Point-by-point response to Reviewer #1's comments

(original comments in black, our responses in purple, original manuscript in blue, changes in manuscript in green)

General Comments:

This manuscript provides a comprehensive and timely assessment of India's 2022 $PM_{2.5}$ air pollution using an updated WRF-Chem modeling framework and substantially improved emission inventories, especially for the residential and power sectors. The integration of a plant-level emissions dataset, updated residential fuel-use information, and modified near-surface mixing represent meaningful advances beyond previous national-scale source attribution studies. The authors also perform rigorous model evaluation using 288 surface sites and satellite AOD, providing a relatively robust validation for India. The study contributes useful insights, particularly the finding that industrial emissions surpassed residential sources as the largest domestic contributor to national population-weighted $PM_{2.5}$ in 2022, while transboundary pollution remains the largest overall contributor. These findings are policy-relevant and address an existing gap in the literature.

However, several aspects require clarification or strengthening before publication. These include (1) uncertainties associated with the updated inventories, (2) treatment of nonlinear chemical responses in the source-removal experiments, (3) reconciliation between $PM_{2.5}$ and AOD biases, and (4) clearer articulation of limitations, especially regarding coarse PM and chlorine-containing species, and (5) a more event-focused, daily-resolution evaluation to verify model skill during rapid changes and extreme episodes. Addressing these points will improve interpretability and robustness.

Overall, the manuscript is clearly written, logically structured. Subject to satisfactory revision, it could be considered for publication.

We thank Reviewer #1 for their comprehensive and constructive review of our manuscript. We have carefully considered all their specific comments. Below, we provide a point-by-point response detailing how each issue has been addressed.

Specific comments:

However, several major issues need to be addressed before the manuscript can be considered for publication.

1. Clarification and Quantification of Emission Inventory Uncertainties

The manuscript incorporates substantial updates to residential and power sector emissions (Lines160-270), but the associated uncertainty ranges are not quantified. The authors should provide uncertainty bounds for the

residential fuel-use regression, the coal composition–based emission factor derivation, and the plant-level coal consumption estimation, or alternatively include a table that summarizes the main sources of uncertainty and their likely impacts on simulated $PM_{2.5}$ concentrations.

We thank the reviewer for this suggestion We have addressed the request to estimate uncertainty in two parts: 1) quantifying specific uncertainties within our updated emission inventories, and 2) estimating the uncertainty in simulated source attribution values resulting from our updated emission inventories for the residential and power sectors. We provide a specific estimation for the sectors where we introduced major updates (i.e., the coal-fired power sector and the residential sector). The following information appears in the **Supplementary Information 4** (original manuscript in blue, changes in manuscript in green):

Emission Inventory Uncertainties:

1. **Global Inventories**: We cannot present quantitative uncertainty ranges for sectoral emissions directly adopted from global inventories (CEDS, EDGAR, and FINN), as these datasets do not provide uncertainty estimates for air pollutant emissions in India.
2. **Updated Coal-fired Power Plant Inventory**:
   Activity Data: We directly retrieve plant-level electricity generation and coal consumption reports from the Central Electricity Authority (CEA) of India. We treat this government-reported data as authoritative, and thus focus our uncertainty quantification on emission factors.
   Emission Factors: we establish region-specific uncontrolled emission factors for both domestic coal and imported coal used in India's power plants (**Supplementary Table 1**). We average the calculated emission factor for a given region if multiple coal composition datasets are found, and present one standard deviation from these calculated values as the uncertainty bounds.
   Total Emissions: We estimate annual total emissions from coal-fired power generation across India in 2022 to be 6.2±1.5 Tg for SO_2, 4.6±0.3 Tg for NO_x, and 0.8±0.1 Tg for primary $PM_{2.5}$, with the range reflecting uncertainties in emission factors without mitigation as reflected in the literature.
3. **Updated Residential Emission Inventory**: The residential inventory developed in an earlier study (Velamuri et al., 2024) explicitly conducted an uncertainty analysis using 10,000 Monte-Carlo simulations by sampling emission factors and activity data based on assumed probability distributions for activity levels and emission factors, then extracting 95% confidence bounds from the simulated distribution of total $PM_{2.5}$ emissions. The earlier study assumed the following uncertainties for the residential sector:
   Activity Data: population (±2.5%), fuel penetration ratios (±5%), and fuel consumption statistics (±5%). Source data is census data and national family health survey reports.
   Emission Factors: ±25%. Source data is from The Energy and Resources Institute (TERI)'s emission inventory development report (The Energy and Resources Institute, 2021).
   Total Emissions: ±50% for $PM_{2.5}$.

Impact of emission uncertainties on annual source attribution results

We perform a simplified uncertainty propagation analysis for the power and residential sectors to quantify uncertainties associated with their emissions. We approximate uncertainties in the annual national population-weighted mean $PM_{2.5}$ concentration, assuming a near-linear relationship between annual sectoral total emissions and concentration at national level. While we acknowledge limitations of this approach as it does not fully capture non-linearities in secondary chemistry or aerosol-meteorology feedbacks, it serves as a reasonable first-order approximation for the rough estimation. We applied the uncertainty percentages from our emission inventories (as detailed above) to each attributed $PM_{2.5}$ component for these sectors. We then sum the component-level uncertainties to derive the total uncertainty for the sector's contribution. These estimated uncertainty ranges are presented in **Supplementary Table 7**.

Table S1. Uncontrolled emission factors for coal and lignite from various regions used in India's power sector

Source Region	Fuel Classification	CO_2 (g/kg fuel)	SO_2 (g/kg fuel)	NO_x as NO_2 (g/kg fuel)	$PM_{2.5}$ (g/kg fuel)
Assam	Domestic Coal	2329.0±232.9	10.8±1.1	6.0±0.6	1.8±0.2
Chhattisgarh		1204.7±272.7	6.6±0.1		11.4±1.3
Jharkhand		1493.7±408.3	6.8±2.1		9.7±2.2
Madhya Pradesh		1517.0±237.6	6.6±1.2		7.9±3.6
Maharashtra		1501.3±93.0	10.9±7.3		8.1±2.7
Odisha		1062.8±184.3	5.6±1.5		12.7±1.3
Telangana		1396.7±139.7	6.4±0.6		12.2±1.2
Uttar Pradesh		1294.4±129.4	9.2±0.9		10.6±1.1
West Bengal		1810.3±245.9	6.3±1.8		8.5±3.2
Australia	Imported Coal	1913.4±577.2	10.7±7.0		4.5±3.8
Indonesia		1913.4±577.2	10.7±7.0		4.5±3.8
South Africa		2307.2±230.7	8.3±0.8		3.9±0.4
Gujarat, Rajasthan, and Tamil Nadu	Lignite	1250.8±125.1	5.2±0.5	4.0±0.4	1.1±0.1

Source Data: (Kalenga et al., 2011; Mittal et al., 2012; Yunus et al., 2014; Cheepurupalli et al., 2015; Gogoi, 2018; The Singareni Collieries Company Limited, 2018; Dwivedi and Kumar, 2022; U.S. Environmental Protection Agency)

Table S7 Uncertainty estimation of 2022 national annual population-weighted mean $PM_{2.5}$ component concentrations for the power and residential sectors

Species	Domestic Sources

	Power	Residential
Organic	0.42±0.05	6.60±3.30
BC	0.01±0.00	0.58±0.29
Dust	0.99±0.12	0.13±0.06
Sulfate	2.80±0.67	-0.01±0.00
Nitrate	0.81±0.05	-0.01±0.00
Ammonium	1.11±0.25	-0.01±0.00
Total $PM_{2.5}$	**6.11±1.14**	**7.28±3.65**

Regarding the uncertainty bounds for the residential fuel-use regression, the inventory we adopted did not present a standalone "regression uncertainty bound". However as mentioned above, it includes an emission uncertainty analysis using 10,000 Monte-Carlo simulations that consider uncertainties from regression-dependent terms such as fuel use shares (Velamuri et al., 2024).

To reflect these SI updates in the Main Text, we added one new paragraph in the **Uncertainty and limitation** section (**Page 31 Line 848-854** in the clean version of the revised main manuscript).

"Our annual source attribution results are subject to input uncertainty from emission inventories. Focusing on the sectors with India-specific updates, we estimate annual national population-weighted mean $PM_{2.5}$ contributions of 6.1±1.1 μg/m^3 for the power sector and 7.3±3.7 μg/m^3 for the residential sector. These estimates account for uncertainties in activity data, emission factors, and residential fuel use (see **Supplementary Information 4** and **Supplementary Table 7** for detailed quantification). We do not quantify uncertainties for sectors relying on global inventories (CEDS, EDGAR, FINN) due to the lack of India-specific uncertainty estimates for air pollutants in these datasets."

We believe these clarifications, combined with the revision, address the concern associated with this comment.

2. Source Attribution and Non-linearity (Section 2.4)

The authors use a "zero-out" method combined with a scaling factor (Equation 7) to force the sum of contributions to match the baseline concentration. While this is a common approach to handle non-linearity in chemistry, it can introduce biases. For species in highly non-linear regimes (e.g., nitrate and ammonium, as shown in Figure 10), it is not obvious that linearly scaling the "difference" accurately represents each source's contribution.

We thank the reviewer for this insightful comment. We acknowledge that this linear scaling can introduce biases, particularly for species like nitrate and ammonium. However, we adopted this scaling approach to ensure mass

closure, which is essential for providing policy-relevant source attribution where the sum of all sources' contributions must equal the baseline concentration.

To quantitatively address this concern, we updated **Supplementary Table 6** (attached below). In this table, we now present both the scaled contributions (values outside parentheses) and the raw sensitivity estimates (values inside parentheses, directly derived from baseline minus zero-out simulations) for each $PM_{2.5}$ component and total $PM_{2.5}$.

A comparison between the scaled and raw values indicates that the scaling method results in moderate changes in national annual population-weighted mean concentrations for total $PM_{2.5}$ and its components attributed to a given source. The scaled concentrations moderately differ from the raw calculations by -5% (i.e., total $PM_{2.5}$ from domestic natural dust) to -20% (i.e., total $PM_{2.5}$ from the domestic agriculture sector). While we acknowledge that the scaling modifies the attributed contribution based on the raw sensitivity test, we believe this adjusted value is necessary for a mass-balanced budget. Furthermore, providing both values in Supplementary Table 6 acknowledges the non-linearities and allows readers to compare the raw contribution with scaled contribution.

Table S6. Source contribution to 2022 national annual population-weighted mean $PM_{2.5}$ component concentrations

	Domestic Sources*								**Transboundary Sources**
	POW	**IND**	**RES**	**TRA**	**AGR**	**FIRE**	**DST**	**BVOC**	
Organic	0.4 (0.4)**	2.4 (2.9)	6.6 (7.9)	0.7 (0.8)	0.1 (0.1)	1.3 (1.5)	0.1 (0.1)	<0.1 (<0.1)	2.9 (3.4)
BC	<0.1 (<0.1)	0.9 (1.1)	0.6 (0.7)	0.2 (0.2)	<0.1 (<0.1)	0.2 (0.2)	<0.1 (<0.1)	<0.1 (<0.1)	0.3 (0.3)
Dust	0.9 (1.0)	1.9 (2.2)	0.1 (0.2)	0.5 (0.6)	0.1 (0.1)	1.3 (1.5)	1.8 (2.0)	<0.1 (<0.1)	4.6 (5.3)
Sulfate	2.3 (2.7)	1.5 (1.7)	<0.1 (<0.1)	0.3 (0.3)	0.2 (0.2)	0.1 (0.1)	<0.1 (<0.1)	-0.2 (-0.3)	1.7 (2.0)
Nitrate	1.4 (1.6)	1.1 (1.3)	<0.1 (<0.1)	1.6 (2.0)	2.4 (3.0)	0.5 (0.6)	<0.1 (<0.1)	-0.1 (-0.1)	1.9 (2.2)
Ammonium	1.2 (1.5)	0.8 (1.0)	<0.1 (<0.1)	0.6 (0.7)	0.8 (1.1)	0.2 (0.2)	<0.1 (<0.1)	-0.1 (-0.1)	1.1 (1.3)
Sodium and Chloride	<0.1 (<0.1)	<0.1 (<0.1)	<0.1 (<0.1)	<0.1 (<0.1)	0.1 (0.1)	<0.1 (<0.1)	<0.1 (<0.1)	<0.1 (<0.1)	0.4 (0.4)
Total	**6.1 (7.4)**	**8.6 (10.1)**	**7.3 (8.8)**	**3.8 (4.7)**	**3.7 (4.6)**	**3.6 (4.1)**	**1.9 (2.0)**	**-0.5 (-0.5)**	**12.8 (14.9)**

*Abbreviations and their corresponding full names: POW=Power, IND=Industry, RES=Residential, TRA=Transportation, AGR=Agriculture, FIRE=Smoke from Open Burning, DST=Natural Dust, BVOC=Biogenic.

**Values outside parentheses are scaled contributions calculated using Equation 7, while values inside parentheses represent raw estimates directly derived from (baseline minus individual-source-zeroed-out) model simulations.

We added a new subsection in **Supplementary Information 2.1** to address this comment:

"We scale model simulation results (Main Text **Section 2.4**) to ensure mass closure, which forces the sum of all sources' contributions to equal the baseline concentration (with all sector emissions on) within each WRF-Chem grid. In **Supplementary Table 6**, we present both the scaled contributions and the raw sensitivity estimates (directly derived from baseline minus individual-source-zeroed-out simulations) for each $PM_{2.5}$ component and total $PM_{2.5}$. A comparison between the scaled and raw values indicates that the scaling method results in moderate changes in national annual population-weighted mean concentrations for total $PM_{2.5}$ and its components attributed to a given source. The scaled concentrations moderately differ from the raw calculations by from -5% (i.e., total $PM_{2.5}$ from domestic natural dust) to -20% (i.e., total $PM_{2.5}$ from the domestic agriculture sector). While we acknowledge that the scaling modifies the attributed contribution relative to the raw sensitivity test, we believe this adjusted value is necessary for a mass-balanced budget."

3. The "Simple SOA" Scheme (Section 2.1.2)

The manuscript implements the GEOS-Chem "simple SOA" scheme into WRF-Chem. This scheme uses fixed yields and is computationally efficient, and was originally designed for global, coarser-resolution models. The authors should comment on whether this scheme is sufficiently robust for a regional model at 27 km resolution, particularly in capturing the diurnal variability of SOA in urban hotspots such as Delhi.

We thank the reviewer for highlighting the importance of validating the simplified SOA scheme at a regional scale. We added a new subsection in **Supplementary Information 1.3** to address this comment:

"The Simple SOA scheme is computationally efficient. In addition, it has also been demonstrated to be robust for regional air quality modeling over Asia. For instance, an earlier study applied this scheme at a 50km resolution and found that, even without modification, it captured observed organic carbon concentrations in polluted urban and suburban environments in China, comparable to more complex, process-based SOA schemes (Miao et al., 2021).

To further support the use of this scheme for India at 27-km resolution, we assessed whether our modeled OA concentrations are within a realistic range. Due to the lack of publicly available ground-based $PM_{2.5}$ speciation data for 2022, we compared our modeled OA concentrations with the COALESCE network (Maheshwarkar et al., 2022; Venkataraman et al., 2024), which provides OA observation data from 11 sites across India for 2019. The comparison shows that, on an annual scale, the average OA concentration simulated by WRF-Chem across these 11 sites is 13.0±10.0 $\mu g/m^3$, which is consistent with the observed average of 13.5±8.4 $\mu g/m^3$. Specifically, for the site near Delhi (Rohtak, Haryana), the model simulated an annual OA concentration of 19.3 $\mu g/m^3$ compared to the observed 30.0 $\mu g/m^3$. While the model underestimates OA by ~30% near Delhi, we consider

this performance reasonable given the temporal mismatch (simulating 2022 vs. observing 2019) and the likely decreasing trend in primary OA and SOA precursor emissions under India's National Clean Air Programme (NCAP) launched in 2019.

However, our simulation reports a relatively small annual national population-weighted mean OA concentration (<0.1 μg/m^3) attributed to biogenic emissions from within India. We note that this low estimate likely reflects two factors: (1) The distinct spatial separation between major biogenic sources (e.g., the India-Myanmar border region and Western Ghats) and dense population centers; (2) The inherent limitations of the simple SOA scheme, which has been reported to underestimate the global isoprene-derived SOA budget by ~50% compared to explicit, complex chemistry schemes (Pai et al., 2020). Consequently, our regional simulation provides a conservative lower-bound estimate for biogenic SOA yields in India.

Further evaluation of the simple SOA scheme in India would require contemporaneous observations and the explicit speciation of SOA (distinguishing it from total OA), which is not currently available in the published COALESCE dataset."

Regarding the reviewer's specific comment on diurnal variability, we note that the publicly available COALESCE data is at daily resolution, which precludes a direct validation of the modeled diurnal OA cycle. However, we have evaluated the model's ability to capture the hourly evolution of total surface $PM_{2.5}$ in the Indo-Gangetic Plain (IGP) and Delhi in **Figure 4** (panels b-c attached below). Our baseline simulation (the dark red lines) reasonably reproduces the observed diurnal patterns (the black lines) in both IGP and Delhi, despite underestimating the morning peak likely due to misrepresentation of hourly emissions. Thus, we believe our model, with simple the SOA scheme, adequately represents diurnal variations of OA in polluted environments like IGP and Delhi.

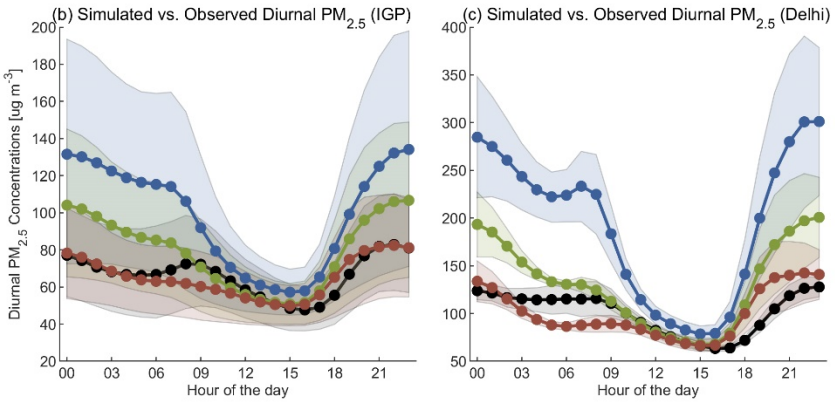

Figure 4. Comparison of $PM_{2.5}$ performances among three emission and model configurations in 2022. The configurations are: (1) Default (in blue)— official WRF-Chem v4.6.1 with the GEOS-Chem simple SOA scheme, driven by the standard CEDS and EDGAR inventories as described in **Section 2.2.1**; (2) Default + Emission Updates (in green)— configuration (1) plus sectoral updates for the residential and power sectors, as well as road dust, as

described in **Sections 2.2.1** and **2.2.2**; (3) Default + Emission Updates + Mixing Updates (in red)— configuration (2) plus improved near-surface mixing of chemical species, this configuration is adopted in the Baseline simulation as mentioned in **Table 1** and **Figures 3** and **5**. Observations from CPCB and US-AirNOW are shown in black. Model performance is evaluated using (**b**–**c**) annual mean $PM_{2.5}$ diurnal patterns for the Indo-Gangetic Plain (IGP) and Delhi, respectively. In **b-c**, shaded areas indicate one standard deviation across available grid cells in each region noted in the panel.

4. Interpretation of Biogenic Contributions (Section 3.3)

The results show a net negative contribution of biogenic emissions to $PM_{2.5}$ due to oxidant depletion (consumption of OH/HO_2) that reduces secondary inorganic aerosol formation. While chemically plausible, this might be confusing for some readers, who could misinterpret a "negative contribution" as implying that biogenic emissions improve air quality. The authors should clarify in the text that biogenic emissions lower secondary inorganic $PM_{2.5}$ by constraining oxidants, but still contribute to organic aerosol loadings, as indicated in Figure 9 where biogenic sources contribute to the organic component, albeit modestly.

We thank the reviewer for this useful comment. We agree that the "net negative" contribution of biogenic emissions to $PM_{2.5}$, driven by oxidant depletion, could be misinterpreted.

We have clarified in the text that biogenic emissions indeed contribute to OA formation, but in our simulations, this mass contribution is outweighed by the reduction in inorganic $PM_{2.5}$. Specifically, isolating domestic biogenic emissions (from within India) reveals a relatively small contribution to the annual national population-weighted mean OA concentration (<0.1 $\mu g/m^3$). We note that this low estimate likely reflects two factors: (1) The distinct spatial separation between major biogenic sources (e.g., the India-Myanmar border region and Western Ghats) and dense population centers; (2) The inherent limitations of the simple SOA scheme, which has been reported to underestimate the global isoprene-derived SOA budget by ~50% compared to explicit, complex chemistry schemes (Pai et al., 2020). Consequently, our regional simulation provides a conservative lower-bound estimate for biogenic SOA yields in India.

In contrast, these domestic biogenic emissions (from within India) drive a 0.5 $\mu g/m^3$ decrease in secondary inorganic $PM_{2.5}$ concentration (annual national population-weighted mean) via oxidant consumption. Thus, the net effect of domestic biogenic emissions in our model framework is a reduction in annual population-weighted $PM_{2.5}$.

However, O_3 substantially increases result from the biogenic emissions (included in the updated **Supplementary Figure 6**, attached below). This suggest biogenic emissions from within India deteriorate O_3 air quality.

Changes are made in the Main Text (**Page 24 Line 653-663** in the clean version of the revised manuscript, original sentences in blue, changes in green) to address this comment:

"Domestic biogenic emissions had a small but net negative contribution to annual PW mean $PM_{2.5}$ concentrations across India (-1%; -0.5 μg/m^3). They contribute to OA formation, but in our simulations, this mass contribution (<0.1 μg/m^3) is outweighed by the reduction in inorganic $PM_{2.5}$ (-0.5 μg/m^3). We find a reduction in all secondary inorganic $PM_{2.5}$ components (i.e., sulfate, nitrate, and ammonium), as well as in the sulfate oxidation ratio and nitrate oxidation ratio, following the inclusion of domestic biogenic emissions in the WRF-Chem model (**Supplementary Figure 6**). The inclusion of biogenic VOCs reduced the oxidation capacity of the atmosphere by consuming OH and HO_2 radicals, thereby decreasing the conversion of SO_2 to sulfate and NO_2 to nitrate, which also reduces ammonium and leads to reductions in secondary inorganic $PM_{2.5}$. Consequently, biogenic emissions from within India resulted in a slight net reduction in total $PM_{2.5}$ concentrations in 2022. However, they significantly enhanced annual mean O_3 concentrations (by up to 20 μg/m^3, **Supplementary Figure 6**), deteriorating O_3 air quality across India."

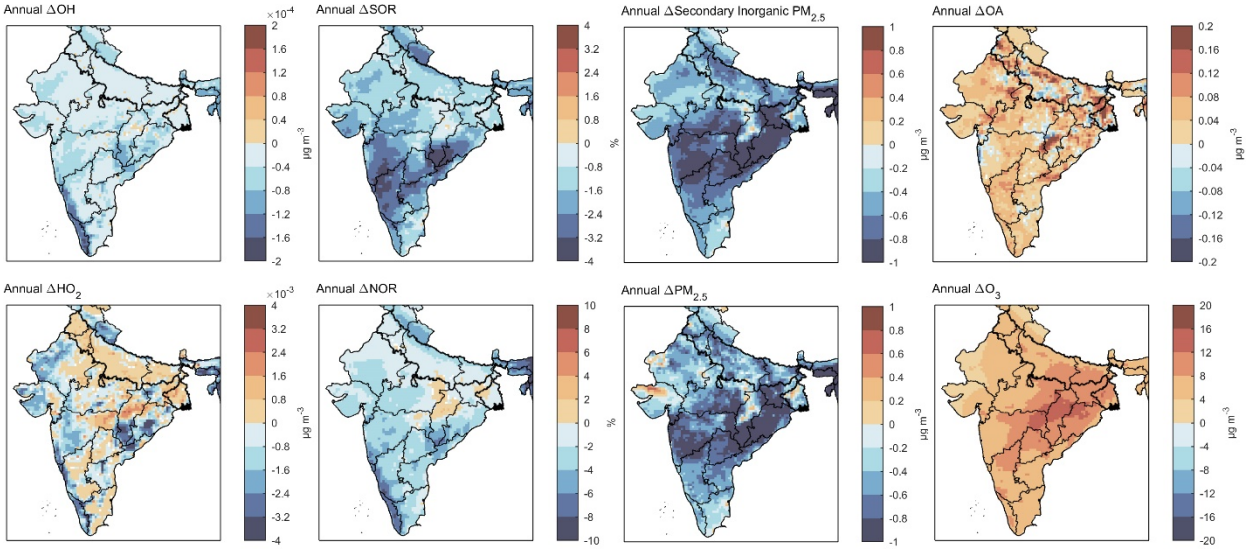

Figure S6. Impact of including Indian BVOC emissions (via turning on the MEGAN module within India) on atmospheric chemistry. All panels are calculated as baseline simulation minus $BVOC_{off}$ simulation. Annual results are calculated as the average of January, April, July, and October simulations. Sulfur Oxidation Ratio (SOR) and Nitrogen Oxidation Ratio (NOR) are calculated as $(SO_4^{2-} + H_2SO_4)/(SO_4^{2-} + H_2SO_4 + SO_2)$ and $(NO_3^- + HNO_3)/(NO_3^- + HNO_3 + NO_2)$, respectively. The thick black line denotes the boundary of the Indo-Gangetic Plain (IGP).

5. AOD-$PM_{2.5}$ Discrepancy

The model strongly underestimates AOD (−29±14%, Lines 510-524) despite achieving reasonably good agreement for surface $PM_{2.5}$. The authors should clarify whether this discrepancy is mainly due to missing coarse PM sources (as suggested on Section 2.2), and whether assumptions related to hygroscopic growth or aerosol optical properties (e.g., the internal-mixing treatment in MOSAIC) may also contribute to the bias.

We have added **Supplementary Information 2.2** to address this comment. Please note that we have consolidated our responses to Comments 5, 6, 7, and 9 into **Supplementary Information 2.2** to provide a unified update.

"We believe that missing coarse PM (PM_{coarse} = PM_{10}-$PM_{2.5}$) is one of the primary drivers for the AOD underestimation, as we find that coarse PM concentrations from our baseline simulation are underestimated by 37.3±25.7 μg/m^3 (59±41%) compared to CPCB measurements across India (**Supplementary Figure 3**). In addition, earlier modeling studies over India also found coarse particles to be responsible for underestimated simulated AOD, and found that the coarse particle AOD had to be scaled by monthly factors ranging from 1 to 12 to bring the simulated AOD closer to observed AOD (David et al., 2018; Singh et al., 2021). In our study, this underestimation of PM_{coarse} (and consequently AOD) is likely attributable to missing emission sources beyond fugitive road dust. While our updated inventory includes fugitive road dust (1.1 Tg/yr of PM_{coarse}), it does not currently account for dust from construction activities or industrial fugitive emissions. For context, the updated SMoG inventory for 2019 reports total fugitive $PM_{2.5}$ of 0.77 Tg/yr for 2019 (Venkataraman et al., 2024), which is more than double our estimate of 0.33 Tg/yr which accounts for road dust only. While SMoG does not explicitly report PM_{coarse}, this fugitive $PM_{2.5}$ discrepancy suggests that the inclusion of construction and industrial fugitive dust for PM_{coarse} would significantly reduce the modeled bias in both surface PM_{coarse} concentrations and AOD.

In addition, the contrast between the model's strong performance of $PM_{2.5}$ on annual/monthly scales and its limitations during daily extreme events suggests that while our total $PM_{2.5}$ emission magnitude is reasonable, the high-frequency temporal allocation (daily/hourly variability) is not fully representative of local episodic patterns:

- **Anthropogenic Emissions (Monthly Resolution)**: The raw inventory provides data at a monthly resolution. In our current configuration, we distribute these monthly totals evenly across each day (with prescribed sectoral diurnal profiles). Consequently, this approach inherently misses high-frequency temporal variability and episodic spikes, such as weekend-weekday patterns, specific festivals (e.g., Diwali), and sudden surges in residential emissions during extreme cold events.
- **Fire Emissions (Daily Resolution)**: We use the satellite-based FINN dataset, which provides daily resolution. While this captures day-to-day variability better than anthropogenic sources, satellite-based inventories can still underestimate emissions from small-scale agricultural fires or be hindered by cloud cover and thick haze during the most intense phase of pollution events.

- **Natural Sources (Online Calculation)**: Both natural dust and biogenic emissions are calculated online within WRF-Chem based on real-time meteorology. While these sources avoid the temporal resolution limitations of offline inventories, they remain sensitive to uncertainties in static input parameters (e.g., land use type, surface erodibility, leaf area index) and biases in simulated meteorology (e.g., wind speed).

We also acknowledge that the internal-mixing assumption used in the MOSAIC aerosol scheme (where all chemical components within a size bin are assumed to be a homogeneous mixture) simplifies the complex reality of aerosol morphology. This approach likely underestimates the absorption enhancement (lensing effect) that occurs when absorbing aerosols (e.g., black carbon) are coated by scattering shells (e.g., sulfate or organics), thereby leading to lower simulated extinction and AOD.

Bias in hygroscopic growth of aerosols likely contribute to the AOD underestimation. Observational studies indicate that high local emissions of hydrochloric acid in India partition into aerosol water, substantially enhancing aerosol water uptake and sustaining particle growth (Gunthe et al., 2021). However, our simulation relies on global emission inventories (CEDS, EDGAR, FINN) that do not currently include these anthropogenic chloride sources. Although this omission was found to have a relatively small impact on dry surface $PM_{2.5}$ concentrations (e.g., contributing to 3–4 $\mu g/m^3$ in $PM_{2.5}$ concentrations in the IGP during January–March 2018) (Patel et al., 2024), it leads to reduced aerosol hygroscopic growth (especially at high relative humidity), contributing to the underprediction of AOD.

Finally, limitations in the representation of the shape of the aerosol size distribution may also lead to AOD bias. In our study, we use the MOSAIC sectional aerosol scheme with four bins (dry diameters: 0.039–0.156, 0.156–0.625, 0.625–2.5, and 2.5–10.0 μm). While this approach avoids the constraints of a single-mode lognormal assumption, the coarse resolution of four bins may be insufficient to resolve specific optical scattering modes. If the ambient aerosol population contains sharp scattering peaks (e.g., specific accumulation mode sizes) that are averaged out within these broad bins, this leads to inaccuracies in the calculated extinction efficiency and AOD, even if the total $PM_{2.5}$ mass is simulated reasonably."

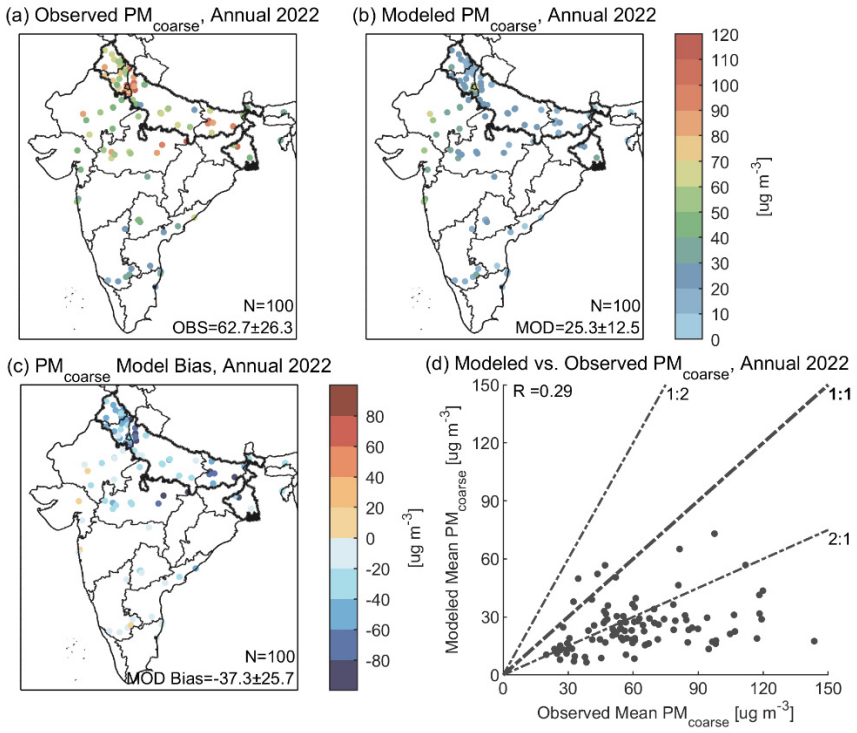

Figure S3. Comparison of observed and modeled surface PM_{coarse} concentrations in 2022 in the baseline simulation. Measurement stations with at least 80% valid hourly data during the four-month period (January, April, July, and October) are selected. Multiple measurements within a single WRF-Chem grid cell are averaged before comparison with WRF-Chem. Annual value is estimated by averaging $PM_{2.5}$ concentrations during the four-month period. In map plots, N denotes the number of grid cells used for evaluation, and the other numbers represent the mean ± one standard deviation across all grid cells. The thick black line denotes the boundary of the Indo-Gangetic Plain (IGP). In the scatter plot in d), R is the Pearson correlation coefficient between observed and modeled annual mean concentrations across all grid cells.

6. Coarse PM_{coarse} Underestimation

The manuscript notes that PM_{coarse} is strongly underestimated compared with CPCB observations (Line 522), but this is not quantified. The authors should explicitly report the magnitude of the PM_{coarse} bias and to discuss potential missing or underestimated sources, such as road dust beyond the adjustments already made for transportation, construction dust, and industrial fugitive emissions.

We thank the reviewer for highlighting the need to quantify the underestimation of coarse particulate matter. To provide the requested quantitative context, we modified the sentence in **Page 19 Line 533-535** in the clean version of the revised manuscript (original manuscript in blue, changes in manuscript in green): "Consistent

with this, our baseline simulation underestimates annual mean surface coarse particulate matter (PM_{coarse} = particles with diameters > 2.5 µm and ≤ 10 µm) by 37.3±25.7 µg/m^3 (59±41%) compared to CPCB measurements (Supplementary Figure 3)."

We agree with the reviewer that this underestimation is likely driven by missing fugitive dust sources beyond road dust. Our current inventory adjustments only account for fugitive road dust from the transportation sector (i.e., 0.33 Tg/yr for $PM_{2.5}$ and 1.1 Tg/yr for PM_{coarse}). However, construction dust and industrial fugitive emissions, are not currently included. The Speciated Multipollutant Generator (SMoG) inventory (IIT Bombay) reports a total fugitive $PM_{2.5}$ emission of 0.77 Tg/yr for 2019 (Venkataraman et al., 2024), which is more than double our road-dust-only adjustment. While SMoG does not explicitly report PM_{coarse}, the scaling implies that if all fugitive sources (construction, industrial, and road) were included, the total coarse dust emissions would be significantly higher than our current input, potentially resolving much of the modeled underestimation in both surface PM_{coarse} concentrations and AOD.

Additional revision in **Supplementary Information 2.2** regarding this response is provided in our reply to the 5th comment.

7. Extreme-Event Underestimation and Time-Varying Emissions

The model underestimates both surface $PM_{2.5}$ and, to an even greater extent, AOD during extreme events. Fires from Myanmar, internal crop burning from within the domain, and severe air pollution events from small industries located outside of city centers are not captured, especially during their most intense phase. The authors should clarify whether this is related to a time-changing emissions dataset that is not currently considered.

We thank the reviewer for this comment. We acknowledge that while the model performs well on annual time scale, it underestimates concentrations or AOD during extreme pollution episodes (e.g., January in Delhi) and high-AOD events (as shown in **Figure 3** and **Supplementary Figure 2**, attached in our reply to the 8th comment).

We clarify that this discrepancy is likely related to the temporal resolution of our emissions dataset. The contrast between the model's strong performance on annual/monthly scales and its limitations during daily extreme events suggests that while our total emission magnitude is reasonable, the high-frequency temporal allocation (daily/hourly variability) is not fully representative of local episodic patterns.

Revision in **Supplementary Information 2.2** regarding this response is provided in our reply to the 5th comment.

8. Daily-Resolution Evaluation

Since the model has daily-resolution data, comparisons should be made with daily data. It is essential to do this, especially during times when aerosol emissions and removal change rapidly (e.g., biomass burning and monsoon arrival).

We refer the reviewer to **Figure 3** (panels e-h attached below) and **Supplementary Figure 2** (timeseries plots attached below), where we include daily evaluation of $PM_{2.5}$ and AOD, respectively.

We refer the reviewer to **Page 17 Line 483-487** in the revised manuscript, where the discussion on $PM_{2.5}$ daily model evaluations is provided (this section remains consistent with the original submission): "WRF-Chem also effectively captures daily $PM_{2.5}$ variations throughout the four-month period (**Figure 3e-h**). In the IGP and Delhi, the Pearson correlation coefficients between modeled and observed regional mean daily $PM_{2.5}$ concentrations are 0.93 and 0.81, respectively. In addition, the twin-peak pattern in diurnal $PM_{2.5}$ concentrations are well reproduced by WRF-Chem, though the morning peak in Delhi is underestimated."

We added one sentence to **Page 19 Line 530-532** in the revised manuscript where the discussion on AOD daily model evaluations is provided: "Evaluation of daily AOD model values demonstrates that while the model performs well in capturing general AOD trends throughout the year, it fails to reproduce the magnitude of extreme AOD events over Delhi and the IGP."

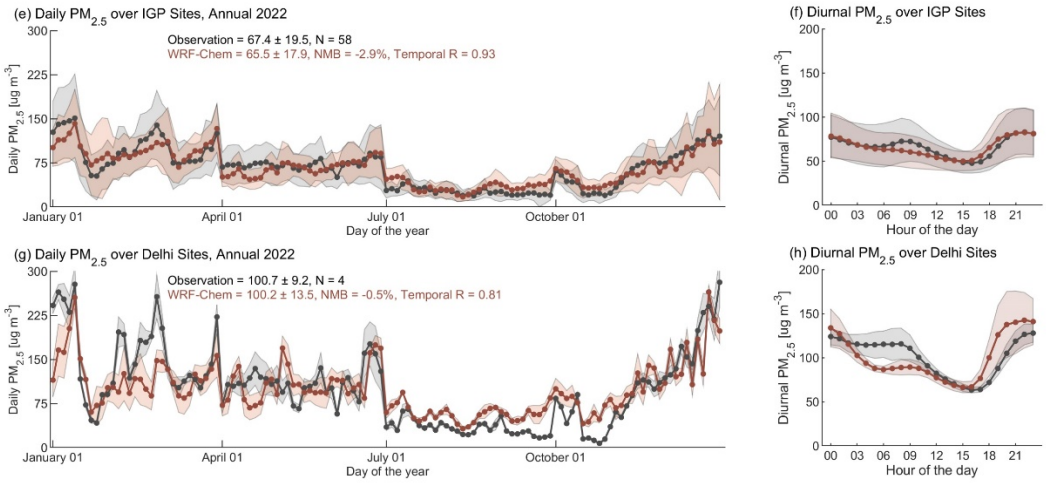

Figure 3. Comparison of observed and modeled surface $PM_{2.5}$ concentrations in 2022. Measurement stations with at least 80% valid hourly data during the four-month period (January, April, July, and October) are selected. Multiple measurements within a single WRF-Chem grid cell are averaged before comparison with WRF-Chem. **e** and **g**, comparison of daily mean surface $PM_{2.5}$ concentrations between observations and WRF-Chem simulations in the IGP (**e**) and Delhi (**g**). **f** and **h** are the same as **e** and **g**, but for annual mean $PM_{2.5}$ diurnal variations. In **e-h**, red and black

lines represent $PM_{2.5}$ concentrations for observations and simulations, respectively, averaged across available grid cells in each region noted in the panel, with shaded areas indicating one standard deviation. Temporal R is the Pearson correlation coefficient between the red and black lines in each panel. See **Supplementary Table 4** for $PM_{2.5}$ model

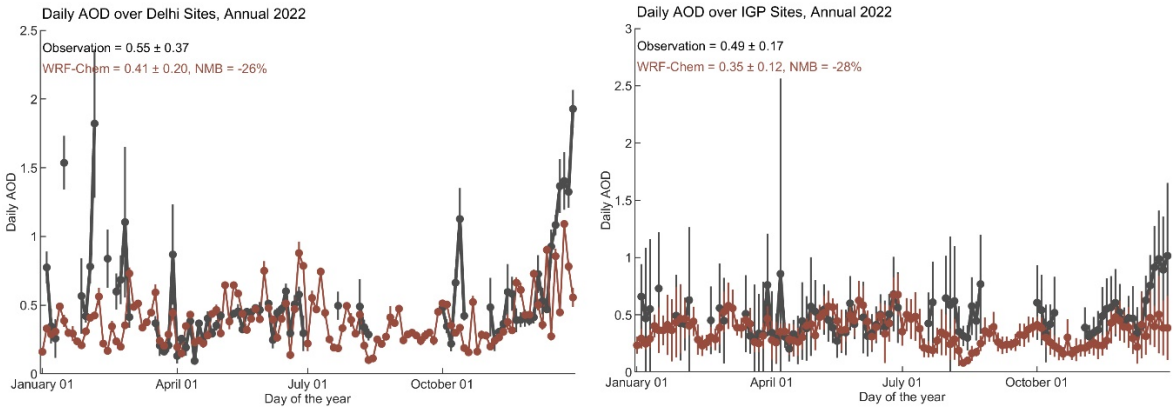

Figure S2. Comparison of WRF-Chem AOD against MODIS satellite-derived AOD at 550 nm in 2022. Daily WRF-Chem AOD is derived using mean AOD during 10:00 to 14:00 local time when same day MAIAC AOD is available. In timeseries plots, we sample MAIAC and WRF-Chem AOD at grids with available surface $PM_{2.5}$ measurements in each region, and present daily AOD data if at least 50% of the grids (with available $PM_{2.5}$ measurements) in that region have valid AOD data.

9. Coarse-Mode/Mixing Discussion: Alternative Possibility

The authors should add to the coarse-mode argument and the mixing argument that another possibility is that the particle sizes are still fine, but have multiple peaks (i.e., not reasonably represented by a single-peaked lognormal, as assumed). The authors should also note that absorption and extinction enhancement due to mixing may contribute to the discrepancy.

We agree with the reviewer that the representation of the aerosol size distribution shape (specifically the presence of multiple modes) is critical for AOD calculation.

We clarify that our simulations use the MOSAIC sectional aerosol scheme, which divides the aerosol size distribution into discrete bins (4 bins in this study: 0.039–0.156, 0.156–0.625, 0.625–2.5, and 2.5–10.0 μm for dry diameter) rather than assuming a fixed single-peaked lognormal distribution (as in modal schemes). Theoretically, this allows our simulation to represent multi-modal distributions (e.g., distinct accumulation and coarse modes).

However, we acknowledge that the discrete bin resolution in our configuration may still limit the model's ability to resolve fine-scale multi-modal features. If the actual ambient distribution contains sharp optical scattering

peaks (e.g., specific accumulation mode sizes) that fall between or are averaged within our model bins, this could indeed lead to an underestimation of extinction efficiency and AOD, even if the total $PM_{2.5}$ mass is simulated reasonably. We have added this to our discussion of AOD uncertainties in **Supplementary Information 2.2:**

Revision in **Supplementary Information 2.2** regarding this response is provided in our reply to the 5th comment.

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