Response to Reviewer 1

"A set of sensitivity studies from a changes to emissions and the ageing factor (conversion rate of hydrophobic to hydrophilic) were presented using RegCM4.6 for BC and organic carbon aerosols over India. This was shown to produce better results and reduce the bias often by factors of two or more over much of India. Most of the discussion and improvements in model performance are presented in terms of default simulations and the soc-called dynamic regional, that includes adjustments to both the emissions and ageing. It is thus hard to understand if most if the improvements are due to emissions or due to changes in the ageing the authors are aware there are numerous papers on sensitivity of the aerosol burden over India to emissions and that is not worth another paper as it adds no new information beyond what we already know."

Response: We thank the reviewer for detailed comments. In this work, we updated the emission and the ageing scheme in RegCM and examined the model improvement. Our simulations also examined the changes in model outputs just because of change in emission and just because of change in ageing scheme. 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). We want to add that this is not just sensitivity study, rather it implements a new ageing scheme in RegCM, and then use the modified model to simulate with better emission estimates to understand how the model performance behaves.

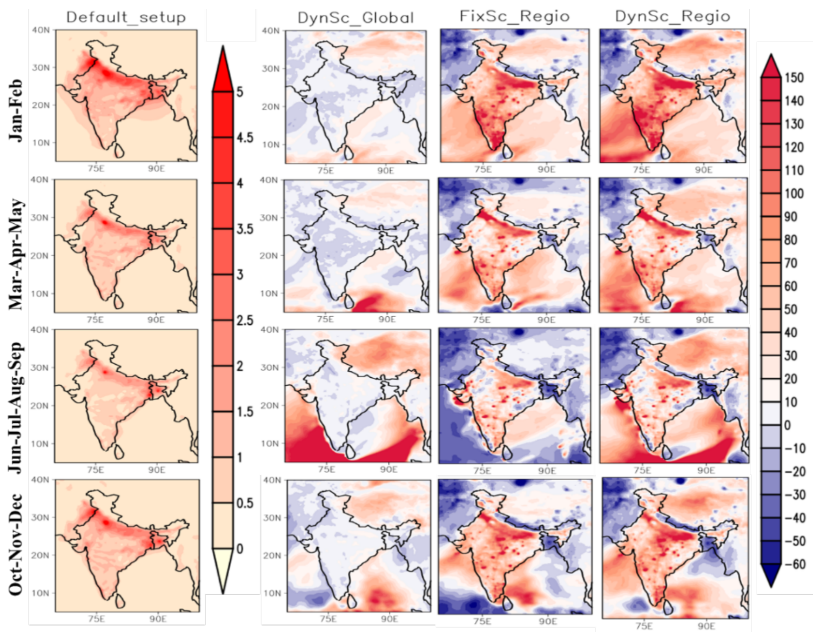


Figure S1.a Spatial patterns of mean seasonal surface BC concentration (µg/m3) over India (1st column) using the default set-up and percentage differences in the (2nd and 3rd columns) modified and (4th column customized configurations relative to the default set-up.

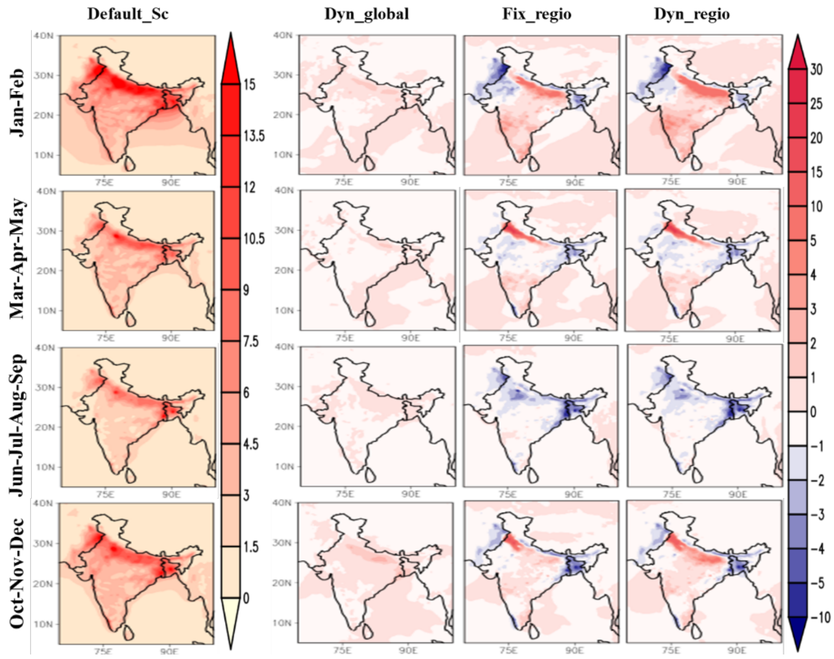


Figure S1.b Spatial patterns of mean seasonal surface BC concentration (µg/m3) over India (1st column) using the default set-up and percentage differences in the (2nd and 3rd columns) modified and (4th column customized configurations relative to the default set-up.

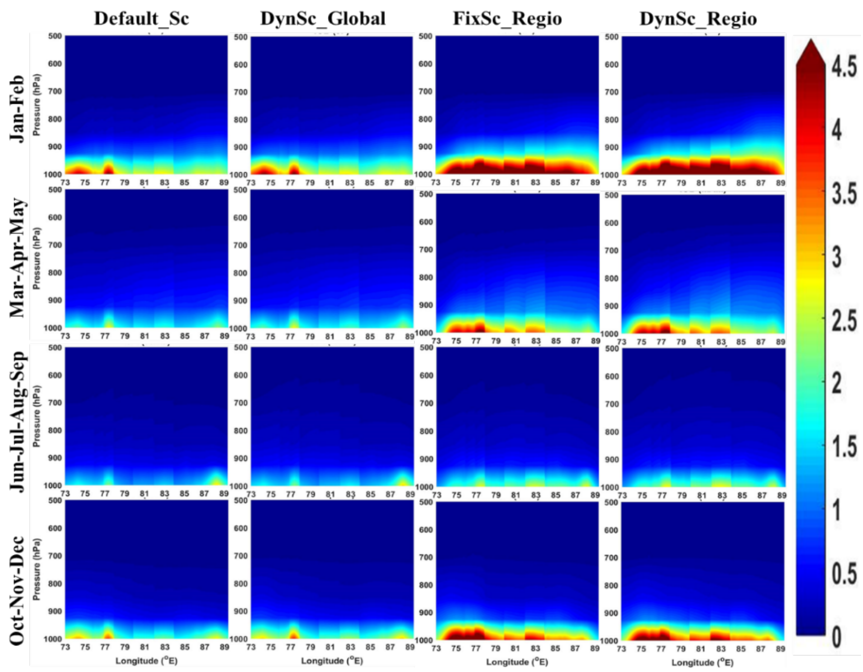


Figure S6. Seasonal variation of vertically distributed mass concentration (µg/m3) of BC over the highly polluted Indo-Gangetic Plain

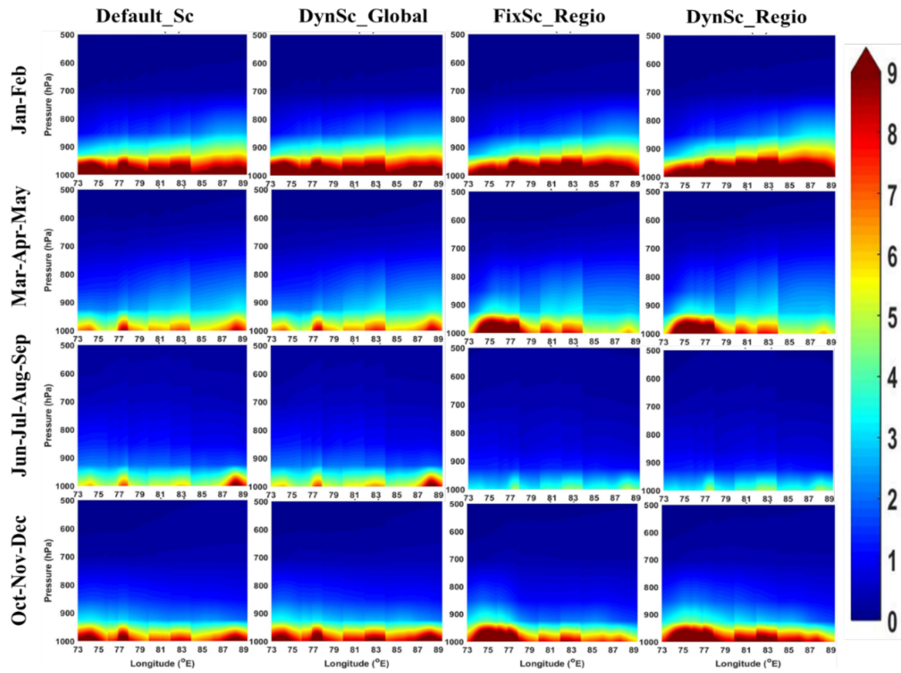
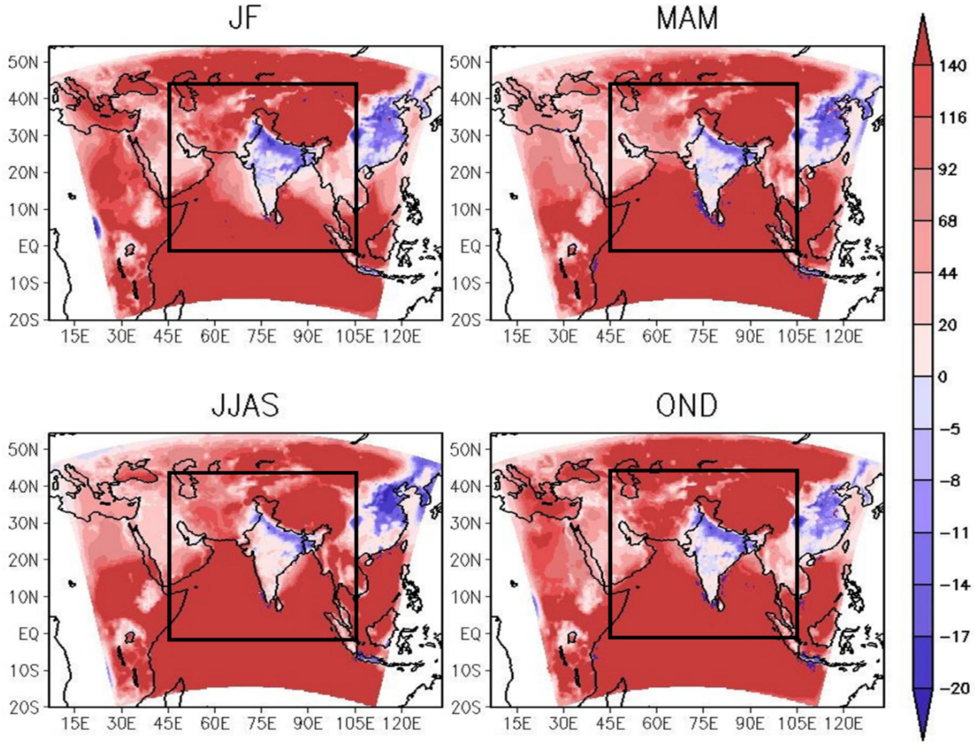


Figure S7. Seasonal variation of vertically distributed mass concentration (µg/m3) of OC over the highly polluted Indo-Gangetic Plain

1. **"The ageing changes should change the ratio of dry deposition/wet deposition in the model and later the ageing the lifetime of aerosols in the model. There are no figures in the paper that show the deposition fluxes and their changes and I am left to wonder if that was even significant in the results? As the primary idea seems to be that this will change the lifetime of BC and OC in the model, why are there no calculations of lifetime of particles regionally, seasonally or annually?"**

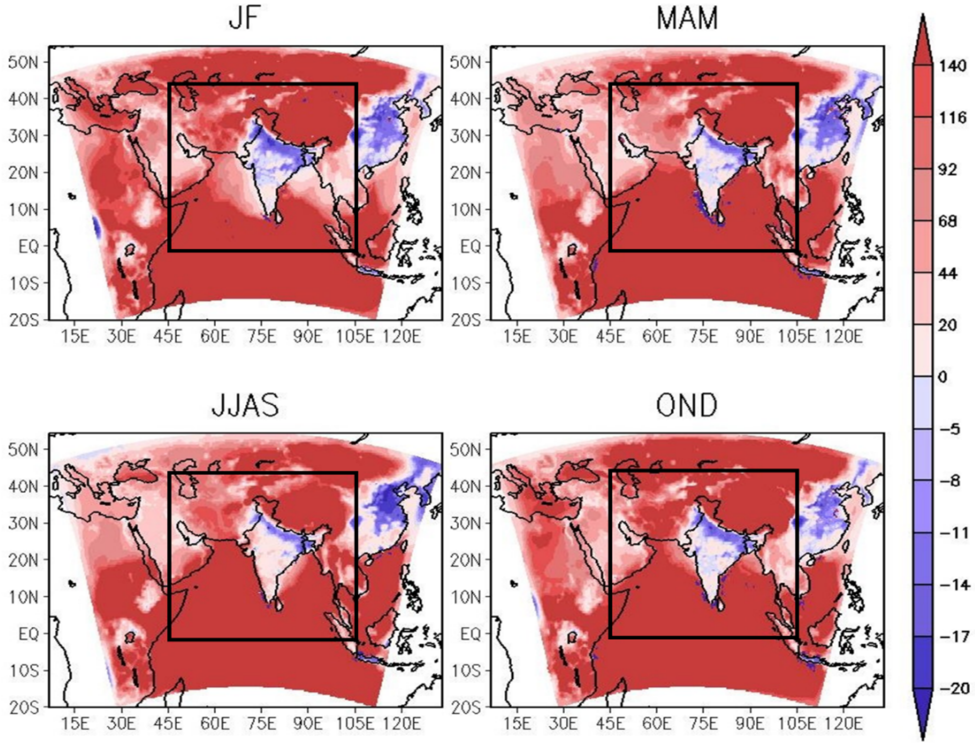
Response: The ageing does change the dry and wet deposition of the tracers and due to this change the ratio of hydrophobic to hydrophilic changes which in turn is altering the atmospheric lifetime of aerosols. The detailed explanation regarding these changes due to ageing alone can be referred from Ghosh et al. 2021. Seasonal variation of lifetime of particles, at the surface and upper atmosphere, due to ageing alone have been already explained in Ghosh et al. 2021.



Seasonal variation of ageing time-scale anomaly (in h) of carbonaceous aerosols at 1000 hPa w.r.t the fixed ageing time-scale of 27.6 h [1.15 day]. The upper level of the colour scale bar has been capped at 140 h (i.e. the aging time scale is (27.6+140) h =167.6 h or 7 days) and the lower limit has been capped at -20 h (i.e. the aging time scale is (27.6 – 20) h = 7.6 h). The figure shows the complete domain of simulation, and the area within the black rectangle represents the study domain. (Ghosh et al. 2021, JGR)

2. **"Are there any changes because of the lifetime changes in particle fluxes over to Indian Ocean that varies by season?"**

Response: Since the focus of this manuscript is Indian landmass only, the changes in aerosol properties over the oceans has not been discussed. The oceanic condition is mostly clean with low concentration of tracers, compared to that over the landmass. There are hardly any emissions over the oceans, shown in the emission plots (supplementary material Fig S2). In Ghosh et al. 2021 it is evident that the ageing time of the carbonaceous aerosols over the oceans is larger than the default ageing timescale.



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3. **"There are only 17 levels in the models, not sure how many of them are in the boundary layer?"**

Response: There are 3 levels (1000, 925, 850 hPa) within the boundary layer and will be added in the revised manuscript.

4. **"How well does the model simulate convection and mixing over the Ganges valley and central plains where the transport to above PBL could be a major factor in increasing the long-range transport of aerosols and hence their lifetimes?"**

Response: The model simulated convective tendency and lateral advection (responsible for long-range transport) are given below. More positive values indicate strong updraft above the surface due to convection. Convection tendency gradually increases from left to right (Figure 1.1). 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 1.2). The magnitude of OC convection tendency is stronger than that of BC particles. This can be due to the higher concentration of available particles.

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 1.3). 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 1.4). 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. 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

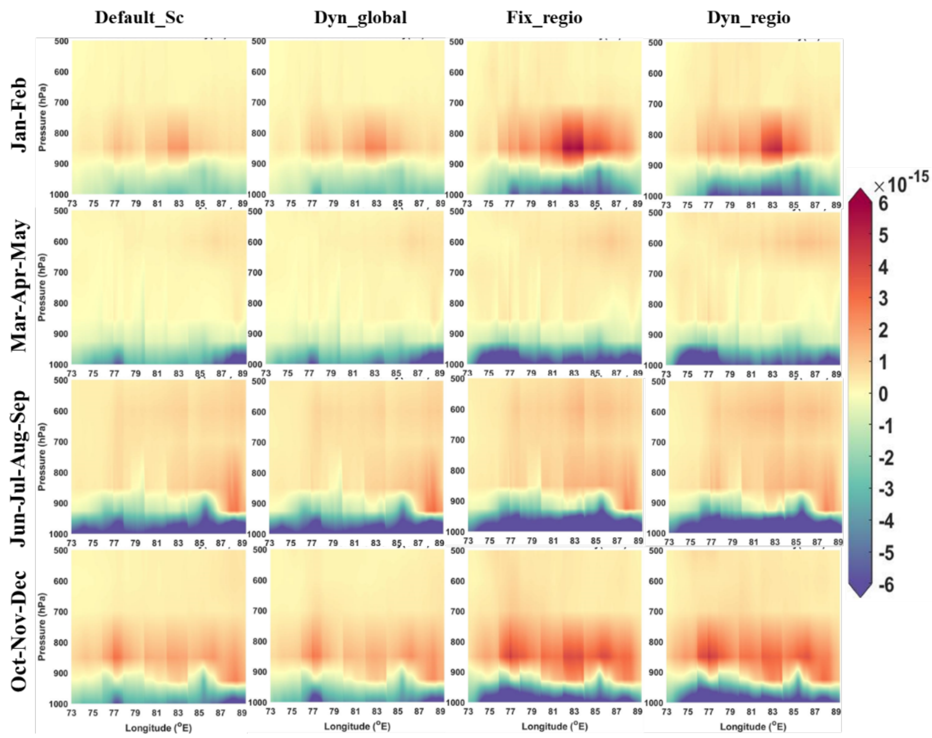

Figure: 1.1: Seasonal distribution of convective tendency (kg/kg/sec) of BC over IGB for four distinct experiments.

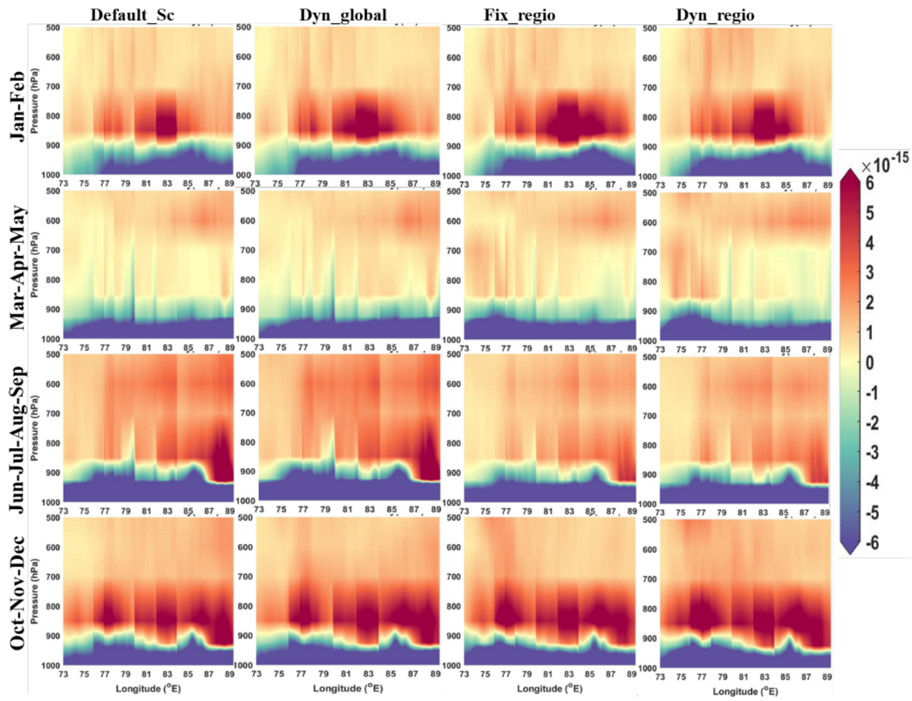

Figure: 1.2: Seasonal distribution of convective tendency (kg/kg/sec) of OC over IGB for four distinct experiments.

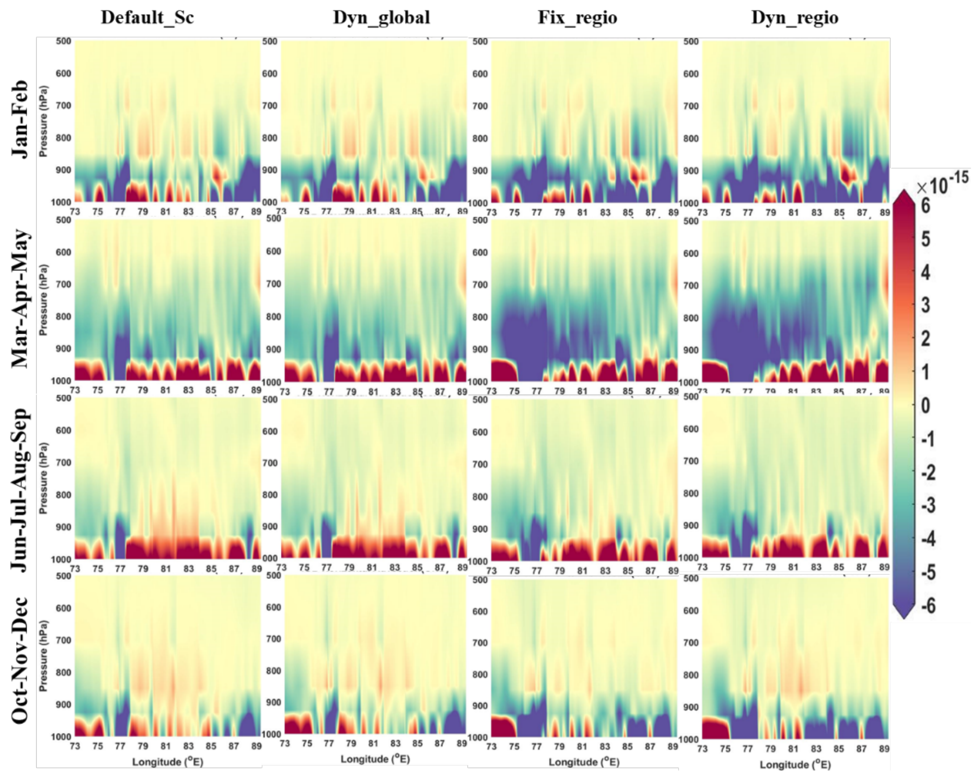

Figure: 1.3: Seasonal distribution of lateral advection (kg/kg/sec) of BC over IGB for four distinct experiments.

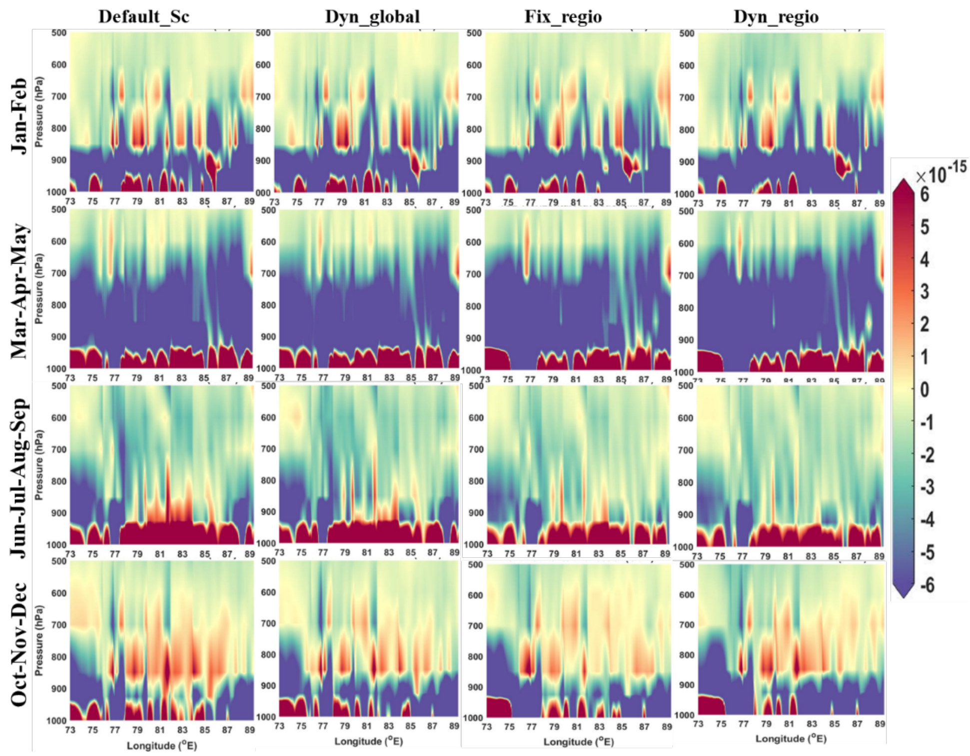

Figure: 1.3: Seasonal distribution of lateral advection (kg/kg/sec) of OC over IGB for four distinct experiments.

5. **"How well does the model simulate column water depths and hence removal rates through wet deposition in the model?"**

Response: The column water depths are not stored as a model output and hence not included in the response document. The wet removal process of the model has been already explained in the submitted manuscript in Section 2.1 (line 120-129):

"Wet deposition in the RegCM4 has been split into "in-cloud" and "below-cloud" terms. The in-cloud removal process starts for large scale clouds if the liquid water is higher than the threshold level (0.01 g m^{-3}) in the model layers where the cloud fraction is more than zero and is a function of the fractional removal rate of liquid water (fraction of precipitating rain over liquid water content of the atmospheric layer, the in-cloud removal rate for cumulus clouds is constant and fixed at 0.001 s^{-1}) and the aerosol solubility. This solubility is different for different species, and thus hydrophilic, and hydrophobic BC/OC have different in-cloud wet deposition rates. The below-cloud washing out of the aerosols is controlled by their effective diameters and densities. Collection efficiency for each aerosol species is computed from the aerosol effective diameter and density, which is different for different species."

Since observation measurements of wet removal are not available, direct comparison to quantify the removal is not possible. The model simulated seasonal distribution of BC_HB and BC_HL removal rates are given below for distinct four experiments and can be added in the revised supplementary document.

It can be observed that for BC_HB, maximum removal can be observed by wet removal during JJAS for all the four experiments with varying magnitudes (highest being in Expt Dyn_reg). During drier months, BC_HB dry removal is observed to be most prominent in Expt Fix_reg and Dyn_reg and clearly due to more emissions. Because in Expt Dyn_global dry removal is lowered due to more conversion to HL (Ghosh et al., 2021). Next in case of BC_HL, removal is highest during JJAS.

However, during drier months, both wet deposition and dry deposition are showing signals due to higher availability. This availability results either due to faster conversion or higher emission or the combined effect. For dry removal process, it is due to the negligible precipitation. But. for wet removal it is due to the fact that even for small amount of precipitation more particles are available for removal. Similar results are available for OC as well but not shown here.

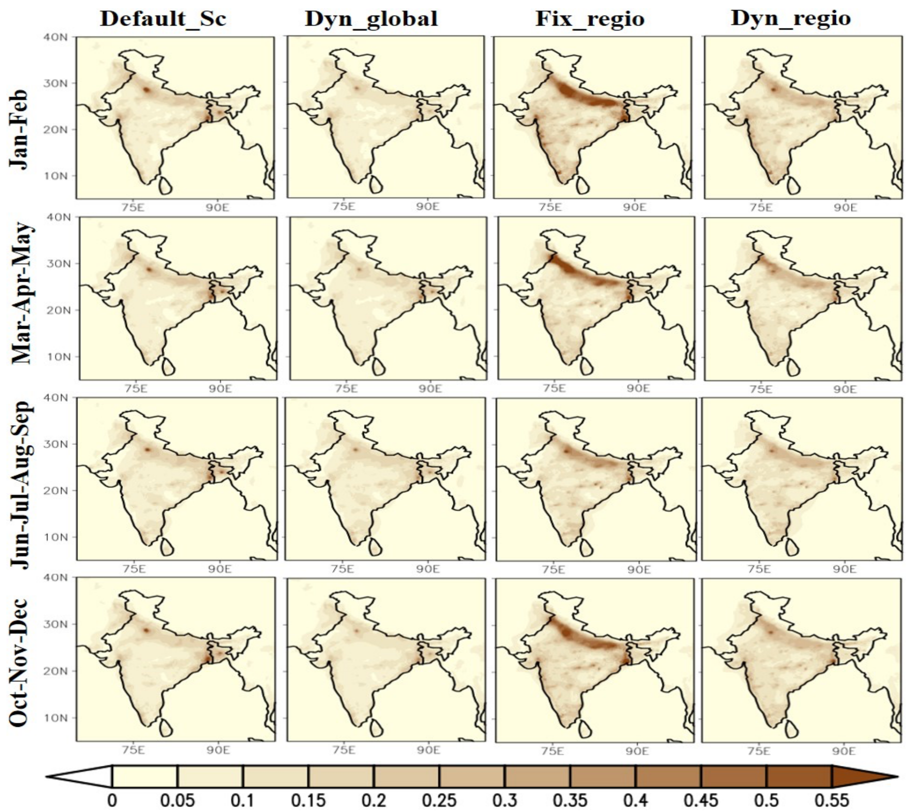

Figure 1.4: Seasonal distribution of BC_HB dry removal for four distinct experiments

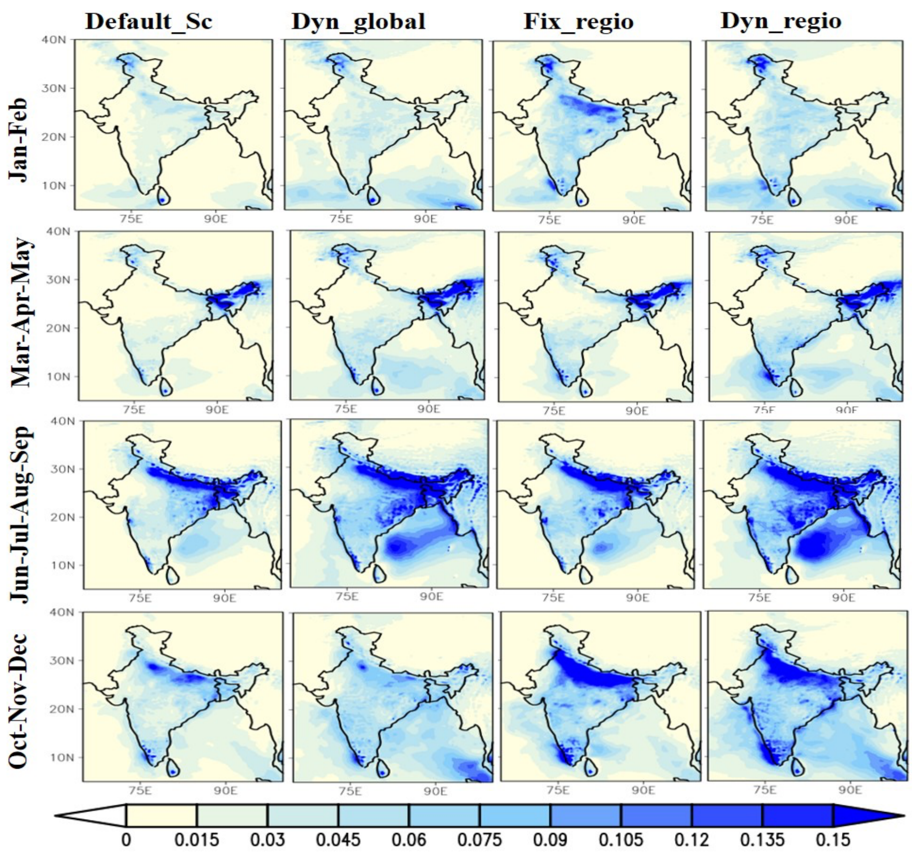

Figure 1.5: Seasonal distribution of BC_HB wet removal for four distinct experiments

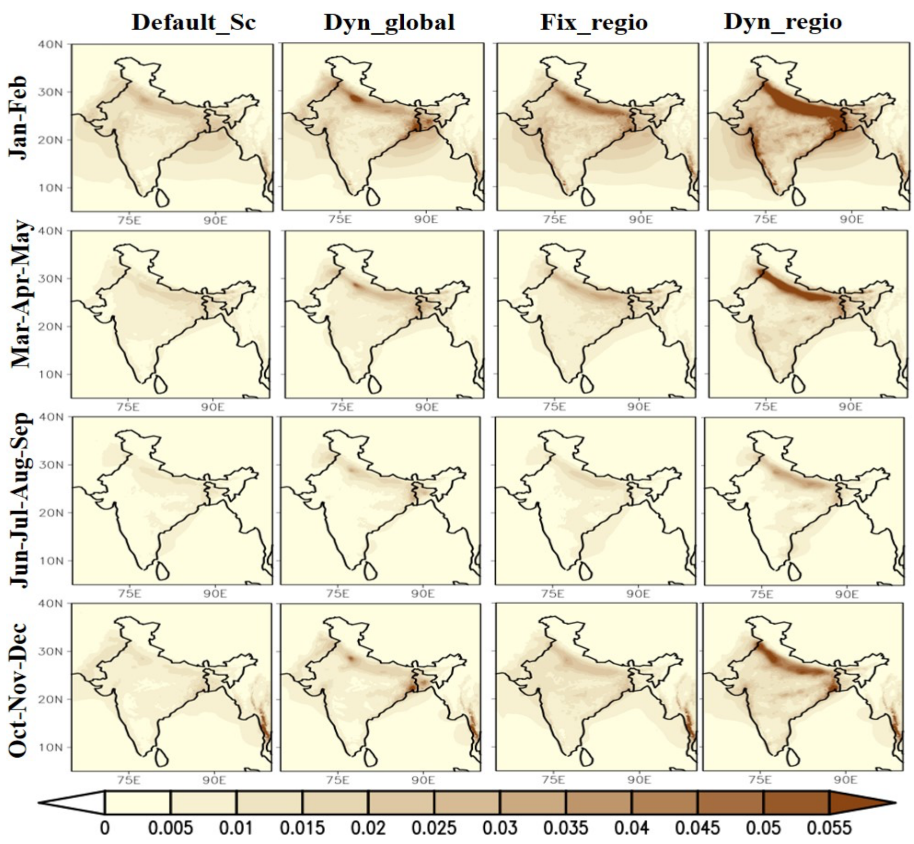

Figure 1.6: Seasonal distribution of BC_HL dry removal for four distinct experiments

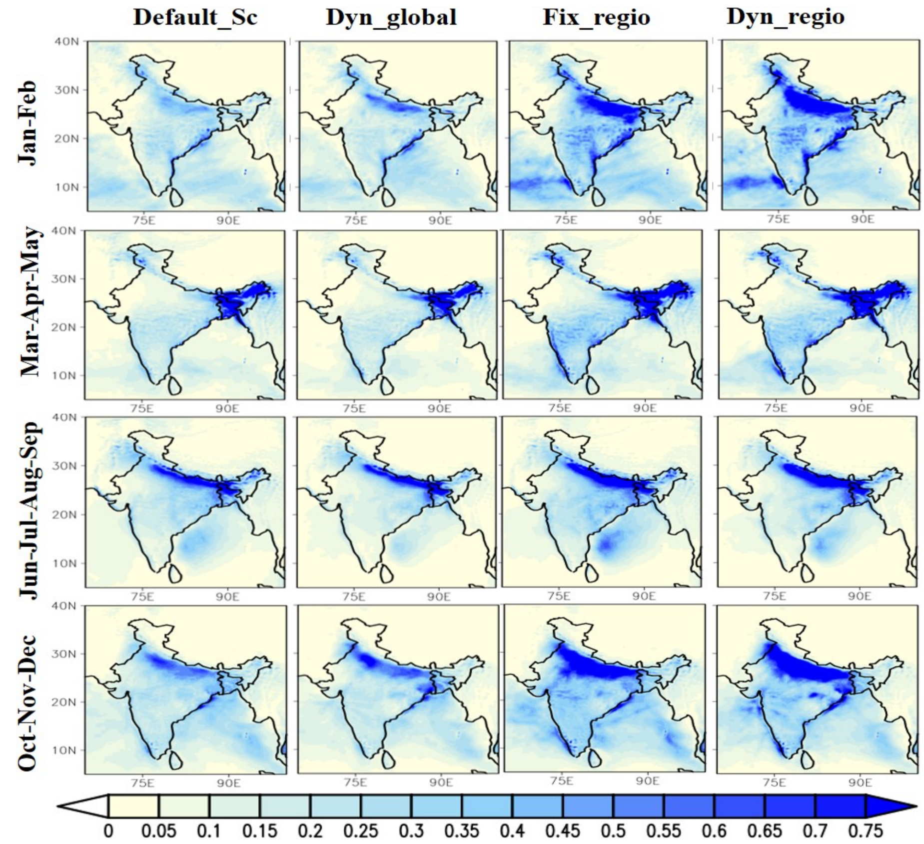

Figure 1.7: Seasonal distribution of BC_HL wet removal for four distinct experiments

6. **"The radiative forcing calculations are for direct radiative forcing one assumes as there is no discussion of aerosol-cloud interactions in the model. If that is the case, did you separate the clear days from cloudy days to perform these forcing calculations are these are seasonal averages for days with and without clouds?"**

Response: Currently the model does not assume aerosol interaction with clouds, therefore the radiative feedback is mainly governed by direct radiative forcing. Cloud parameters in the model change in response to the radiative feedback. Therefore, we didn't separate clear and cloudy days. This limitation will be explicitly mentioned in the revised manuscript.

7. **"How well does the model represent RH in the vertical column?"**

Response: The model is able to capture the both seasonality as well as vertical profile of relative humidity (RH) in Figure 1.9 and Figure 1.10 over IGB and PI respectively. Relative humidity from ERA-interim dataset at 1.5degree resolution has been incorporated to compare the model simulated RH values (Figure 1.8). During monsoon, the RH is highest (>70%) throughout the troposphere up to 500hPa over both IGB and PI. This is followed by post-monsoon (OND), winter (JF) and pre-monsoon (MAM). During, post monsoon the near surface high RH values over IGB facilitate the fog formation (Dey, 2018; Chakraborty et al., 2016). With gradual decrease in atmospheric moisture, RH values decreases in winter and is mainly dominated by the one brought in by western disturbance over IGB. Finally, the lowest values are captured during MAM. Over PI, high RH values have been captured throughout the year with maximum during monsoon (JJAS) followed by post-monsoon. The RH values during monsoon is due to the south-west monsoon and for post-monsoon it is the north-east monsoon. The following figures can be incorporated in the revised supplementary document.

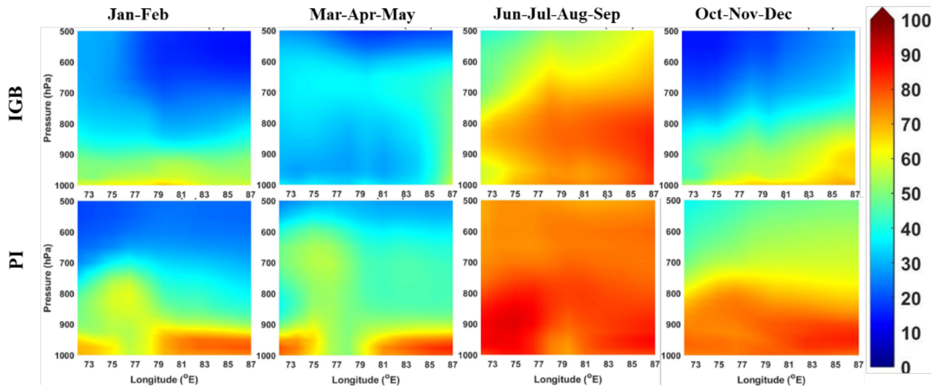

Figure 1.8: Seasonal distribution of relative humidity (%RH) from ERA-interim dataset over Indo-Gangetic Basin (IGB) and Peninsular India (PI)

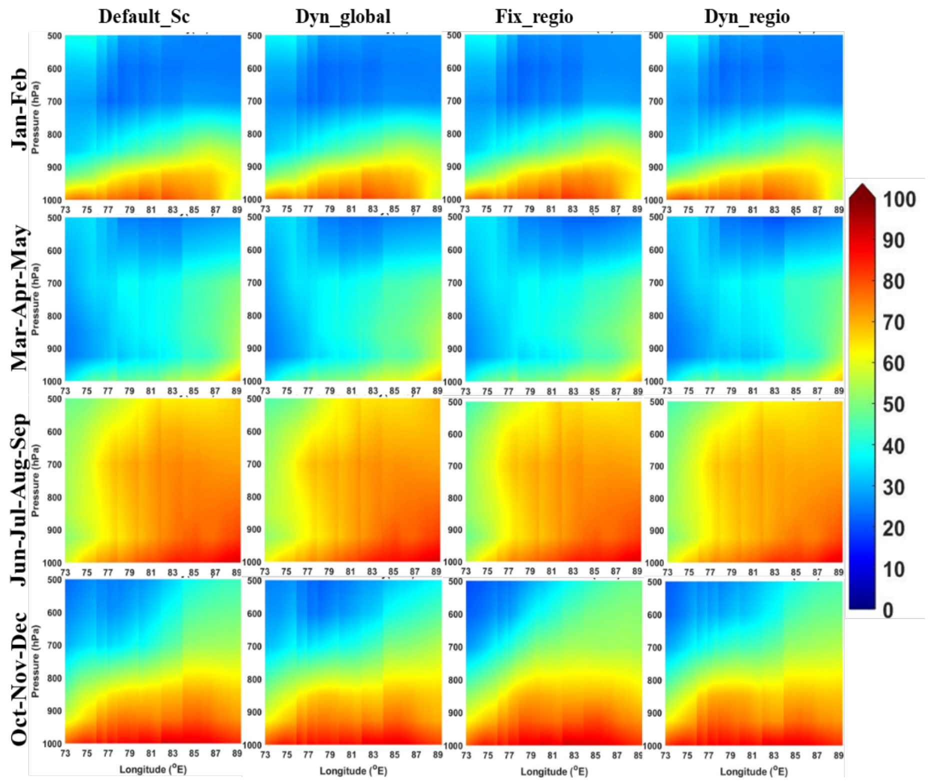

Figure 1.9: Seasonal distribution of relative humidity (%RH) over Indo-Gangetic Basin (IGB) for the four distinct experiments.

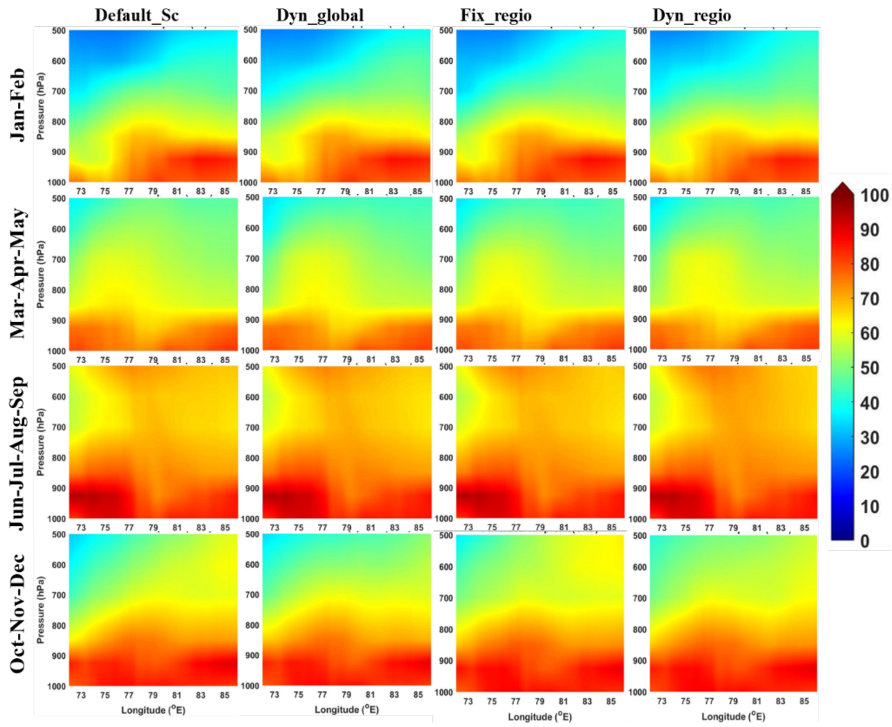

Figure 1.10: Seasonal distribution of relative humidity (%RH) over Peninsular India (PI) for the four distinct experiments.

8. "How much effect does the change in aerosol burden in the column have on the atmospheric profiles and how much of this contributes to changes in optical properties and hence forcing?"

Response: The relative humidity (Figure 1.9 for IGB and Figure 1.10 for PI), temperature (Figure 1.11 for IGB and Figure 1.12 for PI) and vertical wind (Figure 1.13 for IGB and Figure 1.14 for PI) profiles have been shown. In terms of change in temperature profile higher temperatures over IGB during MAM and JJAS facilitated the strong vertical wind movement (negative values in Figure 1.13). But negative convective tendency and positive lateral advection of carbonaceous aerosols during these months lowered their concentrations. This is further supported by the high RH values particularly in JJAS which resulted in higher removal. Exactly opposite is happening during the drier months (JF and OND). Comparatively low temperatures, facilitated more stable wind movement (positive values in Figure 1.13). However, in presence of high emissions, the aerosol pumping effect resulted in strong convective tendency which further facilitated the higher concentrations during these months. The low RH values during these are also conducive of higher aerosol atmospheric lifetime.

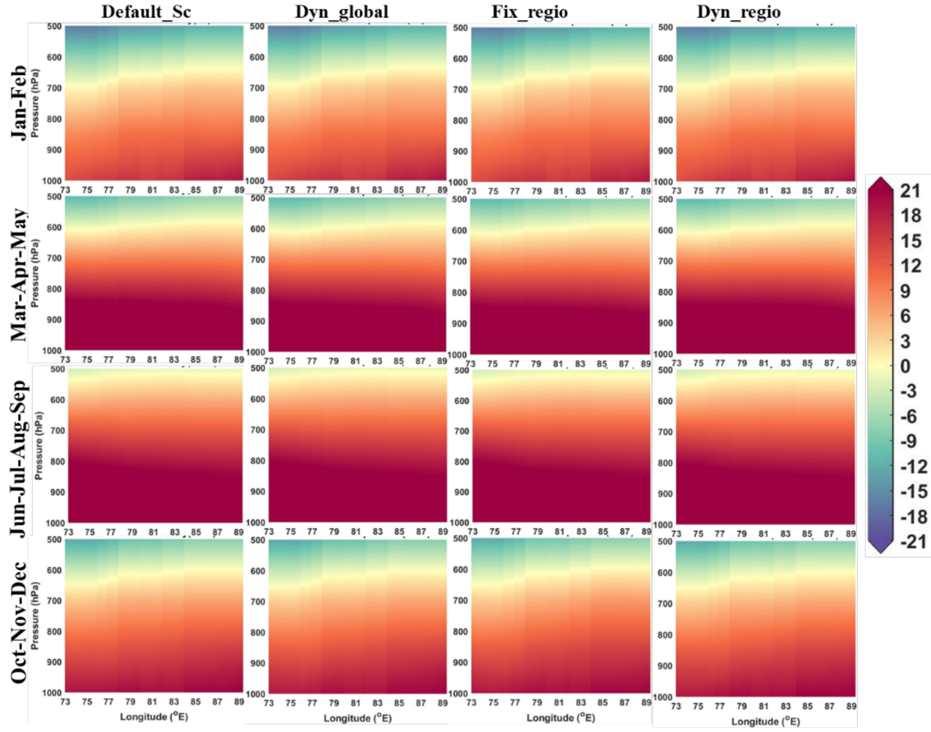

Figure 1.11: Seasonal distribution of temperature (in °C) over IGB for four distinct experiments.

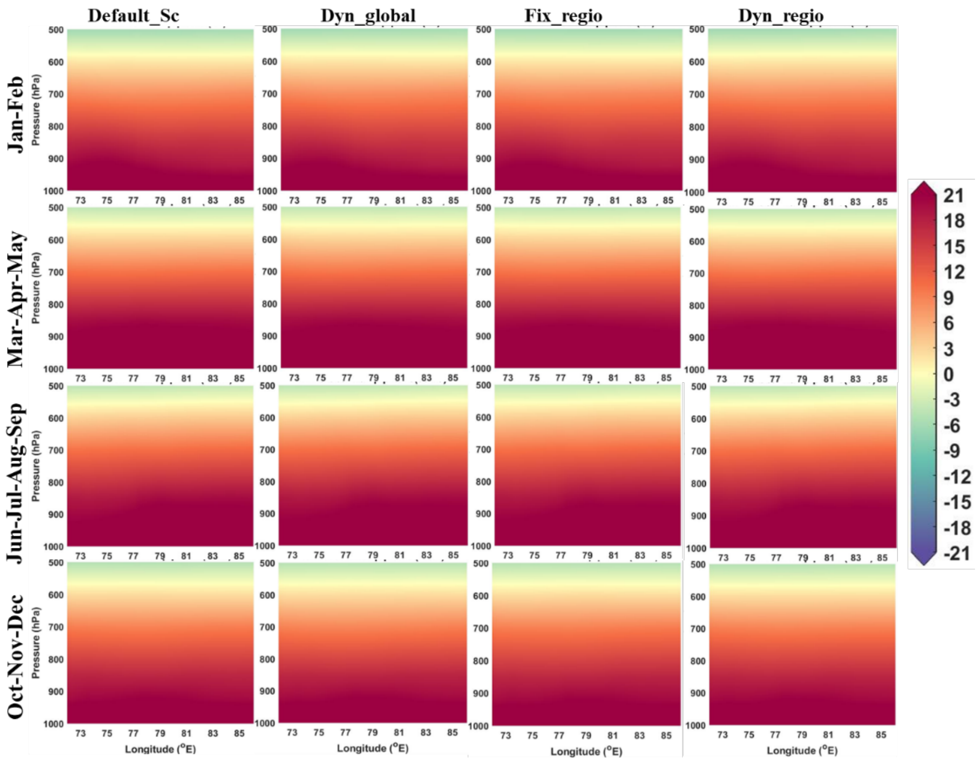

Figure 1.12: Seasonal distribution of temperature (in °C) over PI for four distinct experiments.

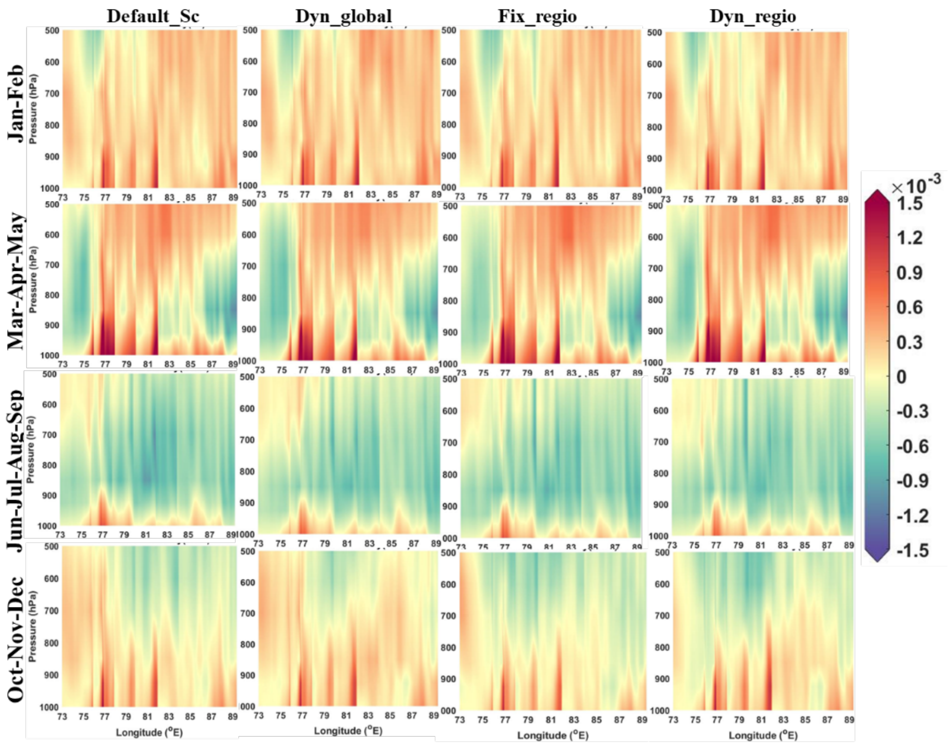

Figure 1.13: Seasonal distribution of vertical wind, omega (in m/sec) over IGB for four distinct experiments.

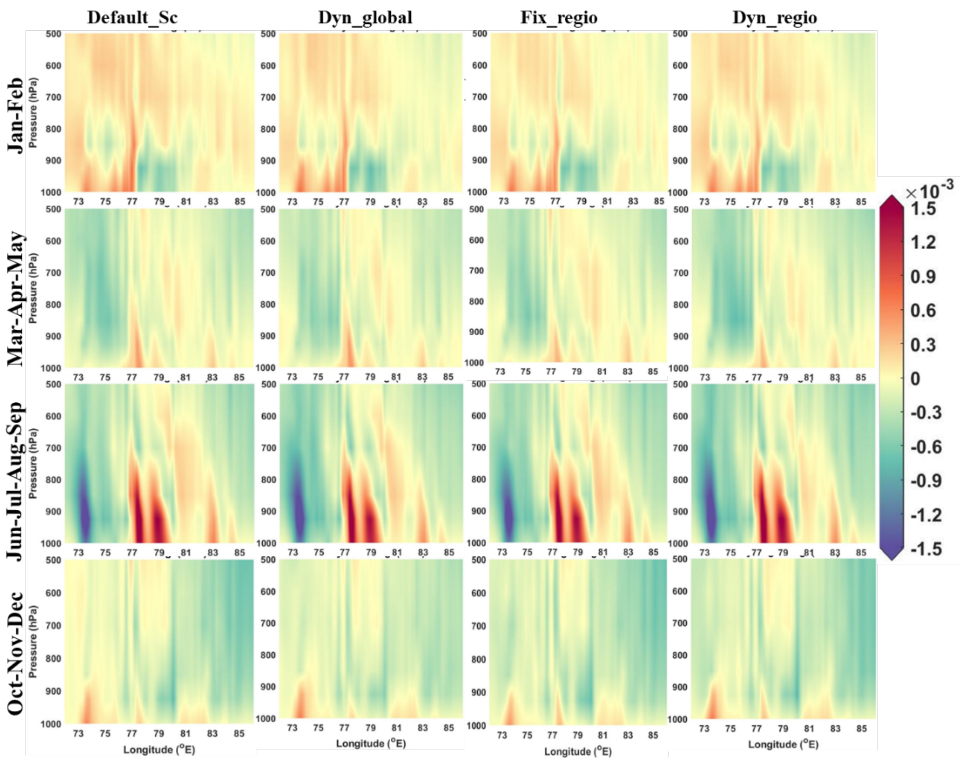

Figure 1.14: Seasonal distribution of vertical wind, omega (in m/sec) over PI for four distinct experiments.

References:

Chakraborty, A., Gupta, T., and Tripathi, S. N.: Combined effects of organic aerosol loading and fog processing on organic aerosols oxidation, composition, and evolution, Science of The Total Environment, 573, 690–698, https://doi.org/10.1016/j.scitotenv.2016.08.156, 2016.

Dey, S.: On the theoretical aspects of improved fog detection and prediction in India, Atmospheric Research, 202, 77–80, https://doi.org/10.1016/j.atmosres.2017.11.018, 2018.

Ghosh, S., Riemer, N., Giuliani, G., Giorgi, F., Ganguly, D., and Dey, S.: Sensitivity of Carbonaceous Aerosol Properties to the Implementation of a Dynamic Aging Parameterization in the Regional Climate Model RegCM, 126, e2020JD033613, https://doi.org/10.1029/2020JD033613, 2021.