This study uses the Weather Research and Forecasting model coupled with chemistry (WRF-Chem) to understand how (a) spatial distribution, mass, and composition of aerosols and (b) formation, duration and dissipation of fog is affected by aerosol-radiation feedback and aqueous-phase chemistry during a winter fog event over the highly polluted Indo-Gangetic Plain (IGP). The paper is well written and well structured. The study is important and within the scope of the journal, but following major and minor comments need to be addressed before I can recommend the paper for publication.

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

1. The motivation to design EXP1, EXP2 and EXP3 is not very clear. Previous studies (for e.g., Mohan and Gupta, 2018) have already confirmed that ACM2 scheme performs better than other parameterizations. Does using a new PBL scheme improves the model performance or nudging the soil moisture improves the model performance?

Response:

The reviewer is correct in that Mohan and Gupta (2018) already showed that the ACM2 scheme performs better than other PBL parameterizations. However, they tested the YSU and ACM2 schemes during the summer time (1-15 June 2010) and focused on the evaluation of temperature, wind speed, PBL height, ozone, and PM_{10}. Since our period focuses on the winter season fog, it was critical for us to ensure that model captures all the relevant meteorological parameters reasonably well including relative humidity. That is why we designed EXP1, EXP2 and EXP3. Furthermore, to investigate both aerosol-radiation and aerosol-cloud interactions, the YSU (and other) PBL schemes in WRF-Chem can be used, but the ACM2 scheme is implemented in WRF-Chem differently than the other schemes making aerosol-cloud interactions not possible to investigate. That was our additional motivation to test the YSU PBL scheme. As shown in the manuscript, the YSU PBL scheme with NCEP initial and boundary conditions (IC/BC) underestimated the relative humidity preventing fog formation. Using the ERA-Interim IC/BC instead of NCEP improved the relative humidity predictions (Figure S1, supplement) but did not simulate fog well (Figure 3 of manuscript). Thus, following Mohan and Gupta (2018), the ACM2 PBL scheme was tested, giving predictions of fog comparable to observations (Figure 3 of manuscript). Including soil moisture nudging improved the surface fluxes and therefore the surface temperature and relative humidity. However, by switching to ACM2, only aerosol-radiation

interactions could be examined. Therefore, our results should be seen as complementing Mohan and Gupta (2018).

2. L225: "Aerosol cloud interactions are not possible"- does that mean fog droplets do not activate? Explain. How realistic is the fog lifecycle in EXP3 without 'Aerosol Cloud Interactions'?

Response:

In WRF-Chem, the aerosol-cloud interactions, that is, the dependence of cloud drop activation on the number of CCN which varies when using a prognostic aerosol scheme, is controlled by the mix-activate scheme that is called from a routine that computes vertical mixing of trace gases and aerosols. Because the ACM2 PBL scheme computes the vertical mixing of trace gases and aerosols and does not call mix-activate, it is not possible to examine the effect of aerosol-cloud interactions with our model configuration. Cloud (fog) water does form but is formed based on a prescribed CCN concentration that does not vary during the simulation.

Several modeling studies have confirmed that aerosol cloud interactions (ACI) plays important role in increasing fog liquid water content and lifetime, sometimes overweighing ARI in polluted environment (Maalick et al., 2016; Yan et al.,2021). Thus, any difference in the fog lifecycle due to ACI is not accounted for in our simulation.

3. **Why Figure S1 only shows 6 stations? On that note, the authors need to explain how the gridded model results are compared to point observations? The wind speed observations are present only in 4 stations. Among that at Amritsar and at Delhi-RKP, wind speeds have large biases. The hypothesis of 'low measurement height and obstructions' needs some evidence. The authors should read previous literature and figure out how WRF-Chem performs in those regions? Are there any systematic biases?**

Response:

We have compared the model output with observation (CPCB) for nine stations over IGP that are also considered for the statistical analysis in the Taylor diagram. However, to compare the time series of the meteorological parameters, only major cities as representative stations in each region of IGP have been shown. There were gaps in the wind speed dataset from CPCB for several stations of IGP, explaining why wind speed from only four stations are shown. At such low wind speeds, WRF-chem in general overestimates wind speed. The analysis code we use finds the WRF-Chem grid point nearest to the station's latitude and longitude. Several earlier studies have reported this bias in wind speed (Mohan and Gupta 2018; Pithani et al.,2019). However, this is unlikely to be the only or the dominant reason for the selective high bias as the photograph of the Amritsar monitoring station shows:

Figure 1: Meteorology monitoring station at Amritsar

For R. K Puram station, we have not been able to obtain a photograph of the station but it is located somewhere in the yellow polygon either along the main road (photo provided) or in the Sector 3&4 park, or in the campus of the National Institute of Health. All these locations have a lot of obstacles to free airflow. There is no large free space anywhere in that area. Moreover, WRF-Chem does not have the capability to represent building meteorology and parameterizes the effects of urban areas on meteorology through roughness length, which likely leads to overestimation of wind speed.
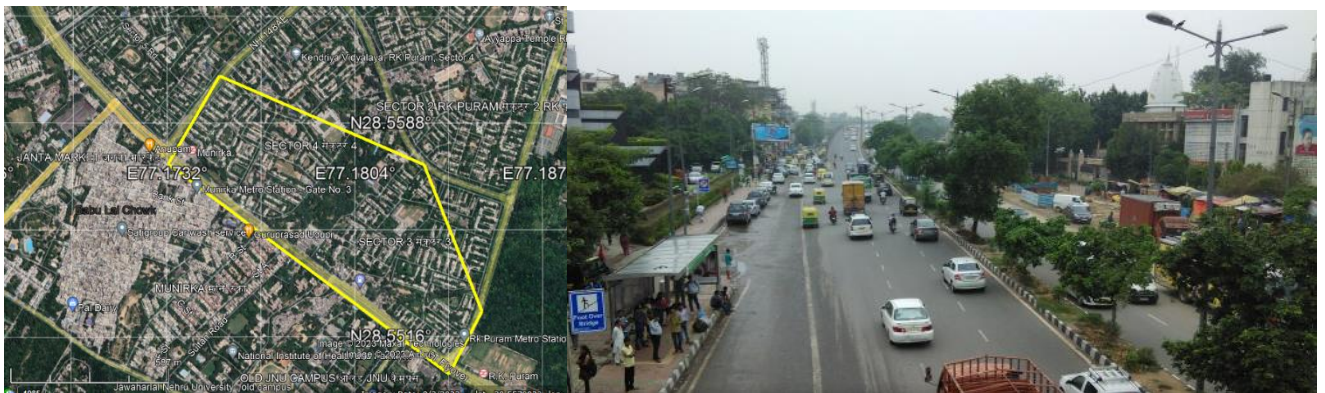

Figure 2: Map showing area around the meteorology monitoring station at RKPuram, Delhi.

4. The model performance in predicting mass and composition of PM$_{2.5}$ is poor. The authors should add a table showing the correlation and error between observations and simulation and compare that with existing literature. Is it possible to design a sensitivity experiment by scaling up the rate of emissions (including the HCl emissions) in the EDGAR-HTAP inventory. Why inorganic ions (other than chloride) are also heavily underestimated and fails to predict the diurnal variations (sulfate for example)?

Response:

Table. Statistics for PM$_{2.5}$ concentrations observed and modelled at four stations.

	Mean_PM$_{2.5}$ CPCB	Stdev	Mean_PM$_{2.5}$ WRFChem	Stdev	MB% NMB	RMSE	NMRSE	r	p
Amritsar	83. 2	32.0	85.0	44.6	-2.17	41.41	0.50	0.45	2.5E-07
Delhi	209	76.0	108	116.	48.49	137	0.65	0.61	2.7E-13
KNP	232	141	109	74.8	53.02	172	0.74	0.52	1.3E-09
Patna	211	78.0	141	98.3	31.92	138	0.65	0.08	0.47

* MB%= (Observed-Simulated)/Observed) *100
* NMRSE=RMSE/ Mean_PM$_{2.5}$ _CPCB

The statistical analysis between WRF-Chem results and observations (Table S1) shows a minimum mean bias for PM$_{2.5}$ at Amritsar (-2.2%) while at other stations it ranges from 48 to 53% similar to the reported range of model bias (underestimated by 40–60%) in winter over IGP by earlier studies (Bran and Srivastava, 2017;Ojha et al., 2020). The RMSE values range from 41 to 138 µg/m^3 (normalized RMSE~0.4 to 0.7) are comparable to the reported values by these studies. The Pearson correlation coefficient (r) for the simulated and observed day-to-day variation in PM$_{2.5}$ lies between 0.4 and 0.7 for all the stations in Fig. 6 except at Patna which lies within the range in these studies. The poor correlation at Patna is due to the loss of PM$_{2.5}$ during fog in the model as discussed earlier.

This is one of the few studies where aerosol composition has been evaluated for the IGP. WRF-Chem fails to predict chemical composition correctly, especially the chloride concentration. It is hypothesized that the aerosol chloride is from trash burning, thus we incorporated trash-burning emissions which include HCl emissions from Chaudhury et al.,(2021). However, the inventory only covers trash burning that occurs due to lack of waste management infrastructure, and does not include the well documented use of trash as wintertime heating fuel (Nagpure et al., 2015). Adding trash burning emissions increased PM$_{2.5}$ concentrations (comparison not shown in manuscript), however, a large bias between simulated and observed PM$_{2.5}$ still exists. The underestimation is likely caused by a low bias in the residential burning emission inventory and a failure of the emission inventory to represent residential sector emissions (including HCl emissions) from both cow dung burning and from the use of trash as cheap heating fuel properly. Since solid fuel is the second largest source for several species including SO$_2$ and NH$_3$ this affects the model measurement agreement of all species. Punjab, being a relatively prosperous state with a comparatively high per capita LPG refilling rate shows the lowest discrepancies between the measurement and the modelled data. In Uttar Pradesh and Bihar, the reliance on cheap solid fuels is highest among all Indian states, and discrepancies are larger. In Delhi missing industrial sources may also add to the model measurement discrepancies. Since the present residential emission inventory lacks residential sector HCl emissions and fails to reproduce current spatial emission patterns, a scaling is

unlikely to solve the problem. Emission inventories still propagate the patterns prevalent in the times before LPG usage became an indicator of social status. Consequently, they have the highest per capita residential sector emissions in prosperous states such as Punjab (where people used to be able to afford 3 meals a day) and lower per capita emissions in poorer states (where cooking used to be restricted to once per day). This spatial pattern has now reversed, but no fundamental inventory emission inventory revision has happened. Instead, old spatial patterns are just scaled to keep pace with population growth.

A study by Pawar et al., (2023) showed an improvement in ammonium prediction by scaling HCl (3xHCl) demonstrating the uncertainties in the chemistry scheme of the model and that model improvements are required to correctly simulate PM composition. To better understand why WRF-Chem poorly predicts aerosol composition, extensive work needs to be pursued by obtaining more observations and conducting other sensitivity simulations like the one suggested by the reviewer. This work (and others e.g. Pawar et al., 2023) is simply pointing out the need for further work to understand the processes affecting aerosol composition in the IGP.

5. **Both CIGP and EIGP regions are foggy. However, for the wFB simulation, in figure S5, the Single Scattering Albedo increases at EIGP but decreases at CIGP near the surface. Why?**

Response: Figures 9 and 11 in the manuscript show the differences in scattering and absorbing aerosols (i.e., sulfate and BC). In EIGP, sulfate concentrations have a larger concentration with AR feedback than without AR feedback with time periods where the difference is >1 ug/m^3 (Fig.11). The BC concentration changes are small (<0.5 µg/m3) in the EIGP, resulting in a higher SSA near the surface with AR feedback in EIGP. In the CIGP, BC concentrations increase while sulfate aerosols decrease within the PBL with AR feedback (Fig.11) compared to the simulation without AR feedback. A decrease in SSA is seen for the CIGP throughout the boundary layer while in EIGP the decrease occurs near the top of the PBL. Also contributing to the higher SSA in EIGP is the increase in RH (Fig. 9) due to AR feedback which favours the growth of aerosols in size by uptake of water and the production of secondary aerosols such as SO_4^{2-} and NH_4^+.

6. **When composition of different inorganic ions is discussed, is it an average over the entire sub-region? In that case a standard deviation value should also be reported. Are there previous similar studies from where the authors can compare their results? Inside CIGP and EIGP, are there any differences in composition between the foggy and non-foggy grid boxes?**

Response: Timeseries of inorganic ions in Fig 13 is an average over the region bounded by the box in Fig 12a. The standard deviation has been added to the figure as suggested.

Gupta and Mandariya.,(2013) showed a decrease in PM$_1$ in fog due to wet scavenging by fog droplets (referred in the discussion on timeseries of PM$_{2.5}$). The box plot in Fig.3 below shows the difference in the inorganic composition (SO_4^{2-} and NH_4^+) with and without aqueous phase chemistry in foggy and

non-foggy grids of CIGP and EIGP. The inorganic ions concentration is higher with aqueous phase chemistry than without aqueous phase chemistry in both the foggy and non-foggy grids in both the regions. The difference is larger in foggy grids compared to the non-foggy grids for both SO_4^{2-} and NH_4^+. However, SO_4^{2-} shows large variability in foggy grid points and with aqueous phase chemistry, likely due to both the formation and loss processes occurring in fog as shown in Fig 13 and discussed in section 6 of manuscript. This indicates that SO_4^{2-} is largely formed in fog through aqueous phase chemistry.
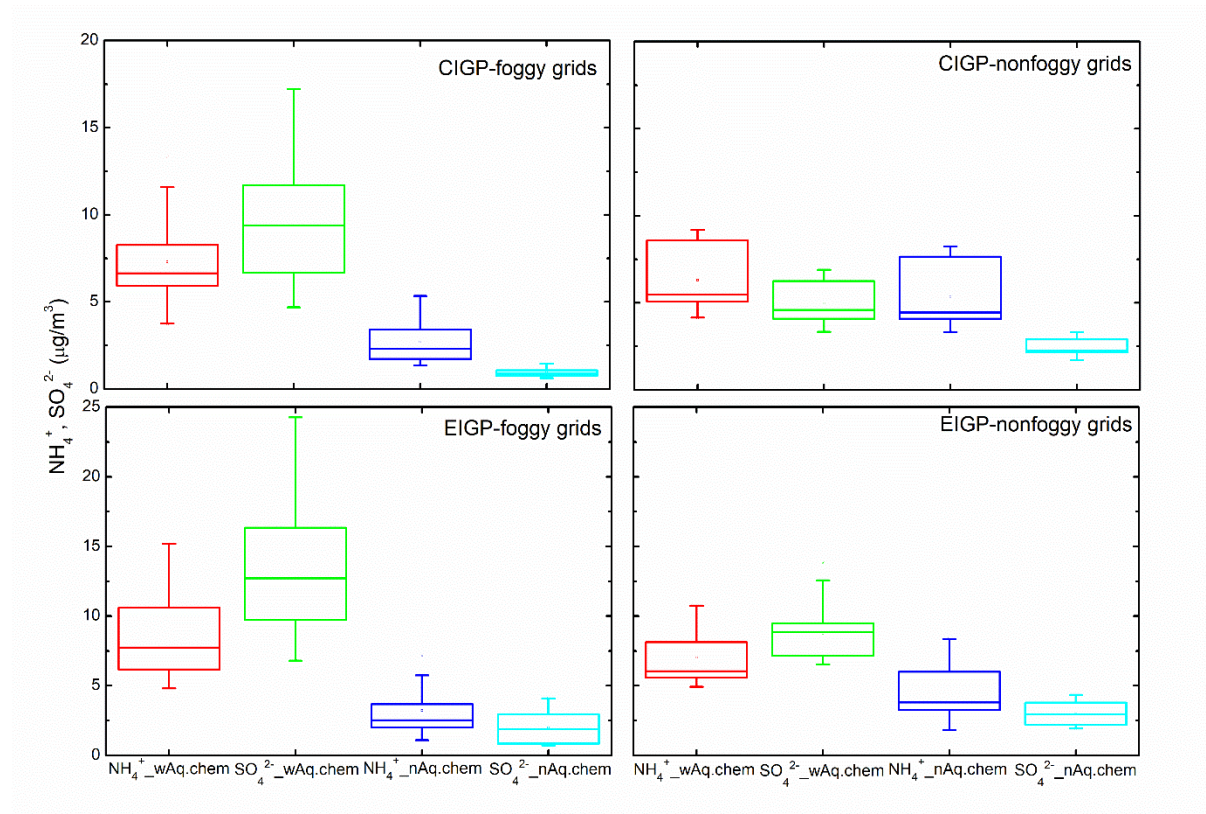

Figure 3: Box plot showing SO_4^{2-} and NH_4^+ with and without aqueous phase chemistry included in the model, averaged for the region CIGP and EIGP for foggy and non-foggy grids.

7. **Why fog deposition is extremely important in the area marked by the box in Fig. 12a? and less important in other areas? Are the meteorological conditions different?**

Response: The region bounded by the box has dense fog because of increased RH with the AR feedback with a maximum difference of ~ 4.5% compared to the surrounding region (<2%). As discussed in the manuscript, the inclusion of aqueous phase chemistry produces and adds sulfate to the aerosol mass, which will increase the PM$_{2.5}$ fog deposition flux. and increase the role of PM$_{2.5}$ on radiation for the simulation with AR feedback. Increase in PM$_{2.5}$ contributes to AR feedback and, consequently, fog intensity increases. Also note that deposition of aerosol with cloud water increases in dense fog.

8. **Why is the fog water content higher when aqueous chemistry is included?**

Response: It is discussed in section 6.

We observe an increase in $PM_{2.5}$ (esp. SO_4^{2-} and NH_4^+) when aqueous phase chemistry is included in the model. The aqueous chemistry increases both the mass of $PM_{2.5}$ and the size of the aerosols, both of which contribute to AR feedback, thus increasing RH and PBL stability. The increase in RH also saturates the air, promotes aerosol growth by water uptake, and thus favors fog formation.

9. **Figure 15 needs to be modified and variations at three separate IGP regions need to be shown with a median and interquartile range, taking foggy grid boxes. Observations from the WiFex Campaign should also be added.**

Response: A box plot is shown below for WRF-Chem simulated QCloud (QC) taking foggy grid boxes with AR feedback, no AR Feedback, and no Aqueous phase chemistry experiments over CIGP and EIGP. We do not have fog over NWIGP in the model for the study period. Each box plot represents the median (50th percentile), with the box indicating the interquartile range (IQR) between the 25th and 75th percentiles. QC is comparatively higher with AR feedback than without AR feedback and without aqueous phase chemistry for both CIGP and EIGP. IQR is larger for simulation with and without AR feedback than without aqueous phase chemistry in CIGP showing large variability in the LWC. Whereas in EIGP the variability in LWC is greater in simulation with AR feedback compared to the other two experiments. We also observe an increase in fog LWC with AR feedback and aqueous phase chemistry compared to without AR feedback and the absence of aqueous phase chemistry (Fig.5).
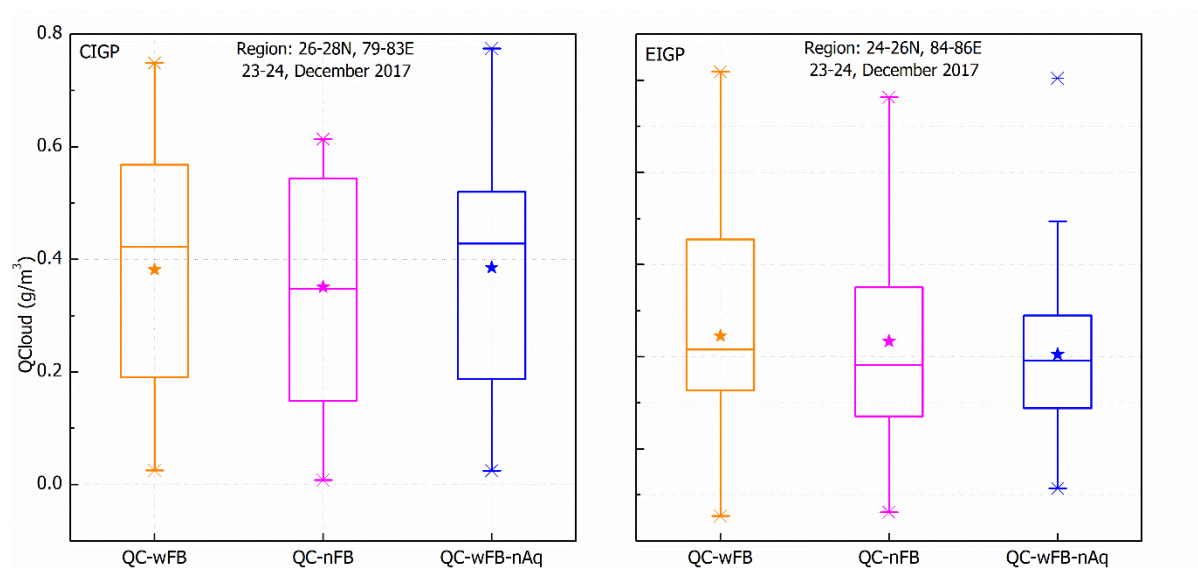
Figure 4: Box plot showing LWC at the surface for the three experiments, with and without AR feedback and without aqueous phase chemistry for CIGP and EIGP. NWIGP does not have fog during the study period.

WiFEx has observation for aerosols and fog at Delhi, while the WRF-Chem simulation does not have fog in NWIGP. Therefore, the WiFEx observations are not included in the plot.
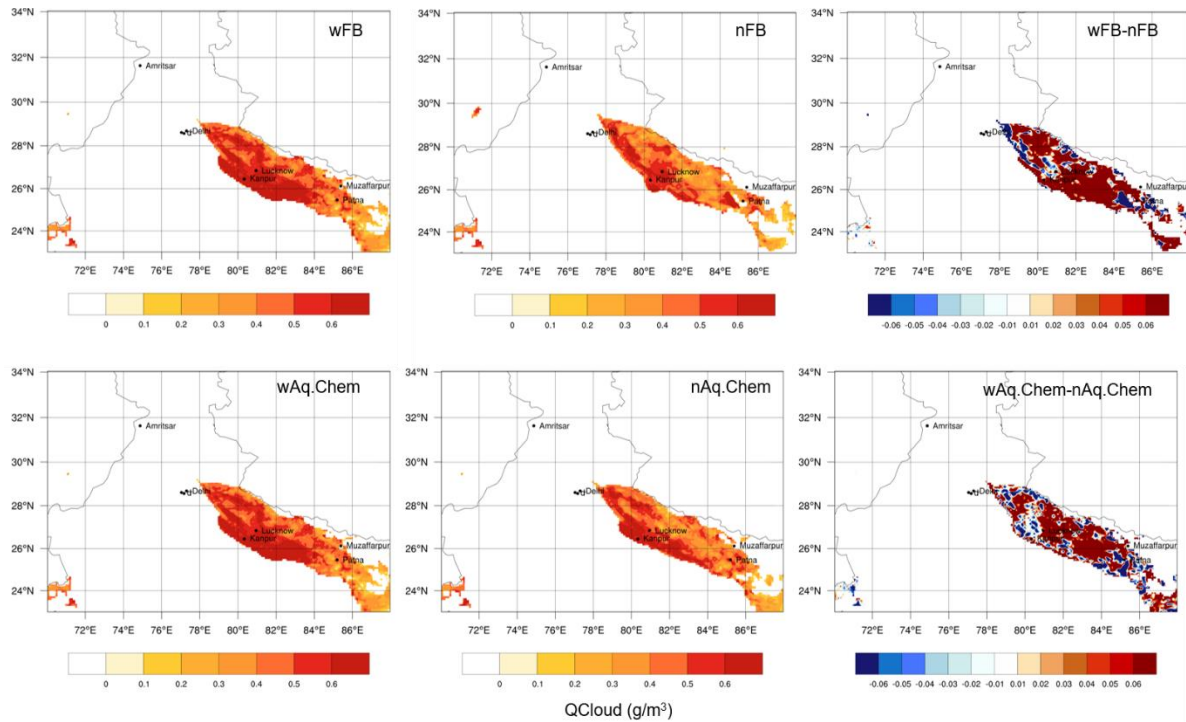

Figure5. Map showing LWC at the surface for the three experiments, with and without AR feedback, without aqueous phase chemistry and their anomalies.

10. L559 mentions that Aerosol-Radiation feedback affects the timing of the fog. However, L584 mentions that aqueous phase chemistry together with radiation feedback promotes early fog formation. This is confusing. How does aqueous chemistry affect fog formation? Explain.

Response: The aqueous chemistry increases both the mass of $PM_{2.5}$ and the size of the aerosols, both of which contribute to AR feedback. Consequently, the AR feedback affects the PBL dynamics affecting the T2 and RH near the surface. This further affects the fog formation and dissipation. We noted in Table 1 and 2 (in response to minor comments 7) that the timing of fog with AR feedback, without AR feedback, and without aqueous phase chemistry are different with formation occurring earlier in the simulation with AR feedback which also includes aqueous phase chemistry. It is likely due to the contribution of added $PM_{2.5}$ to AR feedback when aqueous phase chemistry is included.

Minor Comments

1. The abstract needs to be shortened. The goal of the paper on L14 needs to be more specific. 'Aerosol-Radiation Interactions' and 'Aqueous Chemistry' should be mentioned in the beginning of the abstract.

Response: Abstract have been modified in the manuscript.as suggested.

2. L116: 'it is divided into three areas...'- Describe the areas. What are the spatial extends (latitude, longitude)? Which states/major cities included in each of the three areas?

Response: The three regions are defined by the following latitude-longitude range which has also been added in the text.

'it is divided into three areas, northwest (NWIGP: latitude-longitude range, 27°N-32°N,75°E-79°N), central (CIGP: latitude-longitude range, 25°N-28°N,79°E-83°E), and east (EIGP: latitude-longitude range, 24°N-27°N, 83°E-87°E)'. Discussion on major cities have been added to the Observations section as suggested by Reviewer 2.

3. L128: 'the WRF-Chem model version 4.0.3 has been used for this study'-cite a few studies who have used WRF-Chem or similar models to study aerosol radiation feedback in fog.

Response: WRF-Chem has been widely used to study the role of aerosol radiation (AR) on air quality and haze episodes. References have been added in the text as follows:

"Earlier studies have successfully used WRFChem to predict fog (Pithani et al., 2019) and in the study of aerosol-radiation feedback on air quality (Kumar et al., 2020; Bharali et al., 2019) and the study of aerosol-fog interactions (Shao et al., 2023)."

4. L170: Avoid Italics. 'wFB-nFB' needs a bit more explaining. Write the expression in a way to make it more understandable to a general audience: for example: (Properties/ Parameters in wFB - Properties/ Parameters in nFB)

Response: Modified as suggested:

Impact of radiation feedback=Parameters in wFB- Parameters in nFB

Impact of aqueous phase chemistry= Parameters in wAq.chem- Parameters in noAq.chem

5. Figure1: True color MODIS reflectance map doesn't confirm presence of fog. Could they be low level clouds? Also, a different color (other than black) should be used to mark the stations.

Response: True color MODIS reflectance map represents low cloud indicative of likely fog. These cloud layers align with the fog imagery from INSAT 3D; therefore, we used the image to represent fog. Color of text marking stations changed from black to yellow.

6. The Taylor's diagram needs to be explained more carefully. How are the standardized normalized deviations calculated (include an equation)?

Response: The following equation to calculate standardized deviations in Taylor diagram has been added:

The centered RMS difference, the correlation, and the standard deviation are related by the following formula:

$E'^2 = \sigma_o^2 + \sigma_m^2 - 2\sigma_o\sigma_m R$

where R is the correlation coefficient between the model-simulated and observed fields, E' is the centered RMS difference between the fields, and σ_m^2 and σ_o^2 are the variances of the model-simulated and observed fields, respectively. The correlation(R), centered RMS difference (E') and standard deviations of the model simulated and observed fields are calculated by the following formulas:

$$R = \frac{\frac{1}{N}\sum(M_n - \overline{M})(O_n - \overline{O})}{\sigma_m \sigma_o} \qquad (1)$$

$$E'^2 = \frac{1}{N}\sum[(M_n - \overline{M}) - (O_n - \overline{O})]^2 \qquad (2)$$

$$\sigma_m^2 = \frac{1}{N}\sum(M_n - \overline{M})^2 \qquad (3)$$

$$\sigma_o^2 = \frac{1}{N}\sum(O_n - \overline{O})^2 \qquad (4)$$

where the overall mean of a field is indicated by an overbar.

7. Add a table showing timings of the fog and liquid water content for all the sensitivity studies, for three different regions (with a mean and standard deviation). Also, add mean and standard deviations of observations from all stations (for each area).

Response: A table has been added in the manuscript listing the start and end time of two fog events with LWC for the sensitivity experiments, with AR feedback, no AR feedback and no Aqueous phase chemistry. The table includes fog for the two regions CIGP and EIGP along with two stations in each region. NWIGP does not have fog during the study period. The table along with the box plot of fog in response to major comment 9 above shows that fog intensity and duration changes with AR feedback compared to that without AR feedback and without aqueous phase chemistry for both Fog1 and Fog2. Fog forms earlier with AR feedback compared to the simulation without AR feedback in CIGP as well as EIGP, while it dissipates earlier with AR feedback in CIGP and delayed in EIGP compared to that without AR feedback and without aqueous phase chemistry. In both the regions, the fog lifetime increases with AR feedback. All the stations, however do not show the same pattern, for example, fog 1 in Luckow forms and dissipates at the same time for simulations with AR feedback and without aqueous phase chemistry, and fog2 forms later with AR feedback than without AR feedback. Patna shows no difference in fog2 formation in all the three experiments. We recommend that simulations at higher spatial and temporal resolutions would be better to study the fog dynamics at point locations.

Table 1: Table showing the start and end time of fog1 on 23-24 December 2017 with LWC for the sensitivity experiments, with AR feedback, no AR feedback and no Aqueous phase chemistry

Fog 1 (December 23-24, 2017)

	EXP-wFB			EXP-nFB			EXP-nAq.Chem		
	Start time (IST)	End time (IST)	Duration of Fog	Start time (IST)	End time (IST)	Duration of Fog	Start time (IST)	End time (IST)	Duration of Fog
CIGP	16:30	15:30	**23h**	18:30	17:30	**23h**	18:30	15:30	**21h**
LWC (g/m^3)	0.034	0.036±0.032		0.141±0.154	0.068±0.005		0.184±0.138	0.034 ±0.021	
Kanpur	05:30	13:30	**8h**	05:30	12:30	**7h**	05:30	12:30	**7h**
LWC (g/m^3)	0.334± 0.487	0.017		0.458±0.357	0.173± 0.071		0.533	0.025± 0.0123	
Lucknow	23:30	14:30	**15h**	00:30	14:30	**14h**	23:30	14:30	**15h**
LWC (g/m^3)	0.269±0.145	0.087±0.040		0.232±0.132	0.029±0.024		0.139±0.084	0.025±0.012	
EIGP	21:30	12:30	**15h**	23:30	10:30	**11h**	21:30	10:30	**13h**
LWC (g/m^3)	0.099±0.092	0.007		0.198±0.188	0.084±0.060		0.026±0.008	0.153±0.119	
Patna	00:30	12:30	**12h**	04:30	10:30	**6h**	02:30	10:30	**8h**
LWC (g/m^3)	0.100±0.090	0.007		0.009±0.005	0.038±0.041		0.196±0.198	0.166±0.130	
Muzzafarpur	05:30	11:30	**6h**	06:30	10:30	**4h**	06:30	09:30	**3h**
LWC (g/m^3)	0.112±0.146	0.043±0.057		0.051±0.041	0.003		0.142±0.151	0.157±0.064	

Table 2: Table showing the start time of fog 2 on 23-24 December 2017 with LWC for the sensitivity experiments, with AR feedback, no AR feedback and no Aqueous phase chemistry

	Fog 2 (December 24, 2017) Start time (IST)		
	EXP-wFB	EXP-nFB	EXP-nAq.Chem
CIGP	**19:30**	**20:30**	**21:30**
LWC (g/m^3)	0.025	0.008±0.007	0.025
Kanpur	21:30	22:30	23:30
LWC (g/m^3)	0.041±0.007	0.298±0.218	0.482±0.398
Lucknow	21:30	20:30	00:30
LWC (g/m^3)	0.203±0.165	0.005	0.229±0.209
EIGP	**00:30**	**01:30**	**01:30**
LWC (g/m^3)	0.024±0.030	0.072±0.088	0.014±0.009
Patna	03:30	03:30	03:30
LWC (g/m^3)	0.030± 0.046	0.018	0.060
Muzzafarpur	04:30	No fog	No fog
LWC (g/m^3)	0.159±0.038		

• Fog still exists when the simulation ended on 25 December 2017, 00UT (5:30 IST)

8. Figure 2: x and y-axis need proper labels. Also, can the authors use a color other than blue for EXP2?

Response: Figure 2 has been modified as suggested

9. What is 'ddmass'. Write clearly in figure captions.

Response: 'ddmass' is dry deposition flux

10. 'ug/m3' should be changed to 'μg/m3'.

Response: Corrected in the MS

11. L522: Need citations for PM2.5 compositions.

Response: Citations have been added in the MS

12. L567: change '01 UTC' to '01:00 UTC' and L 568: change '10 UTC' to '10:00 UTC'.

Response: Time format has been changed in the MS