

## Interactive comment on “Amplification of South Asian haze by water vapour-aerosol interactions” by Vijayakumar S. Nair et al.

*This paper uses a regional climate model, RegCM4, to assess the contribution of aerosol hygroscopic growth to the total aerosol optical depth and examines various effects resulted from the aerosol-water vapor interactions in South Asia in winter. They include the positive feedbacks between aerosol hygroscopic growth-surface cooling-PBL relative humidity, the drying effects in the free troposphere, and the worsening of air quality. These effects are assessed by a set of designed model experiments targeted to isolate the change of AOD, temperature, and RH due to the radiative forcing due to the aerosol hygroscopic growth and due to the meteorological feedback. Several interesting results are presented, including the change of AOD and surface radiation due to the aerosol hygroscopic growth and resulting change of surface temperature and relative humidity. It is also interesting to see the opposite trends of RH in the PBL and in the free troposphere that might be explained, at least partially, by the feedbacks from the interactions between aerosol and water vapor. Having said that, there are several major concerns regarding the quality of the paper. My major and specific comments are presented below. Therefore, I recommend a major revision of the paper before it can be published on ACP.*

*We thank the reviewer for the evaluation summary and the extensive comments to improve the manuscript.*

### Major comments:

1. *The paper is particularly focusing on the effects of aerosol hygroscopic growth on two meteorological variables, T and RH, which is fine. But attributing the amplification of the winter haze in South Asia to the aerosol-H<sub>2</sub>O interaction is rather narrow-minded, because there is another important factor, the effects of aerosol-radiation interactions due to the presence of absorbing aerosols, which is probably a more dominant factor in amplification of the South Asian haze but was not addressed at all in the manuscript. I suggest adding one more model experiment, that is to assume all aerosols are not absorbing (e.g., single scattering albedo =1, or omit black carbon), to address that issue.*

*We agree with the reviewer that the amplification of South Asian haze during winter is multifaceted. There are several factors, such as shallow boundary layer, weak/calm winds, high humidity and high concentration of absorbing aerosols, which contribute to this effect. Several modelling and observational studies that addressed the effect of aerosol-boundary layer interaction and direct radiative effects of composite aerosols over the IGP (Nair et al., 2007; Nair et al., 2016; Bharali et al., 2018). However, the contribution of the*

hygroscopic growth of aerosols to AOD and visibility and its implications on the regional meteorology are largely unexplored.

We agree with the reviewer that aerosol-radiation interaction is the more dominant factor over this region due to the high concentration of absorbing aerosols. Experiments 1 and 3 are designed to estimate the aerosol-radiation interaction (Table 1 and Eqn. 3), which include absorbing and scattering aerosols. For that matter, we have validated the model simulated BC mass concentration (shown in Figure 2) using a network of ground-based observatories spread across the region. The total effect of aerosol-radiation interaction on meteorological parameters is discussed in Figure 5 and 6. We have explicitly mentioned in the revised manuscript that radiative effects of hygroscopic-growth of aerosols are the subset of aerosol-radiation interaction of composite aerosols (Eqn. 3 and 5).

As suggested by the reviewer, we have carried out one more simulation where we set the SSA of all the aerosols similar to that of sulphate aerosols ( $SSA \sim 1$ ). The change in temperature and relative humidity for this simulation is shown below. Compared to the ambient aerosol effect shown Figure 5c & d, it is seen that the surface temperature decreases and relative humidity increases in this idealized simulation where no absorbing aerosols are present in the atmosphere.

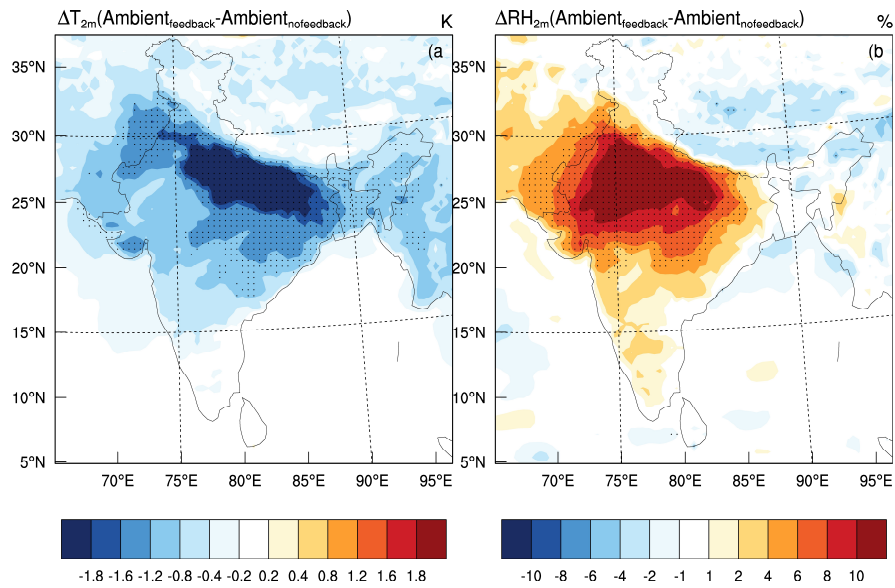


Figure: The change in 2m temperature and relative humidity for completely scattering aerosol system.

2. *Feedbacks: The only met fields dealt with in this paper are surface temperature and RH. Other important met fields, such as clouds, precipitation, winds, and PBL height, are omitted. The*



authors argue that this is because some previous studies have explored these fields. However, at least these other effects should be mentioned and summarized in the context of comparing the magnitude of various effects on haze and air quality.

Complied with. We have revised Figure 5 and included the change in cloud fraction due to radiative effects of (Fig 5c) hygroscopic growth of aerosols and (Fig 5f) due to total aerosols. The discussion on change in boundary layer height and winds are included in the supplementary materials (Figure S6).

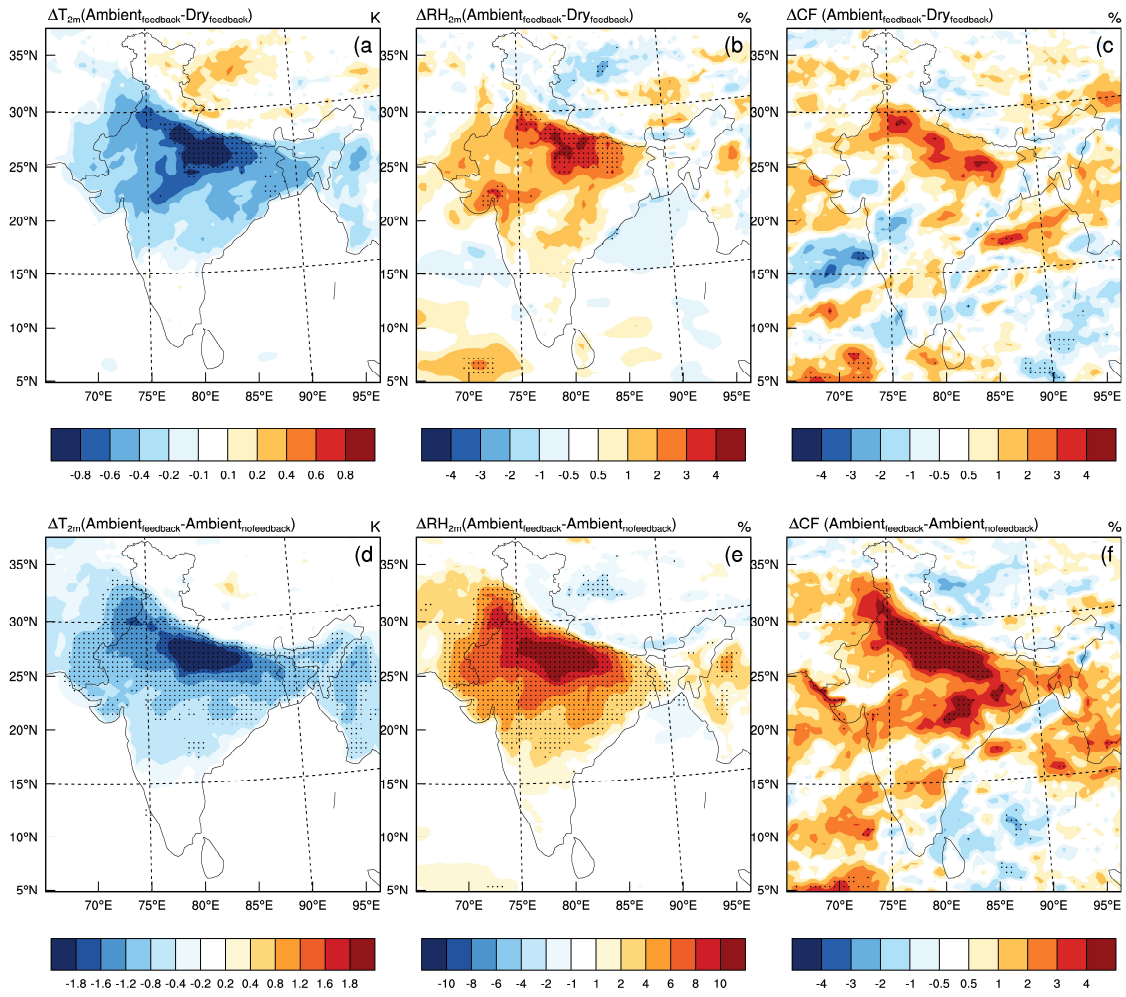


Figure 5: Change in (a) 2m air temperature, (b) relative humidity and (c) cloud fraction solely due to the hygroscopic growth of aerosols (ambient-dry). Change in temperature and relative humidity (RH) due to the total effect of aerosols (dry + hygroscopic growth) is shown (c), (d) and (f). Dots indicate statistical significance of Student's t-test at a 90% level.

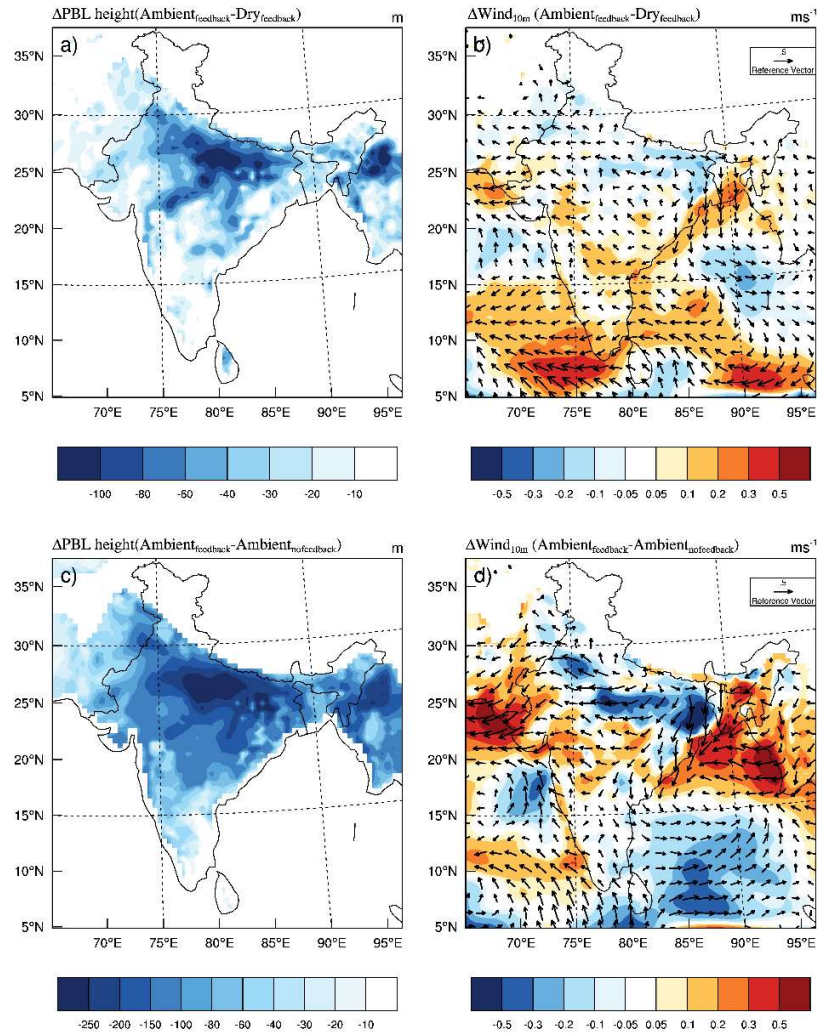


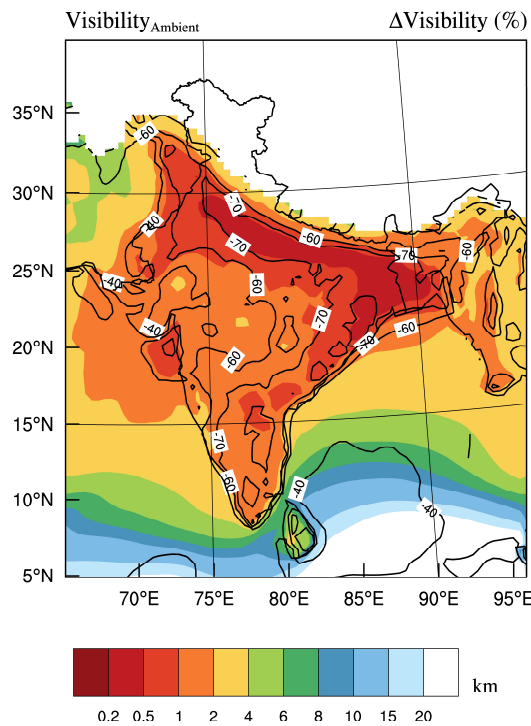
Figure S6: Change in (a) boundary layer height and (b) wind due to hygroscopic growth and meteorological feedback of aerosols. (c) and (d) are similar to figure (a) and (b) but for the total effect of aerosols.

3. The title is about the amplification of South Asian haze, but there is no assessment of the haze condition in the paper, for example, the change of visibility or PM2.5 concentrations. Since the change of AOD does not equal to the change of haze conditions, it is necessary to assess the air quality in more direct way than what was presented in the paper.

This is an important concern raised by the reviewer. We have revised the manuscript with the change in visibility and mass concentration of organics, carbonaceous and PM2.5 to address the impact on air quality.

We agree that AOD may not represent the haze conditions near the surface, especially when the long-range transport of aerosols occurs at high altitudes. Since, the aircraft and lidar observations over the IGP clearly showed that aerosols are mostly confined within the boundary layer during the winter season with lower contributions from the free tropospheric aerosols (Nair et al., 2016), we have used AOD and surface-level parameters like PM<sub>2.5</sub>, and visibility to investigate the implications of aerosol-induced forcing on air quality and visibility.

We have estimated the visibility using the aerosol extinction coefficient and is shown in Figure below. The low visibility conditions represent the poor air quality and haze conditions over the region. We find a 60% reduction in visibility because of the hygroscopic growth of aerosols. The frequency of occurrence of visibility shows a clear decrease for ambient aerosols, this is mostly attributed to the enhancement in aerosol extinction due to hygroscopic growth and an increase in relative humidity due to aerosol-induced surface dimming.



*Figure: Spatial variation of mean visibility over the Indian region for ambient feedback simulation. The percentage variation of visibility changes due to hygroscopic growth with respect to visibility due to dry aerosols is given as line contour.*

Figure below shows the frequency distribution of the daily mean visibility estimated from the aerosol extinction coefficient averaged over the Indo-Gangetic Plain. When

hygroscopic growth of aerosol is activated, the distribution peak shifts toward the lower value to a large extent, which means that there is an increase in the occurrence of the days with lower visibility.

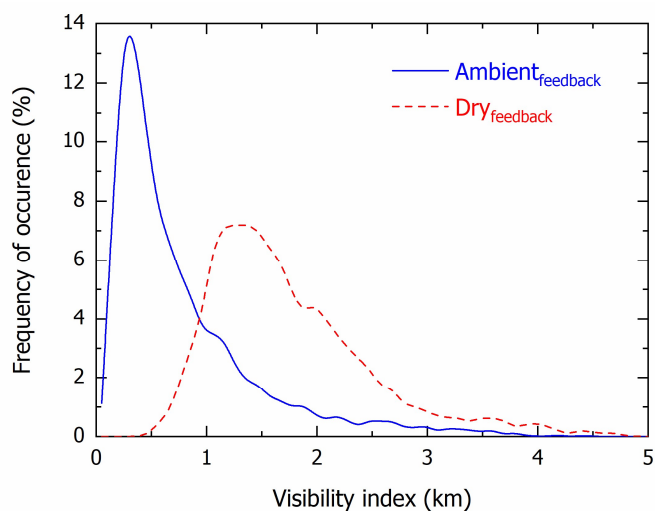
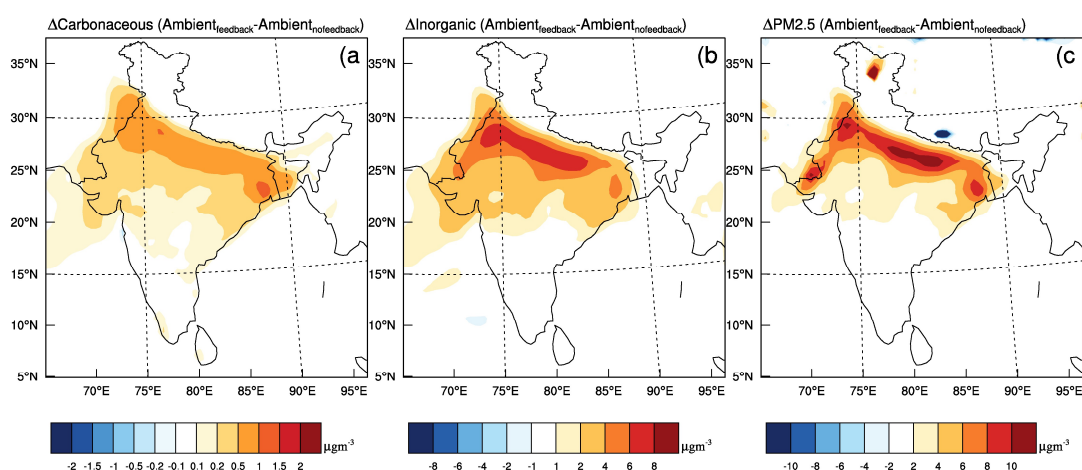


Figure S7: Frequency distribution of spatially averaged visibility over Indo Gangetic Plain for *Ambient<sub>feedback</sub>* and *Dry<sub>feedback</sub>* simulations.

As shown in the figure below, there also exists an enhancement in the PM<sub>2.5</sub> mass concentrations due to the suppression of the boundary layer height induced by the meteorological feedbacks from the aerosol radiation interaction. This results in further degradation of the air quality of this region.



*Figure: Change in near-surface mass loading ( $\mu\text{g m}^{-3}$ ) due to the meteorological feedback of total aerosol radiative forcing for (a) carbonaceous aerosols, (b) inorganic aerosols and (c) PM<sub>2.5</sub>.*

This analysis further confirms that the air quality problems in the IGP are associated not only with the aerosol source strength but also with the ambient humidity conditions and its (aerosol) radiative forcing and meteorological feedback.

Nair, V. S., S. S. Babu, M. M. Gogoi, and K. K. Moorthy (2016), Large-scale enhancement in aerosol absorption in the lower free troposphere over continental India during spring, *Geophys. Res. Lett.*, 43, 11,453–11,461, doi:10.1002/ 2016GL070669.

4. *The analysis and explanations are often confusing. It is not clear what the findings are from the present study and what are from other studies, and how consistent or different they are.*

We are sorry for this ambiguity. We have found that results from the present study and earlier reports got jumbled, especially on page 9. We have revised the manuscript.

**Specific comments:**

*Line 10-11: “This positive feedback mechanism plays an important role in the prevalence of wintertime fog and low air quality conditions over South Asia” – this point has not been demonstrated in the paper.*

The enhancement in the AOD due to the hygroscopic growth of aerosols results in further solar dimming at the surface, reduction in surface temperature, increase in relative humidity and decrease in wind speed. The suppression of the boundary layer height due to these meteorological feedbacks and weaker winds hinders the PM<sub>2.5</sub> dispersion resulting in the enhancement in the mass concentration and thus contributes to the degradation of the air quality over this region. This aspect is addressed in the revised manuscript and please see the reply to the major comment no: 3.

*Line 70-74: These two sentences read like that so far, “there are very limited efforts to understand the implications of aerosol hygroscopicity on regional air quality”, “in this study we use a regional climate model to assess the contribution of the aerosol hygroscopic growth to the total aerosol optical depth over the IGP”. If the purpose is to fill the gaps on very limited efforts on the effects of aerosol hygroscopicity on air quality, assessing the contribution to total AOD does not help much in that regard.*



The sentence is revised as “there are very limited efforts to understand the implications of aerosol hygroscopicity on regional meteorology, visibility and air quality”

We agree with the reviewer that, in the present study the effect of hygroscopic-growth of aerosols on air quality is (PM<sub>2.5</sub>) not investigated in detail. Increase in PM<sub>2.5</sub> (Figure 9c) for aerosol forcing and meteorological feedback are relatively smaller compared to AOD. So, we have revised the section and please see the answer given for major comment 3 regarding air quality.

*Line 76-76: “we quantify the effects and feedbacks of the aerosol hygroscopic growth on the low air quality conditions over the region”: This part is lacking. No quantification on such effects on air quality is specifically addressed in this paper.*

Sentence revised as “Further, we quantify the effects of the aerosol hygroscopic growth on the regional meteorology, visibility and low air quality conditions over the region.”

We thank the reviewer for pointing out this drawback. In the revised manuscript, we have addressed this issue (refer the reply to major comment 3).

*Line 83-84: 50-km horizontal resolution and 18 vertical levels: This is a very coarse resolution regional model. What is the model top?*

We agree with the reviewer that this domain has a coarse spatial resolution. The model top is at 50 hPa. We have carried out higher resolution runs but they did not add much value to the problem being addressed in the paper. Considering the computation cost and memory requirements we have optimized this configuration.

*Line 89-90: Please spell out the acronyms and provide references of each scheme.*

Complied with.

“The parameterization schemes used in the simulations are (1) boundary layer: University of Washington PBL scheme, (2) Convection over land and ocean: Tiedtke scheme, (3) Radiative transfer: CCM3 (Community Climate Model) scheme, (4) cloud microphysics: Subgrid Explicit Moisture Scheme (SUBEX) and (5) Land surface: Biosphere-Atmosphere Transfer Scheme (BATS) scheme.”

*Line 113-114: The growth factor is 1.37 and 1.49 for hydrophilic BC and OC, respectively. What are the growth factors for sulfate and nitrate?*

Complied with.



“at 80% relative humidity, the particle mass extinction cross-section of sulphate and nitrate increases by a factor of 3 compared to its dry value.”

*Line 120: “climate feedbacks” is used here loosely. For the limited seasonal study like this one, “meteorological feedback” is more appropriate.*

We agree with the reviewer. Complied with.

*Line 121-122: How the clouds are formed in RegCM without aerosol “feedbacks”? Does aerosol provide CCN for cloud formation?*

RegCM4 doesn't have explicit aerosol-cloud interaction parameterization scheme. Clouds are formed in the model depending on the meteorological conditions and the selection of convection parameterization schemes. Actually, this indirect effect of aerosols is currently being implemented in the model.

*Line 142: “RegCM simulates reasonably well: : :” This is a subjective statement. How well is reasonably well? How well is “reasonably well”? Within 20%? 50%? 100%? compared to what? Needs more quantitative description.*

Sorry for these qualitative statements regarding the model validation of aerosol and meteorological parameters. That sentence was intended to mention that several earlier studies validated the meteorological parameters with direct observations over the Indian region (Usha et al., 2020; Ajai et al., 2019; Nair et al., 2012; Giorgi et al., 2012). These authors fine-tuned the model parameterization schemes and carried out a sensitivity analysis in detail.

What could be the reasonable range within which one can accept the model simulations is highly relative. For example, several GCMs simulate BC mass loading over the Indian region by almost 2 to 5 times lower than the observations and some models fail to reproduce the basic monsoon rainfall characteristics (Pan et al., 2015; Wang et al., 2018). However, we have considered the simulation as “reasonably good” if the bias is within the standard deviation of the measurements.

The sentence is revised as “Several earlier studies used RegCM to simulate the regional meteorology of South Asia (Ajay et al., 2019; Giorgi et al., 2012; Nair et al., 2012; Usha et al., 2020).”

Pan, X., Chin, M., Gautam, R., Bian, H., Kim, D., Colarco, P. R., Diehl, T. L., Takemura, T., Pozzoli, L., Tsigaridis, K., Bauer, S., and Bellouin, N.: A multi-model evaluation of aerosols over South Asia: common problems and possible causes, *Atmos. Chem. Phys.*, 15, 5903–5928, doi:10.5194/acp-15-5903-2015, 2015.

Wang, Z., Li, G. & Yang, S. Origin of Indian summer monsoon rainfall biases in CMIP5 multimodel ensemble. *Clim Dyn* 51, 755–768 (2018). <https://doi.org/10.1007/s00382-017-3953-x>

*Line 146-147, Comparisons between RegCM simulated aerosols with measurements: Does the simulation include the feedbacks? Figure 2 shows that the modeled aerosols are in general lower than observations taken in the earlier years. What does this mean considering the fast increase of pollutant concentrations and AOD (see intro section)? Does it mean the model underestimations would be even more severe if the comparisons are done for the same years?*

All the comparisons shown in Figure 2 include aerosol feedback.

As far as the effect of the increasing trend in AOD and aerosol concentrations are concerned, since most of the measurements considered in this study are taken during the last 15 years, the contribution of the long-term trend in the difference between model and measurements are within the standard deviation of the data. For example, the increase in AOD during the last decade ( $0.06 \text{ decade}^{-1}$  reported by Moorthy et al., (2013)) is small compared to the mean AOD over the region (less than 10% of mean AOD over the IGP). Broadly, Sulfate is increasing over the region with a small growth rate, whereas BC at surface level is decreasing trend at  $\sim 240 \text{ ng m}^{-3} \text{ yr}^{-1}$ . The bias due to the asynchronous model data and observations are rather small compared to the uncertainties associated with (i) emission inventories, (ii) boundary layer variation, (iii) hygroscopicity parameterization, and (iv) scavenging processes. However, compared to the earlier studies, where simulated BC mass loading was 2 to 10 times lower than the observed values, the present study simulates values within the standard deviation of the actual observations.

*Line 165: “A good agreement” – again, this is a subjective phrase. With the figure, you should be able to quantify the agreement, e.g., the model calculated RH is 5-20% higher than the observed values. Are these differences good enough for this study?*

Complied with. The sentence is revised as

“The RegCM simulated relative humidity values are approximately 10% higher than the measured RH, with a mean absolute error of 6.6% and RMSE of 7.3%. The good agreement between measured and simulated (with aerosol feedback) relative humidity over the IGP

(Fig. 2e) is important for the estimation of aerosol optical properties from simulated speciated aerosol mass concentrations.”

*Line 171: another “reasonably well”. Fig. 2f shows that the differences between simulated and observed PBLH range from 600m lower and \_200m higher than observations. Is this reasonable?*

In the case of PBL intercomparison, the model bias ranges from -57 m over Bhopal to 352 m over Ranchi with a mean absolute error of 200 m (26% of mean PBLH) and RMSE of 240 m. The mean PBL height at 6 locations over the study region is  $793 \pm 323$  m and the standard deviation of the PBL height observations varied from 100m to 670m over Guwahati and Bhopal respectively. Considering this large day to day and diurnal variations of the PBL height, the simulated PBL heights are reasonably good.

*Line 185-186: “The mean observed (ambient, dry) AOD” – how to observe dry AOD?*

Sorry for that mistake. We cannot “observe” the dry AOD, but we can estimate. The sentence is modified as “The mean observed AODs (simulated ambient and dry AODs) for Jaipur, Kanpur, Pune and Gandhi College are 0.53 (0.51, 0.26), 0.83 (0.81, 0.34), 0.48 (0.32, 0.15) and 1.15 (0.92, 0.36) respectively.”

*Line 186, sites: It would be better to follow the orders of a, b, c, d in Figure 3, i.e., Jaipur, Kanpur, Pune, and Gandhi College.*

Complied with. “The mean observed AODs (simulated ambient and dry AODs) for Jaipur, Kanpur, Pune and Gandhi College are 0.53 (0.51, 0.26), 0.83 (0.81, 0.34), 0.48 (0.32, 0.15) and 1.15 (0.92, 0.36) respectively.”

*Line 193, the statement of Jaipur not showing much hygroscopic growth: The ambient AOD (0.51) doubles the dry AOD (0.26) in Jaipur (see line 186). This is quite a lot growth, certainly is not “not much”.*

We agree with the reviewer. We have modified the sentence. “Observations close to the Thar Desert (Fig. 1a, Jaipur) show relatively lower hygroscopic growth (43%) compared to other IGP stations because of the dominance of dust aerosols and relatively low humidity conditions prevailing there.”

*Line 203: “matches well” – how well is well? Please give some quantitative measures, including correlation coef., bias, RMSE, for example.*

Complied with. The mean absolute error of 0.12 (~17% of mean AOD) and RMSE of 0.15.

Line 206-207: 30%. “since the emission fluxes of anthropogenic aerosols are seasonally invariant”: this is from the model or the actual facts in the real world?

Since we used monthly emission data, the model emission fluxes are constant for the entire month. Though may not be the case in the real condition, we don’t have high resolution (temporal and spatial) emission data over the region. The sentence is modified as “All the meteorological conditions, associated processes (winds, chemistry, deposition, and transport) and anthropogenic emissions remained the same for dry and ambient AOD simulations, except the hygroscopic growth of AOD with relative humidity for the ambient AOD.”

Line 207-208: “most of the observed variability of AOD is due to the variability in relative humidity”: This is unjustified to attribute the variability to the RH, because you have not assessed the AOD variability due to the changes of winds, chemistry, deposition, and transport.

Figure 3 clearly shows that ambient AOD has more variability (standard deviation or the coefficient of variation) than dry AOD. All the meteorological conditions, associated processes (winds, chemistry, deposition, and transport) and anthropogenic emissions remained the same for dry and ambient AOD simulations except the hygroscopic growth of AOD with RH for the latter case. Hence it is clear that relative humidity contributes to the relatively high day-to-day variability of AOD over the region.

Line 213: What is the  $AOD_{dry}/AOD_{ambient}$  ratio over W India? The contour lines started at 0.3 and ends at 0.6. I suggest show the full range of the ratio with the contour lines.

Dry AOD contributes more than 50 to 70% of total AOD over W India. We have modified the figure and shown below.

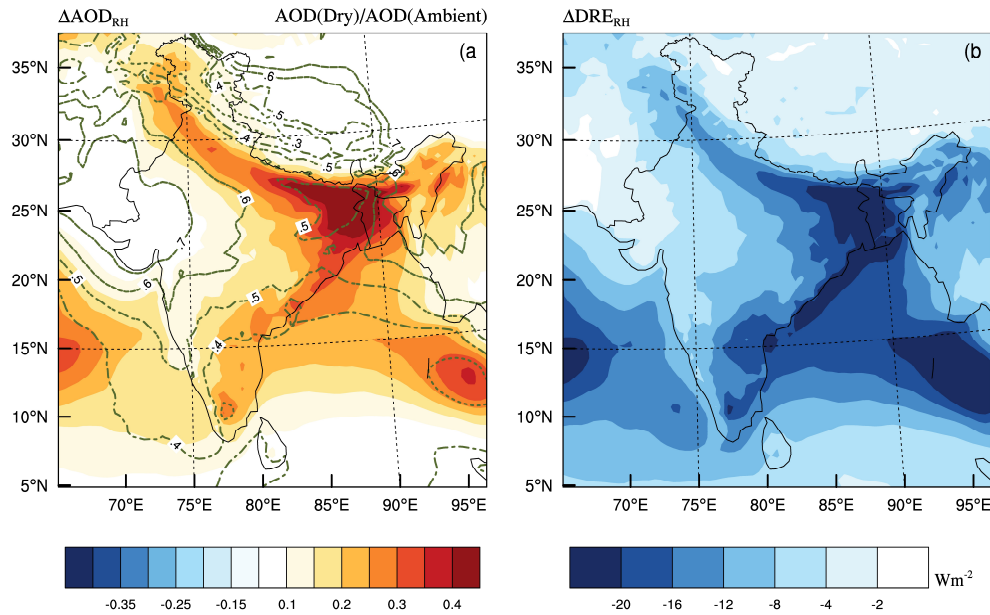


Figure 4: (a) Aerosol optical depth (AOD) simulated using RegCM4 for dry and ambient humidity conditions. Colour map indicates  $AOD(ambient) - AOD(dry)$  for without meteorological feedback and line contours show  $AOD(dry)/AOD(ambient)$  (b) Difference in aerosol direct radiative effects (DRE) at the surface for dry and ambient humidity conditions (without meteorological feedbacks).

Line 210-228: This paragraph is somehow confusing – it mixes the results from this study with findings from other studies, and it is difficult to unravel the message. It should state what are the findings from this study and how they compared with previous studies.

We are really sorry for this confusion. We have revised that paragraph as

“The AOD due to hygroscopic growth ( $\Delta AOD_{RH}$ ) estimated using Eqn. 1 is shown in Fig. 4a. The  $\Delta AOD_{RH}$  values are high over IGP, especially the eastern IGP and Bangladesh ( $\Delta AOD_{RH} > 0.3$ ). The ratio of  $AOD(Dry_{nofeedback})$  to  $AOD(Ambient_{nofeedback})$  shown as contours in Figure 4a indicates that dry aerosols contribute 50 to 60% of the AOD over most of the Indian sub-continent (Fig. 4a), except over western India, where the contribution of hydrophobic dust to the total AOD is high even during winter. The contribution of the aerosol hygroscopic growth is more than 50% of the total AOD over the northern Indian Ocean, which is attributed to the dominance of hydrophilic aerosols (sea salt) and high relative humidity. It is interesting to note that the increase in AOD solely due to hygroscopic growth ( $\Delta AOD_{RH} = AOD(ambient) - AOD(dry)$ ) is  $\sim 0.23$  over the IGP (Fig. 4a), which is higher than the global mean AOD ( $0.155 \pm 0.018$ ) reported by Watson-Parris et al., (2020) using various satellite observations. Mandariya et al., (2020) have reported

that wintertime aerosols over central IGP show an increase in particle diameter by  $1.34 \pm 0.07$  times as that of the dry particle at 85% relative humidity. This increase in particle diameter corresponds to a doubling of the scattering coefficient for anthropogenic aerosols (McInnes et al., 1998). In the present study the mean dry AOD increases almost 1.7 times due to relative humidity over IGP, which is slightly lower than the estimated AOD for the hygroscopic factor reported by Mandariya et al., (2020). Since organic aerosols dominate the aerosol system over IGP (Ram et al., 2012), there could be a substantial decrease in the hygroscopic growth of particles as discussed by several studies. Mandariya et al., (2020) have reported that 150 nm particles are more hygroscopic than 100 nm particles due to the higher organic fraction present in the 100 nm particle mass concentrations. In addition, although most of the observational studies consider 40% as the reference humidity level (dry) for estimating the hygroscopic growth, the model estimates the dry AOD for zero relative humidity. Titos et al., (2016) reported that the assumption of no growth at 40% humidity may contribute higher uncertainty in the estimates of measurement-based hygroscopic growth.”

*Line 245: How much is the cloud cover change? This should be included in Figure 5.*

Complied with. Figure 5 is revised as shown below. “The cloud cover increases by 3 to 5 % over IGP due to ambient aerosol forcing (Figure 5f).”



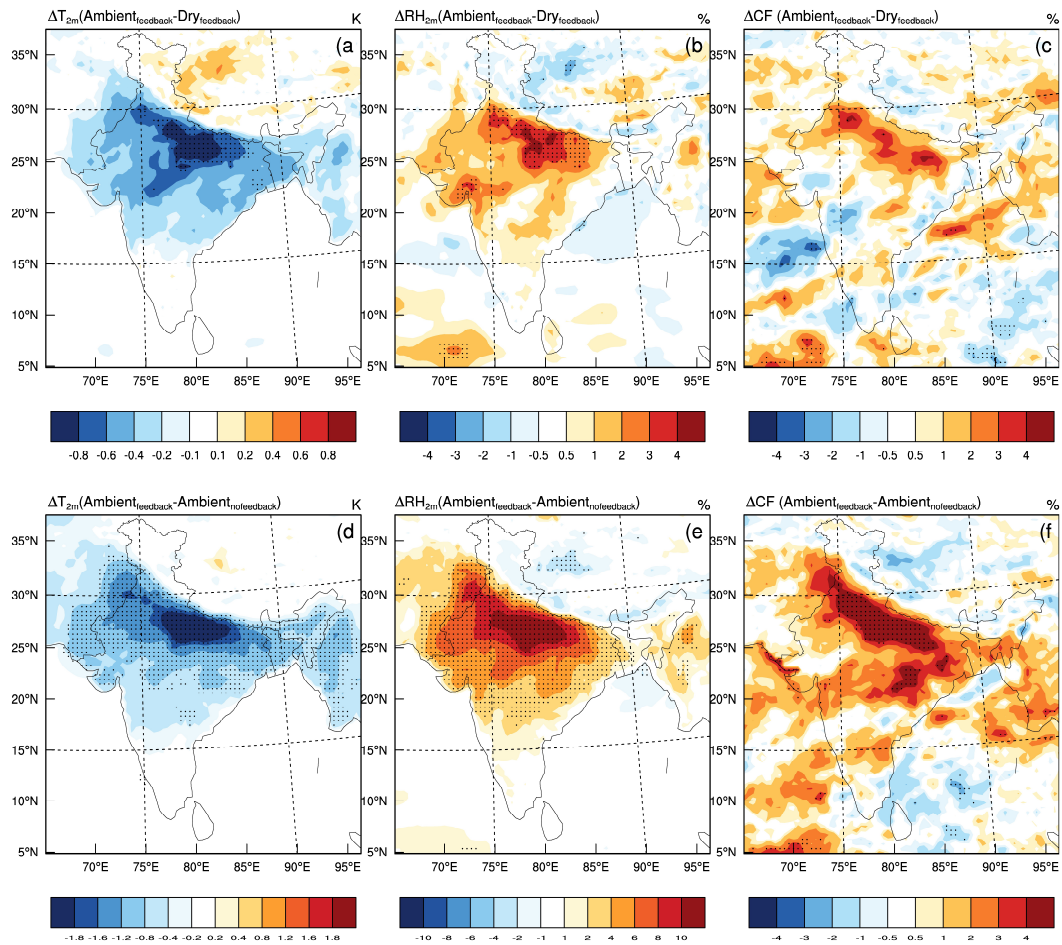


Figure 5: Change in (a) 2m air temperature, (b) relative humidity and (c) cloud fraction solely due to the hygroscopic growth of aerosols (ambient-dry). Change in temperature and relative humidity (RH) due to the total effect of aerosols (dry + hygroscopic growth) is shown (c), (d) and (f). Dots indicate statistical significance of Student's t-test at a 90% level.

Line 249: RH increases by 2-4% in eastern India. Is this insignificant? The student t-test shows that it is statistically significant. Please clarify what “significant” means.

We have mentioned inline 249 that “Although AOD and  $\Delta AOD_{RH}$  are high, the change in relative humidity due to hygroscopic growth of aerosols is negligible over Bangladesh and Eastern IGP.” This inference is made based on Figure 5b, which shows <1% change in RH and it is statistically insignificant as shown by Student t-test. However, for the total aerosol effect (Figure 5d) there is a 2-4% increase in RH and it is statistically significant also (as mentioned by the reviewer). We assume that the reviewer considered Figure 5d (new figure 5e) instead of figure 5b. To avoid this ambiguity, we have revised the sentence as

“Although AOD and  $\Delta\text{AOD}_{\text{RH}}$  are high, the change in relative humidity due to hygroscopic growth of aerosols (Figure 5b) is negligible over Bangladesh and Eastern IGP.”

*Line 249-251, “Hence: :” This statement is not evident from the paper. So far the figures and text only show the effects of aerosol hygroscopicity on RH and T, but not the pathways from land-atmosphere interactions.*

The aerosol induced decrease in surface-reaching solar radiation and its influence on sensible and latent fluxes (Bowen ratio) is already discussed in Bharali et al., (2019), hence not repeated here. Our results also showed a similar change in heat fluxes over IGP during winter. We have modified the sentence as “Bharali et al., (2019) have reported that the strength of land-atmosphere interactions through the exchange of heat and moisture fluxes plays a major role in deciding the aerosol induced dimming effects on meteorology (more details are available in Li et al., 2017)”

*Line 253-254, “In other words: :” But why the effects in the eastern India and Bangladesh is much weaker than in the middle IGP where the dimming effect is the strongest (Fig 4)? Need some better explanation.*

We thank the reviewer for this very important point which we missed in the initial analysis. It is interesting to note that aerosol loading and clear-sky radiative forcing showed very high values over the eastern IGP. In contrast, the change in surface temperature and relative humidity due to aerosol induced surface dimming was high over central IGP with a moderate change over eastern IGP. To further understand this, the change in solar radiation reaching at the surface for with and without aerosol conditions is shown in the figure below. Though the AOD and aerosol direct radiative forcing are high values over the eastern IGP, the presence of relatively high cloud fraction over the eastern IGP mask the aerosol-induced surface cooling. So, the change in surface temperature did not show a similar pattern as that of AOD. Whereas the change in shortwave flux at the surface (due to clouds and aerosols together) showed an almost similar pattern as that of change in temperature and humidity.

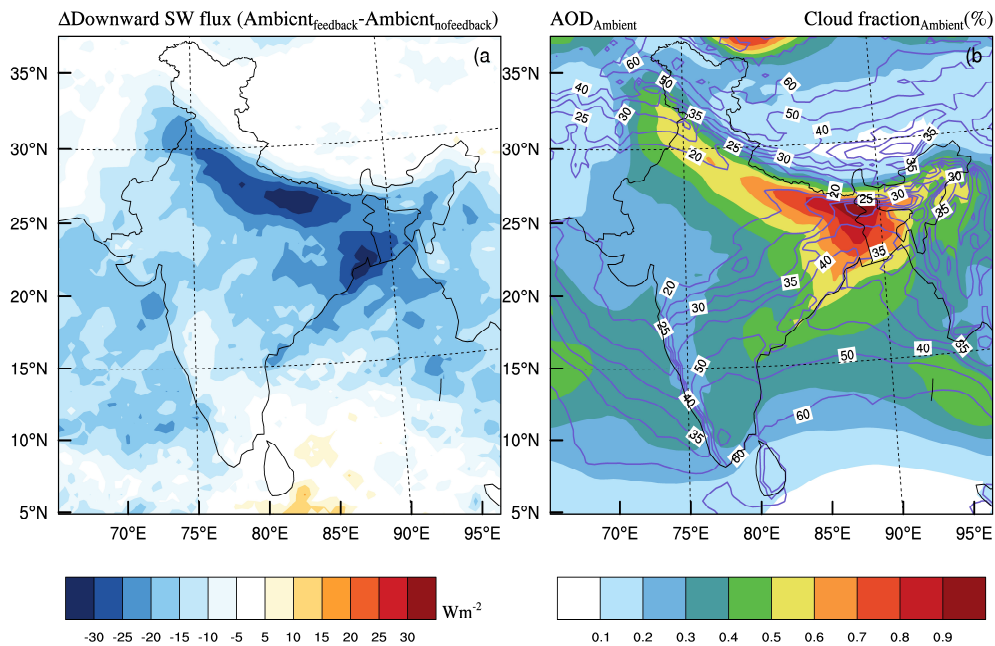


Figure S4: (a) Spatial variation of change in surface-reaching solar radiation for with and without aerosol conditions. (b) variation of AOD (color) and cloud fraction (line contour) for aerosol feedback simulation.

Line 261, “along with the weak entrainment of dry airmass from the free troposphere”: Where is the evidence? Does RegCM show the weakening of the entrainment? What is the cause of that?

The figure below shows the change in the model vertical wind due to aerosol effects. The large-scale entrainment decreases above 2 km due to aerosol feedback. The convection also weakens due to the decreased surface temperature following the solar dimming which reduces the energy required for the upward motion the air parcel.

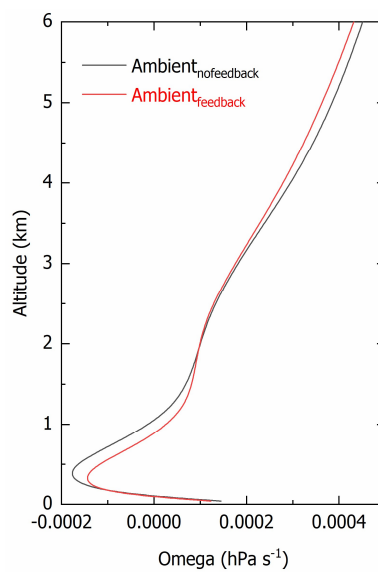
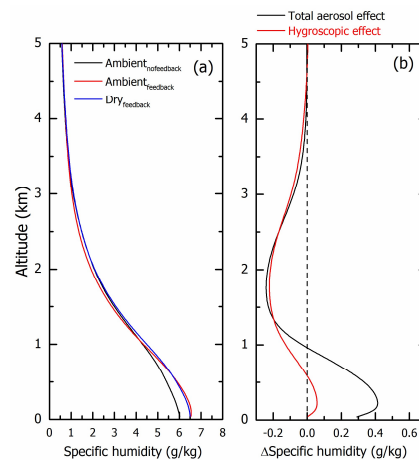


Figure S5: Vertical profiles of change in specific humidity due to the total aerosol effect ( $Ambient_{feedback} - Ambient_{nofeedback}$ ) (black line) and only due to hygroscopic growth of aerosols ( $Ambient_{feedback} - dry_{feedback}$ ) (red line).

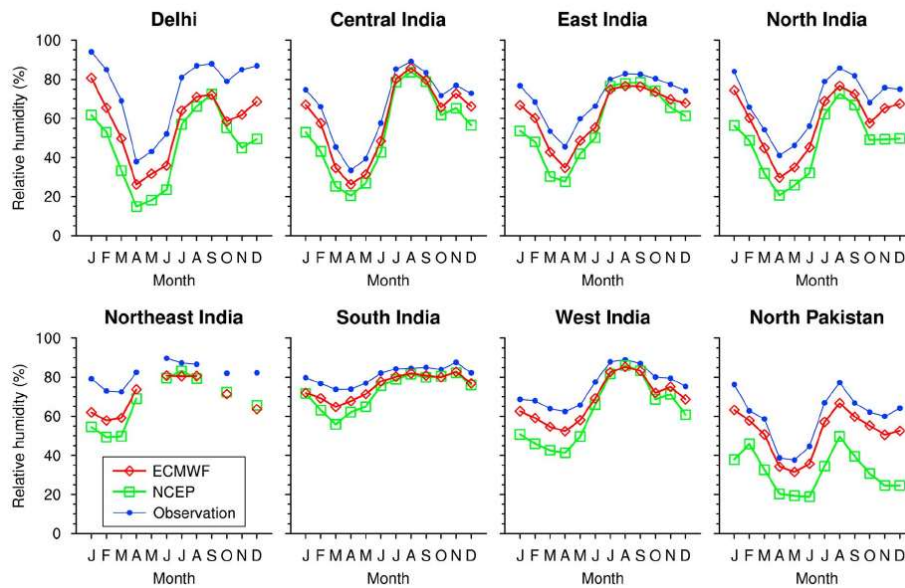
Line 261-262, “leads to an increase in relative and specific humidity at the surface”: The paper only shows an increase of RH, not specific humidity.

Sorry for taking it granted. We have included this figure in the new supplementary file (Figure S5).



Line 267-268, “Most reanalysis datasets have a negative bias in simulating RH during winter”: The “reanalysis” datasets are based on the observation that should include all effects, unless the observations in South Asia are not input into the reanalysis and the RH is solely from the underline model simulations. Please explain.

As we mentioned in the manuscript, Chatani and Sharma, (2018) have clearly shown that NCEP and ECMWF underestimate relative humidity over the IGP. Figure 3 of Chatani and Sharma, (2018) is shown below for the reviewer’s reference. It indicates that RH measurements from this region are not being accepted by the stringent data assimilation criteria.



The figure is taken from Chatani and Sharma, (2018).

Line 272, “drying in the 1-3 km”: Fig 6 shows the RH increased by 3% at 1 km, not decreased, due to total aerosol effect.

We agree. Revised as “drying in the 1.5 to 3.5 km”

Line 272-273, “warming of the top of the boundary layer”: Where is the top of PBL? Fig 6 shows a cooling, not warming, from surface to almost 2 km due to total aerosol effect.

Revised the sentence.

“In contrast to boundary layer moistening, drying in the lower free troposphere (1.5 to 3.5 km) by 1 to 2% was attributed to the aerosol induced warming, which decreases the humidity and suppresses the mixing of air mass between the free troposphere and boundary layer.”

Line 287: “surface temperature increases”: But you have been showing the temperature decreases, not increases. This is so confusing.

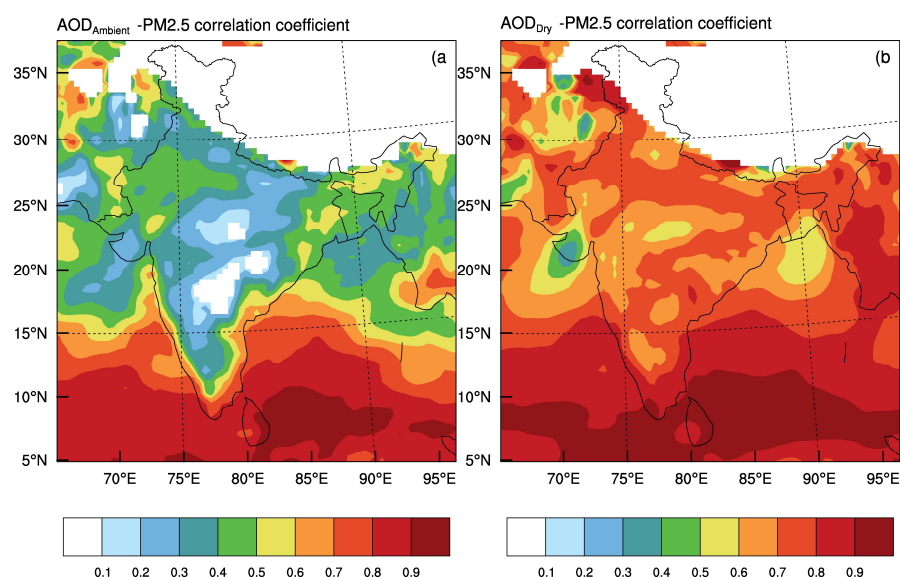
Sorry for that mistake. Corrected.

Line 287-288, increase in water vapour content: :reported by Mukhopadhyay (2017)”: Does that study use the same model with the same settings for wintertime simulation? What is your finding in this study?

Mukhopadhyay et al., (2017) used in-situ measurements of humidity over the Indian region from a network of observatories spread across the country to study the trend in relative and specific humidity for all the seasons. Our model simulations suggest that the aerosol-radiation interaction and associated meteorological feedback could be partially contributing to the wetting of boundary layer (a positive trend in RH) and drying of the lower free troposphere (a negative trend in RH) reported by the Mukhopadhyay et al., (2017).

Line 346-347, “present study also showed poor association with ambient AOD and PM2.5 but higher correlation is observed between the dry AOD and PM2.5”: Where are the associations/correlations are shown in this paper?

Sorry for that omission. Figure below shows the correlation between the wet and dry AOD with PM2.5. Please note that PM2.5 is not sensitive to humidity change or PM2.5 is generally estimated as dry mass loading of particle below 2.5  $\mu\text{m}$  size. So, it is expected that dry AOD has a better association with PM2.5 than ambient AOD. This result assumes importance, for the estimation of PM2.5 from the satellite observations or to find an empirical relation to covert AOD to PM2.5 for air quality applications. It is clear from this figure that high relative humidity deteriorates the association between AOD and PM2.5, which is a very significant contributor over the IGP. The accurate assessment of hygroscopic growth factor is essential to retrieve PM2.5 over the IGP using satellite sensors.



Line 350, “the mass concentration is dominated by the contribution from the coarse mode aerosols”: But PM2.5 is not coarse mode aerosols!



We mentioned there that, generally optical properties of aerosols are more sensitive to the fine mode aerosols, whereas mass concentration is more contributed by coarse mode aerosols. We have removed that general sentence from the manuscript.

*Line 306-375, “section 3.3 Implications on air quality”: This entire section is very weak. There are no quantifications on the haze condition and/or PM<sub>2.5</sub> concentration change due to the aerosol hygroscopic growth and meteorological feedbacks. Maps of change of air quality (haze and PM<sub>2.5</sub>), similar to Figure 5 for T and RH, are needed to illustrate the implications.*

We agree with the reviewer. This aspect is discussed in the reply to major comment 3.

*Line 383: The increase in PM<sub>2.5</sub> along the IGP has not been unequivocally demonstrated.*

We have removed “and PM<sub>2.5</sub>” from that sentence.

## Interactive comment on “Amplification of South Asian haze by water vapour-aerosol interactions” by Vijayakumar S. Nair et al.

### Referee #1

*This paper assesses the contribution of the hygroscopic growth of aerosols to the total AOD and demonstrate that the increased surface cooling due to the hygroscopic effects of aerosols further increases the humidity in the boundary layer and thus enhances the confinement of pollutants through aerosol-boundary layer interactions. This study is a timely contribution to our growing understanding of the chemistry-weather interactions. The paper is well written and easy to follow. However, the paper lacks a comprehensive evaluation exercise necessary to allow the readers lend confidence in the scientific findings presented here. My major and minor concerns are listed below.*

**We thank the reviewer for the encouraging comments.**

*Major comment: Specifically, a qualitative model evaluation is not enough considering the importance of this topic. While I understand that chemical composition measurements are limited over India but the lead author is a part of the Aerosol Radiative Forcing over India (ARFI) project which has done an excellent job in collecting hourly AOD and black carbon at more than 30 sites in India since early 2000s. I will suggest comparing model AOD and BC from the “Dry” and “Ambient” experiments with daily (hourly if possible) measurements of AOD and BC performed at the ARFI sites (similar to Fig. 3 for the AERONET sites). This evaluation will highlight where and when the hygroscopic properties play the most important role in the Indian context.*

**We are sorry for being qualitative in model validation. We have included the mean absolute bias and RMSE of the modelled parameters compared to direct observation. As suggested by the reviewer, RegCM4 simulated AOD and BC are compared with the ARFINET measurements from December 2016 to February 2016. Modelled ambient AOD values are close to the observed values over all the station as shown in the below Figure. The measurement-based estimation of dry AOD is still challenging over this region because of the lack of information on the vertically resolved chemical composition and/or hygroscopicity of aerosols. BC mass concentration over Agra, Jaisalmer, Udaipur and Hyderabad also showed good association with observed values. This comparison clearly shows that the model is able to simulate the aerosol loading over the Indian region. As reviewer correctly pointed out, a synergy of more dedicated experiments and modelling is essential to delineate the various pathways of aerosol-climate interaction over the region.**

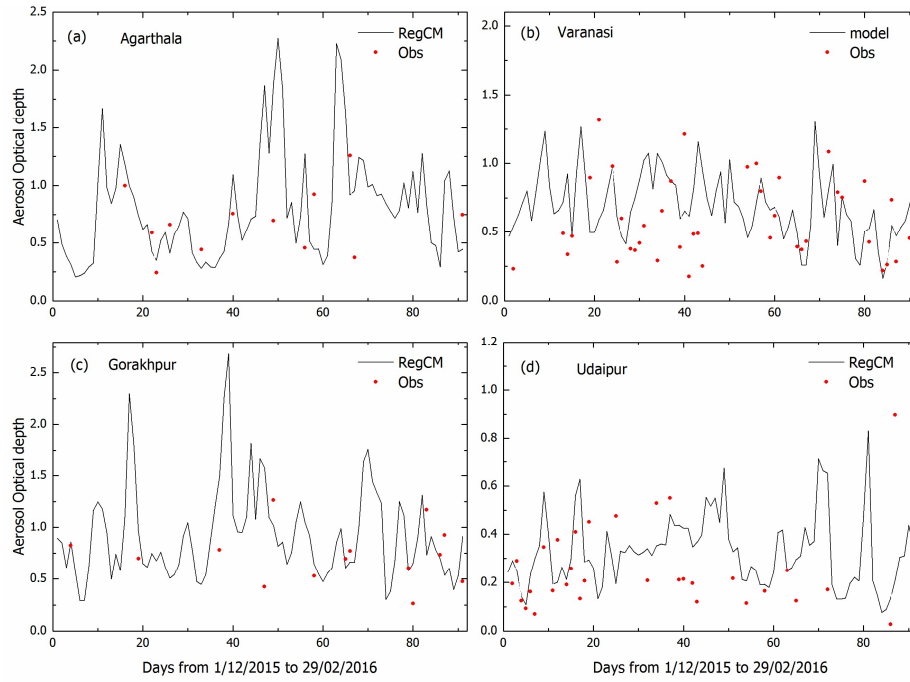


Figure S2: Variation of aerosol optical depth measured using multi-wavelength radiometer installed as a part of Aerosol Radiative Forcing over India Project (ARFINET) and modeled using RegCM4 from December 2015 to February 2016.

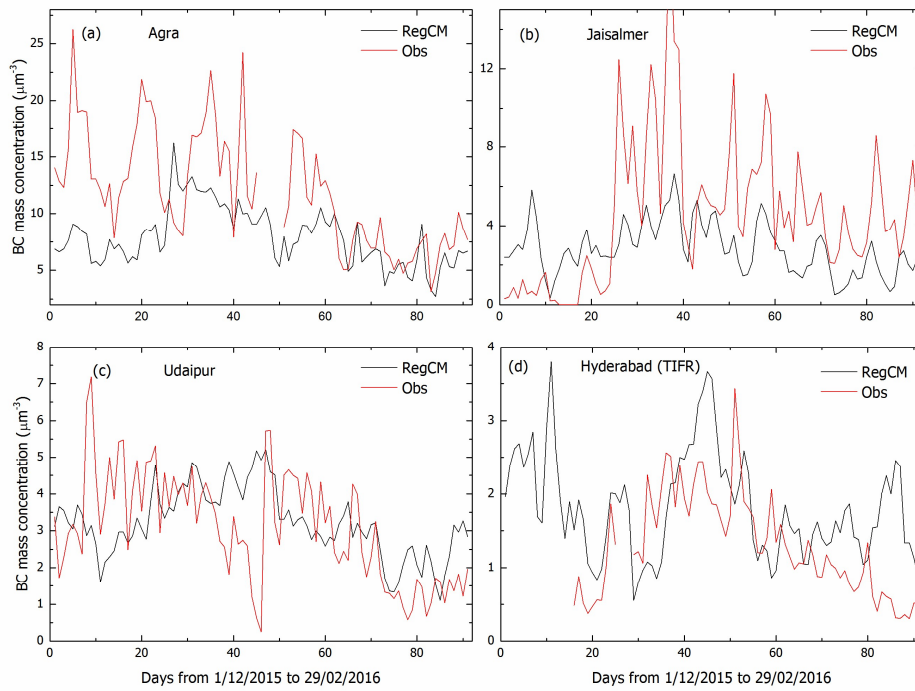


Figure S3: Temporal variation of BC mass loading measured at 4 ARFINET stations and modeled using RegCM4 over Agra, Jaisalmer, Udaipur and Hyderabad from December 2015 to February 2016.

*In addition, the authors should also evaluate the model performance against available CPCB PM2.5 measurements because understanding the processes leading to poor air quality episodes is of utmost importance in this part of the world as also stated in the Introduction of this paper. I think a detailed PM2.5 evaluation during three seasons would provide a robust insight into the implications of aerosol hygroscopicity for air quality.*

As reviewer suggested, PM2.5 simulated using RegCM is validated using the CPCB measurements made at 19 urban centres over the Indian region. In general, the model underestimates PM2.5 values, especially at Delhi, Muzaffarpur, Gaya, Thiruvananthapuram, Lucknow, Varanasi, Agra and Guwahati. Since CPCB measurements are carried out at the urban hotspots and close to the city pollution, models simulations at coarse spatial resolution (50 km) may not simulate the magnitude of city pollution accurately.

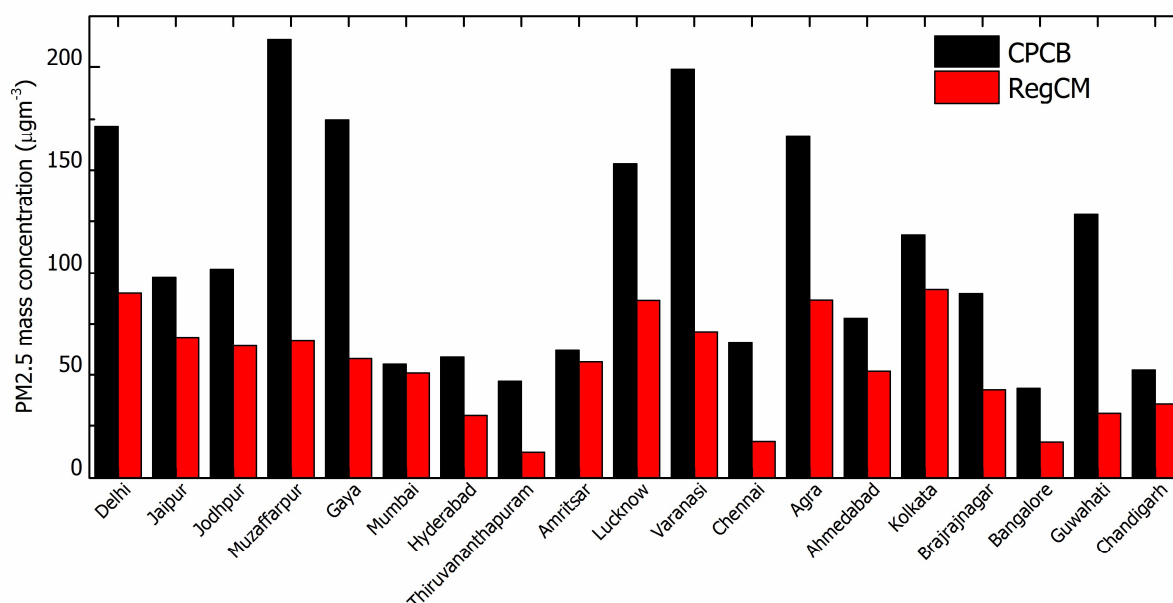


Figure S1: Inter-comparison of PM2.5 simulated using RegCM4 with measurements from 19 stations maintained by Central Pollution Control Board (CPCB) India ([www.cpcb.nic.in](http://www.cpcb.nic.in)).

*Furthermore, diurnal profiles of surface RH and temperature should also be evaluated. It is particularly important to understand if the model is able to capture nighttime increase in RH which is one of the key factors in haze and fog formation.*

As suggested by the reviewer the diurnal variation of the temperature and relative humidity over the 4 stations in northern India is given below. As shown in the figure, the modelled RH shows a positive bias and temperature shows a negative bias over all the stations (Agra, Delhi, Patna and Lucknow). Though there exists a positive bias in the

modelled RH, RegCM4 could capture the diurnal variation with daytime low and nighttime high over the region. As Reviewer correctly pointed out the early morning high in the RH is one of the key factors for the haze and fog conditions prevailing over the IGP.

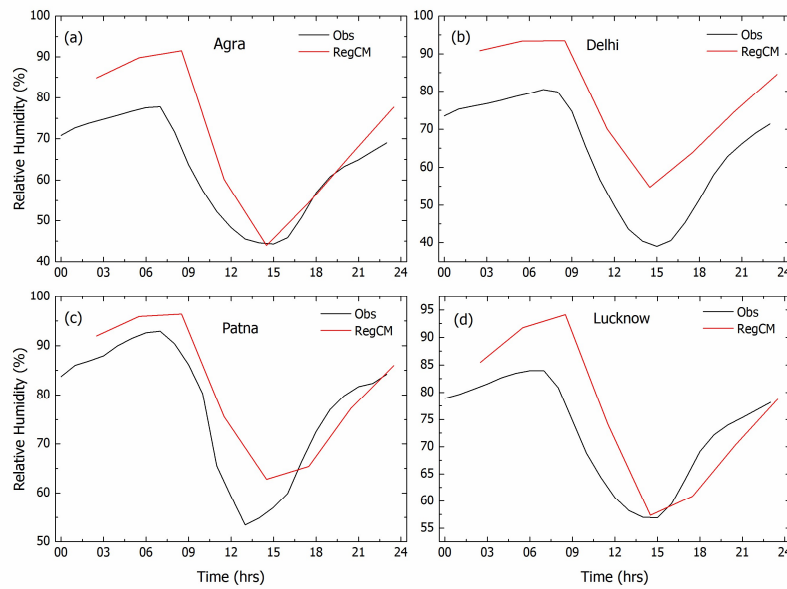


Figure: Diurnal variation of measured (black) and modelled (red) relative humidity at (a) Agra, (b) Delhi, (c) Patna and (d) Lucknow.

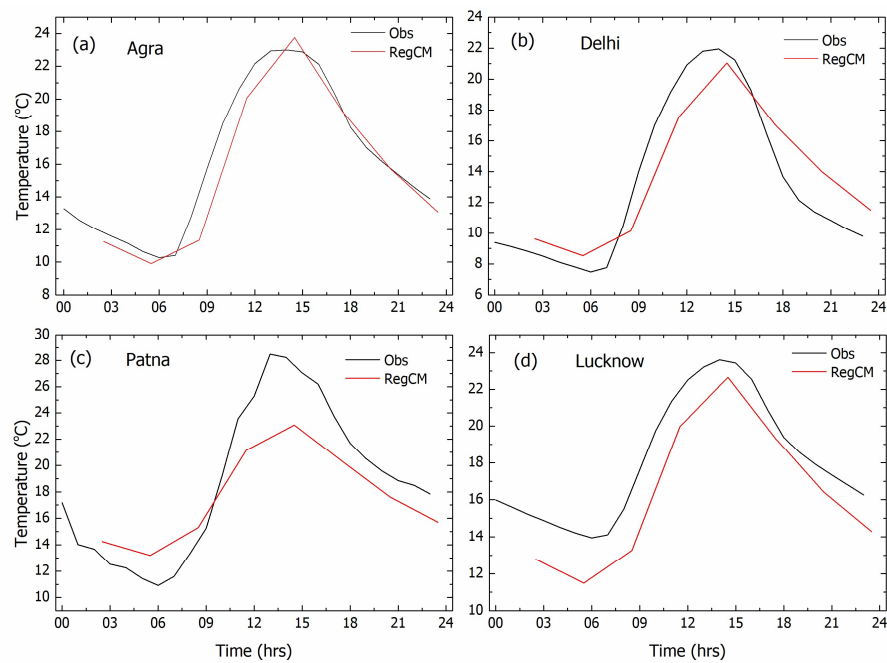


Figure: Diurnal variation of measured (black) and modelled (red) 2m temperature at (a) Agra, (b) Delhi, (c) Patna and (d) Lucknow.

*Therefore, I recommend a major revision before the paper can be accepted for publication in ACP.*

**We thank the reviewer for the encouraging comments.**

Minor comments:

*Line 11: Change low to poor.*

**Complied with.**

*Line 35: Change boundary layers to boundary layer.*

**Complied with.**

*Line 66: Does all aerosol optical properties (AOD, SSA, and asymmetry parameter) increase by a factor of 2 at RH > 80%?*

**Sorry for that mistake. AOD shows a 2-fold increase, whereas SSA and asymmetry parameters show relatively weak dependence on RH.**

**Text revised as “The optical properties of aerosol (AOD/extinction coefficients) is enhanced by more than 2 times at higher relative humidity (>80%) conditions, which has a strong dependence on the relative dominance of organic and inorganic species (chemical composition) and size of the particle.”**

*Line 84: How many levels do you have in the boundary layer and are these sufficient to resolve the PBL processes?*

**RegCM4 has 5 levels below 1km and model cannot resolve the PBL processes explicitly.**

**University of Washington (UW) PBL scheme is used in RegCM4 for the representation of the PBL processes. The layer below which the buoyancy flux cannot be more negative than -0.5 of the layer-mean buoyancy fluxes is estimated as the PBL top height. Which means that the vertical profile of the virtual temperature in the lower atmosphere stabilizes in such a way that at the boundary layer top the buoyancy flux is opposite and half of the layer mean buoyancy flux (Elguindi et al., 2014).**

*Line 100-104: Does the model also include secondary organic aerosols?*

**RegCM4 doesn't have an explicit SOA scheme, rather OC is multiplied with 1.25 to account for secondary organic carbon.**

*Line 190: Are not organic carbon aerosol also emitted as hydrophobic?*



The OC is emitted as hydrophobic and then it transformed to hydrophilic at an ageing time of 1.15 days.

*Line 194-195: If aerosol hygroscopic growth is not a major factor at Jaipur, what is the reason behind large differences between Ambient and Dry AOD at Jaipur in Figure*

Sorry for the ambiguity. We did not rule out the RH effect over Jaipur, where hygroscopic growth was relatively smaller compared to the other IGP locations. Aerosols over Jaipur are not as hygroscopic as the central and eastern IGP aerosols.

We have modified the sentence. "Observations close to the Thar Desert (Fig. 1a, Jaipur) depict relatively lower hygroscopic growth (43%) compared to other IGP stations because of the dominance of dust aerosols and relatively low humidity conditions prevailing there."

*Line 209: I am not convinced that most of the AOD variability can be simply attributed to variability in RH. Since Ambient AOD is higher, surface temperature will be lower in Ambient experiment compared to the Dry. Consequently, PBL will be lower and lead to accumulation of aerosols in the PBL. The winds may also respond to aerosol induced thermodynamic changes and lead to different emissions of dust aerosols. All these aspects should be discussed.*

Sorry for this ambiguity.

We have considered ambient and dry simulations without climate feedback (exp 1 & 2) to estimate the coefficient of variation of  $AOD_{dry}$  and  $AOD_{ambient}$ . Since the climate feedback is switched off, meteorology (PBL, wind, precipitation and RH) is invariant or not affected by the aerosol forcing. All the meteorological conditions, associated processes (winds, chemistry, deposition, and transport) and anthropogenic emissions remained the same for dry and ambient AOD simulations except the hygroscopic growth of AOD with RH for the latter case. Hence it is clear that relative humidity contributes to the relatively high day-to-day variability of AOD over the region.

*Lines 249-255: It is really interesting to note that changes in temperature and RH are not located in the same place as the changes in AOD. Could you show the distribution of solar radiation reaching at the surface as well to corroborate your explanation because the largest changes in solar radiation reaching at the surface should coincide with the largest changes in AOD.*

Complied with. It is interesting to that aerosol loading and clear-sky radiative forcing showed very high values over the eastern IGP. In contrast, the change in surface temperature and relative humidity due to aerosol induced surface dimming was high over central IGP with a moderate change over eastern IGP. To further understand this, the

change in solar radiation reaching at the surface for with and without aerosol conditions is shown in the figure below. Though the AOD and aerosol direct radiative forcing (clear sky) are high values over the eastern IGP, the presence of relatively high cloud fraction over the eastern IGP mask the aerosol-induced surface cooling. So, the change in surface temperature did not show a similar pattern as that of AOD. Whereas the change in shortwave flux at the surface (due to clouds and aerosols together) showed an almost similar pattern as that of change in temperature and humidity.

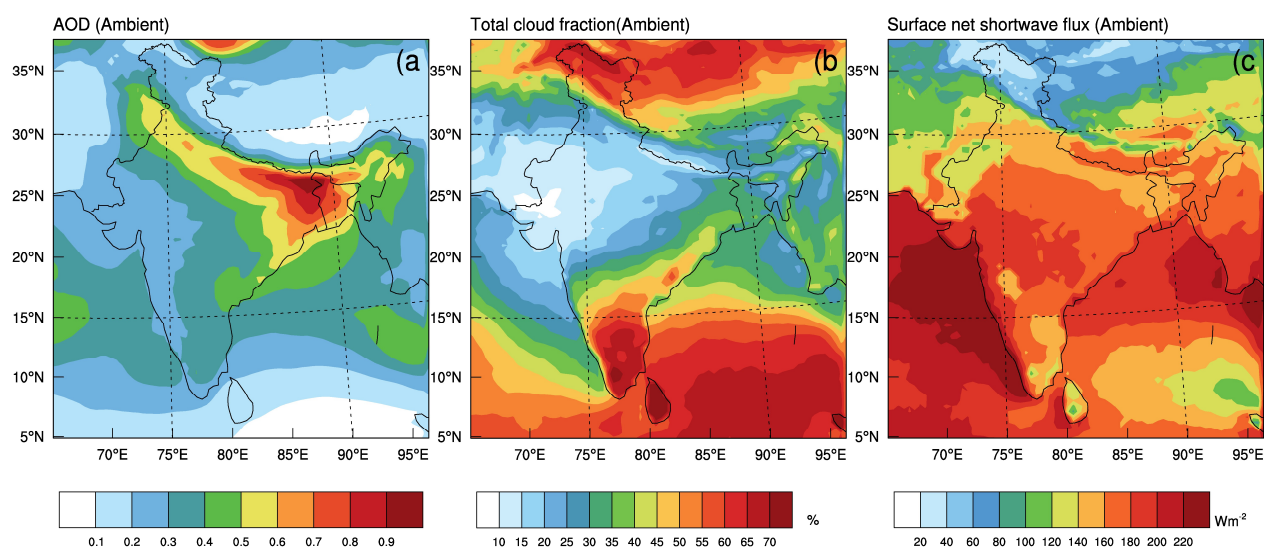


Figure: The spatial variation of (a) AOD, (b) cloud fraction and (c) net shortwave flux at surface.

Line 314: What is the difference between radiative and climatic feedback?

Replaced “radiative and climatic feedback” with “meteorological feedback”

Fig 8b: I think it is delta AOD-RH relationship. Please add delta symbol to the figure title. Why is deltaAOD-RH correlation highest over the Thar Desert?

We thank the reviewer for pointing out this mistake. Figure caption is corrected now.

Line 338-339: I think PM<sub>2.5</sub> response will depend largely on how the PBL height changes and how aqueous-phase production of SO<sub>4</sub> will change. Can you add some discussion on these points?

As reviewer correctly pointed out, the variation in PM<sub>2.5</sub> is mostly associated with the change in PBL height change due to aerosol-induced surface cooling and other meteorological feedback. We have included the variation of carbonaceous, organics and PM<sub>2.5</sub> aerosol mass concentration due to aerosol forcing in the below figure. The aerosol

concentration increases along the IGP due to aerosols due to aerosol induced weak ventilation over the region. This further confirms that the air quality problems of IGP have not only associated with aerosol source strength but also to its (aerosol) radiative forcing and meteorological feedback.

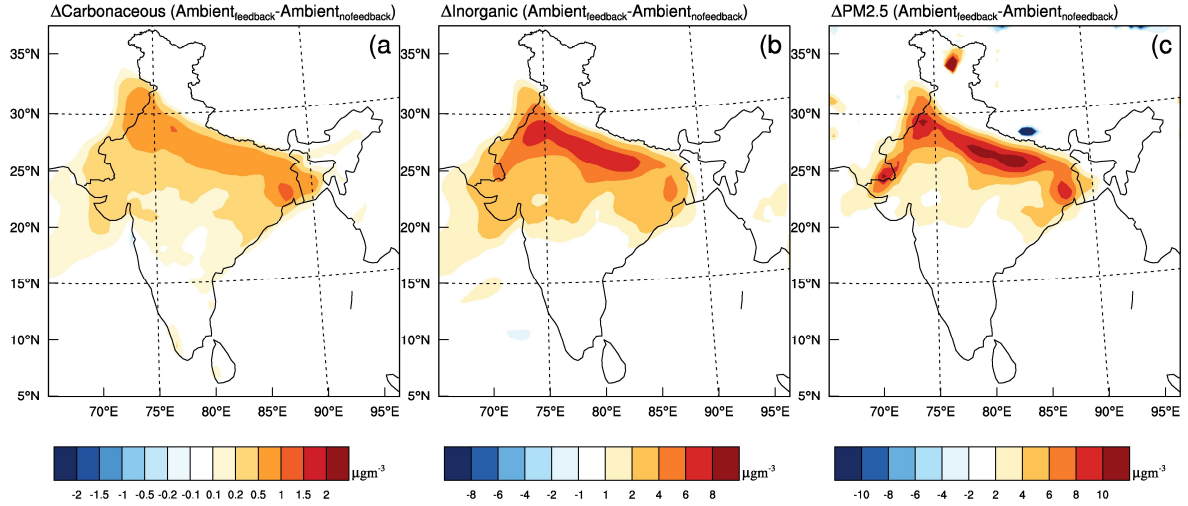


Figure: Change in near surface mass loading ( $\mu\text{g m}^{-3}$ ) due to the meteorological feedback of total aerosol radiative forcing for (a) carbonaceous aerosols, (b) inorganic aerosols and (c) PM2.5.

# Amplification of South Asian haze by water vapour-aerosol interactions

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**Abstract.** Air pollution and wintertime fog over South Asia is a major concern due to its significant implications on air quality, visibility and health. Using a coupled regional climate model with on-line chemistry, we assess the contribution of the hygroscopic growth of aerosols (wet-dry) to the total aerosol optical depth and demonstrate that the increased surface cooling due to the hygroscopic effects of aerosols further increases the humidity in the boundary layer and thus enhances the confinement of pollutants through aerosol-boundary layer interactions. This positive feedback mechanism plays an important role in the prevalence of wintertime fog and **poor** air quality conditions over South Asia, where water vapor contributes more than half of the aerosol optical depth. The aerosol-boundary layer interactions lead to **moistening** of the boundary layer and drying of the free-troposphere, which amplifies the long-term trend in relative humidity over the Indo-Gangetic Plain during winter. Hence, the aerosol-water vapor interaction plays a decisive role in the formation and maintenance of the wintertime thick fog conditions over South Asia, which needs to be considered for planning mitigation strategies.

## 1 Introduction

South Asia experiences regularly severe air pollution events during the winter season. Widespread haze and fog over the northern parts of the Indian sub-continent, especially the Indo Gangetic Plain (IGP), are associated with anthropogenic activities and are noticeable even from space during the winter season (Ali et al., 2019; Gautam and Singh, 2018; Ghude et al., 2017). Poor air quality and visibility persisting throughout the winter period has been a major concern for more than 900 million people living in the IGP (Gautam and Singh, 2018; Gurjar et al., 2008; Lelieveld et al., 2015). Indeed, various studies have shown that the winter concentrations of fine particles (PM<sub>2.5</sub>) and gaseous pollutants exceed national/international air quality standards over most of the IGP region and are considered extremely hazardous for human health (Ali et al., 2019; Bharali et al., 2019; Ghude et al., 2017; Gurjar et al., 2008; Kumar et al., 2015a; Nair et al., 2007; Rengarajan et al., 2011; Safai et al., 2008). Recent estimates of premature mortality related to air pollution over India is ~0.65 million per year, mostly found to occur in the IGP due to residential emissions (Lelieveld et al., 2015). Traffic disruptions and air quality alerts are quite frequent during the winter season over megacities such as Delhi, Kolkata, Karachi, Lahore and Dhaka (Ghude et al., 2017; Gurjar et al., 2008). During the last few decades, the intensity of fog and haze events

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have increased over the region by a factor of three (Ghude et al., 2017; Syed et al., 2012), which is in-line with the observed increase in aerosol optical loading (2.3% per year from 1985 onwards) and surface dimming over South Asia reported from a network of radiometer observations (Babu et al., 2013).

The sources of primary emission of anthropogenic aerosols include residential, transport, industrial and agricultural sectors (Kumar et al., 2015b). The extensive burning of stubble in agricultural fields has been reported to be a major contributor to the formation of haze at the beginning of winter. Mineral dust transport from west Asia and Thar deserts also deteriorates air quality over the region. In addition to the emission sources, low temperatures, shallow boundary layer and low wind speed conditions hinder the vertical and horizontal mixing (ventilation) of aerosols, which favours the accumulation of pollutants within the boundary layer (Bharali et al., 2019; Nair et al., 2007). Even though the aerosol loading within the boundary layer is very high over the IGP, measurements onboard aircraft show relatively clean free troposphere conditions. The vertical extent of this high aerosol loading is thus limited and mostly confined to the first 2 km near the surface with an exponential decrease above the boundary layer (Babu et al., 2016). Mass concentrations of fine particles (size < 2.5  $\mu\text{m}$ , PM<sub>2.5</sub>) near the surface measured by the Central Pollution Control Board (CPCB) clearly indicate the poor air quality conditions prevailing over the IGP. The seasonal mean PM<sub>2.5</sub> values estimated over urban centers such as Delhi, Kanpur, Lucknow and Patna are higher than 150  $\mu\text{g m}^{-3}$  and even higher values (> 300  $\mu\text{g m}^{-3}$ ) are reported during fog events (Bharali et al., 2019). High aerosol loadings over the IGP prevail during most of the winter period, with more than 60% of days being foggy (Ghude et al., 2017). The seasonal mean value of PM<sub>2.5</sub> reported at Delhi is three times higher than the national standard of 60  $\mu\text{g m}^{-3}$  for satisfactory levels of ambient air quality and the aerosol optical depth (AOD) exceeds 1.0 quite frequently over most of the IGP region.

In contrast to the numerous studies on the effects of aerosols on the Indian summer monsoon rainfall characteristics (e.g. see references in Li et al., (2016)), the impact of wintertime high aerosol loadings on regional haze and fog conditions have been little explored, although some studies have focused on the implications of regional emission sources on air quality or aerosol loading over the region. For example, Nair et al., (2007) showed that the aerosol variability over the IGP has strong relations with the boundary layer variability, whereas Bharali et al., (2019) reported that the aerosol forcing strongly influences the boundary layer evolution, thus strengthening the accumulation of pollutants near the surface. Hence, boundary layer variability affects the aerosol loading and the aerosol forcing influences the evolution of the boundary layer as reported by several studies over polluted urban centres (Bharali et al., 2019; Ding et al., 2016; Huang et al., 2014; Li et al., 2017; Tie et al., 2017). Similarly, several observational studies investigated the correlation between meteorological parameters, especially temperature and humidity, with aerosol loadings (Kumar et al., 2015a). However, these observational studies have inherent difficulties in separating the aerosol forcing on meteorological conditions from the effects of meteorology on the aerosol loading. Generally, low temperature and high humidity are observed over the IGP during winter, when the climate over the region is influenced by the western disturbances originating from the Mediterranean and bringing cold-moist air over the aerosol-laden IGP. In addition, irrigation, water bodies (rivers and lakes) and farming activities enhance evapotranspiration and relative humidity in the lower troposphere which, together with high aerosol loadings, lead to

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extensive and frequent fog events over the region (Gautam et al., 2007; Ghude et al., 2017; Goswami and Sarkar, 2015; Syed et al., 2012).

The effect of aerosols on regional climates depends on the aerosol-radiation interactions, which are influenced by the water affinity of particles and the ambient relative humidity. The optical properties of aerosol (AOD/scattering and extinction coefficient) are enhanced by more than 2 times at higher relative humidity (>80%) condition, which has a strong dependence on the relative dominance of organic and inorganic species (chemical composition) and size of the particles. However, the direct measurements of hygroscopic growth functions for physical (size) or optical (scattering coefficient) properties of aerosols are limited over the Indian landmass (Mandariya et al., 2020). Even though several studies addressed the implications of aerosols on air quality, health, and regional and global climate, there are very limited efforts to understand the implications of aerosol hygroscopicity on regional meteorology, visibility and air quality.

Based on these considerations, in this study, we use a regional climate model (the RegCM4, Giorgi et al., (2012)) coupled to air chemistry and aerosol models to assess the contribution of the aerosol hygroscopic growth to the total aerosol optical depth over the IGP. Since the total radiative impacts of aerosols on surface temperature, cloud properties and precipitation through various forcing pathways are more comprehensively explored by several studies (eg: Li et al., (2016)), we focus only on the effects of the increase in AOD due to relative humidity. Further, we quantify the effects and feedbacks of the aerosol hygroscopic growth on the regional meteorology, visibility and low air quality conditions over the region. The model configuration and experimental details are given in the next section.

## 2 Materials and methods

### 2.1 Regional Climate Model (RegCM4)

For our study, we use the regional climate model version 4 (RegCM4) coupled with atmospheric chemistry and aerosol module. RegCM4 is a limited area model with a hydrostatic dynamical core and sigma vertical pressure coordinates (Giorgi et al., 2012). The South Asia-CORDEX domain is used in this study (Figure 01) with 50 km horizontal grid spacing, 18 vertical levels and model top at 50hPa. The meteorological initial and lateral boundary conditions for our simulations are provided by the ERA-interim re-analysis (Dee et al., 2011) for the three winter seasons (December to February) starting from November 2014 to March 2017, where the first month of simulation (November) of each year is discarded from the analysis as model spin-up. Optimum interpolation weekly sea surface temperature data from the National Oceanic and Atmospheric Administration (NOAA) are used as lower boundary condition over the Ocean. The parameterization schemes used in the simulations are (1) boundary layer: University of Washington PBL scheme, (2) Convection over land and ocean: Tiedtke scheme, (3) Radiative transfer: Community Climate Model (CCM3) scheme, (4) cloud microphysics: Subgrid Explicit moisture scheme and (5) Land surface: Biosphere-Atmosphere Transfer Scheme. More details on the model configuration and physics are already discussed in earlier papers (Ajay et al., 2019; Giorgi et al., 2012; Usha et al., 2020). Note that in the

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validation and analysis of the model output we always consider the average over the three simulated seasons unless otherwise specified.

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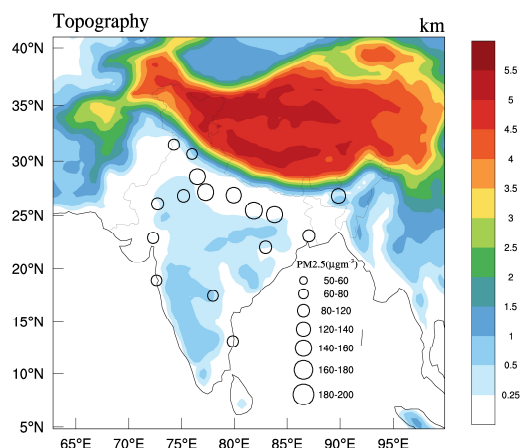


Figure 01: Study domain centred over South Asia. Colormap shows the topography (km) and circles indicate the near-surface mass loading ( $\mu\text{g m}^{-3}$ ) of particles below  $2.5 \mu\text{m}$  size range (PM2.5). PM2.5 data is taken from [the](#) Central Pollution Control Board (CPCB), India.

RegCM4 has an online chemistry module, which is extensively used for understanding aerosol-climate interactions (Nair et al., 2012; Shalaby et al., 2012; Solmon et al., 2015). The aerosol and trace gas emission fluxes are adopted from the IIASA dataset, and chemical boundary conditions are from the global model MOZART (model of ozone and related chemical tracers). Gas-phase chemistry is based on the CBMZ (carbon-bond mechanism version Z) scheme and the ISORROPIA II is used for inorganic aerosols (Shalaby et al., 2012). The aerosol scheme includes sulphate, nitrate, ammonia, sea salt (2 size bins), mineral dust (4 size bins), black carbon (hydrophilic and hydrophobic) and organic carbon (hydrophilic and hydrophobic). The sources, sinks, atmospheric processes and transport of the aerosol species are detailed in Solmon et al., (2006). The mass concentration of each aerosol species is converted into optical properties using a mass extinction cross-section, which depends on the ambient relative humidity and species-specific hygroscopic growth functions (Solmon et al., 2006). We use the growth function described by Kiehl et al., (2000) for sulphate, nitrate and ammonia aerosols, in which, the growth function increases exponentially with humidity. For example, at 80% relative humidity, the particle mass extinction cross-section increases by a factor of 3 compared to its dry value. The model emits carbonaceous aerosols as hydrophobic and the optical properties of this carbonaceous aerosols are invariant with relative humidity. These particles changes from hydrophobic to hydrophilic due to ageing at a fixed time scale of 1.15 days ( $\sim 27.6$  hours, Solmon et al., (2006)). The

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hydrophilic part of the carbonaceous aerosols (black carbon and organic carbon) has a weak affinity to water compared to sulphate aerosols (Kasten, 1969). At 80% relative humidity, the growth factor is 1.37 and 1.49 for hydrophilic black carbon and organic carbon, respectively. Mineral dust is considered as fully hydrophobic. The sea salt size increases by 2 times at 80% relative humidity compared to dry conditions, but this is of little importance over the IGP during winter. In general, most of the aerosols grow with relative humidity and this enhances the optical depth.

## 2.2 Experiments

RegCM4 has been widely used to investigate aerosol-hydroclimate interactions and the impacts of various forcings on monsoon characteristics (e.g. Solmon et al., (2015) and Usha et al., (2020)). The model can be run with and without radiative feedbacks of aerosols. In the former case, aerosols affect the radiation balance and change the surface temperature and meteorology through direct and indirect pathways. In the latter, the aerosol radiative forcing is calculated but it does not have effects on the meteorology, surface temperature and atmospheric thermodynamics. In the present study, model simulations with (ambient) and without (dry) hygroscopic growth of aerosols are named as experiments 1 and 2 respectively. Experimental details are summarized in Table 1. In the case of the dry aerosol simulations, we forced the aerosol hygroscopic growth functions to be equal to 1 in the model code. The model runs without meteorological feedbacks (Exp1 and Exp2) are considered as control runs for the wet and dry aerosol cases. Since the effect of aerosol-radiation interactions on land and meteorological parameters are switched off (no meteorological feedback), the difference between these two experiments provides the contribution of hygroscopic growth to the aerosol properties ( $\Delta AOD_{RH}$ )

$$\Delta AOD_{RH} = AOD(Ambient_{nofeedback}) - AOD(Dry_{nofeedback}) \quad (1)$$

Experiments 3 and 4 are similar to Exp 1 and 2 but with meteorological feedbacks (Table 1). The difference between the Exp3 and 4 provides the net effect of the hygroscopic growth of aerosols on the regional climate, which includes (i) changes due to the radiative forcing of the aerosol hygroscopic growth and (ii) its meteorological feedback. For example, the change in temperature (T) due to the meteorological feedback of  $\Delta AOD_{RH}$  is estimated as

$$\Delta T_{RH} = T(Ambient_{feedback}) - T(Dry_{feedback}) \quad (2)$$

The effect of the total aerosol system on meteorological variables (for eg: Temperature) is estimated as

$$\Delta T_{aerosol} = T(Ambient_{feedback}) - T(Ambient_{nofeedback}) \quad (3)$$

The  $\Delta T_{RH}$  is a subset of  $\Delta T_{aerosol}$ . The change in AOD solely due to the meteorological feedback of the total aerosol system (dry + hygroscopic growth) is estimated as

$$\Delta AOD_{feedback} = AOD(Ambient_{feedback}) - AOD(Ambient_{nofeedback}) \quad (4)$$

Hence the  $AOD(Ambient_{feedback})$  is the sum of dry AOD, its hygroscopic growth and the meteorological feedback

$$AOD(Ambient_{feedback}) = AOD(Dry_{nofeedback}) + \Delta AOD_{RH} + \Delta AOD_{feedback} \quad (5)$$

Table 1: Experimental details.

Exp. Name	Hygroscopic growth	meteorological feedback
-----------	--------------------	-------------------------

Exp1: Ambient <sub>nofeedback</sub>	Yes	No
Exp2: Dry <sub>nofeedback</sub>	No	No
Exp3: Ambient <sub>feedback</sub>	Yes	Yes
Exp4: Dry <sub>feedback</sub>	No	Yes

### 2.3 Model Validation

Several earlier studies used RegCM4 to simulate the meteorology of South Asia (Ajay et al., 2019; Giorgi et al., 2012; Nair et al., 2012; Usha et al., 2020). In general, the model captures the basic features of seasonal precipitation (monsoon), though it is sensitive to the convection schemes used and the extent of the ocean-atmosphere coupling (Ajay et al., 2019; Di Sante et al., 2019). Studies on the validation of chemical constituents simulated by the model are rather limited over South Asia (Nair et al., 2012; Solomon et al., 2015; Usha et al., 2020). A qualitative inter-comparison of the chemical composition of aerosols simulated by RegCM4 with in-situ measurements at distinct locations over the Indian region is shown in Figure 2. The measurements of column aerosol optical depth are taken from AERONET (Aerosol Robotic Network) radiometer observations at Jaipur, Kanpur, Gandhi College and Delhi. The black carbon measurements were carried out using Aethalometers installed under the ARFI (Aerosol Radiative Forcing over India) project. Since the measurements of aerosol composition are sparse over the Indian region, we have taken values reported from earlier studies for the winter season, and therefore the organic carbon and sulphate measurements are for different years than those of the model simulations (Ali et al., 2019; Aswini et al., 2019; George et al., 2008; Ram et al., 2010, 2012; Rengarajan et al., 2011; Safai et al., 2008; Satsangi et al., 2012). Despite these shortcomings, compared to earlier studies, the present model configuration simulates black carbon mass loading and AOD which are closer to the observed values (Nair et al., 2012). Though RegCM4 has a simple scheme for organic aerosols, the organic carbon mass concentration broadly matches the observed seasonal mean values. In fact, most climate and chemical transport models fail to capture the high aerosol loading over this region, a problem which has been mostly attributed to the parameterization of stable boundary layer conditions (Nair et al., 2012), unaccounted emissions (eg: Nair et al., (2012), references are therein) and biases in simulating relative humidity and precipitation (Chatani and Sharma, 2018; Feng et al., 2016). In the present study, AOD and the near-surface mass concentrations of black carbon, organic carbon, sulphate and dust simulated by the model are in the range of values reported by several studies over the region. One of the main challenges in simulating the AOD over the region is the accurate simulation of relative humidity, which affects the hygroscopic growth (Chatani and Sharma, 2018). The mean relative humidity estimated from several re-analysis datasets and measurements in different locations of the IGP varies from 60 to 80% (Chatani and Sharma, 2018; Gautam et al., 2007; Ghude et al., 2017; Goswami and Sarkar, 2015). A good agreement between measured and simulated (with aerosol feedback) relative humidity over the IGP (Fig. 2e) is important for the estimation of aerosol optical properties from simulated speciated aerosol mass concentrations. The RegCM4 simulated relative humidity values are approximately 10% higher than the measured RH, with a mean absolute error of 6.6% and root

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mean square error (RMSE) of 7.3%. During winter, aerosols are confined within the boundary layer, and changes in the boundary layer height play a major role in the dilution (ventilation) of aerosols over the IGP (Bharali et al., 2019; Nair et al., 2012). Shallow and stable boundary layers prevailed during the simulated winter seasons. A comparison with IGRA (Integrated Global Radiosonde Archive) radiosonde data indicates that the UW scheme is able to simulate the boundary layer height over the region reasonably well (Fig. 2f) with a mean absolute error of 200 m (26% of mean boundary layer height) and RMSE of 240 m. Further intercomparison of modelled and measured PM<sub>2.5</sub>, AOD and black carbon (BC) mass concentration at various locations over the Indian region is shown in supplementary Figure S1, S2 and S3.

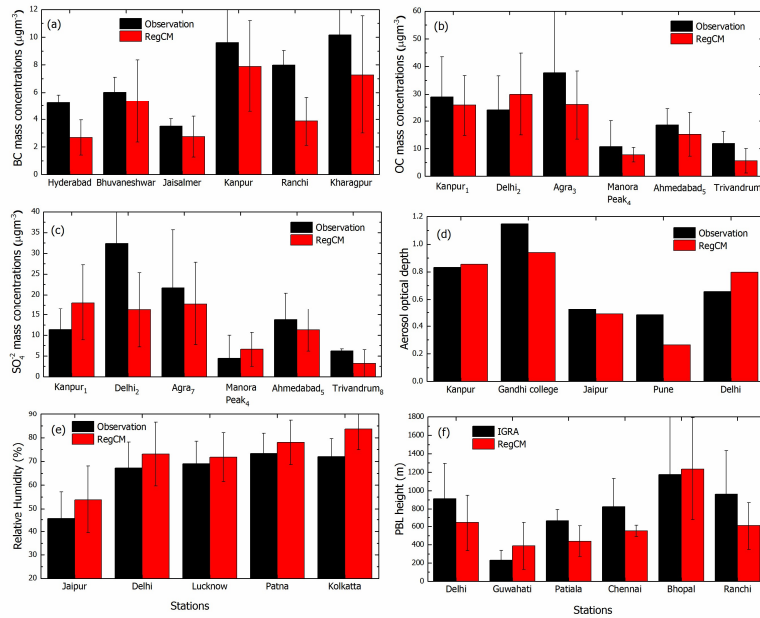
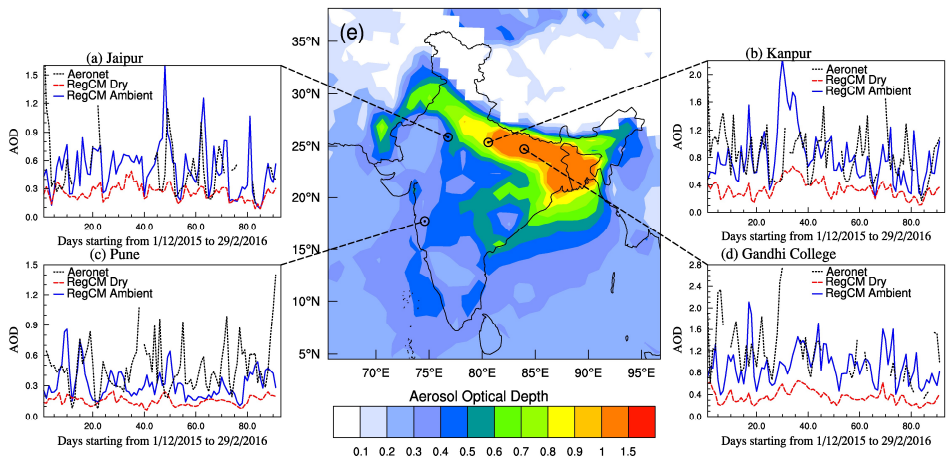


Figure 2: Validation of RegCM4 simulated (Exp 3) aerosol parameters ((a) black carbon (BC), (b) organic carbon (OC), (c) sulphate, (d) AOD), (e) relative humidity and (f) boundary layer height with measurements over different locations in India. The measurements of OC and sulphate mass loading are taken from the earlier measurements (Kanpur<sub>1</sub> - Ram et al., (2012); Delhi<sub>2</sub> - Ali et al., (2019); Agra<sub>3</sub> - Satsangi et al., (2012); Manora Peak<sub>4</sub> - Ram et al., (2010); Ahmedabad<sub>5</sub> - Rengarajan et al., (2011); Trivandrum<sub>6</sub> - Aswini et al., (2019); Agra<sub>7</sub> - Safai et al., (2008); Trivandrum<sub>8</sub> - George et al., (2008)) and these values represent the winter season but for different years than those of the model simulations.

3 Results and discussions

3.1 Hygroscopic growth of aerosols

The particulate mass loadings (PM<sub>2.5</sub>) measured over most cities in the IGP during winter are well above the air quality standards, as shown in Figure 1. The total column AOD at ambient humidity simulated by RegCM4 with meteorological feedbacks show very high values over the IGP, mostly consisting of boundary layer aerosols. The comparison of measured and simulated AOD for dry and ambient humidity conditions (Fig. 3) over the distinct environments of the IGP indicates an almost two-fold increase in AOD due to the hygroscopic growth of aerosols and its meteorological feedback. The mean observed AODs (simulated ambient and dry AODs) for Jaipur, Kanpur, Pune and Gandhi College are 0.53 (0.51, 0.26), 0.83 (0.81, 0.34), 0.48 (0.32, 0.15) and 1.15 (0.92, 0.36) respectively. In general, both the magnitude and large day-to-day variability in AOD (Fig. 3) are captured by the model when the hygroscopic growth ( $\Delta AOD_{RH}$ ) and meteorological feedback ( $\Delta AOD_{feedback}$ ) are included in the simulation. Except for dust and freshly emitted black carbon, most of the aerosol species have an affinity towards water vapour, with the hygroscopic growth of inorganic aerosols such as sulphate, ammonia, nitrate and sea salt being more pronounced compared to organic carbon (Solmon et al., 2006). Due to ageing, a fraction of hydrophobic black carbon also becomes slightly hydrophilic (Solmon et al., 2006) and contributes, though not to a large extent, to the AOD at ambient humidity. Observations close to the Thar Desert (Fig. 1a, Jaipur) show lower hygroscopic growth (43%) compared to other IGP stations because of the dominance of dust aerosols and relatively low humidity conditions prevailing there.



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Figure 3: Wintertime AOD measured using Aeronet radiometers and simulated with RegCM4 for ambient and dry humidity conditions with meteorological feedback at (a) Jaipur, (b) Kanpur, (c) Pune and (d) Gandhi College. Stations (b) and (d) are located into the IGP while (a) and (c) are located outside the IGP. One season (December 2015 - February 2016) data is shown here for inter-comparison; same is the case for other years. (e) Spatial distribution of satellite (MODIS) retrieved aerosol optical depth at 550 nm.

Our study shows that the large day-to-day and spatial variability in AOD over the IGP is mostly contributed by relative humidity (Fig. 3), with lower contributions from changes in source characteristics and synoptic-scale circulation. The simulated AOD at ambient relative humidity matches well with the measured AOD (mean absolute error of 0.12 and RMSE of 0.15) whereas the dry AOD is significantly lower and less variable in time. The standard deviation of measured and simulated AOD at ambient humidity conditions is nearly half of the mean AODs (coefficient of variation of 50%) (Fig. 3), whereas, for dry aerosols, the standard deviation of the AOD is low (0.05 to 0.1), with a coefficient of variance ranging between 20 to 30%. All the meteorological conditions, associated processes (winds, chemistry, deposition, and transport) and anthropogenic emissions remained the same for dry and ambient AOD simulations, except the hygroscopic growth of AOD with relative humidity for the ambient AOD. Compared to the dry AOD, the ambient AOD shows large variability associated with the variability of humidity. The AOD due to dry aerosols has 10 to 15 % less variability compared to that of the total AOD.

The AOD due to hygroscopic growth ( $\Delta AOD_{RH}$ ) estimated using Eqn. 1 is shown in Fig. 4a. The  $\Delta AOD_{RH}$  values are high over the IGP, especially the eastern IGP and Bangladesh ( $\Delta AOD_{RH} > 0.3$ ). The ratio of  $AOD(Dry_{nofeedback})$  to  $AOD(Ambient_{nofeedback})$  shown as contours in Figure 4a indicates that dry aerosols contribute 50 to 60% of the AOD over most of the Indian sub-continent (Fig. 4a), except over western India, where the contribution of hydrophobic dust to the total AOD is high even during winter. The contribution of the aerosol hygroscopic growth is more than 50% of the total AOD over the northern Indian Ocean, which is attributed to the dominance of hydrophilic aerosols (sea salt) and high relative humidity. It is interesting to note that the increase in AOD due to hygroscopic growth ( $\Delta AOD_{RH} = AOD(ambient) - AOD(dry)$ ) is  $\sim 0.23$  over the IGP (Fig. 4a), which is higher than the global mean AOD ( $0.155 \pm 0.018$ ) reported by Watson-Parris et al., (2020) using various satellite observations. Mandariya et al., (2020) have reported that wintertime aerosols over central the IGP show an increase in particle diameter of  $1.34 \pm 0.07$  times as that of the dry particles at 85% relative humidity. This increase in particle diameter corresponds to a doubling of the scattering coefficient for anthropogenic aerosols (McInnes et al., 1998). In the present study, the mean dry AOD increases almost 1.7 times due to relative humidity over the IGP, which is slightly lower than the estimated AOD for the hygroscopic factor reported by Mandariya et al., (2020). Since organic aerosols dominate the aerosol system over the IGP (Ram et al., 2012), there could be a substantial decrease in the hygroscopic growth of particles as discussed by several studies. Mandariya et al., (2020) have reported that 150 nm particles are more hygroscopic than 100 nm particles due to the higher organic fraction present in the 100 nm particle mass concentration. In addition, although most observational studies consider 40% as the reference humidity level

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(dry) for estimating the hygroscopic growth, the model estimates the dry AOD for zero relative humidity. Titos et al., (2016) reported that the assumption of no growth at 40% humidity contributes to higher uncertainty in the estimates of measurement-based hygroscopic growth.

In general, the mean AOD over the IGP stations (Kanpur and Gandhi College) is  $\sim 0.7$ , indicating significant surface dimming of solar radiation (Fig. 4b). The increase in AOD due to the hygroscopic growth alone reduces the surface solar flux by 10 to 20  $\text{Wm}^{-2}$  over the IGP. Nair et al., (2016a) reported a mean surface forcing of  $-28.2 \pm 12 \text{ Wm}^{-2}$  due to total aerosol system over the entire Indian region during winter. The surface dimming due to aerosol-radiation interactions results in the reduction of surface temperature, heat fluxes (sensible and latent) and boundary layer height (Bharali et al., 2019; Ding et al., 2016; Li et al., 2017). As a result, aerosol forcing leads to the accumulation of pollutants in the boundary layer (as discussed later). Since water vapor contributes significantly to the total AOD, most of the effects of aerosols on regional climate, visibility and air quality are strongly associated with meteorological conditions.

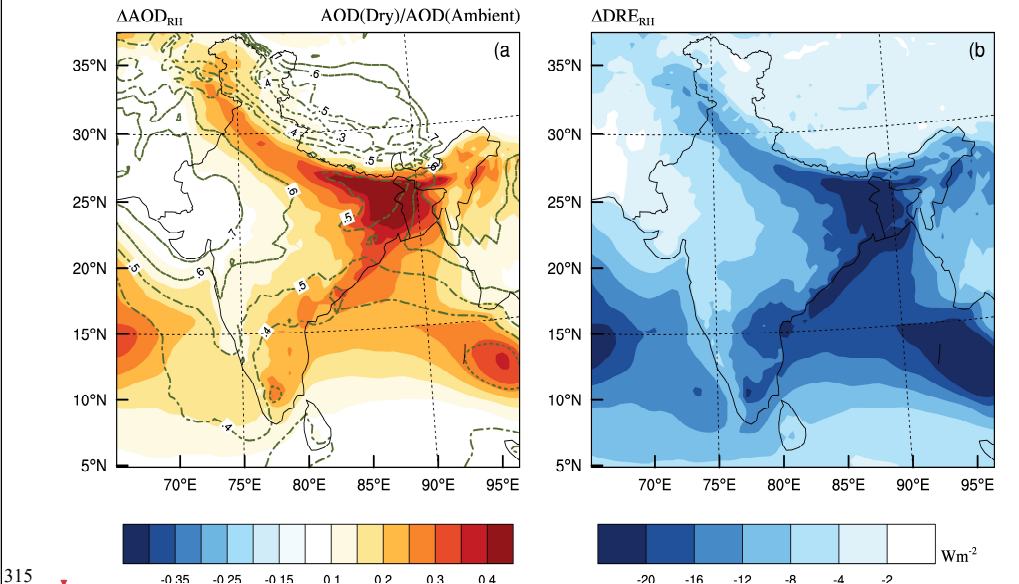
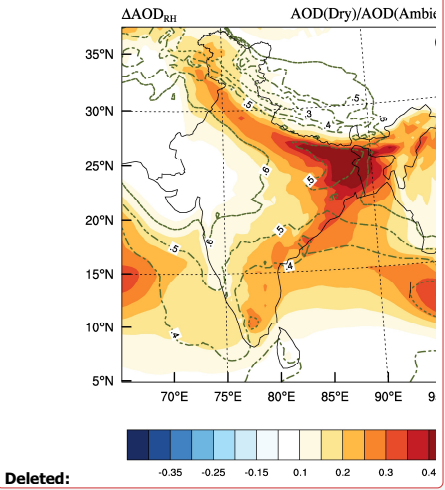


Figure 4: (a) Aerosol optical depth (AOD) simulated using RegCM4 for dry and ambient humidity conditions. Colour map indicates  $AOD(ambient) - AOD(dry)$  for without meteorological feedback and line contours show  $AOD(dry)/AOD(ambient)$  (b) Difference in aerosol direct radiative effects (DRE) at the surface for dry and ambient humidity conditions (without meteorological feedbacks).

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### 330 3.2 Effects on regional meteorology

The effects of the increase in AOD solely due to the hygroscopic growth of particles ( $\Delta AOD_{RH}$ ) on surface temperature, relative humidity and cloud fraction (Eqn. 2), is illustrated in Figure 5 (top panel), which shows a surface cooling of about 0.5°C and a 3% increase in relative humidity over the IGP during winter. This is mostly due to the direct solar dimming at the surface and to a cloud cover change (up to 3%) induced by the  $\Delta AOD_{RH}$  (Figure 5c). The total effect of aerosols on temperature, relative humidity and cloud fraction (Eqn. 3) is shown in Figures 5d & 5e. Nearly 40 to 50% of the aerosol-induced surface cooling is contributed by water vapor through the hygroscopic growth of particles. The cloud cover increases by 3 to 5 % over the IGP due to ambient aerosol forcings (Figure 5f). These effects are more prominent over the IGP and central India, whereas high  $\Delta AOD_{RH}$  and surface dimming is seen over the eastern IGP (Fig. 4). Although AOD and  $\Delta AOD_{RH}$  are high, the change in relative humidity due to the hygroscopic growth of aerosols (Figure 5b) is negligible over Bangladesh and Eastern IGP. The presence of relatively high cloud fraction over the eastern IGP mask the aerosol-induced surface cooling which is attributed to the contrast in the spatial pattern of surface temperature and AOD. The change in shortwave flux at the surface showed an almost similar pattern as that of change in temperature (Figure S4). Bharali et al., (2019) have reported that the strength of land-atmosphere interactions through the exchange of heat and moisture fluxes plays a major role in determining the aerosol induced dimming effects on meteorology (more details are available in Li et al., 2017). At the surface, an observed decrease in evapotranspiration (and associated rainfall) due to solar dimming further confirms a slowing down of the hydrological cycle induced by the aerosol hygroscopic growth (Liepert et al., 2004; Ramanathan, 2001). In other words, water vapour itself reduces the evaporation through dimming associated with the aerosol hygroscopic growth.

The mean vertical profiles of change in temperature and relative humidity over the IGP due to total aerosol and hygroscopic effects are shown in Figure 6. Significant cooling and associated moistening are noticeable in the boundary layer followed by a weak drying in the lower free troposphere (1 to 3 km) and a negligible influence above (>3 km). Based on extensive measurements from Delhi during wintertime fog campaigns, Ghude et al., (2017) have reported more than 70% humidity within the boundary layer and dry conditions (<20%) above 3 km. A similar pattern is observed for specific humidity, with an increase of 0.6 g kg<sup>-1</sup> at the surface, which confirms that the hygroscopic growth is the single largest contributor to the total aerosol effects over the IGP during winter (Figure S5).

Surface cooling, along with the weak entrainment of dry air mass from the free troposphere, leads to an increase in relative and specific humidity at the surface (Fig. 5a). Based on extensive measurements of aerosol and humidity profiles, Feng et al., (2016) reported up to 30% underestimation of relative humidity within the boundary layer and 20% overestimation in the free troposphere by the WRF-Chem model over a north Indian station (Nainital) during the winter season. They attributed this dry bias in boundary layer humidity as one of the major reasons for the underestimation of AOD by climate models over the region. Most re-analysis datasets (NCEP and ECMWF) have a negative bias (underestimate) in simulating relative humidity over this region, especially during winter (Chatani and Sharma, 2018; Feng et al., 2016), which

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significantly affects the simulation of aerosol optical depth, aerosol radiative forcing and fog prediction. The present study shows that the negative bias (underestimation) of relative humidity could be reduced, especially at a high relative humidities (RH > 70%), by including the meteorological feedback of ambient aerosols (Exp 4). In contrast to boundary layer moistening, drying in the lower free troposphere (1.5 to 3.5 km) by 1 to 2% was attributed to the aerosol-induced warming, which decreases humidity and suppresses mixing of air mass between the free troposphere and boundary layer. Compared to dry conditions, the warming of the top of the boundary layer occurs at a lower altitude for ambient aerosol forcing.

We analysed re-analysis data and in-situ observations for the long-term trend in wintertime relative humidity within the boundary layer and free troposphere over the IGP during the last four decades (Fig. 7). In contrast to the global scenario, relative humidity increased over the Indian region at the rate of 1% per decade, together with a concurrent increase in aerosol loadings (Babu et al., 2013) and the number of foggy days (Ghude et al., 2017; Syed et al., 2012). Based on the above discussions (Fig. 6), it can be argued that the aerosol forcing contributes significantly to the observed increasing trend of wintertime moistening of the boundary layer and enhanced occurrence of fog events over the IGP. The aerosol radiative forcing increases the stability of the atmosphere and decreases the transport of moisture from the boundary layer to the free troposphere, which favours the drying trend in the lower free troposphere shown in Figure 7b. Earlier studies also attributed the humidification of the boundary layer and drying of the free troposphere during winter to the aerosol forcing (Li et al., 2017; Tie et al., 2017). Our analysis shows that aerosol feedback processes significantly increase the near-surface relative humidity due to weak turbulent diffusivity in the stratified boundary layer (Bharali et al., 2019; Li et al., 2017). The increase in relative humidity, when surface temperature decreases, is due to the increase in water vapour content in the atmosphere as seen in the specific humidity trend reported by Mukhopadhyay et al., (2017). In contrast to the negative trend in relative humidity in the lower free troposphere during winter (Fig. 7b), Mukhopadhyay et al., (2017) show a positive trend for annual mean humidity, which is largely dominated by the positive trend during summer. Global analyses have shown that most continental regions exhibit a long term warming trend and decrease in relative humidity (Dai, 2006), which has strong implications for the land-ocean warming contrast (Hodnebrog et al., 2019). The long-term trend in the moistening of the boundary layer and drying of the free troposphere over the Indian region during winter is further amplified by the aerosol radiative forcing, primarily through the hygroscopic growth. However, the masking effects of surface warming by aerosols and the contribution of aerosols to the increasing trend in humidity is yet to be quantified.

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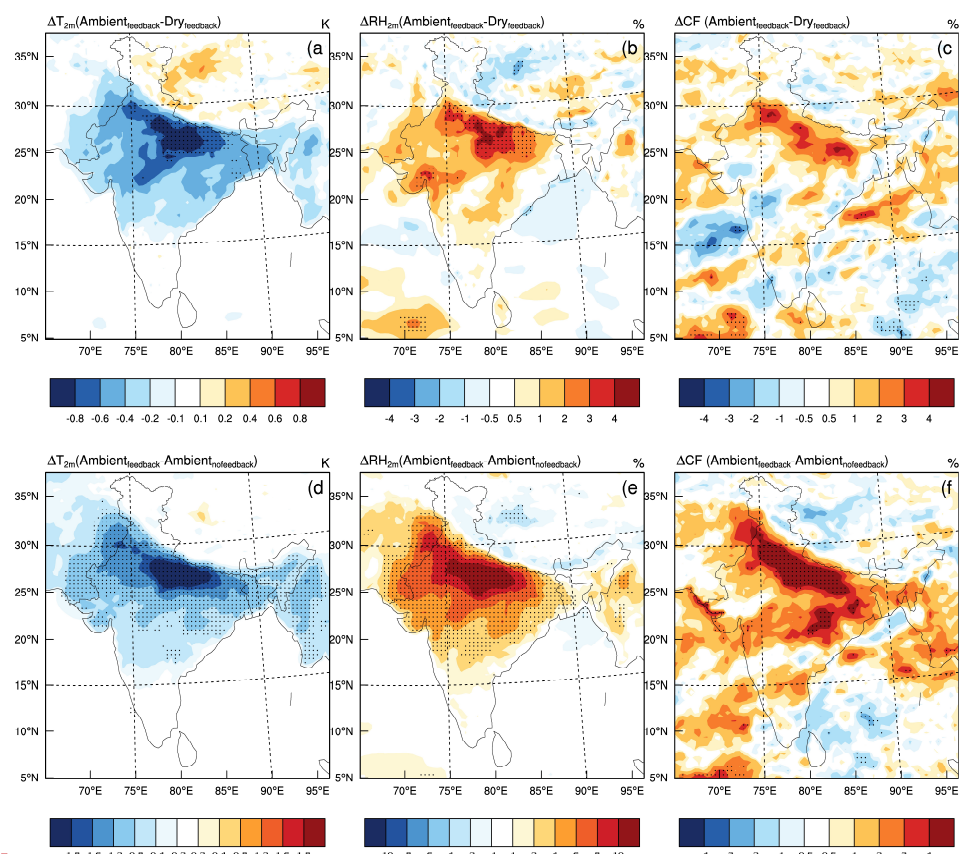
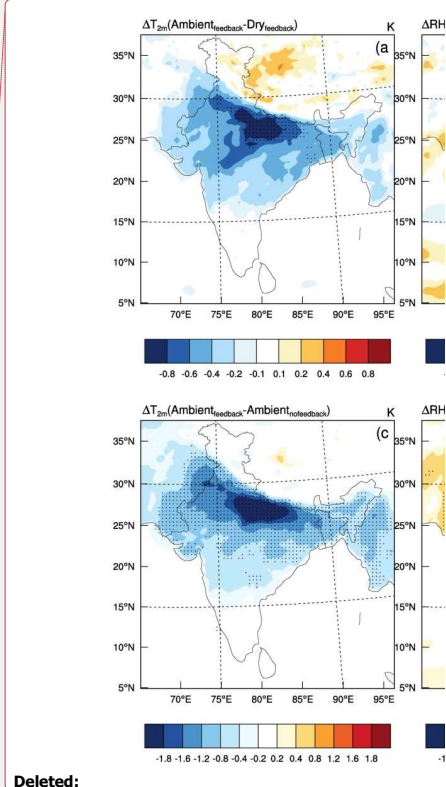


Figure 5: Change in (a) 2m air temperature, (b) relative humidity and (c) cloud fraction solely due to the hygroscopic growth of aerosols (ambient-dry). Change in temperature and relative humidity (RH) due to the total effect of aerosols (dry + hygroscopic growth) is shown (c), (d) and (f). Dots indicate statistical significance of Student's t-test at a 90% level.



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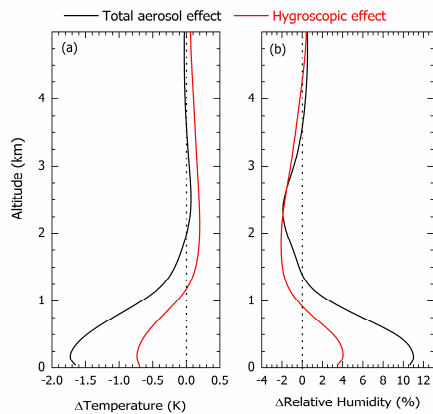


Figure 6: Vertical profiles of change in relative humidity and temperature due to the total aerosol effect ( $Ambient_{feedback} - Ambient_{nofeedback}$ ) (black line) and only due to hygroscopic growth of aerosols ( $Ambient_{feedback} - dry_{feedback}$ ).

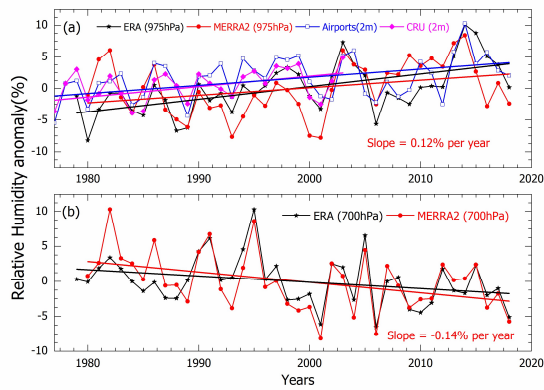


Figure 7: (a) Relative humidity in the boundary layer (975hPa) from re-analysis (ERA, MERRA and CRU) and in-situ observations (b) relative humidity in the free troposphere (700 hPa) taken from re-analysis data.

### 3.3 Implications for visibility and air quality

A regional mean AOD increase of  $\sim 0.23$  is contributed by the hygroscopic growth of aerosols (Figure 3), which further increases the AOD through meteorological feedbacks ( $\Delta AOD_{feedback}$ ) (Figure 8a), whose contribution can be

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estimated using Eqn. 4. Overall, the hygroscopic growth ( $\Delta AOD_{RH} \sim 0.23$ ) and associated meteorological feedback ( $\Delta AOD_{feedback} \sim 0.17$ ) contribute significantly to the total AOD (0.72) over the IGP. This positive feedback is stronger over the central IGP than the eastern IGP, though the eastern IGP has higher AOD and relative humidity. The moisture content in the atmosphere increases the AOD directly by its hygroscopic effect (Fig. 3) while high AOD increases the surface relative humidity through its radiative and meteorological feedbacks (Fig. 8a). Even though the spatial patterns of change in AOD and relative humidity due to aerosol feedbacks are slightly different, the anomalies in AOD and relative humidity show a significant correlation over most of the Indian sub-continent, especially over the IGP (Fig. 8b). Since, the aircraft and lidar observations over the IGP clearly showed that aerosols are mostly confined within the boundary layer during the winter season with lower contributions from the free tropospheric aerosols (Nair et al., 2016b), we have used AOD and surface-level parameters like PM<sub>2.5</sub>, and visibility to investigate the implications of aerosol-induced forcing on air quality and visibility.

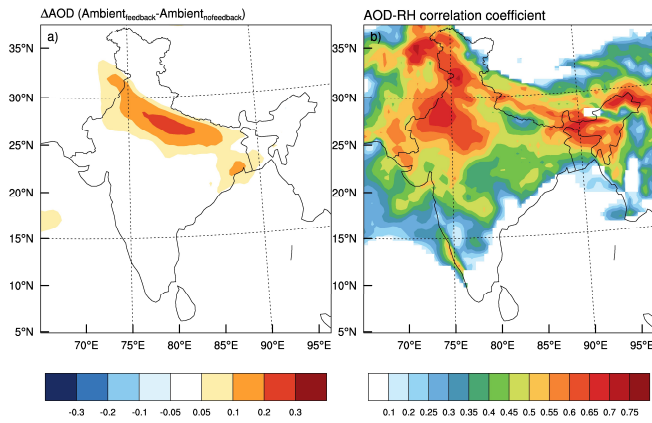


Figure 8: Change in AOD due to the meteorological feedbacks of the hygroscopic growth of aerosols ( $(\Delta AOD_{feedback} - \Delta AOD_{nofeedback})$ ). Correlation coefficient of AOD and relative humidity due to the meteorological feedback of hygroscopic growth of aerosols.

The frequency of occurrence of AOD and PM<sub>2.5</sub> for dry and ambient humidity conditions with meteorological feedback over the IGP is shown in Figure 9. The inclusion of meteorological feedback and hygroscopic aerosol growth ( $\Delta AOD_{RH}$ ) produces an increase in AOD along the IGP (Fig. 5). The narrow frequency distribution for dry AOD broadens substantially when including the effect of ambient humidity, which implies an increase in the number of days with hazy skies, high AOD and significant dimming at surface. The frequency distribution of the ratio of AOD for ambient and dry humidity with and without meteorological feedback is shown in Figure 9b. The no-feedback simulation shows a mean ratio

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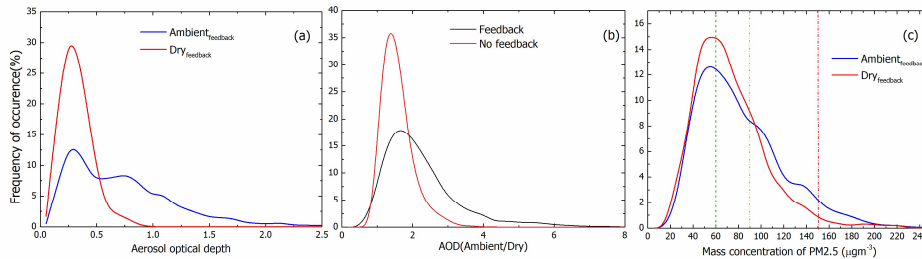
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of 1.72, which increases to 2.3 times at ambient humidity with meteorological feedbacks. Due to the positive feedback as mentioned above, AOD at ambient humidity increases more than 3 times of its dry AOD, depending on the strength of the land-atmosphere and aerosol-boundary layer interaction. The magnitude of the meteorological feedback due to the total aerosol system ( $\Delta AOD_{\text{feedback}}$ ) is comparable to  $\Delta AOD_{RH}$  over the IGP. This clearly indicates that not only the hygroscopic growth of aerosols but also the meteorological feedback is also important over the IGP. Therefore, the aerosol feedbacks with regional meteorology need to be investigated further to better understand and predict the fog events over IGP (Bharali et al., 2019). Although the dominance of organics in the aerosol mass loading over the IGP is expected to decrease the hygroscopic growth of pure inorganic aerosols (Mandariya et al., 2020), the meteorological feedback processes (Eqn. 3) increase the AOD by moistening the boundary layer ( $\Delta RH_{\text{aerosol}}$ ). The frequency of occurrence of high values of PM2.5 mass concentration for ambient humidity shows deterioration of air quality due to aerosol-radiation interaction and the aerosol forcing itself increases the aerosol loading through aerosol-boundary layer interaction.

In fact, generally calm winds ( $< 2 \text{ ms}^{-1}$ ), low temperature, shallow boundary layer and slow descending motion prevail over the IGP during winter, which favours the accumulation of aerosols within the boundary layer (Ghude et al., 2017; Kumar et al., 2015a; Nair et al., 2007). In addition, the aerosol effects further weaken the horizontal and vertical circulations and decrease the boundary layer height (Figure S6), which results in more accumulation of aerosols and thus poorer air quality (Bharali et al., 2019; Ding et al., 2016). Although wintertime aerosols are mostly confined within the boundary layer, near-surface PM2.5 and columnar aerosol loading show poor correlation over the IGP as reported by Kumar et al., (2015a). Our study also shows a poor correlation between ambient AOD and PM2.5, however, a high correlation is found between the dry AOD and PM2.5. This result has implications for the estimation of PM2.5 from the satellite observations or for finding an empirical relationship between AOD and PM2.5 for air quality applications, since the high relative humidity deteriorates the association between AOD and PM2.5. By and large, the offline filter-based PM2.5 measurements have issues associated with sampling aerosols at ambient humidity conditions. This also points to the need for dedicated field experiments focusing on the size segregated hygroscopic growth functions of physical and optical properties of aerosols over the IGP (Mandariya et al., 2020).



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Figure 9: Frequency of occurrence of (a) AOD and (c) PM2.5 for dry and ambient relative humidity conditions with meteorological feedback over the Indo-Gangetic Plain (IGP). (b) The frequency distribution of the ratio of AOD for ambient and dry humidity with and without meteorological feedback. Dotted vertical lines on PM2.5 frequency of occurrence shows the breakpoint concentration for air quality index category for satisfactory ( $60 \mu\text{g m}^{-3}$ ), moderately polluted ( $90 \mu\text{g m}^{-3}$ ) and poor ( $150 \mu\text{g m}^{-3}$ ) conditions as proposed by Central Pollution Control Board (CPCB), India.

As shown in Figure 10, there also exists an enhancement in near-surface aerosol mass loading (carbonaceous, inorganics and PM2.5) due to the suppression of boundary layer height induced by the meteorological feedbacks from the aerosol radiation interaction. This results in further degradation of air quality of this region. The aerosol concentration increases along the IGP due to aerosol induced weak ventilation. Figure 11 shows the estimated visibility using the aerosol extinction coefficient. Due to high aerosol loading and high humidity conditions, visibility is low over the region. Compared to the dry aerosols, there is a 60% reduction in visibility because of the aerosol hygroscopic growth (supplementary figure S7). In addition, the enhancement in relative humidity due to aerosol forcing which again decreases the visibility. This analysis further confirms that the air quality problems in the IGP are associated not only with the aerosol source strength but also with the ambient humidity conditions and its (aerosol) radiative forcing and meteorological feedback.

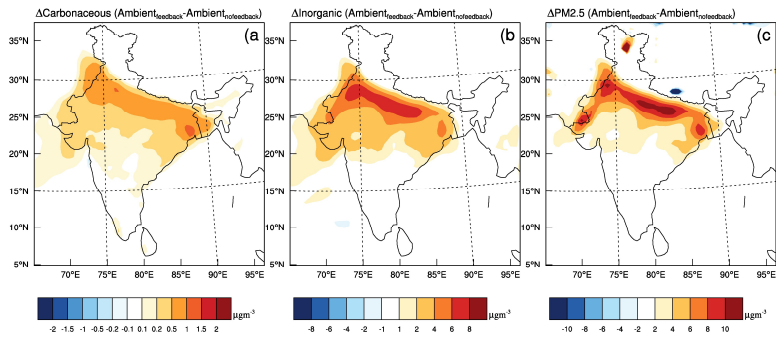


Figure 10: Change in near surface mass loading ( $\mu\text{g m}^{-3}$ ) due to the meteorological feedback of total aerosol radiative forcing for (a) carbonaceous aerosols, (b) inorganic aerosols and (c) PM2.5.



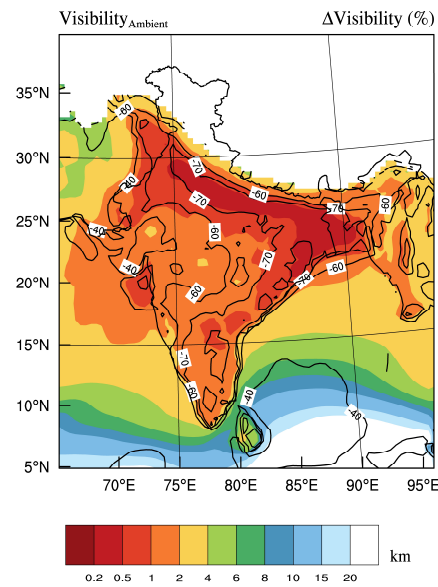


Figure 11: Spatial variation of mean visibility over the Indian region for ambient feedback simulation. The percentage variation of visibility changes due to hygroscopic growth with respect to visibility due to dry aerosols is given as line contour.

As discussed above, the AOD increase via hygroscopic growth of aerosols under high relative humidity conditions further decreases the incoming solar radiation at the surface, which results in enhanced surface cooling (compared to dry aerosols) and a decrease of the exchanges between the boundary layer and free troposphere. Aerosol-radiation interactions lead to a significant increase in cloud cover over the IGP region (2 to 8%), which is further increased by the hygroscopic effect of aerosols. The increase in cloud cover leads to more sulphate formation and thus again influence the radiative balance. Hence, the hygroscopic effects of aerosols significantly strengthen the observed aerosol-boundary layer interactions over the region (Bharali et al., 2019). Recent studies have shown that the high aerosol concentrations at the surface over the urban centres are strongly related to positive feedback processes associated with the boundary layer and water vapour (Ding et al., 2016; Huang et al., 2014; Li et al., 2017; Tie et al., 2017). The surface solar dimming and atmospheric warming due to black carbon in the upper boundary layer decrease the height of the mixed layer (increases stratification) and increase the accumulation of aerosols within the boundary layer (Bharali et al., 2019; Ding et al., 2016; Nair et al., 2007). The aerosol-

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induced increase in relative humidity (1-10 %) over most land regions (Fig. 5), especially the central IGP, further increases the AOD. Tie et al., (2017) have reported that a decrease in the dispersion of water vapour leads to a self-amplifying feedback mechanism through which an increase in relative humidity further increases the AOD due to hygroscopic growth ( $\Delta AOD_{RH}$ ). All these pathways further increase moisture in the boundary layer and, consequently, the AOD (Tie et al., 2017). These feedback processes significantly deteriorate air quality conditions during winter (Li et al., 2017).

#### 4 Conclusions

In this work, the regional climate model RegCM4, interactively coupled with atmospheric chemistry and aerosols is used to investigate the contribution of the hygroscopic growth of aerosols to the total aerosol optical depth and its meteorological feedback over the Indian sub-continent. Our analysis shows that the aerosol hygroscopic growth can contribute up to 40% of the total AOD and that feedback processes significantly increase near-surface relative humidity and decrease lower free troposphere humidity. This might strengthen the long-term trend in boundary layer moistening and free tropospheric drying over the IGP during winter. We also show that the inclusion of the meteorological feedback due to the hygroscopic growth of aerosols ( $\Delta AOD_{RH}$ ) produces an increase in AOD along the IGP, especially over the central IGP regions. The moisture content in the atmosphere not only increases the AOD directly by its hygroscopic effect but also indirectly through its radiative and meteorological feedbacks. Briefly, aerosol forcing due to dry aerosols and their hygroscopic growth at high relative humidity conditions increase the relative humidity in the boundary layer, which further increases the AOD through positive feedback. Our analysis demonstrates that the aerosol-moisture interaction is the most significant contributor favouring and strengthening the high aerosol conditions (poor air quality) prevailing over the IGP during winter.

Our study also highlights the need to increase understanding of aerosol/climate/air quality interactions over the India sub-continent through: i) the inclusion of hygroscopic growth and related feedbacks in climate/chemistry models; ii) direct measurements of hygroscopic growth functions of aerosols, which are rather limited over the region (Mandariya et al., 2020); and iii) measurements and model description of the effect of ageing and mixing state on the water affinity of hydrophobic aerosols. Some model limitations specific to this study are worth noting here. The RegCM4 has a simple organic aerosol module and a single growth function for the hundreds of organic species present in the atmosphere, which are characterized by a wide range of affinity towards water vapour. The model does not include the effects of organic aerosols on the water affinity of inorganic aerosols, and in addition the effect of aerosols on cloud characteristics (indirect aerosol effect) is not included. All these aspects of model development are underway in the next version of the RegCM4 modelling system.

Notwithstanding these limitations, our study clearly shows that understanding the interactions of natural factors (moisture fluxes and relative humidity) with the anthropogenic aerosols (organic and inorganic) is essential for predicting fog and haze events over the IGP and devising appropriate pollution mitigation strategies. In this regard, the aerosol-water vapour interaction is a unique example of the amplification of anthropogenic forcing (aerosols) by natural agents (water

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vapour) leading to significant changes in regional climate and air quality. To date, low air quality and visibility events over the IGP have been considered essentially as a problem of emission sources and transport of particles, however, our study highlights the important, and in fact sometimes dominant, [contributions](#) of atmospheric water vapour to these events, and thus the need to consider this natural factor in air quality assessments and related policymaking.

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**Data and materials availability:** Data are available upon request from the contact author, Vijayakumar S. Nair ([vijayakumarsnair@gmail.com](mailto:vijayakumarsnair@gmail.com)).

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