

We appreciate the reviewer's thorough review and constructive comments on our manuscript. Below, we provide a point-by-point response to your concerns. Your comments appear in the regular font and our responses are given in italics font.

Thank you for responding to my comments. The responses have significantly improved the manuscript. Based on the responses, I have additional comments listed below.

- Supplementary Table S1: Please show the statistics for all the 9 stations separately. Normalized Mean Bias percentage is defined as:

- $$NMB (\%) = \frac{\sum(\text{Model}-\text{Observation})}{\sum \text{Observation}} \times 100\%.$$

- Please update the equation and recalculate the numbers. Kindly change 'MB %' to 'NMB %' in the header. Update the main text accordingly. Why is the p value for Patna so high compared to other locations? Kindly include the number of datapoints used to calculate the statistics. Change 'NMRSE' to 'NRMSE'.

Response: We missed the summation symbol in the equation. Thank you for bringing this to our attention. The table has now been updated.

The following discussions have been added to section 4 of the manuscript

The poor correlation at Patna (and Muzaffarpur) is due to the low modeled PM_{2.5} concentrations which are caused by increased dry deposition of aerosol particles activated as fog droplets during fog periods, as discussed in section 4 of the manuscript. Furthermore, the fog events in WRF and observations have somewhat different time periods causing WRF-predicted PM_{2.5} and the observed PM_{2.5} concentrations to decrease at different times.

- A sensitivity study was conducted by including trash burning emissions. Please add a line commenting on the NMB value for PM_{2.5} concentrations in that study.

Response: The following text has been added in section 4 of the manuscript.

This was accomplished by incorporating trash-burning emissions in the model simulation, which improved the PM_{2.5} prediction, increasing NMB by ~4-8% in IGP

Stations	NMB%-with Trash emissions	NMB%-no Trash emissions	Difference in NMB%
Amritsar	2.17	-2.52	4.69
Dwarka (Delhi)	-48.49	-52.71	4.22
IHBAS(Delhi)	31.93	24.71	7.22
RKP(Delhi)	-40.44	-45.7	5.22
Kanpur	-53.01	-57.65	4.63
Lucknow	-30.14	-32.49	2.35
Patna	-32.31	-40.69	8.37
Muzaffarpur	-36.46	-40.45	4

- Including the equations for the Taylor diagram is helpful. I further suggest the authors to pick any station: for example, pick station 2 for EXP2 and guide the readers about what specific information can be obtained from Figure 3.

Response: Discussion added as suggested in the manuscript.

"For example, simulated RH at Dwarka (4) and Lucknow (7) for EXP2, and IGI Airport (2), IHBAS (3), Lucknow (7), and Patna (8) for EXP3 show good agreement with observation, with $r > 0.7$, standard deviation within ± 0.25 and mean bias within 10%. Among these stations, the model performs better for Dwarka (4) and Lucknow (7) for EXP2, IGI Airport (2), and IHBAS (3) for EXP3 with a smaller centered

RMSE (<0.75). “

“For example, simulated T2 agrees best with observation at IHBAS (3) for EXP1 and IGI Airport (2) for EXP2, with smaller centered RMSE and standard deviation, and bias <5%.”

4. The equation for centered RMS is repeated twice. Please fix. What is the significance of the centered RMS?

Response: Revised in the manuscript.

The centered RMS difference is between the modelled and observed datasets proportional to the distance of a point in the Taylor diagram to the point “OBS” on the x-axis, indicating the extent to which the simulated datasets compare with the observed dataset. It is calculated by centering both the datasets around their respective means.

5. The authors claim that a PBL scheme which works fine for summer over IGP, might not work for winter, as a motivation for designing EXP1, 2, and 3: Are there any known seasonal biases in the PBL schemes used for this study? A short description is needed. The authors might also add a few lines (or a table) briefly describing the differences between different PBL schemes used in this study? (See Xie et al., 2012, for example. They recommended using ACM2 PBL scheme for both summer and winter, for a different, but highly polluted region)

Response: *The following paragraph has been added to the manuscript.*

Although earlier studies (Gunwani and Mohan, 2017; Mohan and Bhati, 2011; Mohan and Gupta, 2018; Xie et al., 2012) recommend using the nonlocal ACM2 PBL scheme for air quality prediction for IGP, there is still seasonal, day-night and regional biases in the PBL schemes. Gunwani and Mohan, (2017) showed that ACM2, QNSE, and MYJ schemes work well in predicting temperature, humidity, and wind speed in different regions. ACM2, MYNN and MYJ work best for Chennai (in South India), New Delhi (NWIGP), and Kolkata (EIGP) respectively for PBL height during summer whereas for winter MYJ works best for Chennai and QNSE for New Delhi and Kolkata. Regarding the prediction of fog, Mohan and Bhati, (2011) found that using ACM2 PBL scheme with Pleim Xiu surface physics improved wintertime meteorology estimates in Delhi indicating its potential in fog predictions, whereas Pithani et al.,(2019) recommend using the local PBL scheme MYNN2.5 with WSM3, WSM6, and Lin microphysics. Shin and Hong, (2011) found that a non-local (e.g., ACM2, YSU) scheme is favorable in unstable conditions and a local scheme (e.g., MYJ, Boulac) in stable conditions. All these studies suggest the need for careful consideration of the above-mentioned biases while selecting a PBL scheme.

The YSU and ACM2 PBL schemes are both nonlocal schemes, however, studies report differences in their performance particularly in the convective daytime boundary layer, with a deeper boundary layer height using ACM2 compared to YSU (Hariprasad et al., 2014; Xie et al., 2012). This is likely due to their different formulations e.g. defining the critical bulk Richardson number (Xie et al., 2012).

6. A few lines should be added in the conclusion/discussion section about the inability of the model to simulate the aerosol-fog interactions and the potential affects it might have on the outcome on the paper. On that note, why the exchange co-efficient of heat is needed to calculate the activation fraction? Kindly refer to relevant equations/literature for better clarity.

Response:

The following text was added to the conclusions.

Aerosol-cloud interactions were not investigated in this study due to the limitation of the ACM2 PBL scheme in providing necessary information with other modules in WRF. Previous studies of aerosol-fog interactions have found that ACI also promotes early onset of fog formation and increases fog duration (Maalick et al., 2016; Yan et al., 2021). While these previous studies were applied to midlatitude fog events, it is likely that ACI also plays a dominant role in IGP fogs, suggesting that future studies are needed to fully understand aerosol effects on IGP fog events.

The mixactivate module in WRF-Chem computes the activation fraction for aerosol mass and number based on when the maximum supersaturation of the air entering the cloud exceeds the critical supersaturation to form cloud droplets based on Kohler theory (Abdul-Razzak and Ghan, 2000, 2002). The maximum supersaturation relies on the mean vertical velocity and the turbulent velocity spectrum as input. The turbulent velocity spectrum depends on the heat exchange coefficient, which causes the spectrum of vertical velocities. Thus, the cloud droplet activation scheme relies on information from the PBL scheme. Unfortunately, the ACM2 PBL scheme does not provide the heat exchange coefficient to other parts of the WRF code, so aerosol-cloud interaction using ACM2 is not possible.

We added text to the model description to note why the exchange coefficient of heat is needed.

7. How do the authors identify whether a fog event is Radiation Fog or an Advection Fog (Both from the observations and the model)?

Response: Radiation fog is formed when the surface cools and humidity levels reach 100%, particularly at night under the clear sky and calm winds (Lakra and Avishek, 2022). Radiation fog is usually categorized based on the onset time of fog, which occurs after sunset and before sunrise. Advection fog on the other hand occurs when horizontally warm, moist air moves over cooler surfaces. Earlier studies categorized advection fog based on visibility and wind speed, where reduced visibility was accompanied by wind speeds exceeding 2.5 m/s, followed by a sudden visibility decrease, indicating an advection-type fog event (Deshpande et al., 2023; Pithani et al., 2019). The fog event in our study is a wintertime fog that starts to form at ~20:00 LT. The nighttime wind speeds were <2.5 m/s at the stations shown in Figure S1, further supporting that the fog events studied were radiation fog. The majority of fog events in the IGP during December-January are radiation fog (Deshpande et al., 2023; Ghude et al., 2023) formed due to radiative cooling of the surface, with longer-duration events compared to other regions of the world (Deshpande et al., 2023).

Lines 120-121 rephrased in section 2. Methodology of the manuscript

8. L397: Fix grammar. Change Mean Bias to Normalized Mean Bias.

Response: Sentence corrected in the manuscript.

9. Figure 1c: I think the units are not required for the title. Please change the title to: “Anthropogenic PM_{2.5} Emissions”. Please increase the gap between texts: ‘Kanpur’ and ‘Lucknow’. Change kg/m²/s to kg m⁻² s⁻¹.

Response: Suggested changes are implemented in the Figure 1 in the manuscript.

10. In Line 349, and in the caption of Figure 3, please clarify if the cloud water mixing ratios are grid average or in-cloud (i.e. divided by cloud fraction). Kindly incorporate the same change throughout the manuscript.

Response: Cloud water mixing ratios are grid average and it is defined in the manuscript text and Figure 3.

11. L667: change “diagnostic output..” to “diagnostic output.

Response: Change implemented in the manuscript.

12. L445: Typically, what percentage of PM_{2.5} mass is secondary in the IGP? How much is nitrate?

Response: PM_{2.5} composition varies across the Indo-Gangetic Plain (IGP). For example, Sharma and Mandal, (2017) reported that secondary aerosols contribute to 23% of PM_{2.5} mass in Delhi, whereas Behera and Sharma, (2010) found that 50% of PM_{2.5} is secondary aerosols, 34% of secondary inorganic aerosol (SIA) and 17% of secondary organic aerosol (SOA) in Kanpur. Another study estimated that the

total secondary aerosols contribute to $42 \pm 10\%$ of $PM_{2.5}$ in winter and $23 \pm 6\%$ in summer (Nagar et al., 2017) Nitrate constituted 9-13% of $PM_{2.5}$ mass in Delhi (Lalchandani et al., 2021; Sharma and Mandal, 2017).

13. L487: I recommend changing $\mu\text{g}/\text{m}^2/\text{hr}$ to $\mu\text{g m}^{-2} \text{hr}^{-1}$. How accurate is the dry deposition flux in the model? Cite previous work, if available.

Response:

Units revised in the manuscript

We do not have observations to validate dry deposition flux. Dry deposition of gases and aerosols is a process that needs continued evaluation and is a focus of some past and future studies. In WRF-Chem, the dry deposition of gas species is calculated following Wesely, (1989) while aerosol dry deposition follows Binkowski and Shankar, (1995). Ryu and Min, (2022) found higher dry deposition velocity for coarse mode particles in the model, resulting in the underestimation of surface PM_{10} concentration. The updated dry deposition scheme by Ryu et al., (2022) significantly increased surface PM_{10} concentrations but showed minimal impact on $PM_{2.5}$ levels. Although this study was done for another region, it is reasonable to assume that the dry deposition flux of $PM_{2.5}$ over IGP is within acceptable limits. In addition, the AQMEII project is currently conducting an evaluation of dry and wet deposition (Galmarini et al., 2021). This model intercomparison study will provide valuable information on deposition model parameterizations.

14. L603: Fix grammar.

Response: Sentence corrected in the manuscript.

15. L672: Please explain “more CCN are expected with aqueous chemistry”

Response: The aqueous chemistry adds sulfate to the aerosol mass increasing the mass of $PM_{2.5}$. Increased $PM_{2.5}$ further contributes to AR feedback, thus increasing RH. Increased RH favors the growth of aerosol size which then promotes the availability of aerosols as CCN.

We have explained the phrase in section 7 of the manuscript

16. Kindly change ug/m^3 to $\mu\text{g m}^{-3}$ or $\mu\text{g}/\text{m}^3$ throughout the manuscript.

Response: Units revised throughout the manuscript.

17. WRF-Chem simulations does not have fog in NWIGP, and hence could not be compared with the WiFEx campaign data: Please add a few lines in the conclusion/discussion section.

Response: Suggested lines added in the manuscript.

18. L12: Improve the sentence structure.

Response: Sentence corrected in the manuscript

19. How are the representative stations selected? Are there data available only from the 9 stations across the IGP during the study period?

Response: Representative stations were selected based on the availability of data. CPCB (Central Pollution Control Board of India) has a large network of stations throughout the country, particularly Delhi in the IGP. However, most of the stations in other parts of IGP experienced gaps in data during the winter of 2017. At present, CPCB data availability has improved, and it can be verified at the CPCB website

Text rephrased in section 2.2 Observations of the manuscript

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