## **Response to Reviewer 2**

1. After reading a manuscript, it is unclear whether the authors are arguing that replacing the emission inventory or dynamical aging scheme or combined effect causes a better representation of carbonaceous aerosols in the RegCM4.6. It will be more appropriate to present or at least discuss (a) how the use of new inventory to represent carbonaceous aerosols improved the simulations? (b) How did implementing the dynamical aging scheme improve aerosols' representation with default emission inventory and (c) the combined effect? It is recommended that authors design the experiments to address the concerns above.

**<u>Response</u>**: We thank the reviewer for detailed comments. However, we want to mention that the experiments designed for the manuscript are already as per the suggestion by the reviewer in the section 1 (lines 85-90):

"We carry out four sets of simulations for the year 2010 - (1) control simulation with the default (fixed) ageing scheme and global inventory (hereafter Default\_Sc), (2) simulation with the dynamic ageing scheme and global inventory (Dyn\_global), (3) simulation with the default ageing scheme and regional inventory (Fix\_Regio) and (4) simulation with the dynamic ageing scheme and regional emission inventory (Dyn\_Regio)."

We did not include these intermediate results in our earlier version and our main message was that climate models require both emission and aerosol processes to be updated in order to improve their performances. However, as pointed out by this reviewer and other reviewers, we agree that it is important to show these intermediate results (now shown in supplementary figures 1, 6 and 7) and discuss them.



Figure S1.a Spatial patterns of mean seasonal surface BC concentration ( $\mu$ g/m3) over India (1st column) using the default set-up and percentage differences in the (2nd and 3rd columns) modified and (4th column customized configurations relative to the default set-up.



Figure S1.b Spatial patterns of mean seasonal surface OC concentration ( $\mu$ g/m3) over India (1st column) using the default set-up and percentage differences in the (2nd and 3rd columns) modified and (4th column customized configurations relative to the default set-up.



Figure S6. Seasonal variation of vertically distributed mass concentration ( $\mu g/m3$ ) of BC over the highly polluted Indo-Gangetic Plain



Figure S7. Seasonal variation of vertically distributed mass concentration ( $\mu$ g/m3) of OC over the highly polluted Indo-Gangetic Plain

2. From Figures 2 and 3, it is evident that both default and changed models underestimate the surface BC compared to in-situ observations even though the augmented model has better skill than the default one. It is not clear that the better representation of the surface of aerosols (Figure 1) causes more concentration of BC due to convection (vertical mass flux) or lateral advection at higher levels from sources at higher elevations in the IGB region (Fig 4) and why there are no appreciable changes seen over PI region. The in-depth analysis is required to quantify the changes in the vertical of the results in terms of changes in the vertical mass fluxes and vertical velocity vs. mass advection simulated in the model to quantify the impact of on distribution of aerosols.

**<u>Response</u>**: The augmented model displayed better skill in simulating surface BC over IGB. This is due to the dual role of higher convective tendency and lower lateral advection and vice-versa depending on the season.

Convective tendency is an important indicator for upper air transport of aerosols. More positive values indicate strong updraft above the surface due to convection. Convection tendency gradually increases from left to right (Figure 2.1). Particularly, in the drier seasons since more particles are available in absence of washout. During winter, augmented model (Dyn\_regio) is showing lesser pumping effect over IGB than that when only emissions have been changed (Fix\_regio). This can be due to the fact that, in presence of dynamic ageing a greater number of hydrophilic tracers are available for removal (evident from the removal plot of BC\_HL) even for small amount of precipitation from western disturbances. However, during post-monsoon (OND), due to negligible precipitation over IGB, removal rates of hydrophilic tracers are comparable and hence the pumping effect also follows the same trend. Similar trend in convective tendency is also shown by OC particles (Figure 2.2). The magnitude of OC convection tendency is stronger than that of BC

particles. This can be due to the higher concentration of available particles. Figures for BC convective tendency and lateral advection can be added in the main revised manuscript and that for OC can be added in the revised supplementary document.



Figure 2.1: Seasonal distribution of convective tendency (kg/kg/sec) of BC over IGB for four distinct experiments.



Figure 2.2: Seasonal distribution of convective tendency (kg/kg/sec) of OC over IGB for four distinct experiments.

Lateral advection on the other hand is an indicator of horizontal long-range aerosol transport. More positive values indicate strong flow along the surface due to advection. Advection shows strong seasonality (from top to bottom – Figure 2.3). In drier months (JF and OND) horizontal transport is comparatively less than pre-monsoon (MAM) and monsoon (JJAS). Therefore, vertical convection is more prominent in dry seasons while horizontal advection is dominant for MAM and JJAS, irrespective of the choice of schemes. Consequently, the observed BC concentration is due to convection in JF and OND and due to advection in MAM and JJAS. Same logic can be applied for OC concentration distribution due to lateral advection (Figure 2.4). However, the positive advection signal is stronger than that of BC particles. This can be again due to the higher concentration of available particles for transport to other regions.



Figure 2.3: Seasonal distribution of lateral advection (kg/kg/sec) of BC over IGB for four distinct experiments.



Figure 2.4: Seasonal distribution of lateral advection (kg/kg/sec) of OC over IGB for four distinct experiments.

Over PI, the lower concentration can be primarily because of the lower emissions for both BC and OC. Convective tendency as well as lateral advection for BC is not playing any major role (as can be seen in Figure 2.5 and 2.7), hence concluding the role of lower emissions. In case of OC, lateral advection (Figure 2.6 and 2.8) and comparatively lower emissions (Figure in the actual supplementary document) than IGB can be the predominant factors for lower concentration over PI in presence of negative convective tendency.





Figure 2.5: Seasonal distribution of convective tendency (kg/kg/sec) of BC over PI for four distinct experiments.

Figure 2.6: Seasonal distribution of convective tendency (kg/kg/sec) of OC over PI for four distinct experiments.



Figure 2.7: Seasonal distribution of lateral advection (kg/kg/sec) of BC over PI for four distinct experiments.



Figure 2.8: Seasonal distribution of lateral advection (kg/kg/sec) of OC over PI for four distinct experiments.

3. The result in Lines 371-3, "Due to the model improvements (forcing estimates 371 with the default model are shown in Figure S8), the TOA forcing changes by -72.75%, and the 372 surface dimming increases by 39.73% over the IGP and by -23.94% and 34.35%, respectively," should be cross-checked with the amount of clouding simulated model and reflections from clouds at TOA due to them vs. the effect of surface dimming as mentioned in the manuscript to be sure. Alternatively, these differences in the simulations can be attributed to the amount of cloudiness simulated (secondary effects) by default and the augmented model.

**<u>Response</u>**: Currently the model does not assume aerosol interaction with clouds, therefore the radiative feedback is mainly governed by direct radiative forcing. Therefore, secondary effects due to aerosols cannot be considered for the observed values. Since the model does not consider aerosolcloud interactions, it is explicitly mentioned in the manuscript. However, the amount of cloudiness simulated by the model in four distinct experiments is given below. It can be seen that the model is able to capture the seasonal variability of total cloud cover for each of the four experiments. The amount of cloud cover is maximum during monsoon. The cloud cover is low during other months. Thus, the reflection from clouds will also be lower. As a result, contribution to the observed anthropogenic AOD due to cloud reflections (in supplementary figure S8) will also be lower. Therefore, AAOD distribution over IGB is primarily responsible for the surface dimming effect and the resulting atmospheric heating.



Figure 2.9: Seasonal distribution of total cloud fraction (in %) for four distinct experiments.

4. The manuscript will be more readable if the same terminology is used in the revision to specify model setup (augmented model or customized setup).

*Response:* The terminology has been updated to "augmented model" throughout the manuscript as per the suggestion.

5. Line 402-403 "Our work demonstrates that even the improvement of some aspects of the aerosol representation can lead to substantial enhancements in the model performance." The sentence requires to be rewritten with more quantification and elaboration.

Response: In the supplementary figure S3, the quantification of model performance has been already shown. At most of the in-situ sites out of 24, only dynamic ageing implementation resulted in 5-10 % improvement. But when both regional emissions along-with ageing is implemented, the model representation of BC surface concentration ( $\mu$ g/m3) increased by 60-120%, particularly for polluted sites like Patiala, Kanpur, Varanasi.



Figure S3. Locations of the 24 cities where BC concentrations were measured during the study period and used to evaluate the customized model performance. The colour of the circles indicates the percentage increase in BC concentrations due to the implementation of the dynamic scheme and the size of the circles indicate the percentage increase in BC concentrations due to the combined impact of ageing scheme and regional inventory in the customized model.

6. More justification is needed to conclude that " a dynamic aging scheme and a regional emission inventory substantially improve the model performance over the Indian subcontinent." and "The BC and OC surface concentration and column burden increase due to the model improvements, more so as a combined effect of the two factors than because of the individual ones."

Response: The following figure further justifies the conclusion: " a dynamic aging scheme and a regional emission inventory substantially improve the model performance over the Indian subcontinent." In each of the season particularly in winter (63.54%), the mean BC burden in 2.10(a) is showing maximum improvement for Dyn\_reg experiment w.r.t to default. Similar, increments are visible for BC surface concentration as well (winter is showing maximum change of 61.46%).



Figure 2.10: Seasonal distribution of % change of (a) BC burden (mg/m2) and (b) BC surface concentration ( $\mu$ g/m3) for each sensitivity experiment w.r.t the default set-up where De = Default, DG = Dyn\_global, FR = Fix\_reg and DR = Dyn\_reg.