

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 NH_3 -rich environment (**Figure 10**), where nitrate availability limits secondary inorganic aerosol formation. This suggests that partial removal of NH_3 is less effective—defined as concentration decrease per unit emission reduction—in mitigating $PM_{2.5}$ than substantial NH_3 emission reductions, especially in northern India.”

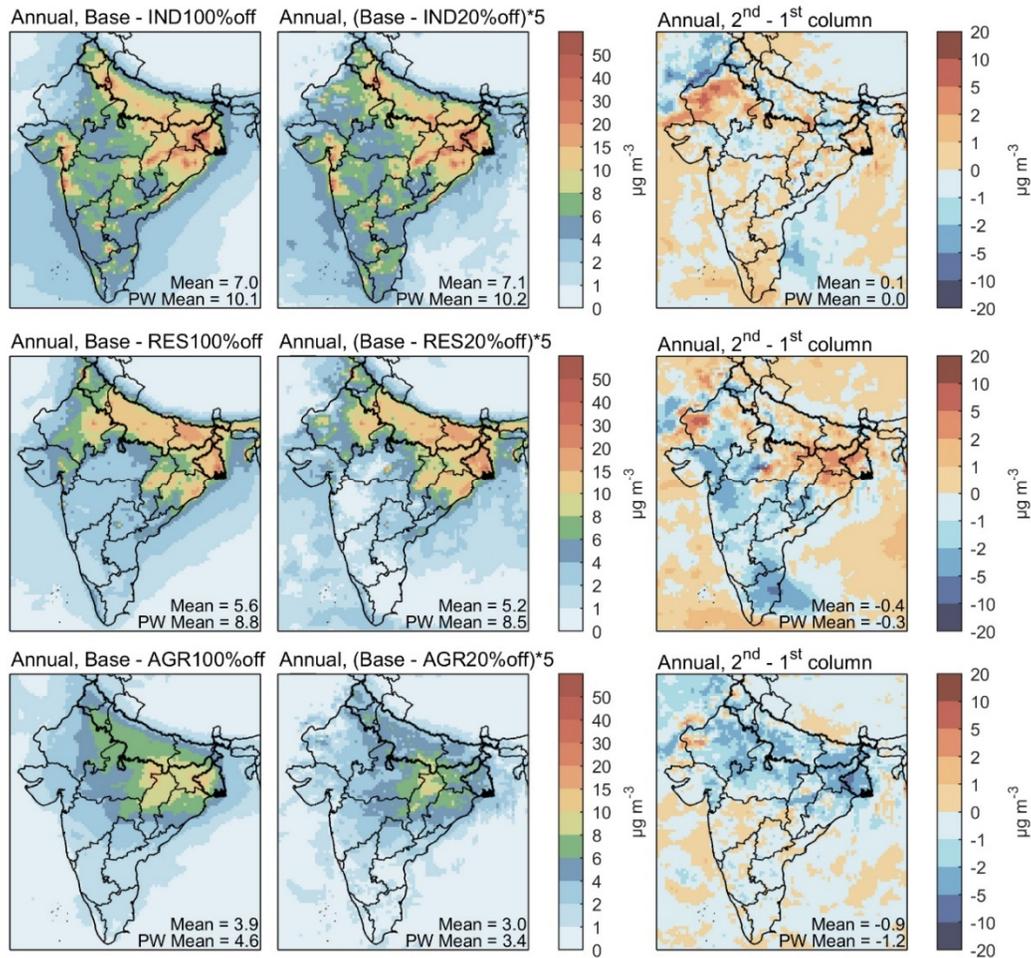


Figure S9. Comparison of PM_{2.5} concentration changes (presenting as absolute values) between a 100% emission reduction and a fivefold scaling of 20% reductions. We show results for three sectors: industry (IND), residential (RES), and agriculture (AGR). The thick black line denotes the boundary of the Indo-Gangetic Plain (IGP). Unlike results presented in Main text figures, we **do not scale concentrations in this figure using Equation 7.**

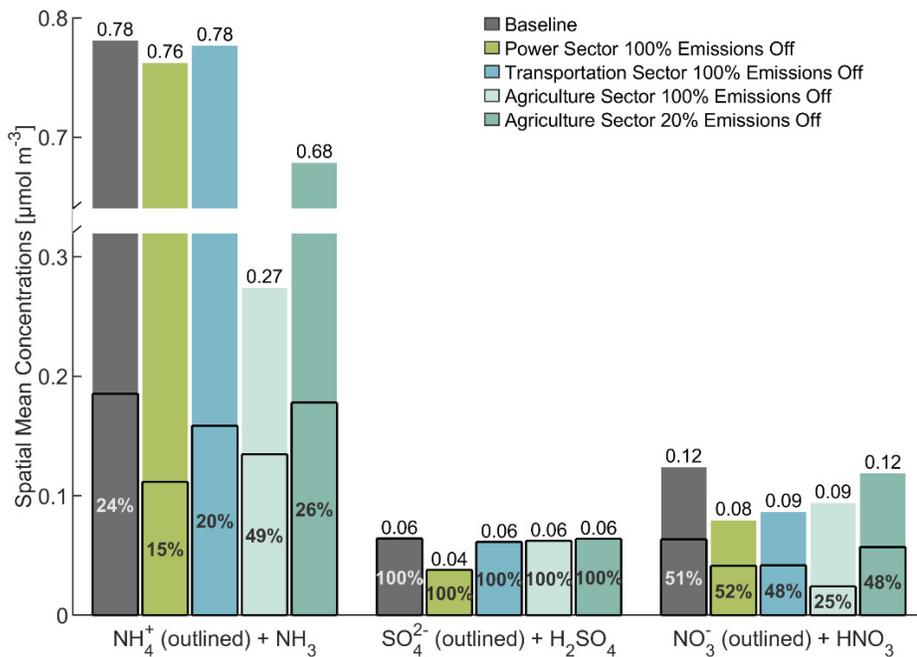


Figure 10. Partitioning between secondary PM_{2.5} components (NH₄⁺, SO₄²⁻, and NO₃⁻) and their relevant precursors (NH₃, H₂SO₄ and HNO₃) for the baseline simulation and following the removal of individual sectoral emissions. Annual spatial mean concentrations (in µmol/m³) across India in 2022 are shown 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 NH₄⁺, SO₄²⁻, and NO₃⁻ aerosol particles, while the upper portions of each bar (above the outlined boxes) represent the concentrations of NH₃, H₂SO₄, and HNO₃. 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 NH₄⁺, SO₄²⁻, and NO₃⁻ in the total concentration of NH₄⁺+NH₃, SO₄²⁻+H₂SO₄, and NO₃⁻+HNO₃, 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.

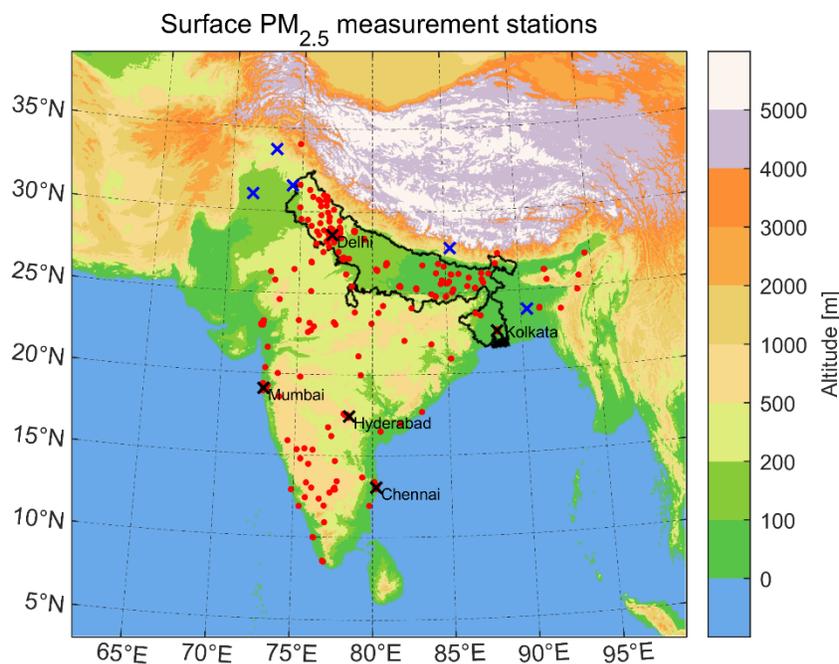


Figure 1. WRF-Chem modeling domain and surface $PM_{2.5}$ measurement stations utilized in this study. The base map shows the WRF-Chem modeling domain, with colors representing the terrain height in the model. The 288 surface $PM_{2.5}$ measurement sites used in this study are marked as follows: Red 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 $PM_{2.5}$ 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).”

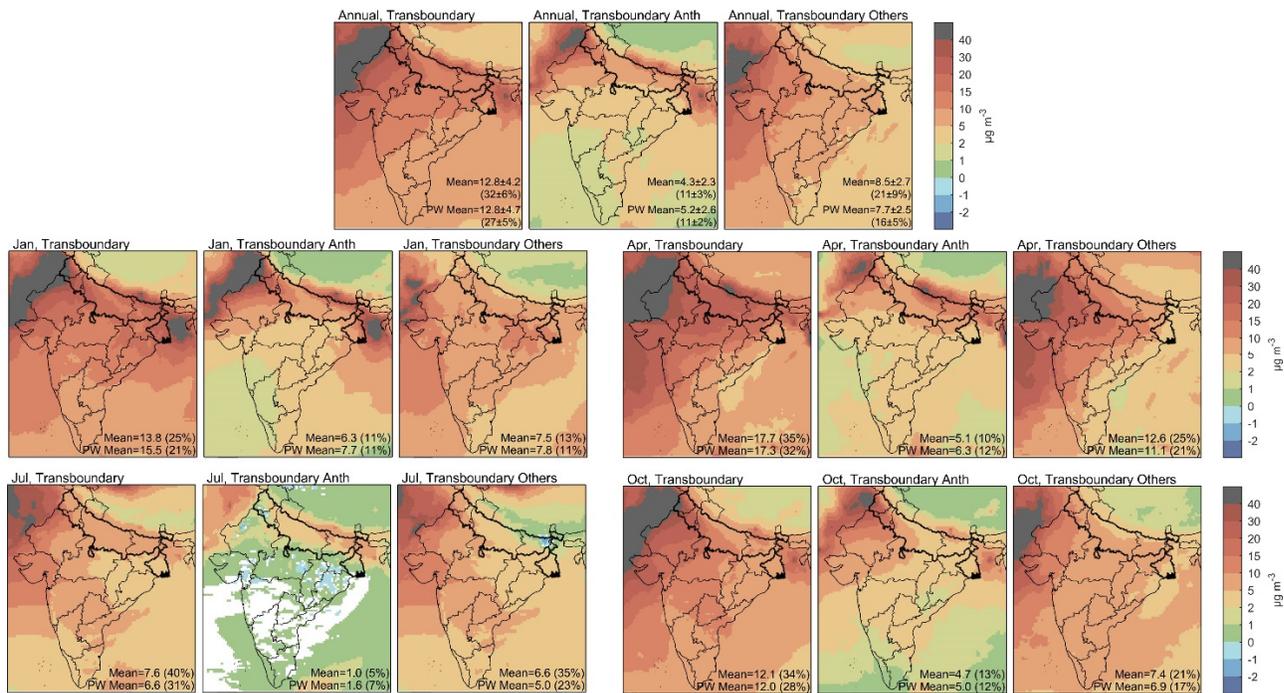


Figure S8. Contributions of transboundary sources to monthly and annual $\text{PM}_{2.5}$ concentrations in 2022. Panels are in five groups: Annual, January, April, July, and October. Within these groups the *Left panels*: Total transboundary sources, including both emissions originating outside India 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:

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 $\text{PM}_{2.5}$, 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 $\text{PM}_{2.5}$ 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±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.”

Reference in this reply:

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