

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 (Lines 160-270), 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 SO₂, 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
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	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/m³ for the power sector and 7.3±3.7 µg/m³ 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 $13.0 \pm 10.0 \mu\text{g}/\text{m}^3$, which is consistent with the observed average of $13.5 \pm 8.4 \mu\text{g}/\text{m}^3$. Specifically, for the site near Delhi (Rohtak, Haryana), the model simulated an annual OA concentration of $19.3 \mu\text{g}/\text{m}^3$ compared to the observed $30.0 \mu\text{g}/\text{m}^3$. 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 ($<0.1 \mu\text{g}/\text{m}^3$) 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 $\sim 50\%$ 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 $\text{PM}_{2.5}$ 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.

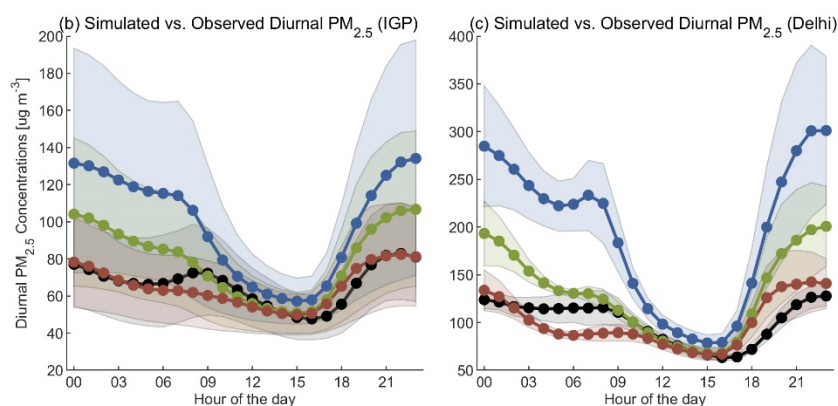


Figure 4. Comparison of $\text{PM}_{2.5}$ performances among three emission and model configurations in 2022. The configurations are: (1) Default (in blue)— official WRF-Chem v4.6.1 with the GEOS-Chem simple SOA scheme, driven by the standard CEDS and EDGAR inventories as described in **Section 2.2.1**; (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/HO₂) 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 µg/m³). 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 µg/m³ 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, O₃ substantially increases result from the biogenic emissions (included in the updated **Supplementary Figure 6**, attached below). This suggest biogenic emissions from within India deteriorate O₃ 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/m³). They contribute to OA formation, but in our simulations, this mass contribution (<0.1 µg/m³) is outweighed by the reduction in inorganic PM_{2.5} (-0.5 µg/m³). 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 HO₂ radicals, thereby decreasing the conversion of SO₂ to sulfate and NO₂ 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 O₃ concentrations (by up to 20 µg/m³, Supplementary Figure 6), deteriorating O₃ air quality across India.”

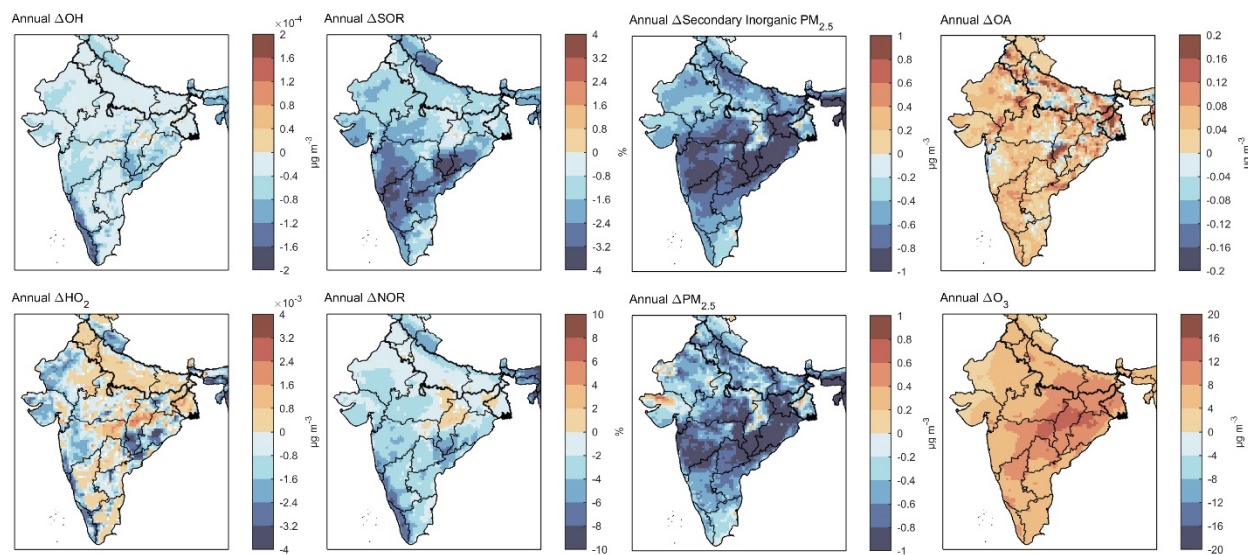


Figure S6. Impact of including Indian BVOC emissions (via turning on the MEGAN module within India) on atmospheric chemistry. All panels are calculated as baseline simulation minus BVOC_{off} simulation. Annual results are calculated as the average of January, April, July, and October simulations. Sulfur 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 ($-29\pm 14\%$, Lines 510-524) despite achieving reasonably good agreement for surface $\text{PM}_{2.5}$. 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 ($\text{PM}_{\text{coarse}} = \text{PM}_{10} - \text{PM}_{2.5}$) is one of the primary drivers for the AOD underestimation, as we find that coarse PM concentrations from our baseline simulation are underestimated by $37.3\pm 25.7 \mu\text{g}/\text{m}^3$ ($59\pm 41\%$) 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 $\text{PM}_{\text{coarse}}$ (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 $\text{PM}_{\text{coarse}}$), 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 $\text{PM}_{2.5}$ 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 $\text{PM}_{\text{coarse}}$, this fugitive $\text{PM}_{2.5}$ discrepancy suggests that the inclusion of construction and industrial fugitive dust for $\text{PM}_{\text{coarse}}$ would significantly reduce the modeled bias in both surface $\text{PM}_{\text{coarse}}$ concentrations and AOD.

In addition, the contrast between the model's strong performance of $\text{PM}_{2.5}$ on annual/monthly scales and its limitations during daily extreme events suggests that while our total $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ concentrations (e.g., contributing to 3–4 $\mu\text{g}/\text{m}^3$ in $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ mass is simulated reasonably.”

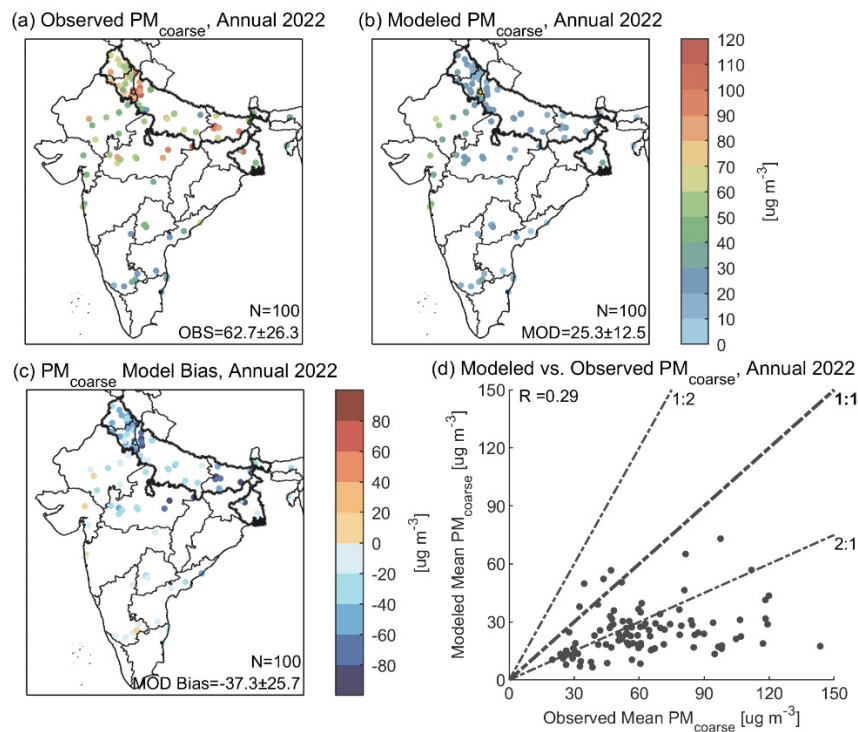


Figure S3. Comparison of observed and modeled surface PM_{coarse} concentrations in 2022 in the baseline simulation. Measurement stations with at least 80% valid hourly data during the four-month period (January, April, July, and October) are selected. Multiple measurements within a single WRF-Chem grid 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 $\pm$ 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 $> 2.5 \mu m$ and $\leq 10 \mu m$) by $37.3 \pm 25.7 \mu g/m^3$ ($59 \pm 41\%$) 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.”

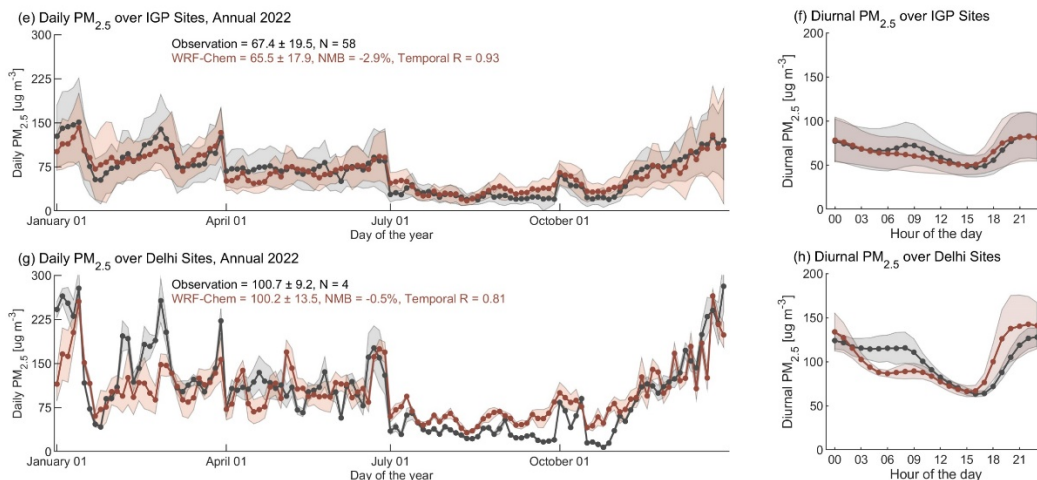


Figure 3. Comparison of observed and modeled surface $PM_{2.5}$ concentrations in 2022. Measurement stations with at least 80% valid hourly data during the four-month period (January, April, July, and October) are selected. Multiple measurements within a single WRF-Chem grid cell are averaged before 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

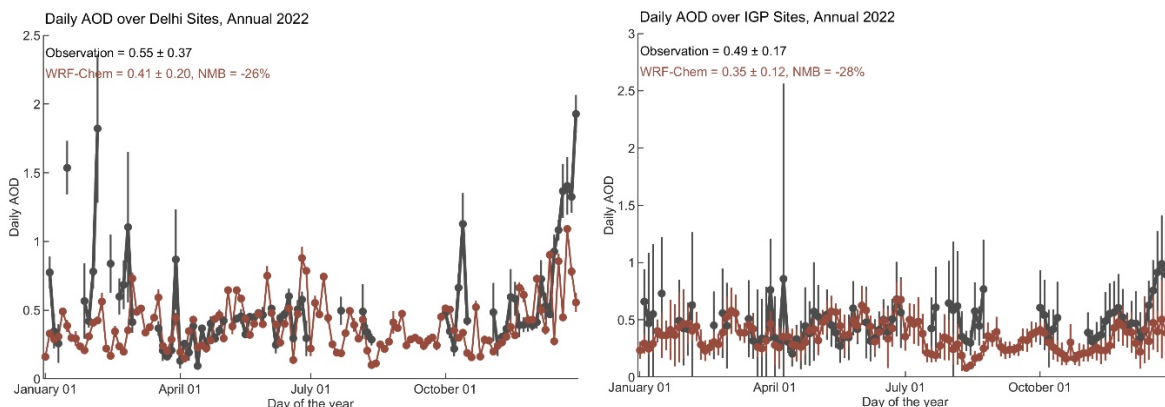


Figure S2. Comparison of WRF-Chem AOD against MODIS satellite-derived AOD at 550 nm in 2022. Daily WRF-Chem AOD is derived using mean AOD during 10:00 to 14:00 local time when same day MAIAC AOD is available. In timeseries plots, we sample MAIAC and WRF-Chem AOD at grids with available surface PM_{2.5} 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.

Point-by-point response to Reviewer #2's comments

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

General Comments:

This study presents a comprehensive regional air quality modeling study for India in 2022 using WRF-Chem, incorporating updated sectoral emissions (notably residential fuel transition and a new plant-level coal power inventory), selected model physics/chemistry improvements, and extensive evaluation against surface PM_{2.5} and satellite AOD observations. The finding that industry has become the largest domestic contributor to population-weighted PM_{2.5}, is potentially impactful and very timely for understanding the changes in India's PM source structure, especially post COVID. I think the paper is suitable for publication after several clarifications.

We thank Reviewer #2 for their positive feedback and their appreciation of our research. We have carefully considered all their specific comments and provide a point-by-point response below.

Specific comments:

1. The authors approximate annual PM_{2.5} using simulations from January, April, July, and October. While this approach is common in resource-limited modeling studies, it may not be suitable for India. The seasons of India is typically characterized as pre-monsoon, monsoon and winter seasons, rather than the four seasons that can be roughly represented by the four months. Each season also has unique pollution characteristics. I suggest the authors re-organize their simulation and discussion according to the Indian seasons. This may better reveal the contribution of different sectors to annual PM concentration.

We thank the reviewer for highlighting the unique characteristics of Indian seasonality. Indeed, the categorization of seasons in India can be complex due to the year-to-year variability in monsoon onset and retreat. However, while some analyses may group the year into three main seasons, the air quality research community analyzing India widely recognizes four distinct seasons to capture the full range of meteorological and emission variability: Winter (Dec–Feb), Pre-Monsoon (Mar–May), Monsoon (Jun–Sep), and Post-Monsoon (Oct–Nov). This four-season structure is widely adopted in recent literature (Lan et al., 2022; Venkataraman et al., 2024; Zhou et al., 2024; Xie et al., 2024; Kumar et al., 2025). We also note that while some studies may classify September as Post-Monsoon or early June as Pre-Monsoon or December as Post-Monsoon, the four-season structure remains the prevailing framework in India air quality analysis (Conibear et al., 2018; Kota et al., 2018; Reddington et al., 2019; Maheshwarkar et al., 2022; Pai et al., 2022; Sharma and Mauzerall, 2022; Sharma et al., 2023; Chen et al., 2025).

To justify our selection of these four months and to address the reviewer’s comment, we have revised the **Methods** section (**Page 4-5 Line 89-93** in the clean version of our revised manuscript; original sentences in blue, new sentences in green):

“We conduct simulations for 2022, the most recent year with available emission inventories for India (Section 2.2), using one month to represent each season: January for winter, April for pre-monsoon, July for monsoon, and October for post-monsoon (**Supplementary Information 1.1**). This four-season structure is widely adopted in recent literature to represent the distinct pollution and meteorological characteristics of each season (Lan et al., 2022; Venkataraman et al., 2024; Zhou et al., 2024; Xie et al., 2024; Kumar et al., 2025).”

We also added **Supplementary Information 1.1**, which includes detailed justification for our selection of four months:

“The air quality research community analyzing India widely recognizes four distinct seasons to capture the full range of meteorological and emission variability: Winter (Dec–Feb), Pre-Monsoon (Mar–May), Monsoon (Jun–Sep), and Post-Monsoon (Oct–Nov). This four-season structure is widely adopted in recent literature (Lan et al., 2022; Venkataraman et al., 2024; Zhou et al., 2024; Xie et al., 2024; Kumar et al., 2025). We also note that while some studies may classify September as Post-Monsoon or early June as Pre-Monsoon or December as Post-Monsoon, the four-season structure remains the prevailing framework in India air quality analysis (Conibear et al., 2018; Kota et al., 2018; Reddington et al., 2019; Maheshwarkar et al., 2022; Pai et al., 2022; Sharma and Mauzerall, 2022; Sharma et al., 2023; Chen et al., 2025).

Our simulation months were selected to represent the distinct pollution and meteorological characteristics of each season:

- 1) January (Winter): Captures peak anthropogenic emissions and stagnant weather conditions (e.g., low surface wind speed and strong near-surface temperature inversion) (Zhou et al., 2024).

- 2) April (Pre-Monsoon): Captures dust events, pre-monsoon biomass burning, and higher temperatures.
- 3) July (Monsoon): Captures monsoon-relevant large precipitations and inflow of ocean air.
- 4) October (Post-Monsoon): Captures the unique transition period from the monsoon season to winter season, which is heavily influenced by agricultural crop residue burning (Sembhi et al., 2020; Kumari et al., 2021; Lan et al., 2022).”

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

2. The study used a 100% emission off strategy to estimate the contributions. This is a quite large perturbation, which may result in unrealistic responses of some nonlinear processes. For example, secondary aerosols respond nonlinearly to precursor removal. Some discussion of these potential limitations is needed, and the attribution results should be interpreted as “effective contributions under 100% removal”, and if possible, I recommend the authors add a limited sensitivity test (e.g., 20–30% reduction for one sector) to demonstrate the magnitude of nonlinearity.

We thank the reviewer for this important suggestion. We fully agree that complete emission removal introduces nonlinearities in secondary aerosol chemistry and chemistry-meteorology feedbacks, which must be accounted for when interpreting source attribution results.

We’d like to clarify that we have already conducted the recommended sensitivity tests to address the reviewer’s concern. As detailed in **Section 4 (Discussion)**, attached below with changes), **Supplementary Figure 8** (attached below with only figure title changes), and **Figure 10** (attached below with only caption changes), we performed simulations with a 20% partial emission reduction for the three Indian sectors (Industrial, Residential, and Agricultural sectors) to quantify the magnitude of nonlinearity.

We selected these specific sectors to capture different chemical regimes:

- 1) Industrial and Residential Sectors: Selected to represent sources where the resulting ambient $PM_{2.5}$ concentrations are dominated by primary $PM_{2.5}$ components.
- 2) Agricultural Sector: Selected to represent sources where the resulting ambient $PM_{2.5}$ concentrations are dominated by secondary $PM_{2.5}$ components.

We summarized our findings in **Section 5 (Uncertainty and limitation)** Page 32 Line 873-886 in the clean version of the revised manuscript), with original manuscript in blue and changes in green:

“Nonlinear secondary aerosol chemistry limits the direct application of our results to real-world emission regulations, particularly for sources dominated by $PM_{2.5}$ precursor emissions whose reductions have a nonlinear effect on resulting $PM_{2.5}$ concentrations, since emission control policies typically require partial rather than

complete reductions. To address this limitation, we conduct three additional simulations where we individually reduce emissions from industrial, residential, and agricultural sectors by 20% (**Table 1**). For sources dominated by primary PM_{2.5} components, such as the industrial and residential sectors, the differences between complete removal and scaled partial reductions are small at the national level: national spatial mean and population-weighted mean PM_{2.5} concentration reductions differ by less than 7% and 3%, respectively, between a 100% emission reduction and a fivefold scaling of 20% reductions (**Supplementary Figure 9**). However, for the agricultural sector, a 20% emission reduction results in a 25% smaller reduction in national PM_{2.5} concentrations (after a fivefold scaling) than a 100% emission reduction. This nonlinearity is primarily due to India's overall NH₃-rich environment (**Figure 10**), where nitrate availability limits secondary inorganic aerosol formation. This suggests that partial removal of NH₃ is less effective—defined as concentration decrease per unit emission reduction—in mitigating PM_{2.5} than substantial NH₃ emission reductions, especially in northern India.”

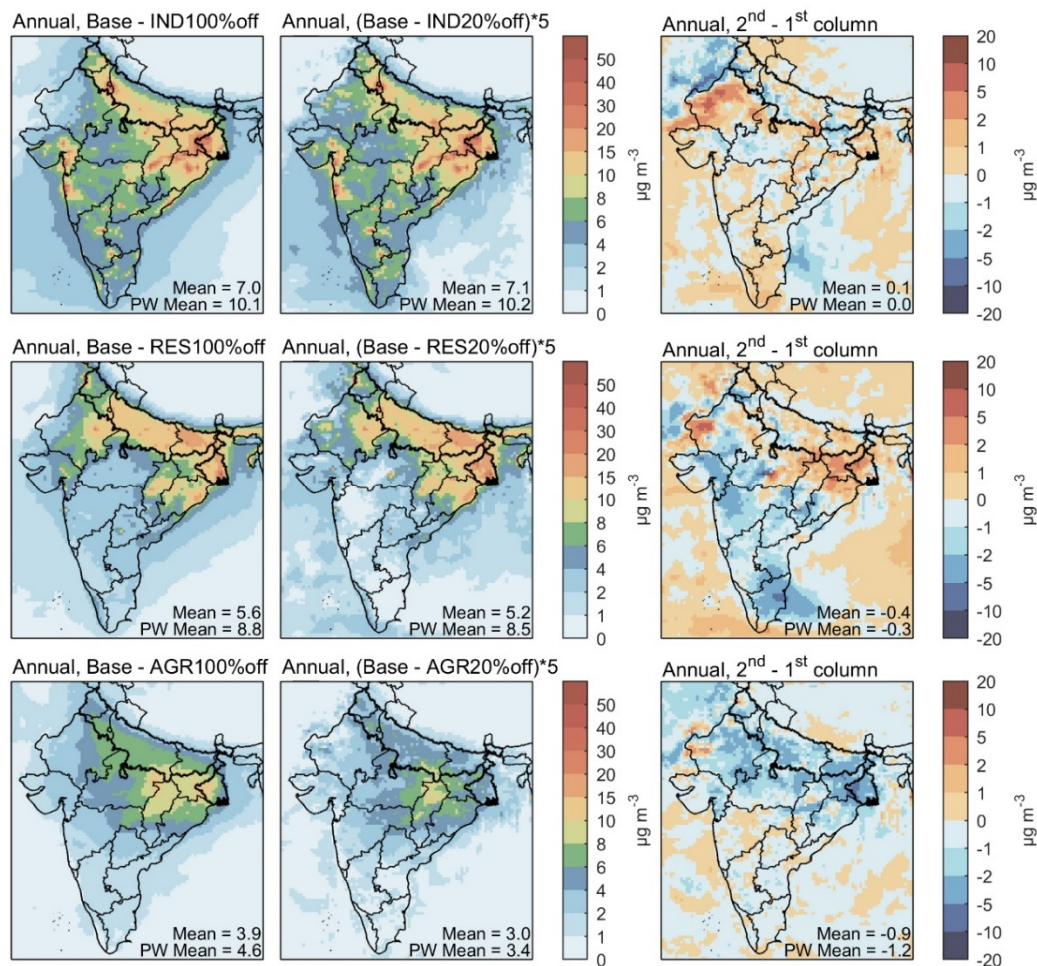


Figure S9. Comparison of PM_{2.5} concentration changes (presenting as absolute values) between a 100% emission reduction and a fivefold scaling of 20% reductions. We show results for three sectors: industry (IND), residential (RES), and agriculture (AGR). The thick black line denotes the boundary of the Indo-Gangetic Plain (IGP).

Unlike results presented in Main text figures, we **do not** scale concentrations in this figure using Equation 7.

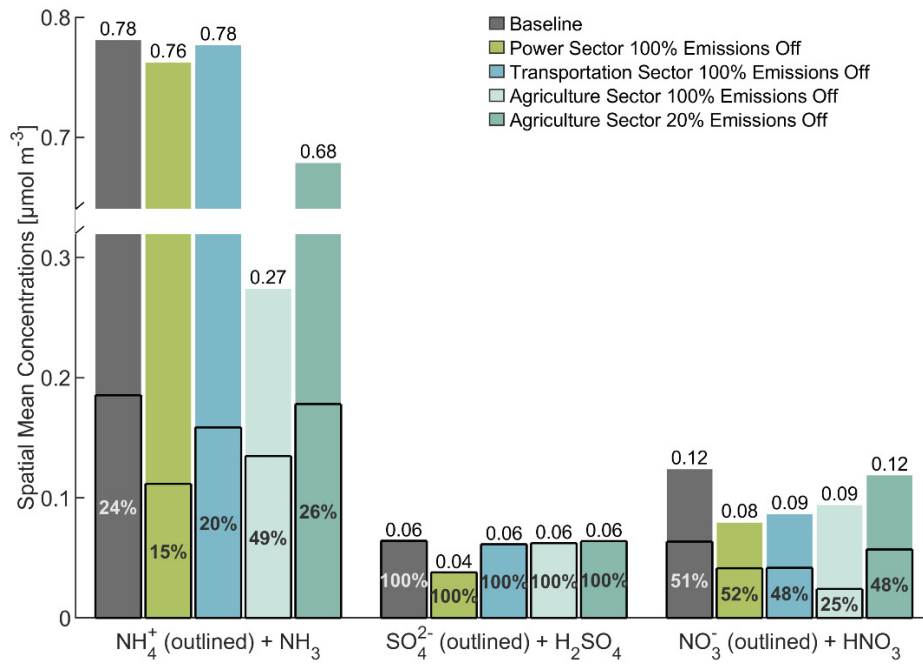


Figure 10. Partitioning between secondary PM_{2.5} components (NH_4^+ , SO_4^{2-} , and NO_3^-) and their relevant precursors (NH_3 , H_2SO_4 and HNO_3) for the baseline simulation and following the removal of individual sectoral emissions. Annual spatial mean concentrations (in $\mu\text{mol}/\text{m}^3$) across India in 2022 are shown for the baseline simulation (grey bars) and for scenarios where individual sector emissions (power, transportation, and agriculture) are removed completely or reduced by 20%. Solid-line-outlined boxes indicate the concentrations of NH_4^+ , SO_4^{2-} , and NO_3^- aerosol particles, while the upper portions of each bar (above the outlined boxes) represent the concentrations of NH_3 , H_2SO_4 , and HNO_3 . Numbers above each bar show the total concentration of the species group on the X-axis for the respective scenario. Percentages inside each outlined box indicate the share of NH_4^+ , SO_4^{2-} , and NO_3^- in the total concentration of $\text{NH}_4^++\text{NH}_3$, $\text{SO}_4^{2-}+\text{H}_2\text{SO}_4$, and $\text{NO}_3^-+\text{HNO}_3$, respectively.

To ensure these findings are more prominent and to address the reviewer’s recommendations on interpretation: We have added the 20% partial emission reduction scenarios explicitly to **Table 1** (original content in blue and new rows in green) to clearly indicate that these sensitivity tests are part of the study design.

Table 1. Emission scenarios for WRF-Chem simulations conducted in this study

Scenarios	Domestic Sources (Emissions inside India)			Transboundary Sources (Emissions outside India) [†]
	Anthropogenic [*]	Dust [‡]	Biogenic [‡]	
Baseline	On	On	On	On
POW _{off}	Power sector <i>off</i> [‡]	On	On	On
IND _{off}	Industry sector <i>off</i> [‡]			

RES _{off}	Residential sector <i>off</i> [‡]			
TRA _{off}	Transportation sector <i>off</i> [‡]			
AGR _{off}	Agriculture sector <i>off</i> [‡]			
FIRE _{off}	Open burning <i>off</i> [‡]			
DST _{off}	On	<i>Off</i>	On	On
BVOC _{off}	On	On	<i>Off</i>	On
TBDY _{off}	On	On	On	<i>Off</i>
TBDY _{anthoff}	On	On	On	Anthropogenic <i>off</i> , others on
POW _{20%off}	Power sector <i>20% off</i> [‡]	On	On	On
IND _{20%off}	Industry sector <i>20% off</i> [‡]			
RES _{20%off}	Residential sector <i>20% off</i> [‡]			

*This includes emissions from six source sectors within India: power, industry, residential, transportation, agriculture (excluding open burning), and open burning.

†We modify WRF-Chem to enable grid-level customization to turn dust and biogenic emission modules on and off.

‡This includes both anthropogenic and natural emissions (i.e., dust and biogenic) originating outside of India but within the WRF-Chem modeling domain, as well as the long-range transport of pollutants from regions beyond the WRF-Chem domain (i.e., the chemical boundary conditions for the model derived from the Whole Atmosphere Community Climate Model).

‡All other sectors' emissions are kept unchanged as the baseline scenario.

We believe these revisions, combined with the existing analysis in Section 4, fully address the concern regarding nonlinear processes and sensitivity testing.

- The authors stated that transboundary sources contribute 27% of national PW mean PM_{2.5}. This seems to be a striking finding, so careful clarification is needed. In particular, the authors did not separate natural vs. anthropogenic transboundary sources, and the quantitative attribution may depend on the domain size and boundary conditions chosen. I think at least a separation between dust and anthropogenic sources should be performed.

We thank the reviewer for this constructive suggestion. We agree that the reported 27% contribution from transboundary sources is a significant finding that warrants further analysis.

We acknowledge that separating transboundary anthropogenic versus natural sources is inherently challenging and dependent on domain size. Specifically, in our regional modeling framework, we cannot differentiate the specific source types (e.g., natural dust vs. anthropogenic pollution) of the pollutants entering our domain via the lateral chemical boundary conditions provided by the global model (WACCM).

However, to address the reviewer's request and provide a more granular breakdown, we conduct one additional

simulation where we specifically turn off anthropogenic emissions (including biomass burning) in the region outside India but within our modeling domain, while keeping the lateral chemical boundary conditions as well as natural emissions unchanged. This will isolate the contribution of regional transport of anthropogenic emissions from neighboring countries within our domain to PM_{2.5} in India. We attach **Figure 1** here to show our modeling domain.

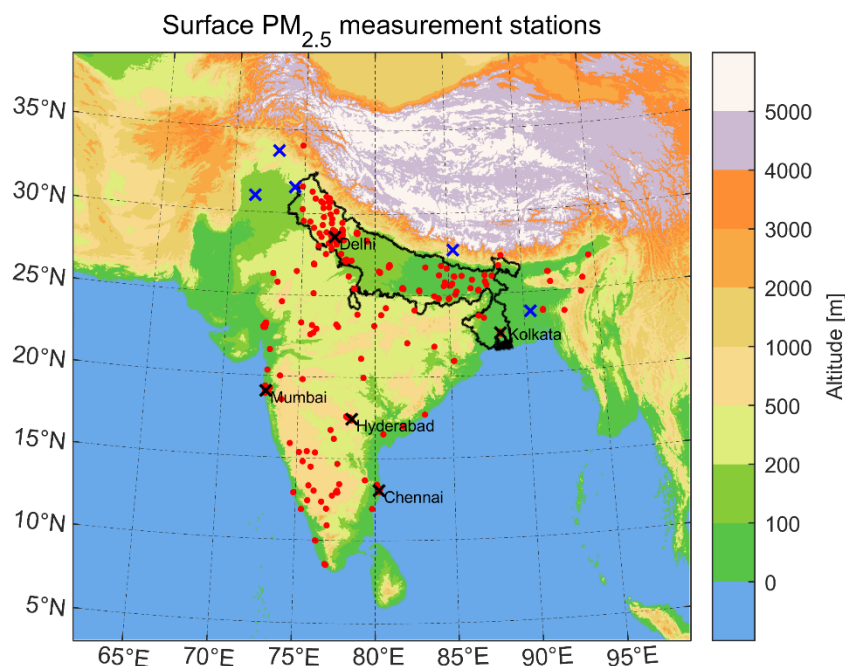


Figure 1. WRF-Chem modeling domain and surface PM_{2.5} measurement stations utilized in this study. The base map shows the WRF-Chem modeling domain, with colors representing the terrain height in the model. The 288 surface PM_{2.5} measurement sites used in this study are marked as follows: Red dots represent the continuous monitoring stations from the Indian Central Pollution Control Board (CPCB) network; the black and blue crosses represent the stations from U.S. Air Now network in India and adjacent countries, respectively. Thick black lines represent the boundary of the Indo-Gangetic Plain (IGP), which includes Delhi, Punjab, Haryana, Uttar Pradesh, Bihar, and West Bengal. We also label the five major cities of Delhi, Mumbai, Kolkata, Hyderabad, and Chennai, where U.S. Air Now PM_{2.5} measurements were available in India.

We have added this new emission scenario explicitly to **Table 1** (with new rows shown in green) to clearly indicate that it is part of the study design. Please see the updated **Table 1** in our response to the second comment of Reviewer #2.

We have added sentences (**Page 25 Line 675-679** in the clean version of the revised manuscript) and **Supplementary Figure 8** (attached below) to discuss our new analysis based on this new simulation:

“Further decomposition reveals that anthropogenic emissions originating from neighboring countries within our

modeling domain accounted for 11% (5.2 $\mu\text{g}/\text{m}^3$) of the 2022 annual national PW mean $\text{PM}_{2.5}$ in India. Meanwhile, other transboundary components, comprising regional natural sources (dust and biogenic) and long-range transport of pollutants entering via model lateral boundary conditions, contributed the remaining 16% (7.7 $\mu\text{g}/\text{m}^3$) (Supplementary Figure 8).”

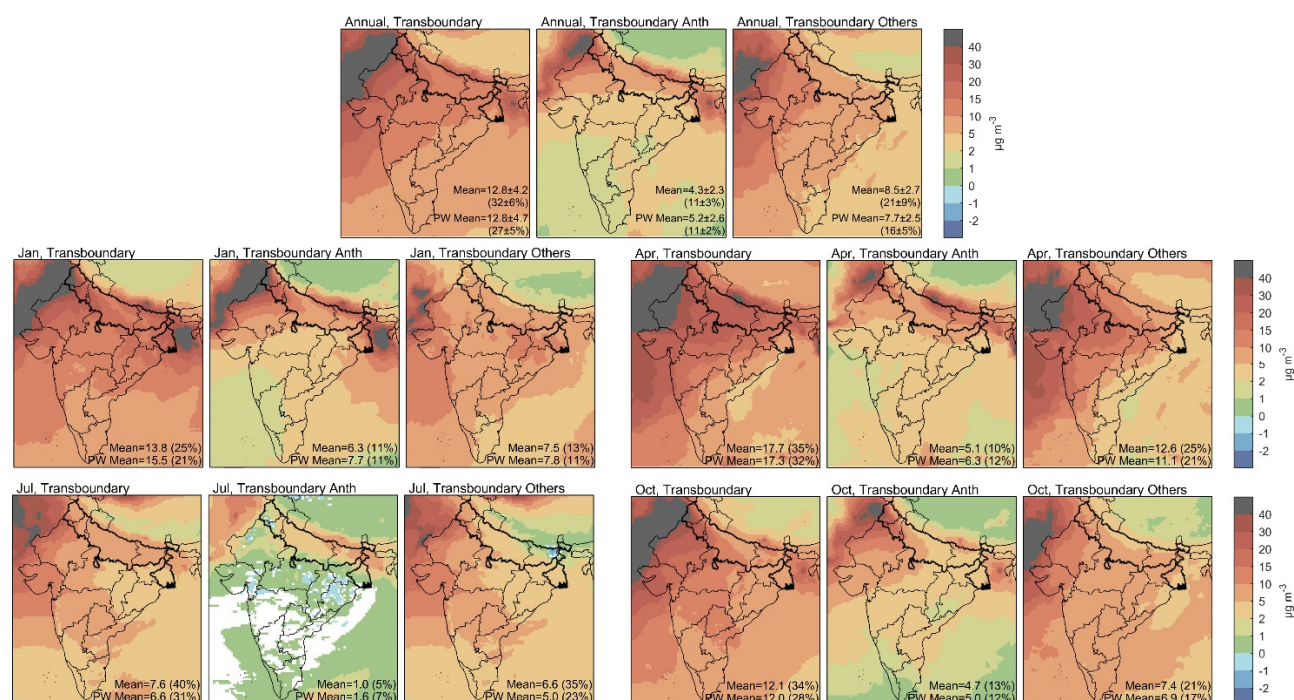


Figure S8. Contributions of transboundary sources to monthly and annual $\text{PM}_{2.5}$ concentrations in 2022. Panels are in five groups: Annual, January, April, July, and October. Within these groups the *Left panels*: Total transboundary sources, including both emissions originating outside India but within the WRF-Chem domain (anthropogenic and natural) and long-range transport from regions beyond the domain (from WRF-Chem boundary conditions). *Middle panels*: Transboundary anthropogenic sources, representing anthropogenic emissions specifically emitted outside India but within the modeling domain. *Right panels*: Other transboundary sources, representing the sum of transboundary natural emissions (dust and biogenic) and long-range transport from lateral boundary conditions. The thick black line denotes the boundary of the Indo-Gangetic Plain (IGP).

4. An uncertainty range (or at least rough estimation) of each attribution number should be provided, especially concerning the uncertainty in the emission inventories.

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 emission uncertainties. 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:

4. **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.

5. **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 $\text{PM}_{2.5}$, with the range reflecting uncertainties in emission factors without mitigation as reflected in the literature.

6. **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 $\text{PM}_{2.5}$ emissions. The earlier study assumed the following uncertainties for the residential sector:

Activity Data: population ($\pm 2.5\%$), fuel penetration ratios ($\pm 5\%$), and fuel consumption statistics ($\pm 5\%$). Source data is census data and national family health survey reports.

Emission Factors: $\pm 25\%$. Source data is from The Energy and Resources Institute (TERI)'s emission inventory development report (The Energy and Resources Institute, 2021).

Total Emissions: $\pm 50\%$ for $\text{PM}_{2.5}$.

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 $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ 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	6.0±0.6	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

To reflect these SI updates in the Main Text, we added one new paragraph in the **Uncertainty and limitation** section (**Page 31 Line 849-855** in the clean version of the revised 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 $6.1 \pm 1.1 \mu\text{g}/\text{m}^3$ for the power sector and $7.3 \pm 3.7 \mu\text{g}/\text{m}^3$ 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.”

Response to R Subramanian’s comments

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

The two anonymous reviewers have already raised some of my concerns, so I will focus on the new residential emissions inventory from Velamuri et al. (2024). As the authors of the current study state: "However, recent research highlights challenges in sustaining LPG usage under PMUY, including high refill costs and subsidy delays (Asharaf and Tol, 2024; Gaikwad et al., 2025), which may result in backsliding to a continued reliance on solid fuels which may not be fully captured in the updated inventory." This can be significant, leading to an overestimate of biofuel replacement by as much as half - based on studies that show LPG refills are consumed at half the rate (or even less) in PMUY households compared to non-PMUY households, e.g. <https://india.mongabay.com/2023/09/lpg-subsidy-can-increase-uptake-but-interventions-needed-to-counter-myths-and-improve-access/>

We are grateful for the public comment on this topic. We agree that fuel stacking (the concurrent use of solid fuels and LPG) and the financial barriers to sustained LPG refills are critical uncertainties in estimating India's residential emissions. As noted in the earlier research (Velamuri et al., 2024), the residential inventory adopted in this study explicitly recognized this challenge and was designed to mitigate this LPG overestimation.

Specifically, LPG emissions were calculated using a top-down approach based on actual district-level consumption, rather than assuming a fixed, theoretical per-capita usage for every household. This approach

utilizes comprehensive survey data from the National Family Health Surveys (NFHS-3: 2005-06; NFHS-4: 2015-16; NFHS-5: 2019-21), which was projected to 2022 levels using a regression analysis. Thus, the inventory inherently accounts for the lower LPG consumption rates observed in recent years (e.g., due to limited refills captured in the 2019-2021 data), thereby reducing the overestimation of clean fuel usage or the underestimation of solid fuel reliance in 2022.

It would also be helpful to compare the new residential EI to SMOG-2015 and the newer COALESCE version. EDGAR has much lower PM_{2.5} and NO_x numbers for this sector compared to SMOG, and the new sectoral EI numbers used in the current manuscript are even lower than EDGAR. The COALESCE SMOG (Venkataraman et al. 2024 <https://doi.org/10.1029/2024JD040834>) captures some of the PMUY transition (2017-2019) but including barriers to uptake, which seems a better approach:

"The SMOG-India_{COALESCE} inventory captures the large fuel transition which has taken place in residential cooking and heating from the launch of the Pradhan Mantri Ujjwala Yojana (PMUY, 2016) for the substitution of biomass fuels with liquefied petroleum gas (LPG), albeit with barriers which lead to significant levels of continued fuel stacking with biomass."

We have included the updated **Figure 2** below, which compares our updated 2022 emission inventory with three global inventories (CEDS: 2015–2022; EDGAR: 2015–2022; HTAP: 2015–2020) and the SMOG inventory for both 2015 and 2019. To discuss the comparison between SMOG and other inventories, we added **Supplementary Information 3**:

“Residential primary PM_{2.5} emissions in other inventories are significantly lower than those in SMOG (Venkataraman et al., 2018; Venkataraman et al., 2024):

- ♦ In 2015: HTAP (2.4 Tg/yr) and EDGAR (1.6 Tg/yr) represent only 73% and 48% of the SMOG PM_{2.5} emissions (3.3 Tg/yr), respectively.
- ♦ In 2019: While HTAP and EDGAR remain relatively flat (2.4 Tg/yr and 1.6 Tg/yr), SMOG reports a substantial increase to 5.4 Tg/yr. Consequently, HTAP and EDGAR represent only 44% and 30% of the SMOG 2019 PM_{2.5} emission, respectively.
- ♦ Current Study: Our updated 2022 residential PM_{2.5} estimate (1.1 Tg/yr) is ~21% of the SMOG 2019 value.

These discrepancies likely stem from fundamental differences in the source data and methodology used to estimate fuel consumption and resulting emissions.

The SMOG 2015 inventory calculates residential emissions based on theoretical energy demand (Pandey et al., 2014). For example, it utilizes survey-based food consumption data to derive end-use energy requirements (estimated at 12 MJ/household-day for cooking) and assumes a theoretical water heating demand (e.g., heating

daily water usage to 35 °C). This 'useful energy' methodology likely overestimates actual fuel usage in low-income households subject to fuel poverty, who may 'need' that energy theoretically but cannot afford the fuel to generate it. While the SMOG 2019 publication (Venkataraman et al., 2024) incorporated recent work characterizing non-cooking residential activities based on updated survey data (Navinya et al., 2023), it did not explicitly detail methodological updates for cooking emissions, which remain the dominant source.

In contrast, the residential emission inventory adopted in this study utilized a top-down approach based on actual district-level fuel consumption, derived from comprehensive survey data including the National Family Health Surveys (NFHS-3: 2005–06; NFHS-4: 2015–16; NFHS-5: 2019–21), which were projected to 2022 levels using regression analysis. By relying on reported usage rather than theoretical demand, this approach implicitly captures the affordability constraints and actual consumption habits that define the residential sector in India.

Furthermore, both HTAP and EDGAR inventories typically rely on the International Energy Agency (IEA) World Energy Balances to estimate fuel consumption for the residential sector in India (Janssens-Maenhout et al., 2019; Kurokawa and Ohara, 2020; Guizzardi et al., 2025),

Thus, the underlying source data and methodology differ significantly among these datasets: SMOG relies on energy demand modeling (idealized consumption); Global Inventories rely on IEA statistics (international aggregates); and the inventory adopted in our study is grounded in domestic household survey datasets (actual reported consumption).”

However, we emphasize that the residential emission inventory adopted in our study was initially built using actual district-level consumption data (Velamuri et al., 2024), and its implementation in WRF-Chem yields improved agreement with surface PM_{2.5} observations from the CPCB network in 2022 (**Figure 4** in the main text, not attached here). Given that SMOG PM_{2.5} residential emissions are nearly 5 times higher than our inputs, adopting the SMOG residential PM_{2.5} emissions (or scaling our residential inventory to similar values) would result in a very large overestimation of surface PM_{2.5} concentrations compared with observations. Therefore, we maintain that our current inventory is more representative of the residential emission patterns required to reproduce observed PM_{2.5} pollution level in 2022. In addition, we have explicitly addressed the uncertainty of residential emission estimates in our response to the subsequent comment.

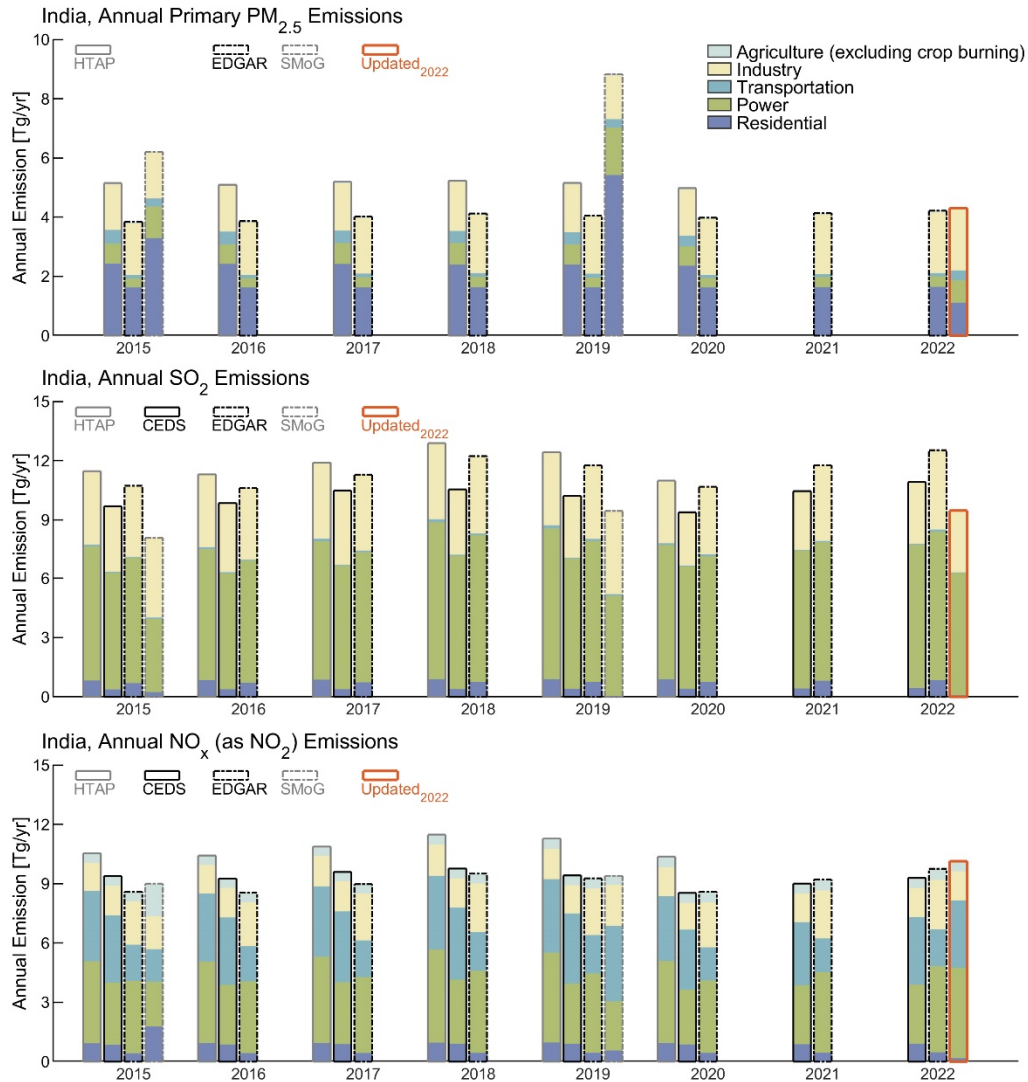


Figure 2. Comparison of annual anthropogenic sectoral total emissions over India among three global inventories, one regional 2015 inventory, and our updated 2022 inventory. For each country and species, CEDS_{v2024-07-08}, EDGAR_{v8.1}, and HTAP_{v3.1} report sectoral total emission across 59, 32, and 16 detailed sectors, respectively. Those detailed sectors are aggregated into five widely used sectors: power, industry, residential, transportation, and agriculture. In this figure, because CEDS_{v2024-07-08} does not include open burning of agricultural waste and wildfire, we exclude it from the agricultural sector emissions in EDGAR_{v8.1} and HTAP_{v3.1} to enable direct comparison among the inventories. Note that CEDS_{v2024-07-08} does not provide primary PM_{2.5} emissions, HTAP_{v3.1} extends only until 2020, and SMOG is only available for 2015 and 2019. Details on the updated inventory are provided in **Section 2.1**.

So at the very least, I would expect an uncertainty analysis that shows the impact of this underestimated biofuel usage (e.g. increase residential emissions by 2x) - the headline finding of this study may look very different.

Regarding the potential impact of residential emission uncertainties on our conclusions, we have included a detailed discussion in **Supplementary Information 4**.

We note that the residential inventory adopted in this study (Velamuri et al., 2024) explicitly included an uncertainty analysis using 10,000 Monte Carlo simulations. This analysis estimated a total uncertainty of $\pm 50\%$ for residential $\text{PM}_{2.5}$ emissions, accounting for uncertainties in population, fuel penetration, and emission factors.

We then performed a simplified uncertainty propagation analysis. Assuming a near-linear relationship between sectoral emissions and resulting concentrations at the national level (a reasonable first-order approximation without additional WRF-Chem simulations), we estimate the residential sector's contribution to the annual population-weighted mean $\text{PM}_{2.5}$ to be $7.3 \pm 3.7 \mu\text{g}/\text{m}^3$ in 2022.

We acknowledge that the upper bound of this estimate ($11 \mu\text{g}/\text{m}^3$) exceeds the baseline contribution of the industrial sector ($8.6 \mu\text{g}/\text{m}^3$). However, the industrial sector estimate is derived from global inventories (gases from CEDS, aerosols from EDGAR) which also carry significant (though unquantified in these inventories) uncertainties for India. Consequently, based on the comparison of mean estimates, we maintain our finding that the industrial sector contribution exceeds that of the residential sector in 2022, while explicitly acknowledging the limitation regarding the unquantified uncertainty of industrial emissions in our paper's **Uncertainty and limitation** Section:

“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 $\text{PM}_{2.5}$ contributions of $6.1 \pm 1.1 \mu\text{g}/\text{m}^3$ for the power sector and $7.3 \pm 3.7 \mu\text{g}/\text{m}^3$ 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 in these datasets. However, we note that the upper uncertainty bound of the residential sector contribution ($11 \mu\text{g}/\text{m}^3$) exceeds the baseline estimate for the industrial sector ($8.6 \mu\text{g}/\text{m}^3$). Consequently, while our central estimates identify India's industrial sector as the largest domestic $\text{PM}_{2.5}$ source in 2022, the definitive ranking of these top two sectors is sensitive to the unquantified uncertainties in the industrial emission inventory.”

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