



1 **Towards an improved representation of carbonaceous aerosols over the**
2 **Indian monsoon region in a regional climate model RegCM4.6**

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19 **Keywords:** RegCM4; emission inventory; carbonaceous aerosols; model customization;
20 Indian monsoon region

21

22 **Abstract**

23 Mitigation of carbonaceous aerosol emissions is expected to provide climate and health co-
24 benefits. The accurate representation of carbonaceous aerosols in climate models is critical for
25 reducing uncertainties in their climate feedbacks. In this regard, emission fluxes and aerosol
26 life-cycle processes are the two primary sources of uncertainties. Here we demonstrate that
27 incorporating a dynamic ageing scheme and emission estimates that are updated for the local
28 sources improve the representation of carbonaceous aerosols over the Indian monsoon region
29 in a regional climate model, RegCM, compared to its default configuration. The mean BC and
30 OC surface concentrations in 2010 are estimated to be 4.25 and 10.35 $\mu\text{g m}^{-3}$, respectively,
31 over the Indo-Gangetic Plain (IGP), in the augmented model. The BC column burden over the
32 polluted IGP is found to be 2.47 mg m^{-2} , 69.95% higher than in the default model configuration
33 and much closer to available observations. The anthropogenic AOD increases by more than



34 19% over the IGP due to the model enhancement, also leading to a better agreement with
35 observed AOD. The top-of-the-atmosphere, surface, and atmospheric anthropogenic aerosol
36 shortwave radiative forcing are estimated at -0.3 , -9.3 , and 9.0 W m^{-2} , respectively, over the
37 IGP and -0.89 , -5.33 , and 4.44 W m^{-2} , respectively, over Peninsular India. Our results suggest
38 that both the accurate estimates of emission fluxes and a better representation of aerosol
39 processes are required to improve the aerosol life cycle representation in the climate model.

40

41 **1. Introduction**

42 Carbonaceous aerosols (organic carbon, OC, and black carbon, BC) emitted from
43 incomplete combustion constitute 20%-50% of the total global aerosol mass (Kanakidou et al.,
44 2005; Putaud et al., 2010), causing substantial air quality degradation (Singh et al., 2021). Due
45 to their ability to absorb solar radiation, carbonaceous aerosols also contribute to global
46 warming (Ramanathan and Carmichael, 2008). Hence, they are considered to be key short-
47 lived climate pollutants (SLCPs) (UNFCCC, 2015), and mitigating their emissions is expected
48 to result in both climate and health co-benefits (Tibrewal and Venkataraman, 2021). Climate
49 models are characterized by large discrepancies in simulating carbonaceous aerosol loadings,
50 their optical properties, and radiative forcing (Ajay et al., 2019), primarily due to uncertainties
51 in emission inventories and limitations in the treatment of aerosol processes in the models
52 (Bond et al., 2013). Unless the representation of the life cycle of carbonaceous aerosols in
53 climate models is improved, their role in climate impacts and air quality degradation cannot be
54 assessed accurately (Riemer et al., 2019).

55 A multi-institutional network program - Carbonaceous aerosol emissions, source
56 apportionment, and climate impacts (COALESCE) was launched by the Government of India
57 to address some of these issues for the Indian monsoon region (Venkataraman et al., 2020).
58 One of the scientific objectives of COALESCE is to understand and reduce uncertainties in
59 representing carbonaceous aerosol life cycle in global and regional climate models, focusing
60 on the Indian subcontinent. The regional climate model, RegCM4, developed at the
61 International Centre for Theoretical Physics (ICTP), Italy (Giorgi et al., 2012), is one of the
62 participating models in COALESCE. RegCM4 was extensively used to examine variability in
63 the Indian summer monsoon (Dash et al., 2006; Rai et al., 2019), to project climate change over
64 South Asia (Pattanayak et al., 2019), and to elucidate the dynamical impacts of aerosols on the
65 Indian summer monsoon in the present (Das et al., 2015; 2016) and future (Das et al., 2020)
66 climate conditions.



67 The aerosol module in the RegCM4 (Solmon et al., 2006; Zakey et al., 2006) considers
68 various aerosol life cycle processes, such as emission (source), advection, horizontal and
69 vertical diffusion, transport, conversion of hydrophobic to hygroscopic species, and wet and
70 dry deposition (sink). Previous studies (Das et al., 2016) have pointed out that the RegCM4
71 underestimates the anthropogenic aerosol loading over the Indian subcontinent, and therefore,
72 the net aerosol impact over the region is dominated by natural aerosols (Das et al., 2020). We
73 recently implemented a dynamic ageing scheme in the RegCM aerosol module (Ghosh et al.,
74 2021), which converts carbonaceous aerosols from hydrophobic to hygroscopic states based on
75 the aerosol number concentration. Compared to the constant conversion rate of 27.6 hours used
76 in the default version of the model, the scheme allowed a faster conversion in the polluted
77 regions than in the clean areas of the South Asia region. This, in turn, affected the aerosol
78 forcing due to the changes in aerosol loadings induced by the new hydrophobic-to-hygroscopic
79 conversion scheme. It was also found that implementing the dynamic ageing scheme alone is
80 not sufficient to fully improve the model performance and hypothesized that much of the model
81 uncertainty was due to the emission inventory.

82 In this work, we examine the changes in carbonaceous aerosol burden and their impact on
83 the radiation budget over the South Asia region due to the combined impact of the new dynamic
84 ageing scheme and a regional emission inventory (Venkataraman et al., 2018) replacing the
85 global emission inventory used in the default model version. We carry out four sets of
86 simulations for the year 2010 - (1) control simulation with the default (fixed) ageing scheme
87 and global inventory (hereafter Default_Sc), (2) simulation with the dynamic ageing scheme
88 and global inventory (Dyn_global), (3) simulation with the default ageing scheme and regional
89 inventory (Fix_Regio) and (4) simulation with the dynamic ageing scheme and regional
90 emission inventory (Dyn_Regio). We analyse and report the improvements in model
91 performance due to the combined impact of incorporating a better emission inventory and a
92 more realistic ageing scheme relative to the default model configuration and investigate these
93 performance changes in terms of the aerosol processes considered in the model.

94 **2. Data and Methodology**

95 **2.1 Model configuration**

96 In this work, we have used a regional climate model, RegCM version 4. It is a compressible,
97 hydrostatic, and primitive equation model with a sigma-p vertical coordinate and a dynamical
98 core from the NCAR Mesoscale Model Version 5 (MM5)(Grell, 1993). We have used the
99 Community Climate Model Version 3 (CCM3)(Kiehl et al., 1996) radiative transfer scheme
100 along with the changes mentioned in the literature (Giorgi et al., 2012). Interactive modules for



101 both natural (dust and sea salt) (Zakey et al., 2006, 2008) and anthropogenic aerosols (Solmon
102 et al., 2006) are integrated into RegCM4. The model is also coupled with a gas-phase chemistry
103 module (Shalaby et al., 2012); however, for the current study, we have only considered the
104 anthropogenic module (Solmon et al., 2006). The choice of parameterisation schemes for our
105 experiments have been provided in the following table:

106

Land surface processes	Biosphere-Atmosphere Transfer Scheme (BATS) (Dickinson et al., 1993)
Planetary boundary layer	University of Washington (UW) scheme (Grenier and Bretherton, 2001; Bretherton et al., 2004; O'Brien et al., 2012)
Cumulus convection scheme	Emanuel (Emanuel and Rothman, 1999) over land and Tiedtke (Tiedtke, 1989) over the ocean
Large scale cloud and moisture process	SUBEX scheme (Pal et al., 2000)
Aerosol module	SUCA (Solmon et al., 2006)
Emission inventories	IIASA and IIT Bombay 2010

107

108 The model was simulated over the South-Asian CORDEX domain (Giorgi et al., 2009) [20°
109 S - 50° N and 10°-130° E] for the year 2010 at 0.25° × 0.25° resolution, while the results are
110 analysed over the Indian subcontinent [5°- 40° N and 65°-100° E] with special focus on the
111 IGP [Figure 1] and PI [8° - 20° N and 72° - 85° E]. The simulations consist of 18 vertical levels
112 with the model top pressure at 50 hPa. ERA-Interim 6-hourly reanalysis dataset, at 1.5°
113 resolution, has been used to generate the initial and lateral meteorological boundary conditions
114 for this study (Dee et al., 2011). The sea surface temperature was derived from the NOAA
115 Optimum Interpolated weekly 1° × 1° gridded data and the chemical boundary conditions from
116 MOZART 6-hourly data. The model was simulated from October 01, 2009, to December 31,
117 2010. The first three months are considered as spin-up and thus are not included in the analysis.

118 Aerosol atmospheric lifetime is governed by dry and wet deposition. The dry deposition
119 velocity depends on the type of surface, while the dry deposition flux variation varies directly
120 with the tracer concentration at the lowest level of the model (around 30 m above the surface).
121 Wet deposition in the RegCM4 is governed by the "in-cloud" and "below-cloud" terms. When
122 the liquid water content exceeds the threshold level (0.01 g m⁻³) of the model layers where the



123 cloud fraction is more than zero, the in-cloud removal process starts for large-scale clouds.
124 This is then expressed as a function of the fractional removal rate of liquid water (i.e., ratio of
125 precipitating rain over the atmospheric layer liquid water content) and the aerosol solubility.
126 The in-cloud removal rate in RegCM4 for cumulus clouds is fixed at 0.001 s^{-1}) while the aerosol
127 solubility differs with the species variants. Thus the in-cloud wet deposition rates for
128 hydrophilic and hydrophobic BC and OC are different. The below-cloud aerosol wet removal
129 is influenced by their respective effective diameters and densities. Aerosol effective diameter
130 and density, which are different for different species, are used to calculate the collection
131 efficiency for individual aerosol species.

132 **2.2 Implementation of a dynamic ageing parameterisation (Ghosh et al. 2021)**

133 The anthropogenic aerosol module in RegCM4 consists of sulphate, hydrophilic and
134 hydrophobic BC, and hydrophilic and hydrophobic OC. The sulphate scheme has been adapted
135 from Qian et al. (2001). These species are considered as externally mixed while tracking their
136 mass concentrations. During simulations, RegCM4 considers emitted carbonaceous aerosols to
137 be 80% hydrophobic and 20% hydrophilic for BC and equal proportions of hydrophobic and
138 hydrophilic OC. The rate of change of mass mixing ratios of hydrophobic and hydrophilic
139 tracers has been described by the chemical transport equation in Solomon et al. [2006].

140 The time taken by hydrophobic tracers to get converted into hydrophilic tracers is called
141 “ageing”. In the default model set-up, ageing is governed by a fixed e-folding time of 1.15 days
142 (27.6 h) (Cooke et al. 1999). In our recently published work (Ghosh et al., 2021) we have
143 replaced this fixed number with a dynamic ageing scheme (equation 1), where the e-folding
144 time (τ_{mix}) changes as a function of coagulation and condensation. The parameterization has
145 been derived by Fierce et al. (2016) using least square regression on the output of the PartMC-
146 MOSAIC model (Riemer et al., 2009; Zaveri et al., 2008).

$$147 \quad \tau_{mix} = \frac{1}{I_{cond} \cdot k_{cond} + N \cdot k_{coag}} \quad (1),$$

148 where I_{cond} is the condensational flux and N is the total number concentration of all the
149 anthropogenic tracers. The detailed calculations of I_{cond} and N in the jargon of RegCM4 have
150 been provided in Ghosh et al. (2021). Constant values, k_{cond} (0.1 nm^{-1}) and k_{coag} ($6 \times 10^{-6} \text{ cm}^3$
151 hr^{-1}) have been derived by implementing the least square regression method on the PartMC
152 model output considering the impact of condensation and coagulation separately (Fierce et al.
153 2016).

154 Implementation of the new dynamic ageing scheme in RegCM4 resulted in faster conversion
155 of the hydrophobic to hydrophilic BC and OC, particularly over the polluted IGP. This, in turn,



156 increased column burden and surface concentrations of the carbonaceous aerosols over the
157 study region in the dry season. On the contrary, it decreased the burden in the wet season due
158 to a more efficient washout (Ghosh et al., 2021) everywhere except the polluted IGP, where
159 precipitation reduced as a result of radiative feedback.

160 **2.3 Emission inventories**

161 In this study, we replace a global emission inventory
162 [https://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html]
163 with a regional emission inventory (Venkataraman et al., 2018). Figure S1 (see the
164 Supplementary Information, SI) represents the seasonal variation of the emissions estimated
165 by the two inventories. The global emission inventory used in the experiments 'Default_Sc' and
166 'Dyn_global' was developed by the IIASA emission inventory at a resolution of $0.5^\circ \times 0.5^\circ$
167 [https://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html].
168 The key emission sectors considered in this inventory are energy, industry, solvent use,
169 transport, domestic combustion, agriculture, open burning of agricultural waste, and waste
170 treatment. The emission estimates were available only at an annual scale with no seasonal
171 variation from 1990-2010.

172 The regional emission inventory used in experiments 'Fix_regio' and 'Dyn_regio' was
173 developed and reported by Venkataraman et al. (2018) at a horizontal resolution of $0.25^\circ \times$
174 0.25° , that too at a monthly scale. Thus the regional emissions have a profound seasonal
175 variability (Figure S1). The key sectors included in the regional inventory are energy (coal +
176 oil + gas), heavy and light industry, brick production, residential cooking, solid biomass fuel,
177 residential cooking (LPG and kerosene), residential lighting (kerosene lamp), residential water
178 heating, residential space heating, informal industry, agricultural residue burning, on-road
179 gasoline, on-road diesel, railway, agricultural diesel pump, agricultural tractors. Among these
180 sectors, residential water heating, residential space heating, and agricultural residue burning
181 sectors have seasonality in emissions.

182 **2.3 In-situ BC data**

183 In-situ BC data for the year 2010 has been procured from 28 sites to evaluate the model
184 performance. Twenty-one of these sites is part of the Indian Space Research Organization's
185 Aerosol Radiative Forcing over India Network, ARFINET (Babu et al., 2013; Gogoi et al.,
186 2021). This network has been measuring columnar AOD and BC for many years. In addition
187 to the ARFINET, BC concentrations were also measured independently at Kanpur (Tripathi et
188 al., 2005) during the entire 2010 except the monsoon season, Gadanki (Gadhavi et al., 2015;



189 Jain et al., 2018), and Delhi (October-December 2010) by individual institutions. In all the
190 sites, BC was measured by an Aethalometer, which measures the amount of attenuation of the
191 light beam passing through the filter where particles get deposited. BC mass concentration is
192 subsequently estimated by the change in optical attenuation given by the rate of BC deposition
193 on the filter tape (Hansen et al., 1984). Dataset from all the sites except Gadanki (monthly
194 values) are available on a daily scale and have been averaged to get the annual concentrations.

195 **2.4 MERRA-2 data**

196 Model simulated BC and OC columnar burden have been evaluated against MERRA-2
197 reanalysis data. MERRA-2 is an updated reanalysis of atmospheric data produced by the NASA
198 Global Modeling and Assimilation Office (Buchard et al., 2017). MERRA-2 consists of
199 parameters that are not available in its predecessor, MERRA. It includes updates of the
200 Goddard Earth Observing System model and analysis scheme in order to give a more realistic
201 view of the ongoing climate analysis beyond MERRA's jargon. This dataset addressed the
202 limitations of MERRA. Various improvements in MERRA-2 include assimilation of aerosol
203 observations and improved representation of stratosphere, including ozone and cryosphere.
204 MERRA-2 data products are freely accessible through the NASA Goddard Earth Sciences Data
205 Information Services Centre. We note that MERRA-2 data are also not observations and direct
206 validation of the MERRA-2 columnar BC and OC burden is not possible.

207 **2.5 MISR aerosol data**

208 MISR on-board Terra satellite crosses the equator around 10:30 hrs local time. It has a high
209 spatial resolution and a wide range of viewing angles. It views the Earth using four spectral
210 bands in each of the nine cameras and has a weekly global coverage between $\pm 82^\circ$. A detailed
211 description is provided in the literature (Diner et al., 1998). MISR-AOD has a correlation
212 coefficient of ~ 0.9 (for maritime sites) and ~ 0.7 (for dusty sites) w.r.t AERONET (Kahn et al.,
213 2005). In the absence of any direct measurement, we use MISR fine AOD (AOD for particles
214 smaller than $0.35 \mu\text{m}$) as a proxy for anthropogenic AOD (Dey and Girolamo, 2010).

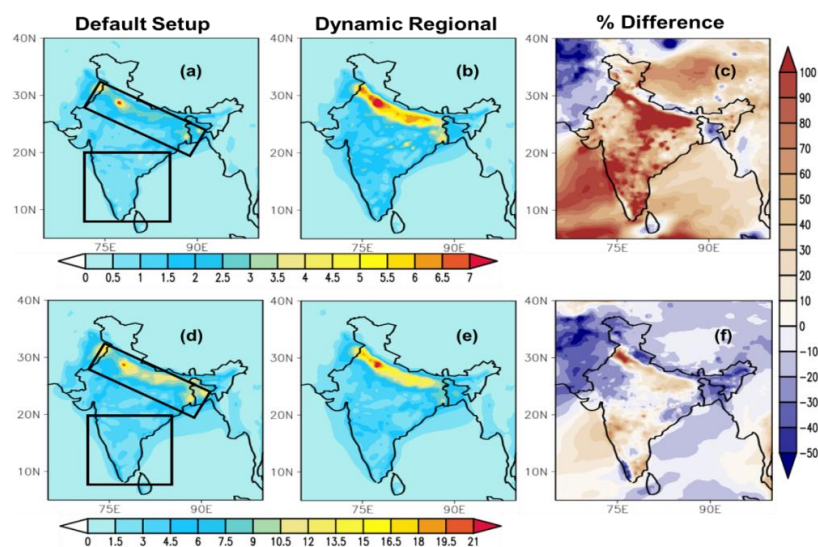
215 **3. Results**

216 **3.1 Spatial distribution of carbonaceous aerosols**

217 Figure 1 shows the spatial distributions of the annual surface concentration for BC and OC
218 using the default and augmented model along with their differences. Several key features are
219 notable. First, the OC concentration is almost three times higher than the BC concentration in
220 the augmented model, consistent with the literature (Priyadharshini et al., 2019). Secondly, the
221 concentrations are 2-3 times higher over the polluted IGP compared to the rest of India in the



222 augmented model. High aerosol loading over the IGP is a result of the combined effects of
223 greater source strength, low topography surrounded by highlands to the north and south, and
224 unfavourable meteorology (Dey and Girolamo, 2010; Srivastava et al., 2012). Thirdly, the BC
225 and OC concentrations increase by >100% and >60%, respectively, over the IGP and by
226 smaller margins elsewhere in the augmented model relative to the default configuration. The
227 increase in the annual concentrations of surface BC and OC is mainly driven by the changes in
228 the concentrations during the winter (JF), and pre-monsoon (MAM) seasons (Figure S2) since
229 during the monsoon, precipitation removes large amounts of aerosols. The relative impact of
230 the switch from the global to regional emission inventory (Figure S2) is greater than the impact
231 of the implementation of the dynamic ageing scheme (Figure S1) on the observed increases in
232 BC and OC mass concentrations in the augmented model.



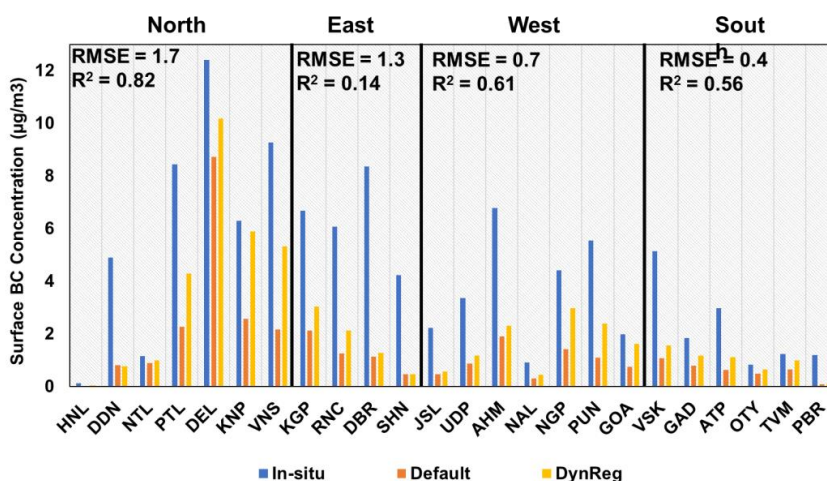
233

234 **Figure 1.** Spatial distribution of surface mass concentration ($\mu\text{g m}^{-3}$) of BC (a, b) and OC (d,
235 e) in 2010 over the Indian subcontinent using (left) the default and (middle) the augmented
236 model configurations. Figures 1c and 1f represent the corresponding percentage differences
237 due to the augmented model set-up (positive values imply an increase in mass concentration).
238 The vertical distributions (shown in Figure 3) are analysed for the IGP and PI sub-regions
239 marked by boxes in the panels of the left column.

240



241 We evaluate the performance of the customized model against BC surface concentrations
 242 measured at 24 sites across India (Figure S3). We note that the in-situ concentrations are point
 243 measurements while the model grids containing these sites are representative of 25 km by 25
 244 km areas. The default model severely underestimates the surface BC compared to the in-situ
 245 observations (mean normalized bias, MNB = -69%). Previous model studies reveal that the BC
 246 concentration is underestimated almost 2-5 times across various sites in India (Nair et al., 2012;
 247 Ramachandran et al., 2021). Though the underestimation persists in the augmented model (by
 248 varying proportions across the sites), the simulated concentration magnitudes are closer to the
 249 observations (MNB = -51%), more so in the urban centres of the polluted IGP (e.g., Delhi,
 250 Kanpur, Varanasi, Kharagpur). In some cities (e.g., Dehradun, Dibrugarh, Ahmedabad) where
 251 the differences in global and regional emission inventories are small (Figure S1), the
 252 improvement in simulated BC concentrations is also limited. This suggests that the problem
 253 could be related to the emission fluxes. In several cities, especially in the North and South
 254 Indian regions (e.g., Goa, Nainital, Ooty, Thiruvananthapuram), the simulated BC using the
 255 augmented model shows an almost perfect match with the observations. Overall, the
 256 augmented model ($R^2=0.66$) performs better than the default model ($R^2=0.6$) in simulating
 257 surface BC concentrations, and the errors shown in Figure 2 could also be amplified by the fact
 258 that the model data refers to a 25 km \times 25 km area as a single grid.



259

260 **Figure 2.** Comparison of simulated BC surface concentration ($\mu\text{g m}^{-3}$) using the default and
 261 augmented model with in-situ measurements from 24 cities across India. Locations of the cities



262 are shown in Figure S2. RMSE (in $\mu\text{g m}^{-3}$) and R^2 between the customized model simulations
263 and surface measurements are also provided.

264

265 Since there are no in-situ measurements of columnar burden available, we compare the
266 simulated columnar burden (Figure 3) with data from Modern-Era Retrospective Analysis for
267 Research and Applications, version 2 (MERRA-2) (Buchard et al., 2017). Similar to the surface
268 mass concentration, the BC burden shows a more pronounced change than the OC burden due
269 to the inclusion of the new model features. Though the simulated burden is still underestimated
270 relative to the MERRA-2 data, the values in the augmented model are much closer to the
271 reanalysis data. During the winter season (Jan-Feb), the percentage difference of model-
272 simulated column burden (w.r.t. MERRA-2) decreases from $>70\%$ to $\sim 35\%$ for BC and from
273 $\sim 63\%$ to $\sim 49\%$ for OC in the augmented model (Figures S4 and S5). A similar improvement
274 is found in the pre-monsoon season (Mar-May). The higher loading results from higher
275 magnitudes of regional emissions coupled with faster ageing and slower removal rate. The
276 percentage difference increases for OC burden over northwest India, decreases over the IGP,
277 and is negligible over the rest of the country. A probable explanation for such OC distribution
278 relies on the emission inventories used since the OC emissions are slightly higher in the global
279 inventory than those in the regional inventory over northwest India and lower in the IGP.
280 Emissions over the PI are roughly similar in the two inventories (Figure S2). We also note that
281 anthropogenic aerosol emissions vary on an annual basis in MERRA-2 (Buchard et al., 2017);
282 hence, there could be larger uncertainties at a seasonal scale.

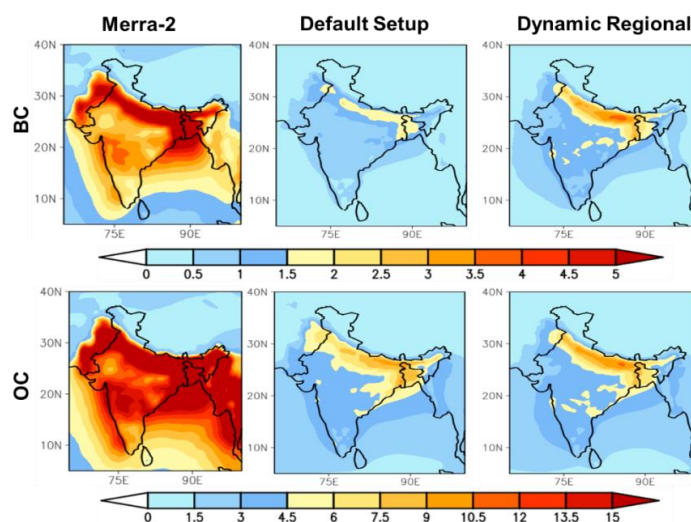
283 During the monsoon season (Jun-Sep), the BC loading increases in magnitude in the
284 augmented model compared to the default set-up (Figure S4), mostly due to the implementation
285 of the regional inventory. The magnitude of the simulated BC column burden is comparable
286 between the Default_Sc and Dyn_sc experiments and between the Fix_Regio and Dyn_Regio
287 (Figure S4), with an opposite pattern found for the OC column burden (Figure S5). Two
288 possible reasons can explain this result. First, the OC emissions in the global inventory are
289 higher than in the regional ones (Figure S1). Second, the model assumes that OC is 50%
290 hydrophobic and 50% hydrophilic at the time of emission (for BC, it is 80% hydrophobic and
291 20% hydrophilic), and therefore the faster conversion to hydrophilic OC due to the dynamic
292 ageing enhances the hydrophilic OC burden, which is subsequently removed by rain. In the
293 post-monsoon season (Oct-Dec), an overall increase in column burden in the augmented model
294 is observed throughout India. Higher emissions (in the case of the regional inventory) result in



295 higher concentrations of available condensing and coagulating particles, which in turn allows
296 faster ageing of hydrophobic to hydrophilic BC leading to accumulation of BC particles in the
297 atmosphere before their removal by dry deposition. The changes in the OC loading are
298 negligible in this season.

299

300



301

302 **Figure 3.** Comparison of spatial patterns of annual (top panel) BC and (bottom panel) OC
303 column burden (mg m^{-2}).

304

305 3.2 Vertical distribution of carbonaceous aerosols

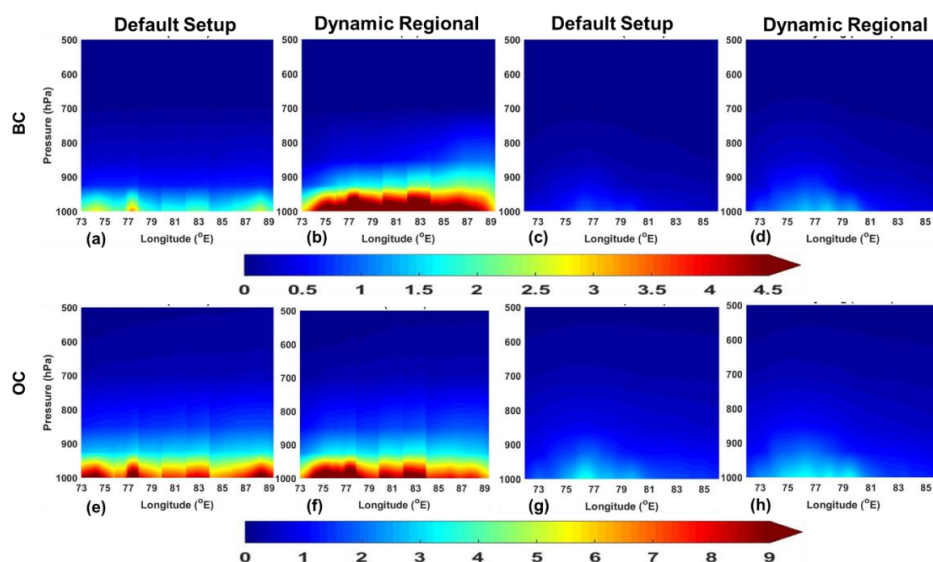
306 In this section, we analyse the effects of the model improvements on the vertical
307 distribution of aerosols over the IGP and compare the results with the contrasting PI region,
308 where the emissions are much lower. The two regions are indicated by the boxes in Figure 1
309 and Figure 4 longitude-altitude cross-sections of BC and OC mass concentration over the
310 regions.

311 The vertically distributed mass concentrations of both BC and OC increase due to the model
312 improvements up to 500 hPa. A larger increase is observed during the winter and post-monsoon
313 seasons over the IGP (Figures S6 and S7). Both the BC and OC concentrations are comparable
314 in the default and Dyn_global configurations, but they increase in the Fix_regio and Dyn_regio
315 set-ups (Figures S6 and S7). While the role of emissions is only predominant during the post-



316 monsoon season, the highest increase due to the combined effect of the two improvements is
317 observed during the winter. In these two seasons, the BC remains confined mostly within the
318 lower troposphere while the OC is distributed up to the mid-tropospheric levels, which is
319 indicative of higher concentrations of OC vertical transport than that of BC. During the pre-
320 monsoon season, the vertical distributions of both BC and OC show responses similar to that
321 of their spatial distributions. In the monsoon season, the tracer concentration is mainly confined
322 to the surface levels, indicating a lower wet removal and slower ageing above 1000 hPa (Ghosh
323 et al., 2021).

324 Over the PI, the concentrations of carbonaceous aerosols are lower than over the IGP, which
325 limits the impact of dynamic ageing because of the lower availability of condensing and
326 coagulating particles (relative to the IGP). This results in a slower ageing and lower
327 accumulation of hydrophilic tracers in the troposphere. However, the vertical pumping effect
328 is quite prominent during the winter season in the augmented configuration (Figures S6 and
329 S7). During the pre-monsoon season, only the BC concentration shows an increment in the
330 lower troposphere, while the OC concentration remains more or less unchanged. The PI
331 receives rainfall during the southwest and northeast monsoon; hence the tracer concentration
332 is further lowered during the monsoon and post-monsoon seasons.



333

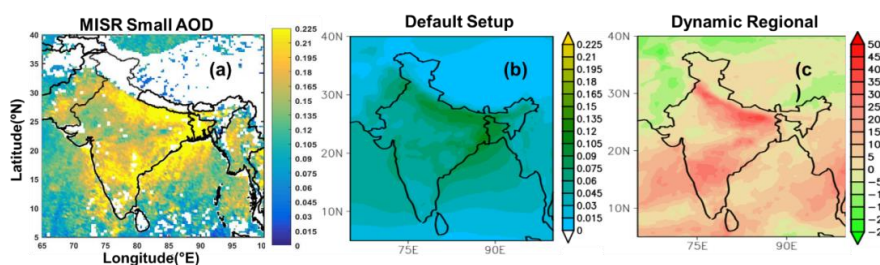
334 **Figure 4.** Longitude (in °E)-altitude (in hPa) cross-sections of (top panel) BC and (bottom
335 panel) OC mass concentration ($\mu\text{g m}^{-3}$) over the IGP (a, b, e, f) and PI (c, d, g, h) for the default
336 and customized model.



337 3.3 Optical and radiative properties of anthropogenic aerosols

338 We now examine the effects of the model improvements on the optical properties of
339 anthropogenic aerosols. In this regard, we note that the changes due to the implementation of
340 the dynamic ageing scheme can alter only BC and OC concentrations, while the changes related
341 to the emission inventory impact the sulphate concentration as well. We consider the AOD due
342 to small particles (radius <math><0.35 \mu\text{m}</math>) from MISR (Kahn and Gaitley, 2015), as a proxy for
343 anthropogenic AOD (hereafter AAOD) since direct measurement of AAOD are not available
344 to evaluate our model performance (Figure 5).

345 The simulated annual AAOD is >50% lower than the MISR small-AOD over the polluted
346 IGP and 30-50% lower over the PI in the default model. This is consistent with the previous
347 studies (Nair et al., 2012). These model underestimations improve by 25-35% over the IGP and
348 parts of PI in the augmented model. The seasonal plots (Figure S8) clearly show an increase of
349 AAOD in all seasons except during the monsoon. This increase in AAOD is due to both the
350 implementation of region-specific emission fluxes and the dynamic ageing scheme. The
351 AAOD still remains underestimated in some regions, which can possibly be addressed by
352 further improvements of the emission estimates, for example, the addition of missing sectors
353 (e.g., crematorium, municipal solid waste burning, etc.), improving sectoral methodologies for
354 informal activities and incorporation of regionally measured emission factors.



355

356 **Figure 5.** Spatial distribution of (a) MISR small mode AOD ('white' color implies 'no data'),
357 (b) AAOD simulated by default_sc, and (c) percentage increase in AAOD simulated by the
358 augmented model w.r.t default_sc for 2010.

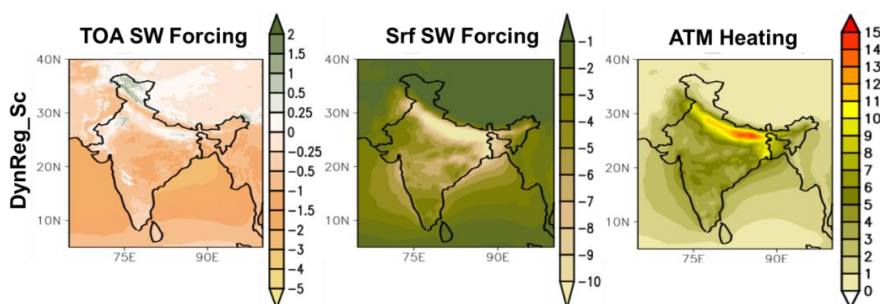
359

360 Spatial patterns of the annual top-of-the-atmosphere (TOA), surface, and atmospheric
361 radiative forcing associated with the anthropogenic aerosols for the augmented model are
362 shown in Figure 6. The TOA aerosol radiative forcing lies in the range -0.5 to -1.5 W m^{-2} over
363 most of the Indian landmass, except the IGP, where it is positive (0.25 to 1 W m^{-2}) due to the



364 higher concentration of carbonaceous aerosols (Figure 3), particularly BC. The TOA forcing is
365 also positive over the Indian desert and snow-covered regions even when the carbonaceous
366 aerosol concentrations are lower or comparable to the rest of India. The high surface albedo in
367 these regions allows for an enhanced interaction of the carbonaceous aerosols with solar
368 radiation, resulting in a warming effect (Ramachandran and Kedia, 2010). The surface radiative
369 forcing is found to be larger than -10 W m^{-2} over the polluted IGP, which is consistent with
370 published results (Ramanathan and Carmichael, 2008). Over the rest of India, the surface
371 forcing values lie between -3 to -8 W m^{-2} . Due to the model improvements (forcing estimates
372 with the default model are shown in Figure S8), the TOA forcing changes by -72.75% , and the
373 surface dimming increases by 39.73% over the IGP and by -23.94% and 34.35% , respectively,
374 over PI. As a result, the atmospheric heating increases by $\sim 9 \text{ W m}^{-2}$ over the IGP. The
375 simulated surface shortwave radiation shows a statistically significant ($p < 0.05$) correlation
376 with the observations from CERES (Su et al., 2005) all-sky and clear-sky radiation throughout
377 the year except in MAM and JJAS clear-sky conditions (Figure S9 and S10).

378



379

380 **Figure 6.** Annual variation of SW radiative forcing (W/m^2) at TOA (left column), at the surface
381 (middle column), and the resultant atmospheric heating (W/m^2) (right column) for the
382 customized set-up.

383

384 4. Discussion and conclusions

385 Accurate estimates of emission fluxes and a better representation of aerosol processes are
386 required to improve the representation of aerosol life-cycle and radiative effects in climate
387 models. Here we augmented the regional climate model RegCM4 by implementing a dynamic
388 ageing scheme and a regional emission inventory and examined the combined impact of these
389 factors on the model performance over the Indian monsoon region. We note that though the



390 aerosol simulations improve due to these model enhancements, some systematic biases persist
391 (underestimation of carbonaceous aerosol concentrations) and need to be further addressed.
392 For example, RegCM has a bulk scheme for anthropogenic aerosols, and thus the number
393 concentration is calculated from the bulk mass concentration (Ghosh et al. 2021). The
394 anthropogenic aerosol module can thus be improved by including a particle size-dependent
395 representation. In addition, the present dynamic ageing timescale depends only on the
396 anthropogenic aerosol number concentration, while it should, in fact, depend on the total
397 (anthropogenic + natural) number concentrations. The simulations presented in this work did
398 not include natural aerosols, which could have impacted the meteorology through dynamic
399 feedbacks, possibly affecting the carbonaceous aerosol burden. This aspect will be examined
400 in future work. Thirdly, though the emission fluxes of BC, OC, and SO₂ are higher in the region
401 than the global inventory, there may still be uncertainty related to missing sectoral sources.

402 Our work demonstrates that even the improvement of some aspects of the aerosol
403 representation can lead to substantial enhancements in the model performance. We also find
404 that over the South Asian monsoon region, particularly over highly polluted regions such as
405 the IGP, the default model significantly underestimates the surface dimming and atmospheric
406 heating, which can have implications for climate studies (Das et al., 2016, 2020) and this
407 problem is substantially ameliorated with our model augmentations.

408 The key conclusions of our work can be summarized as follows.

- 409 1. The conclusion in the model RegCM4 of a dynamic ageing scheme and a regional
410 emission inventory substantially improves the model performance over the Indian sub-
411 continent.
- 412 2. The BC and OC surface concentration and column burden increase due to the model
413 improvements, more so as a combined effect of the two factors than because of the
414 individual ones.
- 415 3. The TOA, surface, and atmospheric radiative forcing are estimated to be -0.3, -5.3, and
416 5.0 W m⁻², respectively, over the polluted IGP using the augmented model, but they
417 could still be underestimated.

418

419 *Data availability.* The model RegCM4.6 code is freely available online from
420 <https://zenodo.org/record/5729783#.YaDifbso-Uk>. The anthropogenic aerosol emissions
421 considered for the simulations are taken from the IIASA inventory. The data used can be easily
422 accessed

423 online http://climadods.ictp.it/Data/RegCM_Data/RCP_EMGLOB_PROCESSED/iiasa/



424 website. Input files for the RegCM4 model are archived in <http://clima->
425 [dods.ictp.it/Data/RegCM_Data/](http://clima-dods.ictp.it/Data/RegCM_Data/) website. MISR data is available freely from <https://www->
426 [misr.jpl.nasa.gov/](https://www-misr.jpl.nasa.gov/) while MERRA-2 data is freely available from the NASA Giovanni site
427 <https://giovanni.gsfc.nasa.gov/giovanni/> .

428

429 *Author contributions.*

430 SG and SD (corresponding author) planned the experiments and wrote the first draft. SG carried
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432 the regional emission inventory. DG and FG provided inputs on experiment design. SNT, SR,
433 TAR, HG, and AKS provided the BC data for evaluation. All authors contributed and edited
434 the manuscript.

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450 **References**

- 451 1. Ajay, P., Pathak, B., Solmon, F., Bhuyan, P. K., Giorgi, F.: Obtaining best
452 parameterization scheme of RegCM 4.4 for aerosols and chemistry simulations over
453 the CORDEX South Asia. *Clim Dyn* 53, 329–352, [https://doi.org/10.1007/s00382-018-](https://doi.org/10.1007/s00382-018-4587-3)
454 [4587-3](https://doi.org/10.1007/s00382-018-4587-3), 2019.
- 455 2. Babu, S. S., Manoj, M. R., Krishna Moorthy, K., Gogoi, M. M., Nair, V. S., Kompalli,
456 S.K., Satheesh, S. K., Niranjana, K., Ramagopal, K., Bhuyan, P. K., and Singh, D.:



- 457 Trends in aerosol optical depth over Indian region: Potential causes and impact
458 indicators, *J. Geophys. Res. Atmos.* 118, 11794–11806,
459 <https://doi.org/10.1002/2013JD020507>, 2013.
- 460 3. Bond, T. C., et al.: Bounding the role of black carbon in the climate system: A scientific
461 assessment, *J. Geophys. Res. Atmos.*, 118, 5380–5552,
462 <https://doi.org/10.1002/jgrd.50171>, 2013.
- 463 4. Bretherton C.S., McCaa, J.R. and Grenier, H. A new parameterization for shallow
464 cumulus convection and its application to marine subtropical cloud-topped boundary
465 layers. I. Description and 1D results. *Mon Weather Rev* 132, 864–882,
466 [https://doi.org/10.1175/1520-0493\(2004\)132<0864:ANPFSC>2.0.CO;2](https://doi.org/10.1175/1520-0493(2004)132<0864:ANPFSC>2.0.CO;2), 2004.
- 467 5. Buchard, V., Randles, C. A., da Silva, A. M., Darmenov, A., Colarco, P. R.,
468 Govindaraju, R., Ferrare, R., Hair, J., Beyersdorf, A. J., Ziemba, L. D., and Yu, H.: The
469 MERRA-2 Aerosol Reanalysis, 1980 Onward. Part II: Evaluation and Case Studies,
470 *Journal of Climate* 30(17), 6851–6872, <https://doi.org/10.1175/JCLI-D-16-0613.1>,
471 2017.
- 472 6. Das, S., Dey, S., Dash, S. K., Giuliani, G., and Solmon, F.: Dust aerosol feedback on
473 the Indian summer monsoon: Sensitivity to absorption property. *J. Geophys. Res.*
474 *Atmos.* 120, 9642–9652, <https://doi.org/10.1002/2015JD023589>, 2015.
- 475 7. Das, S., Dey, S., and Dash, S. K.: Direct radiative effects of anthropogenic aerosols on
476 Indian summer monsoon circulation, *Theor Appl Climatol.* 124, 629–639,
477 <https://doi.org/10.1007/s00704-015-1444-8>, 2016.
- 478 8. Das, S., Giorgi, F., Giuliani, G., Dey, S., and Coppola, E.: Near-future anthropogenic
479 aerosol emission scenarios and their direct radiative effects on the present-day
480 characteristics of the Indian summer monsoon, *Journal of Geophysical Research:*
481 *Atmospheres* 125, e2019JD031414, <https://doi.org/10.1029/2019JD031414>, 2020.
- 482 9. Dash, S. K., Shekhar, M. S. and Singh G. P.: Simulation of Indian summer monsoon
483 circulation and rainfall using RegCM3, *Theor. Appl. Climatol.* 86, 161–172,
484 <https://doi.org/10.1007/s00704-006-0204-1>, 2006.
- 485 10. Dee, D.P., Uppala, S.M., Simmons, A.J., Berrisford, P., Poli, P., Kobayashi, S., Andrae,
486 U., Balmaseda, M.A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A.C.M., van de
487 Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A.J.,
488 Haimberger, L., Healy, S.B., Hersbach, H., Hólm, E.V., Isaksen, L., Kållberg, P.,
489 Köhler, M., Matricardi, M., McNally, A.P., Monge-Sanz, B.M., Morcrette, J.-J., Park,
490 B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N. and Vitart, F.: The ERA-



- 491 Interim reanalysis: configuration and performance of the data assimilation system,
492 Q.J.R. Meteorol. Soc. 137, 553-597, <https://doi.org/10.1002/qj.828>, 2011.
- 493 11. Dey, S., and Di Girolamo, L.: A climatology of aerosol optical and microphysical
494 properties over the Indian subcontinent from 9 years (2000–2008) of Multiangle
495 Imaging Spectroradiometer (MISR) data, *J. Geophys. Res.* 115, D15204,
496 <https://doi.org/10.1029/2009JD013395>, 2010.
- 497 12. Dickinson, R.E., Sellers, H.A. and Kennedy, P.J.: Biosphere atmosphere transfer
498 scheme (BATS) version 1E as coupled to the NCAR community climate model,
499 Technical note NCAR/TN-3871 STR, p 72, DOI:10.5065/D67W6959, 1993.
- 500 13. Diner, D. J., Beckert, J.C., Reilly, T.H., Bruegge, C.J., Conel, J. E., Kahn, R.,
501 Martonchik, J.V., Ackerman, T.P., Davies, R., Gerstl, S.A.W., Gordon, H.R., Muller,
502 J-P., Myneni, R.B., Sellers, R.J., Pinty, B., and Verstraete, M.M.: Multi-angle Imaging
503 Spectro Radiometer (MISR) instrument description and experiment overview, *IEEE*
504 *Transactions on Geoscience and Remote Sensing*, 36 (4), 1072-1087, DOI:
505 10.1109/36.700992, 1998.
- 506 14. Emanuel, K.A. and Rothman, M.Z.: Development and evaluation of a convection
507 scheme for use in climate models, *J. Atmos. Sci.* 56, 1766–1782,
508 [https://doi.org/10.1175/1520-0469\(1999\)056<1766:DAEOAC>2.0.CO;2](https://doi.org/10.1175/1520-0469(1999)056<1766:DAEOAC>2.0.CO;2), 1999.
- 509 15. Fierce, L., Riemer, N., and Bond, T.: Toward Reduced Representation of Mixing State
510 for Simulating Aerosol Effects on Climate, *Bull. Amer. Meteor. Soc.*, 98, 971–980,
511 DOI: 10.1175/BAMS-D-16-0028.1, 2016.
- 512 16. Gadhavi, H. S., Renuka, K., Kiran, V. R., Jayaraman, A., Stohl, A., Klimont, Z., and
513 Beig, G. Evaluation of black carbon emission inventories using a Lagrangian dispersion
514 model - a case study over southern India, *Atmospheric Chemistry and Physics*, 15(3),
515 1447–1461, <https://doi.org/10.5194/acp-15-1447-2015>, 2015.
- 516 17. Ghosh, S., Riemer, N., Giuliani, G., Giorgi, F., Ganguly, D., and Dey, S.: Sensitivity of
517 carbonaceous aerosol properties to the implementation of a dynamic ageing
518 parameterization in the regional climate model RegCM, *Journal of Geophysical*
519 *Research: Atmospheres*, 126, e2020JD033613, <https://doi.org/10.1029/2020JD033613>,
520 2021.
- 521 18. Giorgi, F., Jones, C. and Asrar, G. R.: Addressing climate information needs at the
522 regional level: the CORDEX framework, *WMO Bulletin* 58(3), 175-183, 2009.
- 523 19. Giorgi, F., Coppola, E., Solmon, F., Mariotti, L., Sylla, M. B., Bi, X., Elguindi, N.,
524 Diro, G. T., Nair, V., Giuliani, G., Turuncoglu, U. U., Cozzini, S., Güttler, I., O'Brien,



- 525 T. A., Tawfik, A. B., Shalaby, A., Zakey, A. S., Steiner, A. L., Stordal, F., Sloan, L. C.,
526 and Brankovic, C.: RegCM4: model description and preliminary tests over multiple
527 CORDEX domains, *Clim. Res.* 52, 7-29, <https://doi.org/10.3354/cr01018>, 2012.
- 528 20. Gogoi et al., Response of ambient BC concentration across the Indian region to the
529 nation-wide lockdown: results from the ARFINET measurements of ISRO-GBP,
530 *Current Science* 120(2), 341-351,
531 <https://doi.org/10.18520/cs%2Fv120%2Fi2%2F341-351>, 2021.
- 532 21. Grell, G. A.: Prognostic Evaluation of Assumptions Used by Cumulus
533 Parameterizations, *Monthly Weather Review* 121(3), 764-787,
534 [https://doi.org/10.1175/1520-0493\(1993\)121<0764:PEOAUB>2.0.CO;2](https://doi.org/10.1175/1520-0493(1993)121<0764:PEOAUB>2.0.CO;2), 1993.
- 535 22. Grenier, H. and Bretherton, C.S. A moist PBL parameterization for large-scale models
536 and its application to subtropical cloud-topped marine boundary layers, *Mon. Weather*
537 *Rev.*, 129, 357–377, [https://doi.org/10.1175/1520-0493\(2001\)129<0357:AMPPFL>2.0.CO;2](https://doi.org/10.1175/1520-0493(2001)129<0357:AMPPFL>2.0.CO;2), 2001.
- 539 23. Hansen, A.D.A., Rosen, H. and Novakov, T.: The Aethalometer—An Instrument for
540 the Real-Time Measurement of Optical Absorption by Aerosol Particles, *Science of*
541 *The Total Environment*, 36, 191-196, 10.1016/0048-9697(84)90265-1, 1984.
- 542 24. Jain, C. D., Gadhavi, H. S., Wankhede, T., Kallelapu, K., Sudhesh, S., Das, L. N., Pai,
543 R. U., and Jayaraman, A.: Spectral Properties of Black Carbon Produced during
544 Biomass Burning, *Aerosol and Air Quality Research*, 18(3), 671–679, doi:
545 10.4209/aaqr.2017.03.0102, 2018.
- 546 25. Kahn, R., Li, W., Martonchik, J. V., Bruegge, C. J., Diner, D. J., Gaitley, B. J., Abdou,
547 W., Dubovik, O., Holben, B., Smirnov, A., Jin, Z., & Clark, D.: MISR calibration and
548 implications for low-light-level aerosol retrieval over dark water, *J. Atmos. Sci.* 62 (4),
549 1032-1052, <https://doi.org/10.1175/JAS3390.1>, 2005.
- 550 26. Kahn, R. A., and Gaitley, B. J.: An analysis of global aerosol type as retrieved by MISR,
551 *J. Geophys. Res. Atmos.* 120, 4248– 4281, <https://doi.org/10.1002/2015JD023322>,
552 2015.
- 553 27. Kanakidou, M., Seinfeld, J. H., Pandis, S. N., Barnes, I., Dentener, F. J., Facchini, M.
554 C., Van Dingenen, R., Ervens, B., Nenes, A., Nielsen, C. J., Swietlicki, E., Putaud, J.
555 P., Balkanski, Y., Fuzzi, S., Horth, J., Moortgat, G. K., Winterhalter, R., Myhre, C. E.
556 L., Tsigaridis, K., Vignati, E., Stephanou, E. G., and Wilson, J.: Organic aerosol, and
557 global climate modelling: A review, *Atmos. Chem. Phys.* 5, 1053–1123,
558 <https://doi.org/10.5194/acp-5-1053-2005>, 2005.



- 559 28. Kiehl, J.T., Hack, J. J., Bonan, G. B., Boville, B. A., Briegleb, B. P., Williamson, D.
560 L., & Rasch, P. J.: Description of the NCAR community climate model (CCM3), Tech.
561 Rep. National Center for Atmospheric Research, NCAR/TN-420+STR, P 152,
562 doi:10.5065/D6FF3Q99, 1996.
- 563 29. Nair, V.S., Solmon, F., Giorgi, F., Mariotti, L., Babu, S.S. and Moorthy, K. K.
564 Simulation of South Asian aerosols for regional climate studies, *J Geophys Res.* 117,
565 D04209, <https://doi.org/10.1029/2011JD016711>, 2012.
- 566 30. O'Brien, T.A., Chuang, P.Y., Sloan, L.C., Faloona, I.C. and Rossiter, D.L.: Coupling a
567 new turbulence parametrization to RegCM adds realistic stratocumulus clouds, *Geosci.*
568 *Mod. Dev.* 5, 989–1008, <https://doi.org/10.5194/gmd-5-989-2012>, 2012.
- 569 31. Pal, J. S., Small, E. E. and Eltahir E. A. B.: Simulation of regional-scale water and
570 energy budgets: Representation of subgrid cloud and precipitation processes within
571 RegCM, *J. Geophys. Res.* 105(D24), 29579–29594,
572 <https://doi.org/10.1029/2000JD900415>, 2000.
- 573 32. Pattnayak, K.C, Panda, S.K., Saraswat, V. and Dash, S. K.: Assessment of two versions
574 of a regional climate model in simulating the Indian Summer Monsoon over South Asia
575 CORDEX domain, *Clim Dyn* 50, 3049–3061, [https://doi.org/10.1007/s00382-017-](https://doi.org/10.1007/s00382-017-3792-9)
576 [3792-9](https://doi.org/10.1007/s00382-017-3792-9), 2018.
- 577 33. Priyadharshini, B., Verma, S., Chatterjee, A., Kumar Sharma S. and Kumar Mandal,
578 T.: Chemical Characterization of Fine Atmospheric Particles of Water-Soluble Ions and
579 Carbonaceous Species in a Tropical Urban Atmosphere over the Eastern Indo-Gangetic
580 Plain, *Aerosol and Air Quality Research* 19, 129–147,
581 <https://doi.org/10.4209/aaqr.2017.12.0606>, 2019.
- 582 34. Putaud, J.P., Van Dingenen, R. , Alastuey, A., Bauer, H., Birmili, W., Cyrus, J., Flentje,
583 H., Fuzzi, S., Gehrig, R., Hansson, H.C., Harrison, R.M., Herrmann, H., Hitzenberger,
584 R., Hüglin, C., Jones, A.M., Kasper-Giebl, A., Kiss, G., Koussa, A. , Kuhlbusch, T.A.J.,
585 Löschau, G., Maenhaut, W., Molnar, A., Moreno, T., Pekkanen, J., Perrino, C., Pitz,
586 M., Puxbaum, H., Querol, X., Rodriguez, S., Salma, I., Schwarz, J., Smolik, J.,
587 Schneider, J., Spindler, G., ten Brink, H., Tursic, J., Viana, M., Wiedensohler, A., Raes,
588 F.: A European aerosol phenomenology-3: Physical and chemical characteristics of
589 particulate matter from 60 rural, urban, and kerbside sites across Europe, *Atmos.*
590 *Environ.* 44, 1308–1320, <https://doi.org/10.1016/j.atmosenv.2009.12.011>, 2010.



- 591 35. Qian, Y., Giorgi, F., Huang, Y., Chameides, W. L. and Luo, C.: Simulation of
592 anthropogenic sulfur over East Asia with a regional coupled chemistry-climate model,
593 *Tellus, Ser. B.* 53, 171–191, <https://doi.org/10.3402/tellusb.v53i2.16573>, 2001.
- 594 36. Rai, P. K., Singh, G. P. and Dash, S. K.: Projected Change and Variability Assessment
595 of Indian Summer Monsoon Precipitation in South Asia CORDEX Domain Under
596 High-Emission Pathway, *Pure Appl. Geophys.* 177(7), 3475–3499,
597 <https://doi.org/10.1007/s00024-019-02373-3>, 2019.
- 598 37. Ramachandran, S., and Kedia, S.: Black carbon aerosols over an urban region:
599 Radiative forcing and climate impact, *J. Geophys. Res.*, 115, D10202,
600 doi:10.1029/2009JD013560, 2010 .
- 601 38. Ramachandran, S., Rajesh, T.A. and R. Cherian, R.: Black carbon aerosols over source
602 vs. background region: Atmospheric boundary layer influence, potential source regions,
603 and model comparison, *Atmospheric Research*, 256, 105573, ISSN 0169-8095,
604 <https://doi.org/10.1016/j.atmosres.2021.105573>, 2021.
- 605 39. Ramanathan, V. and Carmichael G.: Global and regional climate changes due to black
606 carbon. *Nat Geos.* 1, 221-227, <https://doi.org/10.1038/ngeo156>, 2008.
- 607 40. Riemer, N., West, M., Zaveri, R. A., and Easter, R. C.: Simulating the evolution of soot
608 mixing state with a particle resolved aerosol model, *J. Geophys. Res.*, 114, D09202,
609 doi:10.1029/2008JD011073, 2009.
- 610 41. Riemer, N., Ault, A. P., West, M., Craig, R. L., & Curtis, J. H.: Aerosol mixing state:
611 Measurements, modeling, and impacts, *Reviews of Geophysics* 57, 187– 249,
612 <https://doi.org/10.1029/2018RG000615>, 2019.
- 613 42. Satheesh, S.K.: Aerosol radiative forcing over land: effect of surface and cloud
614 reflection, *Ann. Geophys.* 20, 2105–2109, [https://doi.org/10.5194/angeo-20-2105-](https://doi.org/10.5194/angeo-20-2105-2002)
615 [2002](https://doi.org/10.5194/angeo-20-2105-2002), 2002.
- 616 43. Shalaby, A., Zakey, A. S., Tawfik, A. B., Solmon, F., Giorgi, F., Stordal, F., Sillman,
617 S., Zaveri, R. A., and Steiner, A. L.: Implementation and evaluation of online gas-phase
618 chemistry within a regional climate model (RegCM-CHEM4), *Geosci. Model Dev.*, 5,
619 741–760, <https://doi.org/10.5194/gmd-5-741-2012>, 2012.
- 620 44. Singh, A., Rastogi, N., Kumar, V., Slowik, J. G., Satish, R., Lalchandani, V., Thamban,
621 N. M., Rai, P., Bhattu, D., Vats, P., Ganguly, D., Tripathi, S.N., and Prévôt, André S.H.:
622 Sources and characteristics of light-absorbing fine particulates over Delhi through the
623 synergy of real-time optical and chemical measurements, *Atmos. Environ.* 252,
624 118338, <https://doi.org/10.1016/j.atmosenv.2021.118338>, 2021.



- 625 45. Solmon, F., Giorgi, F. and Liousse, C. Aerosol modelling for regional climate studies:
626 application to anthropogenic particles and evaluation over a European/African
627 domain, *Tellus B* 58, 51–72, <https://doi.org/10.1111/j.1600-0889.2005.00155.x>, 2006.
- 628 46. Srivastava, A. K., Dey, S., and Tripathi, S. N. Aerosol characteristics over the Indo-
629 Gangetic Basin: Implications to regional climate, *Atmospheric Aerosols-Regional*
630 *Characteristics-Chemistry and Physics* 10, 47782, DOI: 10.5772/47782, 2012.
- 631 47. Su, W., Charlock, T. P. and Rose, F. G.: Deriving surface ultraviolet radiation from
632 CERES surface and atmospheric radiation budget: Methodology, *J. Geophys. Res.* 110
633 (D14209), <https://doi.org/10.1029/2005JD005794>, 2005.
- 634 48. Tibrewal, K. and Venkataraman, C.: Climate co-benefits of air quality and clean energy
635 policy in India, *Nat Sustain.* 4, 305–313, <https://doi.org/10.1038/s41893-020-00666-3>,
636 2021.
- 637 49. Tiedtke, M.: A comprehensive mass-flux scheme for cumulus parameterization in
638 large-scale models, *Mon. Weather Rev.* 117, 1779–1800, [https://doi.org/10.1175/1520-0493\(1989\)117<1779:ACMFSF>2.0.CO;2](https://doi.org/10.1175/1520-0493(1989)117<1779:ACMFSF>2.0.CO;2), 1989.
- 640 50. Tripathi, S. N., Dey, S., Tare, V., Satheesh, S. K., Lal, S., and Venkataramani, S.:
641 Enhanced layer of black carbon in a north Indian industrial city, *Geophys. Res. Lett.*,
642 32, L12802, <https://doi.org/10.1029/2005GL022564>, 2005.
- 643 51. UNFCCC Report 2015
- 644 52. Venkataraman, C., Brauer, M., Tibrewal, K., Sadavarte, P., Ma, Q., Cohen, A.,
645 Chaliyakunnel, S., Frostad, J., Klimont, Z., Martin, R. V., Millet, D. B., Philip, S.,
646 Walker, K., and Wang, S.: Source influence on emission pathways and ambient PM_{2.5}
647 pollution over India (2015-2050), *Atmos. Chem. Phys.* 18, 8017-8039,
648 <https://doi.org/10.5194/acp-18-8017-2018>, 2018.
- 649 53. Venkataraman, C., Bhushan, M., Dey, S., Ganguly, D., Gupta, T., Habib, G., Kesarkar,
650 A., Phuleria, H., & Raman, R. S.: Indian Network Project on Carbonaceous Aerosol
651 Emissions, Source Apportionment and Climate Impacts (COALESCE), *Bulletin of the*
652 *American Meteorological Society*, 101(7), E1052-E1068,
653 <https://doi.org/10.1175/BAMS-D-19-0030.1>, 2020.
- 654 54. Zakey, A. S., Solmon, F. and Giorgi, F.: Development and testing of a desert dust
655 module in a regional climate model, *Atmos. Chem. Phys.* 6, 4687–4704,
656 <https://doi.org/10.5194/acp-6-4687-2006>, 2006.



- 657 55. Zakey, A. S., Giorgi, F. and Bi, X.: Modeling of sea salt in a regional climate model:
658 Fluxes and radiative forcing, *J. Geophys. Res.* 113, D14221,
659 <https://doi.org/10.1029/2007JD009209>, 2008.
- 660 56. Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for Simulating Aerosol
661 Interactions and Chemistry (MOSAIC), *J. Geophys. Res.*, 113, D13204,
662 doi:10.1029/2007JD008782, 2008.
- 663