-range mismatch and, if necessary, revise the comparison accordingly. If such correction is not possible, the manuscript should explicitly discuss how this mismatch may introduce an additional source of uncertainty in the model evaluation and interpretation of the results.

Reply: We thank the reviewer for raising this point, which is an important one for any comparison between an aerosol mass spectrometer and a regulatory  $\text{PM}_{2.5}$  network. We have verified the instrument configuration and we confirm that there is no size range mismatch in our analysis. You are correct that a standard Aerodyne HR-ToF-AMS, fitted with the standard

aerodynamic lens and a standard porous tungsten vaporiser, quantifies the non-refractory submicron fraction, NR-PM<sub>1</sub>. The instrument deployed in this study was not in that configuration. It was equipped with a PM<sub>2.5</sub> aerodynamic lens, which extends the transmission window beyond the submicron range of the standard lens (Williams et al., 2013), and with a capture vaporiser, which greatly reduces particle bounce at the vaporiser surface relative to the standard vaporiser (Hu et al., 2017). The measured and reported quantity is therefore the non-refractory fine aerosol, NR-PM<sub>2.5</sub>, and the organic aerosol mass spectra used for source apportionment correspond to that same size range. The simulated PM<sub>2.5</sub> SOA and OA are consequently compared against observations from the matching size cut, and no size correction or rescaling of the observations is required.

We agree that this was not stated clearly enough in the discussion paper. The revised Sect. 2.2 now specifies the inlet, the lens type and the resulting size cut explicitly, and Text S1 of the Supplement gives the full instrument configuration, calibration and data processing chain. We have also followed the reviewer's instruction to discuss the residual uncertainty. Transmission through an aerodynamic lens is not perfectly flat across the whole accepted size range, and non-refractory material above roughly 2.5 µm vacuum aerodynamic diameter is not transmitted at all, so the reported NR-PM<sub>2.5</sub> should be read as a slight lower bound on the true non-refractory fine mass rather than as an exact closure with a gravimetric or beta-attenuation PM<sub>2.5</sub> measurement. This statement has been added to Text S1.

In the course of verifying the point raised by the reviewer we also identified and corrected a problem in the high-resolution peak fitting that underlies the earlier version of the observational SOA. In the analysis presented in the discussion paper, the m/z calibration of a subset of the runs was poor, and the organic factors were consequently not resolved cleanly. The main consequence was that oxidised biomass-burning organic aerosol was absorbed into the oxygenated factor and interpreted as semi-volatile oxygenated OA, so the oxygenated aerosol assigned to SOA contained a substantial contribution of biomass-burning origin and the reported observed SOA mass concentrations were correspondingly too high. For the revision we repeated the analysis from the raw data: the m/z calibration and baseline were redetermined, the peak width and peak shape were refitted in PIKA, and the SQUIRREL and PIKA processing was repeated before a new PET PMF analysis was carried out step by step.

The corrected four-factor solution resolves a hydrocarbon-like factor (HOA), a primary biomass-burning factor (BBOA), an oxidised biomass-burning factor (O-BBOA) and a more oxidised oxygenated factor (MO-OOA). The observational SOA used in this paper is the MO-OOA factor alone, and the two biomass-burning factors are assigned to primary organic aerosol. The separation of O-BBOA from MO-OOA is the substantive change relative to the discussion paper: the biomass-burning marker ion C<sub>2</sub>H<sub>4</sub>O<sub>2</sub><sup>+</sup> is clearly present in O-BBOA and is at background level in MO-OOA, and the two factors differ in oxidation state and are only weakly correlated in time. As a result, the observed SOA time series has changed and the total SOA mass concentration is lower than in the discussion paper. The diagnostics used to select

the number of factors, the criteria used to identify and name each factor, and the basis for the primary and secondary assignment are set out in full in Text S1 and Tables S1 and S2 of the revised Supplement, with the supporting figures in Figures S1 to S4.

All model evaluation statistics, figures, tables, and the corresponding discussions in the revised manuscript have been updated using the corrected observational SOA data.

Changes made to the manuscript

- Sect. 2.2 now states the PM<sub>2.5</sub> cyclone, the PM<sub>2.5</sub> aerodynamic lens and the resulting NR-PM<sub>2.5</sub> size cut, with the Williams et al. (2013) reference.
- Sect. 2.2 now describes the corrected SQUIRREL, PIKA and PET PMF processing chain, the four resolved factors, and the definition of the observational SOA as the MO-OOA factor.
- Text S1 of the Supplement has been added, covering the instrument configuration and calibration, the high-resolution peak fitting and elemental ratios, the PMF input and error weighting, the selection of the number of factors, the identification of each factor, the consistency of the assignment across the campaign, the separation of primary and secondary organic aerosol, and the data coverage and uncertainty.
- Tables S1 and S2 and Figures S1 to S4 have been added in support of Text S1; the existing supplementary figures and tables have been renumbered accordingly.
- The sentence on aerodynamic lens transmission has been added to Text S1 in direct response to the second part of this comment.
- Based on the revised observed SOA, OA, and BBOA data, the analyses (Tables 2–3), time series plots (Figures 2–4), box plots (Figure 7), and the corresponding discussions in Sections 3.1, “**Performance Evaluation of SOA Parameterization,**” and 3.3, “**Comparison of SOA Formation in MOZCART with Different Chemical Schemes,**” have been revised in the manuscript. Since the parameterization has been revised, the spatial plots (Figure 5) and the corresponding discussion in Section 3.2, “**Spatial Distribution of SOA,**” have also been revised.

”

## 7. Diurnal cycle mismatch deserves deeper analysis

The model peak occurs ~3 hours earlier than observations. The explanation is brief and speculative. The authors should analyze OH diurnal cycle, boundary-layer evolution, transport timing, and possibly nighttime chemistry contributions.

Reply: Thank you for this important comment. Following the revision of the SOA parameterization and the observational SOA data, we have updated the diurnal cycles of the simulated and observed aerosol species. In response to your suggestion, we have also added an

analysis of the diurnal variations in near-surface OH concentrations, planetary boundary layer height (PBLH), and temperature (Figure 6) to help interpret the diurnal variation in SOA and its potential relationship with photochemical activity and boundary-layer evolution. The revised discussion is included in Section 3.3, “Comparison of SOA Formation in MOZCART with Different Chemical Schemes,” alongside the comparison with the other chemical mechanisms. Details of these analyses and revisions are provided in our response to Comment 5. Regarding the contribution of nighttime nitrate-radical chemistry to SOA formation, we would like to emphasize that the default MOZCART chemistry scheme currently does not include nitrate chemistry. Therefore, this process could not be explicitly evaluated in the present study. Work on incorporating a computationally efficient representation of nitrate chemistry into MOZCART is in progress, and the findings will be reported in future studies.

## **8. Missing comparison against inorganic aerosol biases**

Since simulated PM<sub>2.5</sub> improves substantially after adding SOA, it would be useful to know how much of the remaining PM bias is due to inorganic aerosol. A PM<sub>2.5</sub> composition evaluation would be useful.

Reply: Thank you for highlighting this important aspect of PM<sub>2.5</sub> evaluation. We agree that evaluating aerosol composition, particularly secondary inorganic aerosols (SIA), would help quantify their contribution to the remaining PM<sub>2.5</sub> bias. However, the default MOZCART chemistry scheme in WRF-Chem that we are working with does not include SIA chemistry. We are currently working on developing a computationally efficient parameterization of SIA chemistry for MOZCART. Therefore, we are unable to present a comparison of inorganic aerosols to quantify their contribution to the remaining bias. We will address this in our future studies.

## **Minor Comments**

### **1. language issues, typos, and formatting inconsistencies**

Examples include:

- line 281: “practical phase” → “particle phase”

Reply: corrected as suggested

- line 476: “other oxidant like ozone” → “other oxidants such as ozone”

Reply: corrected as suggested

- line 477: “with in” → “within”

Reply: corrected as suggested

- line 520: “fogy conditions” → “foggy conditions”

Reply: corrected as suggested

- inconsistent spacing around units,

Reply: corrected as suggested

- lines 275, 438, 531, Tables 2-3: inconsistent use of  $\mu\text{g}/\text{m}^3$  vs  $\mu\text{g m}^{-3}$ ,

Reply: We have corrected and made it consistent to  $\mu\text{g}/\text{m}^3$  in entire manuscript

- inconsistent notation for OH radicals,

Reply: We have corrected and made it consistent to OH in entire manuscript

- duplicated references (e.g., Cubison et al. 2011 appears twice; DeCarlo et al. 2010 appears twice).

Reply: corrected as suggested

## 1. Figure readability

Several figures are difficult to read:

- labels are too small,
- color contrast is weak,
- time-series are cluttered.

Figures 2–5 especially need larger fonts and clearer legends.

Reply: Thank you for the suggestions. We have revised all the figures using larger fonts and improved colour contrast and removed clutter from the time-series plots.

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