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.

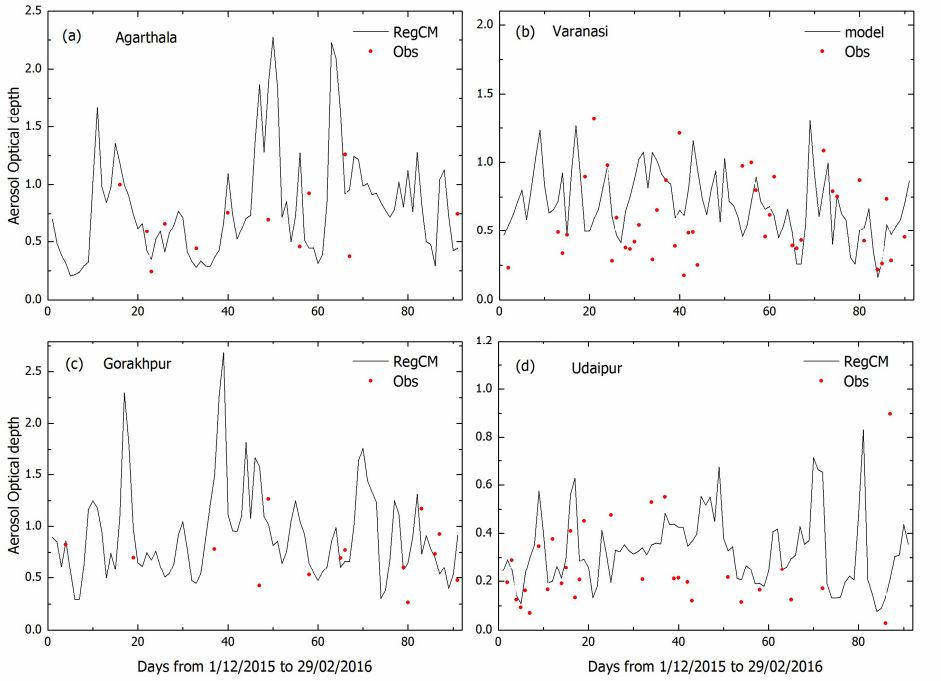

Figure S2: *Variation of aerosol optical depth measured using multi-wavelength radiometer installed as a part of Aerosol Radiative Forcing over India Project (ARFINET) and modeled using RegCM4 from December 2015 to February 2016.*

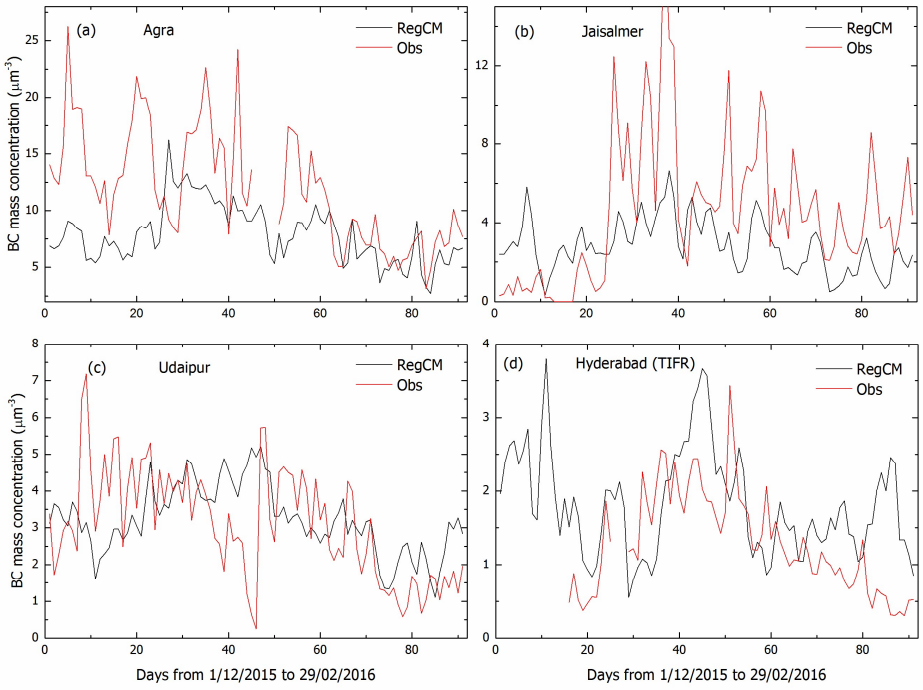

Figure S3: *Temporal variation of BC mass loading measured at 4 ARFINET stations and modeled using RegCM4 over Agra, Jaisalmer, Udaipur and Hyderabad from December 2015 to February 2016.*

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.

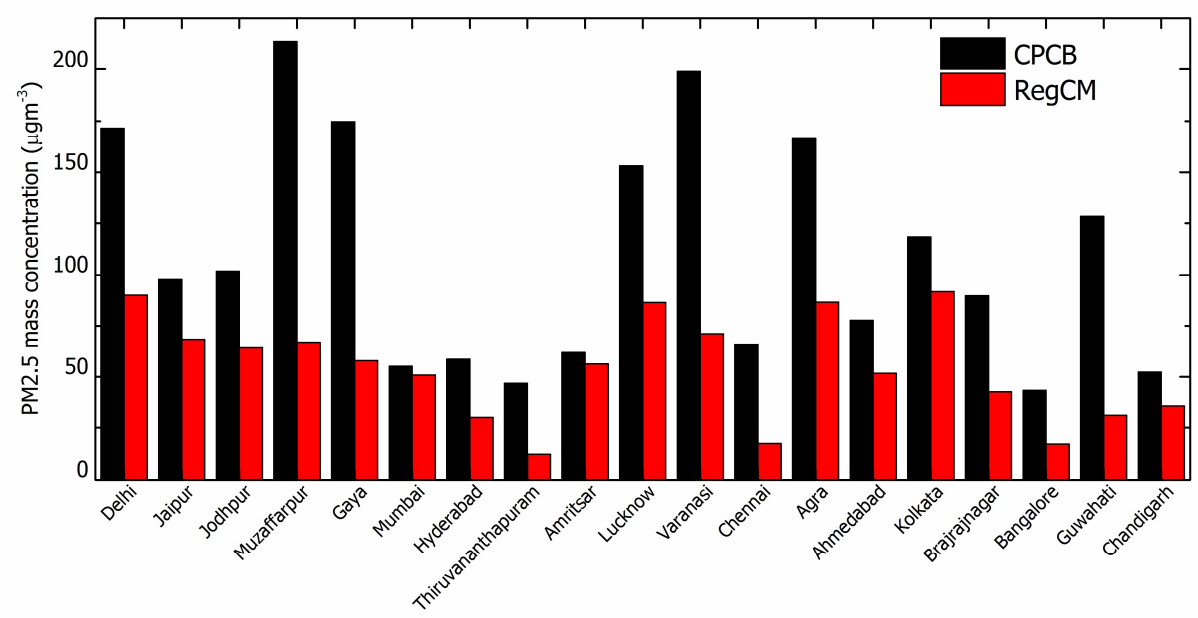

Figure S1: *Inter-comparison of PM2.5 simulated using RegCM4 with measurements from 19 stations maintained by Central Pollution Control Board (CPCB) India (www.cpcb.nic.in).*

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.

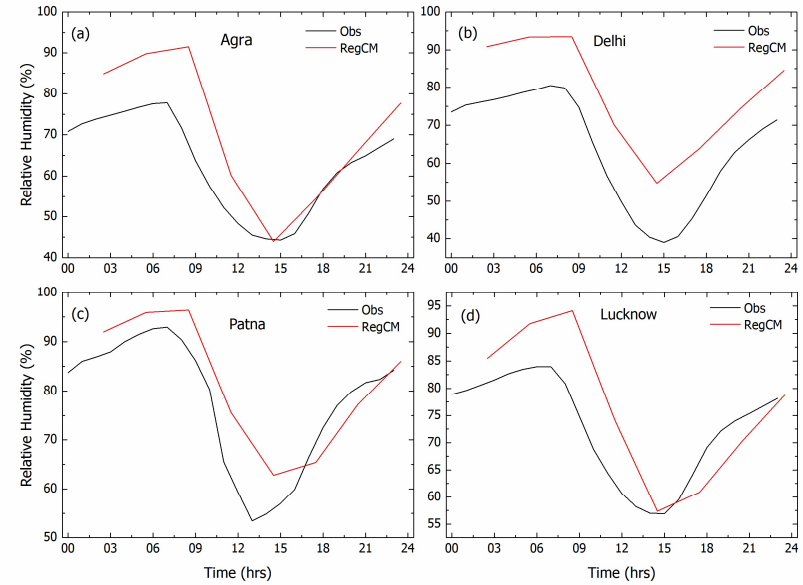

Figure: Diurnal variation of measured (black) and modelled (red) relative humidity at (a) Agra, (b) Delhi, (c) Patna and (d) Lucknow.

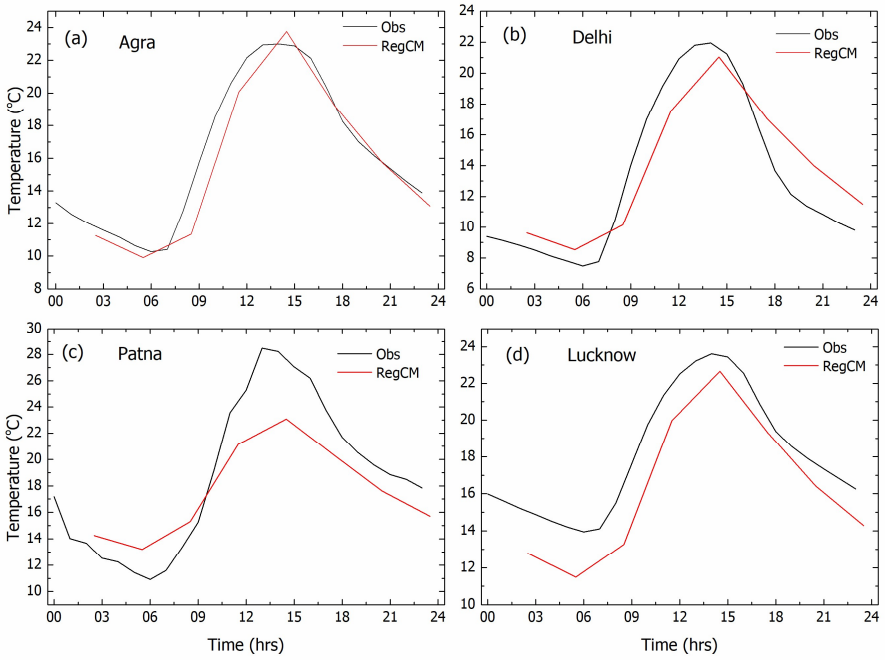

Figure: Diurnal variation of measured (black) and modelled (red) 2m temperature at (a) Agra, (b) Delhi, (c) Patna and (d) Lucknow.

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.

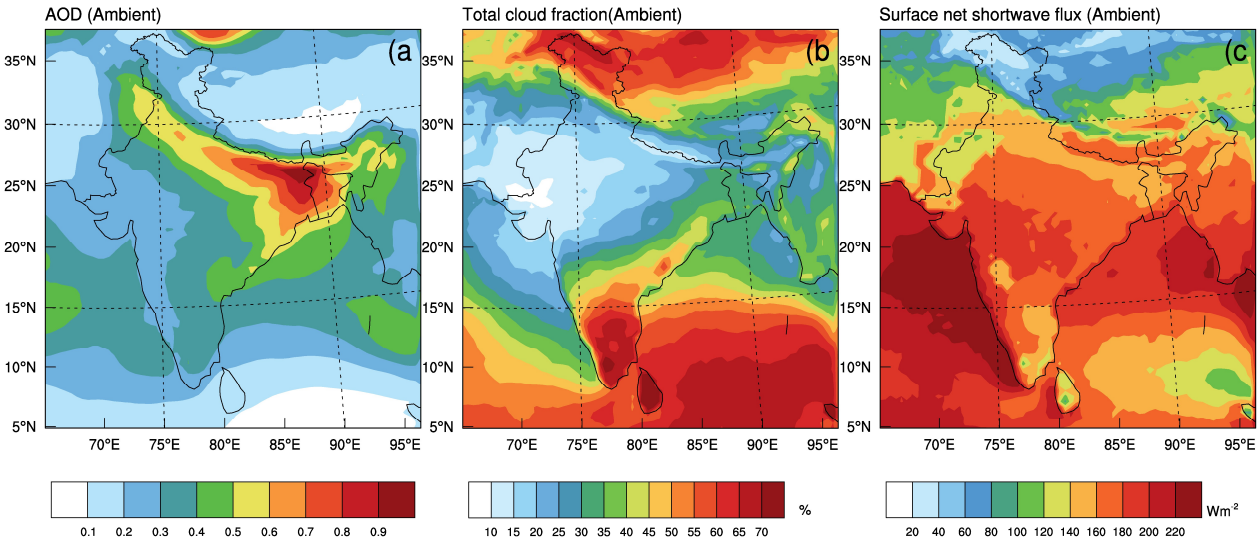

Figure: The spatial variation of (a) AOD, (b) cloud fraction and (c) net shortwave flux at surface.

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 PM2.5 response will depend largely on how the PBL height changes and how aqueous-phase production of SO4 will change. Can you add some discussion on these points?

As reviewer correctly pointed out, the variation in PM2.5 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 PM2.5 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.

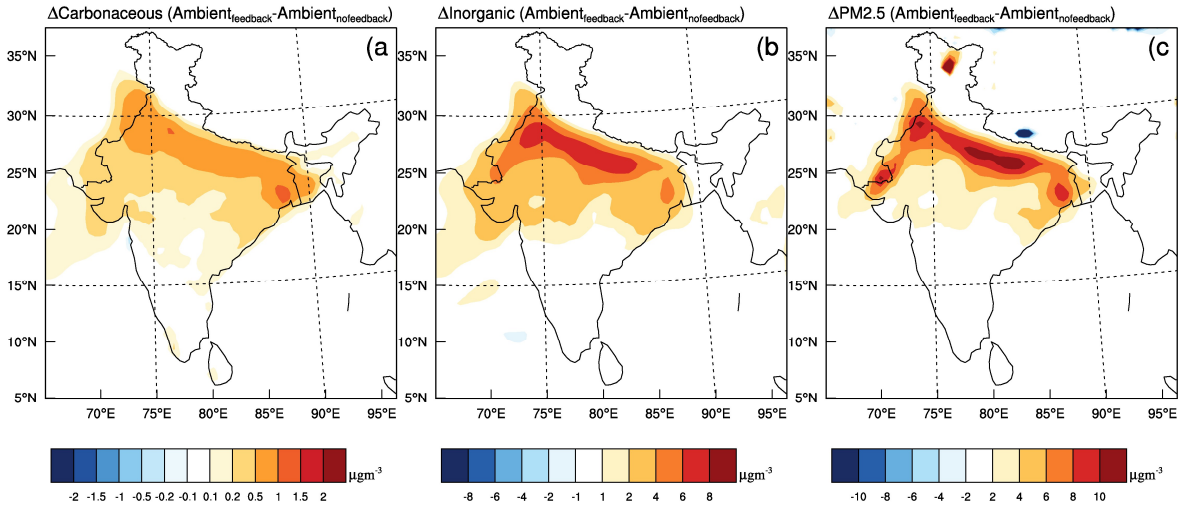

Figure: Change in near surface mass loading (μg m^{-3}) due to the meteorological feedback of total aerosol radiative forcing for (a) carbonaceous aerosols, (b) inorganic aerosols and (c) PM2.5.