Point-by-point response to the editor's comments

(original comments in black, our responses in blue)

We thank the editor for their continued feedback for our manuscript. We attached our revised paragraphs based on their suggestions below.

1. Transboundary seasonal peaks (replace current seasonal representation paragraph in Section 5):

"We further acknowledge that using single months (January, April, July, October) to represent entire seasons may lead to an underestimate of the contribution of transboundary biomass burning and regional haze. In particular, peaks in aerosol optical depth (AOD) and black carbon (BC) loading in the eastern Indo-Gangetic Plain and neighboring countries are observed in February-March (driven by fires in Myanmar and northeastern India) and in December (driven by biomass burning in Bangladesh and western India/eastern Pakistan). Consequently, our annual estimates for organic aerosol and black carbon from these episodic transboundary sources should be considered conservative lower bounds."

To incorporate the content mentioned by the editor, we modify the paragraph as:

"Using single months (January, April, July and October) to represent entire seasons may underrepresent the air quality impacts of episodic emission sources with strong intra-seasonal variability, such as open biomass burning and its transboundary transport. For example, post-monsoon crop-residue burning in northwestern India has shifted later in the season, with peak burning delayed into November (Sembhi et al., 2020; Liu et al., 2022). In addition, open biomass burning in Northeast India and adjacent regions, particularly Myanmar, is most active during the dry pre-monsoon period, with strong fire activity and emissions in March–April (Singh et al., 2020). Our seasonal sampling does not fully capture these episodic peaks (e.g., missing March and November), and the source attribution for open burning and its transboundary transport, particularly at the regional level, should therefore be considered conservative lower bounds."

2. AOD bias attribution (revise Supplementary Information 2.2):

Your current text attributes the model's AOD underestimation primarily to "missing coarse PM mass." This is likely incorrect. Coarse particles (diameter >2.5 μm) have low mass extinction efficiency in the mid-visible range, and cannot explain persistent mid-range visible AOD bias. The missing emissions are most likely in the accumulation mode (approximately 0.25-2.5 μm), including particles near 1 μm diameter, which dominate both mass and extinction. Please revise this paragraph to correctly state that the AOD bias may reflect under-represented accumulation-mode emissions (e.g., from combustion, secondary aerosol formation), not coarse dust. Add the following text:

"Our qualitative attribution of mid-visible AOD underestimation to missing coarse PM mass in the

Supplementary Information should be interpreted as a diagnostic discussion, not as a quantitative source apportionment of AOD bias. Emissions of 1 µm particles are systematically under-represented in current inventories for India, a gap that our study does not address. Furthermore, the vertical allocation of the emissions that are included remains uncertain; incorrect plume heights and vertical mixing can substantially alter column-integrated AOD and surface PM2.5 relationships. Our model's persistent AOD bias likely reflects both missing emission sources and their misallocation in the vertical. Future work should incorporate these constraints."

We agree that attributing the mid-visible AOD underestimation primarily to missing coarse PM mass would be an over-interpretation, because AOD depends strongly on aerosol size distribution, composition, hygroscopic growth and vertical distribution, and fine/accumulation-mode particles generally have higher mass extinction efficiencies than coarse particles. However, our comparison with CPCB measurements shows that coarse PM is substantially underestimated in the baseline simulation across India, by 37.3 ± 25.7 µg m^{-3} ($59 \pm 41\%$). Therefore, missing coarse PM mass may still contribute to the AOD underestimation, although it is unlikely to fully explain the persistent bias on its own. We have revised the Supplementary Information to avoid presenting this as a quantitative attribution of the AOD bias, and instead discuss missing coarse PM as one possible contributor alongside underrepresented fine/accumulation-mode aerosol, secondary aerosol formation, aerosol water uptake, optical properties and uncertainties in vertical allocation.

“We interpret the model’s AOD underestimation as a diagnostic indication of missing or misrepresented aerosol extinction, rather than as a quantitative source apportionment of the AOD bias. Our baseline simulation substantially underestimates surface coarse PM concentrations compared with CPCB measurements across India, by 37.3 ± 25.7 µg/m^3 ($59 \pm 41\%$; **Supplementary Fig. 3**). This suggests that missing coarse particulate mass, including fugitive dust from sources not represented in our inventory, such as construction activities and industrial fugitive emissions, may contribute to the AOD low bias. This interpretation is also consistent with previous modelling studies over India that found improved AOD agreement after scaling coarse-particle AOD (David et al., 2018; Singh et al., 2021). However, missing coarse PM alone is unlikely to fully explain the persistent mid-visible AOD underestimation, because visible extinction depends strongly on aerosol size distribution, composition, hygroscopic growth, optical properties and vertical distribution, and fine and accumulation-mode particles generally have higher mass extinction efficiencies than coarse particles.

The AOD bias may therefore also reflect underrepresented fine and accumulation-mode aerosol mass, including primary combustion particles and secondary inorganic and organic aerosol formation. Aerosol water uptake may further contribute to the bias: high local emissions of hydrochloric acid in India can partition into aerosol water and enhance particle growth (Gunthe et al., 2021), but these anthropogenic chloride sources are not represented in the global emission inventories used here. Although this omission has been shown to have a relatively small effect on dry surface $PM_{2.5}$ concentrations (Patel et al., 2024), it can reduce simulated aerosol hygroscopic growth under high relative humidity and thereby contribute to AOD underestimation. Additional uncertainty arises from the treatment of aerosol optical properties in the MOSAIC scheme, including the internal-mixing assumption and the use of four broad size bins, which may not fully resolve the size ranges most efficient for visible-light extinction.”

3. Temporal representation of episodic sources and scatter plot spread (add to Section 5, after the paragraph on temporal smoothing):

"The substantial spread in our density scatter plot (new Supplementary Figure 2) at medium to high PM2.5 values (observed concentrations >100 µg m-3) indicates that the model systematically underestimates the magnitude of moderate to extreme pollution episodes. As noted above, monthly emission totals for anthropogenic sectors are distributed evenly across days with static diurnal profiles, smoothing episodic variability. This structural limitation is particularly problematic for biomass burning and transboundary sources, which are inherently episodic. The episodic nature of crop-residue burning in the Indo-Gangetic Plain (post-monsoon) and transboundary fires from Myanmar (pre-monsoon) is therefore not fully represented in our source attribution, nor is day-to-day variability in domestic anthropogenic emissions (e.g., holidays, weather-driven demand). Consequently, our annual mean source contributions for open burning and transboundary sources should be interpreted as lower-bound estimates."

We have revised Section 5 to further discuss the substantial spread in the density scatter plot at observed daily $PM_{2.5}$ concentrations above 100 µg m^{-3} and its implications for representing moderate-to-extreme pollution episodes. We also explain how the temporal allocation of anthropogenic emissions smooths day-to-day variability, while noting that biomass-burning emissions are represented using the satellite-based FINN inventory at daily resolution for the simulated months. Nevertheless, we acknowledge that satellite-derived fire emissions may miss small fires or be affected by cloud cover and thick haze. In addition, the discussion of missing episodic biomass-burning emissions and transboundary transport is included in our response #1 and its revision to the manuscript.

"Our baseline emission inventory has limitations in capturing temporal variations of real-world emission patterns. While annual emission budgets at the regional scale are reasonably represented (**Figure 3 a-b**), the model cannot fully reproduce extreme daily episodes (**Figure 3 e-g** and **Supplementary Figure 3**). The substantial spread in the density scatter plot of observed and simulated daily $PM_{2.5}$ concentrations at observed $PM_{2.5}$ values above 100 µg/m^3 indicates larger model uncertainty during moderate-to-extreme pollution episodes, with a tendency to underestimate the magnitude of some high-pollution events, although both under- and overestimation occur (**Supplementary Figure 2**). This limitation partly stems from how temporal allocations of emissions are handled in our model: for anthropogenic sources, including industry, residential combustion, power generation, transport and agriculture, monthly totals are distributed evenly across days with prescribed sectoral diurnal profiles, smoothing episodic spikes (e.g., holidays and weather-driven activity changes). For biomass burning, we use the satellite-based FINN inventory at daily resolution, which better captures day-to-day variability; however, satellite-derived fire inventories can still underestimate emissions from small-scale fires or be degraded by cloud cover and thick haze during intense pollution episodes. Natural dust and biogenic emissions are calculated online within WRF-Chem using real-time meteorology, but carry uncertainties from static input parameters (e.g., land use type, surface erodibility, leaf area index) and biases in simulated meteorological fields (e.g., wind speed)."

Reference:

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[3] Liu, T., Mickley, L. J., Patel, P. N., Gautam, R., Jain, M., Singh, S., et al.: Cascading Delays in the Monsoon Rice Growing Season and Postmonsoon Agricultural Fires Likely Exacerbate Air Pollution in North India, J Geophys Res-Atmos, 127, 2022.
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