The authors simulated a fog event with high aerosol loading over the Indo Gangetic Plains of India, and estimated the role of atmospheric aerosols in severe winter fog through aerosolradiation interaction and aqueous phase chemistry. Overall, this is well-executed study and the topic is very interesting. The authors made great efforts in model evaluation and sensitivity simulation. However, several conclusions in the main text should be further explained and the presentation quality can be improved. I suggest a major revision before it can be accepted.

We appreciate your thorough review and constructive comments on our manuscript. They have significantly contributed to the improvement and refinement of the work. Below, we provide a point by point response to your concerns. Your comments appear in the bold font and our responses are given in regular font.

Major comments:

I am very curious about if the fog event is due to radiation or advection. If this is a radiation fog, then the role of AR feedback could be a major reason. However, if this is an advection fog, the authors may need to pay attention on wind changes.

Response: It is a radiation fog. The majority of fog events in the IGP during December-January are radiation fog (<u>Deshpande et al., 2023</u>; <u>Ghude et al., 2023</u>) formed due to radiative cooling of the surface.

Simulated PM2.5. In L363, the authors mentioned that the observed high PM2.5 can be predicted by model but not for PM2.5 composition (Fig.7). I am very confused about this. The model strongly underestimated observed inorganic PM2.5 composition on 24th Dec; then why was total PM2.5 mass concentration well simulated?

Response: The WRF-Chem model results show that a large percentage of $PM_{2.5}$ is classified as "other inorganics (OIN)", which is usually dominated by $PM_{2.5}$ other than BC and OC. OIN contributes to >50% of $PM_{2.5}$ in Delhi (Fig 12 of manuscript). Emissions of mineral dust from road and construction dust also contribute to aerosol loading in urban areas (Sharma and Mandal.,2023). The model simulates the day-to-day variability in $PM_{2.5}$ considerably well, still, there is a large bias between the measured and observed values due to discrepancy in the simulation of $PM_{2.5}$ composition and other factors discussed in the MS and quantified in the new Table S1.

Decreased PM2.5 in CIGP due to AR feedback. The authors mentioned this issue in L427-436 but didn't explain the reason clearly. How about the changes in winds due to AR, which may be a reason.

Response: Over the CIGP, the AR feedback causes a depletion of surface $PM_{2.5}$, which is likely due to their hygroscopic growth, and then dry deposition (e.g. average dry deposition mass concentration of $PM_{2.5}=331 \ \mu g/m^2/hr$ with AR feedback and 282 $\mu g/m^2/hr$ without AR feedback on 24th December) in

dense fog. The increase in RH with AR feedback favors the growth of aerosols in size by the uptake of water.

Wind speed increases and $PM_{2.5}$ decreases with AR feedback on December 23, whereas wind speed decreases and $PM_{2.5}$ increases on December 24 (Fig.1 below) which is expected due to reduced dispersion in the stable boundary layer. However, there is a reduction in $PM_{2.5}$ (>10 µg/m³) on 24 December between 00:30 and 03:30 hrs, which is likely due to loss via deposition in fog and not due to change in wind speed with AR feedback.

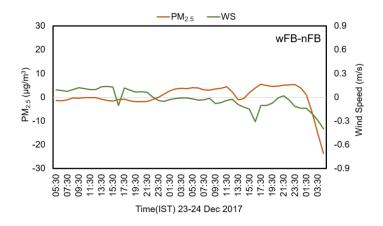


Figure 1. Time series showing the difference in $PM_{2.5}$ and windspeed due to AR feedback over CIGP (79-83E,26-28N).

High particulate Cl. The model can't reproduce the observed high Cl that could come from trash-burning and industrial emissions. I am wondering if trash-burning emissions from Chaudhary et al. (2021) can represent Cl emissions at a city scale.

Response: The reviewer makes a good point in that the trash-burning emissions inventory may not represent trash burning in urban regions that need high horizontal grid spacing to capture variations among neighbourhoods in the city. The trash-burning emissions of <u>Chaudhary et al., (2021)</u> are available at 10 km resolution and capture city-scale emissions of larger megacities like Delhi, which occupy several grid cells. However, the inventory contains annual emissions and fails to resolve the seasonality of trash-burning emissions identified by <u>Nagpure et al., (2015</u>). They suggested almost all the wasteburning emissions in neighbourhoods with higher socioeconomic status in Delhi occur due to the use of waste as cheap heating fuel by individuals such as night watchmen and pavement dwellers. Hence these "rich" neighbourhoods see little to no waste burning for most of the year (≤18 kg/km²-day), except during winter (~89 kg/km²-day). Hence wintertime waste-burning emissions, in central Delhi, may be underestimated in the Chaudhary et al., (2021) inventory, which only considers waste burning that occurs due to lack of collection infrastructure, and at landfills and, therefore, shows a concentration of waste burning emissions around the periphery of the Delhi NCR, but low waste burning emissions in the relatively prosperous City centre. During radiation fog events with very limited transport, such a

discrepancy in the local emissions can indeed drive a model measurement discrepancy. However, such "heating motivated" waste-burning emissions would ideally need to be included in an updated residential sector emission inventory, which should also capture the seasonality of residential sector emissions caused by heating demand and hot water needs.

In addition, emissions from other sources (e.g., industries) are not accounted for in WRF-Chem and the WRF-Chem chemistry option used does not represent halogen chemistry. The results shown in this paper suggest that further work on HCl emissions needs to be conducted in order to improve aerosol composition in the IGP region.

Fog duration. In Fig.15, I find the results from three simulations are quite similar and I suggest the authors to clearly identify their difference.

Response: To better quantify the differences among the three simulations, a box plot and table have been added comparing the three simulations as also suggested by reviewer 1, which show that the average fog intensity and duration of fog increases comparatively with AR feedback than without AR feedback and without aqueous phase chemistry. Discussions added in the MS

Minor comments:

L18-20: the model tends to strongly underestimate observed PM2.5.

Response: The line has been modified as suggested.

L33: "These processes" refers to aerosol-radiation interaction and aqueous phase chemistry? The latter can't change PBL meteorology.

Response: Aqueous phase chemistry cannot directly change the PBL meteorology, however the increased $PM_{2.5}$ concentrations and size of the aerosols caused by the aqueous phase chemistry affect the PBL dynamics through AR Feedback and hence fog formation

L42: "NOx" refers to emission or concentration?

Response: NOx here refers to NO₂ column concentration. It has been rephrased accordingly in the text.

L47&L55: duplication for NAAQS

Response: Duplicate has been removed.

L123-125: I suggest to move this description to Section 2.2 Observations

Response: Moved to Section 2.2 Observations as suggested

L183: please rephrase "aromatic compounds HONO"

Response: rephrased as "aromatic compounds, HONO"

L270-272: again, this information should be given in the Observations part if not.

Response: Removed, as it is already mentioned in Section 2.2 Observations

L300: in Fig.2, I find most of the correlation coefficient (r) for RH is below 0.87. so I am not convinced by the "r2>0.75".

Response: It is the correlation coefficient **r** and not \mathbf{r}^2 . Thank you for bringing it to our attention. "r2>0.75" corrected as "r>0.75".

L308: please change "are" to "is".

Response: Corrected in the MS as suggested

L351-352: I suggest the authors to show the correlation coefficient using daily data to justify this argument.

Response: A table has been added to show the statistics between simulated and measured PM_{2.5}.

Section4: It looks the authors failed to discuss about the uncertainties in chemistry scheme used in the model, which is also an importance source of model biases.

Response: Discussion added in the MS.

L532: In fact, the model can't reproduce the observed aerosol composition. The following discussions on previous studies couldn't be helpful.

Response: We agree with the reviewer that the model could not reproduce the observed aerosol chemical composition. We took several steps to improve the model performance as described below. First, we tested three different meteorological configurations as discussed in the manuscript and used the best configuration for conducting sensitivity simulations focused on exploring aerosol-fog interactions. Second, we included the best estimates of trash-burning emissions to represent anthropogenic chloride aerosols in our configuration. While incorporation of trash burning emissions did improve the model simulations of PM_{2.5} and better captured the day-to-day variability of PM_{2.5}, we still see large underestimation of chloride aerosols in the model. This indicates the need for more work on better quantifying trash burning emissions, which may not only improve particulate chloride in the model but also improve simulations of other aerosol chemical components through aerosol thermodynamics. Despite these challenges, the model was able to simulate the same changes in inorganic composition during fog events as reported by observational studies referred to in the MS. This encouraged us to perform sensitivity simulations described in the manuscript. We anticipate that the challenges identified in our study will provide motivation to both our and other groups working on this part of the world to close this gap between observations and model simulations.