

Inter-comparison and performance evaluation of chemistry transport models over Indian region

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Abstract

Aerosol loading over the South Asian region has the potential to affect the monsoon rainfall, Himalayan glaciers and regional air-quality, with implications for the billions in this region. While field campaigns and network observations provide primary data, they tend to be location/ season specific. Numerical models are useful to regionalize such location-specific data. Studies have shown that numerical models underestimate the aerosol scenario over the Indian region, mainly due to shortcomings related to meteorology and the emission inventories used. In this context, we have evaluated the performance of two such chemistry-transport models: WRF-Chem and SPRINT-ARS over an India-centric domain. The models differ in many aspects including physical domain, horizontal resolution, meteorological forcing and so on. etc. Despite these differences, both the models simulated similar spatial patterns of Black Carbon (BC) mass concentration, (with a spatial correlation of 0.9 with each other), and a reasonable estimates of its concentration, though both of them under-estimated vis-a-vis the observations. While the emissions are lower (higher) in SPRINTARS (WRF-Chem), overestimation of wind parameters in WRF-Chem caused the concentration to be similar in both models. Additionally, we quantified the underestimations of anthro-

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pogenic BC emissions in the inventories used these two models and three other widely used emission inventories. Our analysis indicates that all these emission inventories underestimate the emissions of BC over India by a factor that ranges from 1.5 to 2.9. We have also studied the model simulations of aerosol optical depth over the Indian region. The models differ significantly in simulations of AOD, with WRF-Chem having a better agreement with satellite observations of AOD as far as the spatial pattern is concerned. It is important to note that in addition to BC, dust can also contribute significantly to AOD. The models differ in simulations of the spatial pattern of mineral dust over the Indian region. We find that both meteorological forcing and emission formulation contribute to these differences. Since AOD is column integrated parameter, description of vertical profiles in both models, especially since elevated aerosol layers are often observed over Indian region, could be also a contributing factor. Additionally, differences in the prescription of the optical properties of BC between the models appear to affect the AOD simulations. We also compared simulation of sea-salt concentration in the two models and found that WRF-Chem underestimated its concentration vis-a-vis SPRINTARS. The differences in near-surface oceanic wind speeds appear to be the main source of this difference. In spite of these differences, we note that there are similarities in their simulation of spatial patterns of various aerosol species (with each other and with observations) and hence models could be valuable tools for aerosol-related studies over the Indian region. Better estimation of emission inventories could improve aerosol-related simulations.

Keywords: WRF-Chem, SPRINTARS, BC, AOD, Dust

1. Introduction

Aerosols are tiny (10^{-9}m - 10^{-4}m) solid or liquid particles suspended in air. They are capable of affecting the Earth's radiation budget through direct (Haywood and Ramaswamy, 1998, Haywood and Boucher, 2000, Kaufman et al., 2002, Takemura et al., 2005, Yu et al., 2006, Myhre, 2009) and indirect (Twomey, 1977, Lohmann and Lesins, 2002, Lohmann and Feichter, 2005, Kiran et al., 2009) pathways, besides having severe effects on human health (Krzyzanowski et al., 2005, Janssen et al., 2011). The South Asian region is known to be a hot spot of natural as well as anthropogenic aerosols (Ramanathan et al., 2001, Lelieveld et al., 2001). There has been an in-

11 creasing trend in AOD over this region (Porch et al., 2007, Ramachandran
12 et al., 2012, Babu et al., 2013). Such an aerosol loading over the region is
13 capable of altering the radiation budget and potentially offsetting the large
14 scale monsoonal circulation, resulting in modification in the rainfall distri-
15 bution over the region (Chakraborty et al., 2004, Ramanathan et al., 2005,
16 Lau et al., 2006). Additionally, a few recent studies (Lau et al., 2010, Qian
17 et al., 2011, Yasunari et al., 2010, Gautam et al., 2013) have shown that the
18 build-up of high concentrations of absorbing aerosols over the Indo-Gangetic
19 Plains (IGP) and the Himalayan foothills appears to cause a reduction in
20 snow albedo and subsequent accelerated snow/ice melt in the Himalayas dur-
21 ing the pre-monsoonal months. Combining models and measurements, Nair
22 et al. (2013) have brought out the importance of BC deposits on Himalayan
23 glaciers in impacting the radiation budget through snow albedo forcing. Un-
24 derstanding the similar effects of such aerosols on the regional climate and
25 local air quality thus becomes very important. To understand the regional
26 heterogeneity of the aerosols, the Indian Space Research Organisation (ISRO)
27 under ISRO-GBP (Geosphere Biosphere Programme) has set up a network
28 of surface observatories for the measurement of aerosol related properties un-
29 der the ARFI (Aerosol Radiative Forcing over India) (Moorthy et al., 2009,
30 Moorthy and Satheesh, 2011, Babu et al., 2013) project; has carried out air-
31 craft and high altitude balloon measurement campaigns under the ICARB
32 (Integrated Campaign for Aerosols, gases and Radiation Budget) (Moorthy
33 et al., 2008, Babu et al., 2011) and RAWEX (Regional Aerosol Warming EX-
34 periment) projects respectively. While such observational campaigns help us
35 develop a strong understanding about the aerosol scenario over specific loca-
36 tions, the numerical simulations of aerosols bring out such information for a
37 larger spatial domain. Aerosol model simulations play an instrumental role
38 in understanding the effect of aerosols on regional weather and climate pro-
39 cesses. Studies like Chakraborty et al. (2004), Ramanathan et al. (2005), Lau
40 et al. (2006), Bollasina et al. (2011) etc. have employed aerosol simulating
41 numerical models to examine the interaction between the aerosol burden over
42 the south Asian region and the monsoonal rainfall over the region. However,
43 while the use of models is essential to understand the climate implications, it
44 is equally important to evaluate the model performances against the actual
45 measurements. There have been a few recent efforts in this direction (Reddy
46 et al., 2004, Chin et al., 2009, Ganguly et al., 2009, Henriksson et al., 2011,
47 Goto et al., 2011, Kumar et al., 2011, 2012, Nair et al., 2012, Cherian et al.,
48 2013, Moorthy et al., 2013, Sanap et al., 2014, Pan et al., 2015, Govardhan

49 et al., 2015). Ganguly et al. (2009) have examined the performance of the
50 online-aerosol model GFDL-AM2 over the Indian region. The model under-
51 estimated the total column AOD over the belt of IGP by a factor close to 6
52 and BC mass concentrations by a factor of 10. A study by Govardhan et al.
53 (2015) with WRF-Chem shows that AOD and BC concentrations are under-
54 estimated by this model by a factor of 2 or more during the pre-monsoon
55 and post-monsoon periods. They have identified over- estimation of bound-
56 ary layer height, stronger winds and possible underestimation of emissions
57 as causes for this underestimation. Their results are in agreement with those
58 of Nair et al. (2012), Moorthy et al. (2013) who have evaluated the perfor-
59 mance of RegCM4, GOCART and CHIMERE chemistry transport models
60 over the Indian region in simulating the mass concentration of BC aerosols,
61 and compared these with concurrent measurements. Recently, Pan et al.
62 (2015), examined the performance of 7 chemistry transport models in simu-
63 lating AOD and mass concentrations of different aerosol species over Indian
64 region. They also found under-estimations in AOD and species mass con-
65 centration by the models vis-a-vis satellite and surface observations. They
66 attributed such shortcomings in the model to the unrealistic inventory of
67 biomass-burning emissions and simulations of RH within the models. All
68 these point to the dire need for more such evaluations, primarily to improve
69 the applicability of such models for regional and global studies.

70

71 Driven by the reasons above, in this paper, we evaluate the performance
72 of WRF-Chem simulations over the Indian region with the concurrent simula-
73 tion using another established global chemistry transport model viz. SPRINT-
74 ARS. The 2 chemistry transport models differ in many aspects including a).
75 Horizontal resolution, b). Physical domain, c). Meteorological formulation.
76 SPRINTARS uses grid nudging technique and corrects its meteorological
77 component by nudging it with NCEP-NCAR reanalysis dataset (generally
78 considered a good proxy for actual observations) once every 6 hours. WRF-
79 Chem on the other hand, does not nudge its meteorological fields with ob-
80 servations. The model simulated aerosol parameters like AOD and surface
81 concentrations of BC are inter-compared and evaluated against the concur-
82 rent observational data from satellites, and ground based network measure-
83 ments. We have attempted to understand the reasons behind similarities
84 and the differences in model simulations of AOD and BC. Additionally, with
85 the help of such inter-comparison and evaluation of model simulated BC, we
86 have suggested quantitative modifications in the anthropogenic BC emissions

87 inventories used in these models as well as in three other widely used emis-
88 sions inventories, for the BC emissions over Indian region. Model-to-model
89 comparisons of sea-salt and dust simulations are also done to examine the
90 consistency between the models, especially in view of the absence of direct
91 measurements of these species.

92

93 This paper is organised as follows- Model and simulation details are pre-
94 sented in section 2. Section 3 describes the various observational datasets
95 used in this study. Results can be found in section 4 and conclusions are
96 written in section 5.

97

98 **2. Model and Simulation details**

99 *2.1. WRF-Chem*

100 The online chemistry transport model WRF-Chem used in this study
101 has been developed at NOAA, National Oceanic and Atmospheric Admin-
102 istration. A detailed documentation of WRF is given in Skamarock et al.
103 (2008). These simulations were done over the Indian region (55⁰E-97⁰E, 1⁰N-
104 37⁰N), for 2 selected months, May 2011 (representative of the pre-monsoon
105 season) and October 2011 (representative of the post-monsoon). Lambert-
106 conformal projections were used with 12 km horizontal resolution. Cloud mi-
107 crophysics was parameterized using the Thompson scheme (Thompson et al.,
108 2004) while the Zhang-McFarlane scheme (Zhang and McFarlane, 1995) was
109 employed for cumulus parameterization. The MYJ scheme (Janjić, 2002)
110 was used to parameterize boundary layer processes, while surface processes
111 were modelled using RUC-LSM (Smirnova et al., 1997, 2000). The RRTMG
112 scheme (Mlawer et al., 1997) was used to compute long and shortwave ra-
113 diation. The chemistry in these simulations was modelled using the MOZ-
114 CART chemical mechanism option, which is an alliance between MOZART
115 (Emmons et al., 2010) mechanisms for gas-phase chemistry and the GO-
116 CART bulk aerosol scheme (Chin et al., 2002) for aerosol phase chemistry,
117 with the Fast-J photolysis scheme (Wild et al., 2000). The model consid-
118 ers the following aerosols species: BC1 (Hydrophobic), BC2 (Hydrophilic),
119 OC1 (Hydrophobic), OC2 (Hydrophilic), Dust (5 bins: effective diameters
120 from 0.5 to 8 μ m), Sea-salts (4 bins: effective diameters from 0.1 to 7.5 μ m)
121 and sulfate. The model considers aerosol transport processes like emissions,
122 advection, diffusion and deposition (dry and wet). The direct effects of the

123 aerosols are taken into account by coupling the aerosol scheme with the ra-
124 diation scheme. More details about WRF-Chem can be found in Grell et al.
125 (2005). The Initial and boundary conditions (updated every 6 hours) for me-
126 teorological variables are taken from NCEP FNL (Final) Operational Global
127 Analysis data, interpolated to model resolution. For chemistry, a global
128 chemistry transport model MOZART 4 (Emmons et al., 2010) has been used
129 for initial and boundary conditions.

130

131 We have used the standard emission pre-processor software PREP-CHEM-
132 SRC (version 1, Freitas et al. (2011)) for generating emissions of precursor
133 gases and aerosols over our domain in WRF-Chem simulations. The soft-
134 ware uses chemistry-emissions from three different emission inventories. The
135 RETRO (Schultz et al., 2007) database is used for different greenhouse and
136 precursor gases, EDGAR (Olivier et al., 1996) for emissions of CO, NO, NH₃
137 and VOCs and the GOCART database (Chin et al., 2002) is used for gener-
138 ating emissions OC, BC and SO₂ over the region. This emissions database
139 has been formulated by following the methodology described in Cooke *et al.*
140 (1999).

141 2.2. SPRINTARS

142 SPRINTARS is a global chemistry transport model fully coupled with
143 MIROC an AGCM. The meteorological fields of the MIROC are nudged
144 with NCEP-NCAR reanalysis dataset once every 6 hours. In these simula-
145 tions, SPRINTARS has been run with a horizontal resolution of $1.125^0 \times$
146 1.125^0 globally. It considers the following aerosol species: BC, OC, Dust (6
147 bins : effective radius from 0.1 to 10 μm), Sea-salts (4 bins : effective radius
148 from 0.1 to 10 μm) and sulfate. Aerosol Transport processes viz. emission,
149 advection, diffusion, deposition (dry and wet) are included in the simula-
150 tions. The direct and the indirect effects of the aerosols are incorporated
151 in the model. Takemura et al. (2000, 2009) discuss SPRINTARS in detail.
152 These simulations of SPRINTARS employed emissions from RCP 8.5 scenar-
153 ios (Riahi et al., 2007) for anthropogenic sources and GFED3 (van der Werf
154 et al., 2010) for biomass burning. The emissions of mineral dust, sea-salt
155 and DMS are calculated online within the model framework. More details
156 about the prescribed emissions in SPRINTARS can be found in Takemura
157 (2012). The archived results from the model simulations of SPRINTARS are
158 directly available at <http://sprintars.riam.kyushu-u.ac.jp/archive.html>, and

159 the same have been used in this study.

160

161 **3. Observational Data**

162 *3.1. Surface Observations*

163 The Indian Space Research Organisation (ISRO) has set up a regional
164 network (ARFINET) of several surface aerosol observation stations under
165 the ISRO- Geosphere Biosphere Program (ISRO-GBP). These observatories
166 measure near-real time mass concentrations of aerosol BC and other climatic
167 parameters (Moorthy et al., 2009, Moorthy and Satheesh, 2011, Babu et al.,
168 2013). In this study we compare model-simulated BC mass concentrations
169 with the measurements from ARFINET network stations. We have used
170 the data from 8 ARFINET stations: Bangalore (BLR), Chennai (CHN) and
171 Trivandrum (TVM) from southern India; Hyderabad (HYD) and Ananthpur
172 (ATP) from central-southern India; Ranchi (RNCH), Varanasi (VNS) and
173 Delhi (DEL) from northern India. Fig.1 shows the locations of these obser-
174 vatories.

175

176 *3.2. Satellite data*

177 We have evaluated model simulated AOD by comparing it with that de-
178 rived from MODIS (Moderate Resolution Imaging Spectroradiometer) and
179 MISR (Multiangle Imaging Spectroradiometer) satellite AOD products. For
180 MODIS, we have used the mean of the following level 3 products:- AOD at
181 550 nm from TERRA, AOD at 550 nm from Aqua and Deep blue AOD from
182 Aqua (Deep blue from TERRA is not available for simulation periods), while
183 for MISR, we have used level 3 product of AOD at 555nm from TERRA .
184 MODIS AOD data has horizontal resolution of $1^0 \times 1^0$ while MISR has a
185 spatial resolution of $0.5^0 \times 0.5^0$. The comparisons are done for the monthly
186 mean spatial patterns of AOD during May and October 2011.

187

188 **4. Results and Discussions**

189 *4.1. Black Carbon (BC)*

190 BC aerosols assume critical importance over the south Asian region, as
191 the region is considered to be a global hot-spot of BC aerosols. Previous

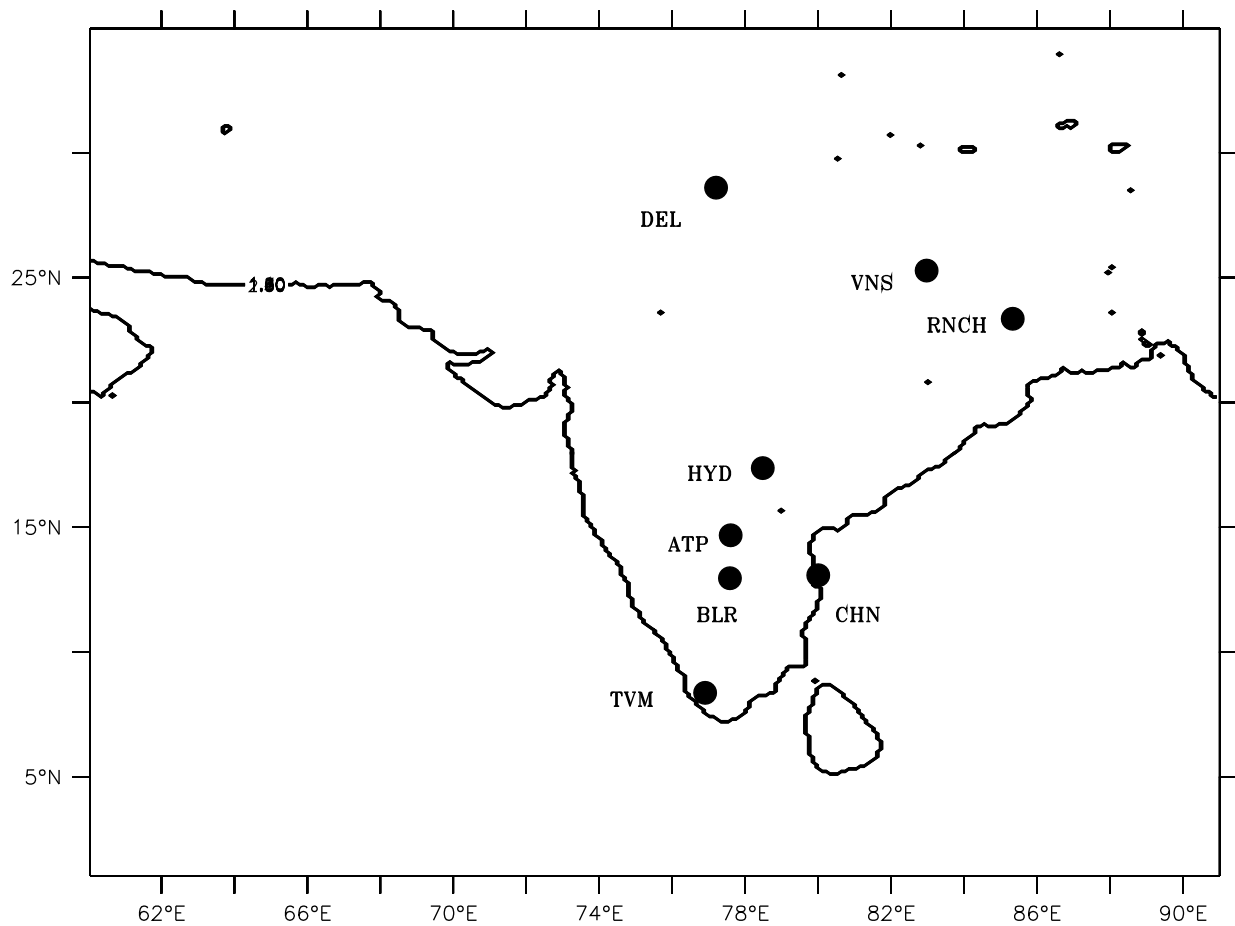


Figure 1: Surface observatories under ARFI network used in this study for measurement of BC mass concentrations.

192 studies indicate that simulations of BC over this region could be affected
193 by shortcomings in model simulated meteorology and emissions' inventories.
194 Here we inter-compare and evaluate the simulated near-surface BC over the
195 region in the two chemistry transport models which differ in many aspects.
196 We attempt to understand the reasons behind similarities and differences in
197 the model simulated BC.

198 *4.1.1. Model vs Observations*

199 A comparison of model simulations and observed near-surface BC mass
200 concentrations has been carried out with ARFINET data from observato-
201 ries at representative locations over India (Moorthy et al., 2009, Moorthy
202 and Satheesh, 2011). The comparisons are done using data at following 8
203 stations- Bangalore (12.96⁰ N, 77.58⁰E), Chennai (13.08⁰N, 80.27⁰E), Hyder-
204 abad (17.37⁰N, 78.48⁰E), Trivandrum (8.37⁰N, 76.9⁰E), Ananthpur (14.68⁰N,
205 77.60⁰E), Varanasi (25.28⁰N, 82.97⁰E), Ranchi (23.35⁰N, 85.33⁰E) and Delhi
206 (28.60⁰N, 77.20⁰E) (Fig.1). This set of stations represents a mixture of urban
207 and semi-urban locations across India. Fig.2 shows the comparison between
208 the daily mean BC near-surface mass concentrations from the models and
209 observations, for the experimental period under consideration. The red dots
210 show comparison between WRF-Chem and observations while the blue dots
211 show such a comparison between SPRINTARS and observations.

212 Both the models underestimate Near-Surface BC mass concentrations
213 (NSBC) vis-a-vis observations across all the measurement stations. The
214 underestimation is larger over Chennai, Trivandrum, Varanasi and Delhi.
215 Additionally, the models underestimate BC over the coastal stations like
216 Chennai and Trivandrum by a larger margin, than that over the in-land sta-
217 tions. The BC underestimation by the models is quantified using a parameter
218 known as Adjustment Factor (AF), which is the ratio of the observed value
219 of BC mass concentration to the model-simulated value. So, by definition,
220 higher the value of AF, higher is the magnitude of underestimation by the
221 model. The AF for WRF-Chem is written in red text, while that for SPRINT-
222 ARS is written in blue text. It appears that the AF varies from location
223 to location. Additionally, over in-land stations like Bangalore, Hyderabad,
224 Anantpur, Varanasi, Ranchi and Delhi, WRF-Chem exhibits higher values
225 of the AF vis-a-vis SPRINTARS, whereas over coastal stations SPRINTARS
226 yields a higher AF. It may be noted that, even the model which employs
227 realistic meteorological parameters (SPRINTARS) also underestimates the
228 NSBC vis-a-vis observations. Also, it can be seen that, the model simulated

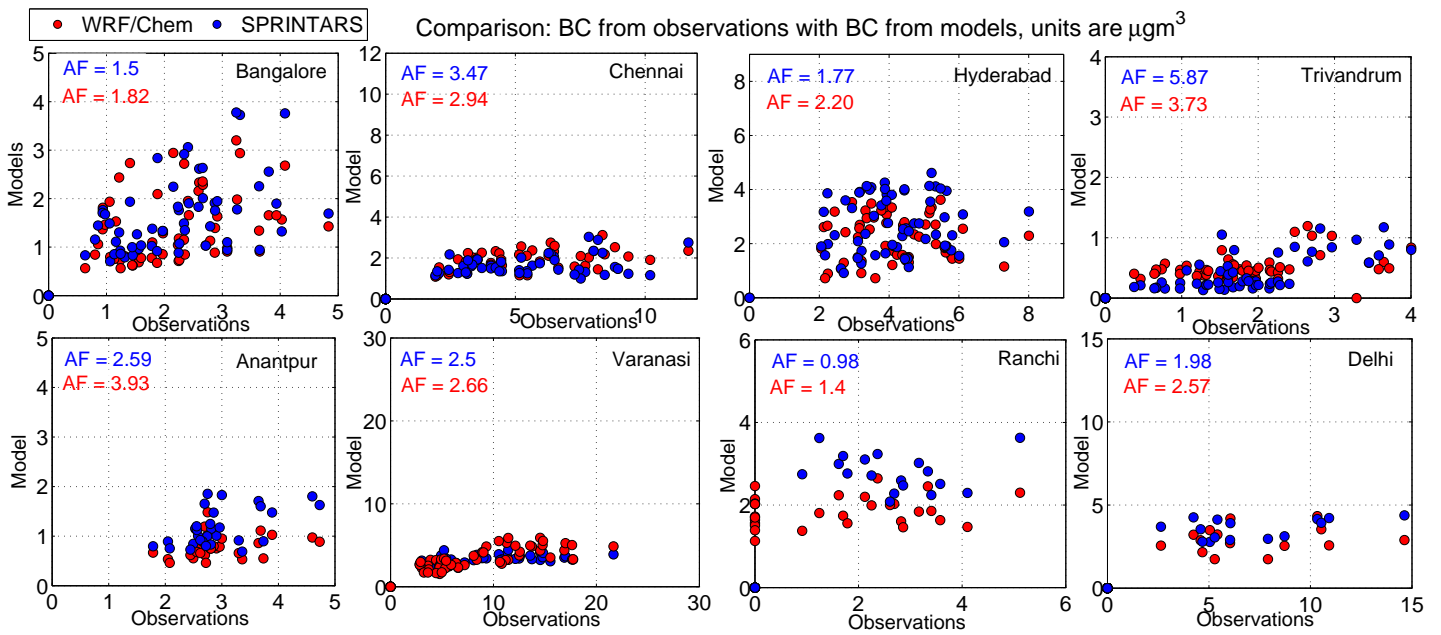


Figure 2: Comparison of model simulated BC with ARFINET daily observational data, for both the months .

229 BC do not differ significantly with respect to each other and both of them
230 largely underestimate the observations by similar margins, despite the over-
231 all differences in the modelling frameworks. To throw more light on this, we
232 inter-compare the model simulated BC in the next section.

233

234 *4.1.2. WRF-Chem vs SPRINTARS BC Simulations*

235 Given the similarities in the model simulated NSBC over the observation
236 locations, we here compare the model simulated NSBC over the entire In-
237 dian region in Fig.3. The panels 3a and 3c show the regional distribution
238 of simulated NSBC during May 2011, by WRF-Chem and SPRINTARS re-
239 spectively. The panels 3b and 3d, present the corresponding scenarios during
240 October 2011. During May 2011, WRF-Chem (Fig.3a) captured the regional
241 BC hot-spot over the IGP region with BC magnitudes going up to 3.5-4 $\mu\text{g m}^{-3}$.
242 The BC maxima occurs over eastern polluted regions like Bengal and
243 north-east India, with magnitudes reaching 4.5 $\mu\text{g m}^{-3}$. The eastern part
244 of India depicts a higher loading of BC (1-2 $\mu\text{g m}^{-3}$) vis-a-vis the western
245 part (less than 0.5 $\mu\text{g m}^{-3}$). Such a gradient would partly be due to the
246 differences in magnitude of BC emissions over these regions and also due to
247 the direction of prevailing winds over the Indian landmass. The low level
248 wind over the Indian region during the pre-monsoon season are predomi-
249 nantly westerlies and those transport the particulates towards the eastern
250 side of India. This causes the Arabian-Sea (AS) to have a lower BC loading
251 in WRF-Chem simulations (Fig.3a), while the Bay of Bengal (BoB) faces
252 a continental BC transport near the coasts of the southern-central parts of
253 India, with BC near-surface values going upto 0.5-1 $\mu\text{g m}^{-3}$. During the
254 same month, SPRINTARS (Fig.3c) also exhibits a regional BC hot-spot over
255 the GP belt, with BC values reaching upto 4-4.5 $\mu\text{g m}^{-3}$. A similar east-
256 west gradient in surface BC mass concentrations is also seen in SPRINTARS
257 simulations for the month of May 2011. A similar continental transport of
258 BC into BoB is simulated by SPRINTARS, but it extended slightly farther
259 into the Bay. In general, the spatial patterns of simulated near-surface BC
260 mass concentrations from both the models were in good agreement with the
261 pre-monsoon month, with a spatial correlation coefficient of 0.92.

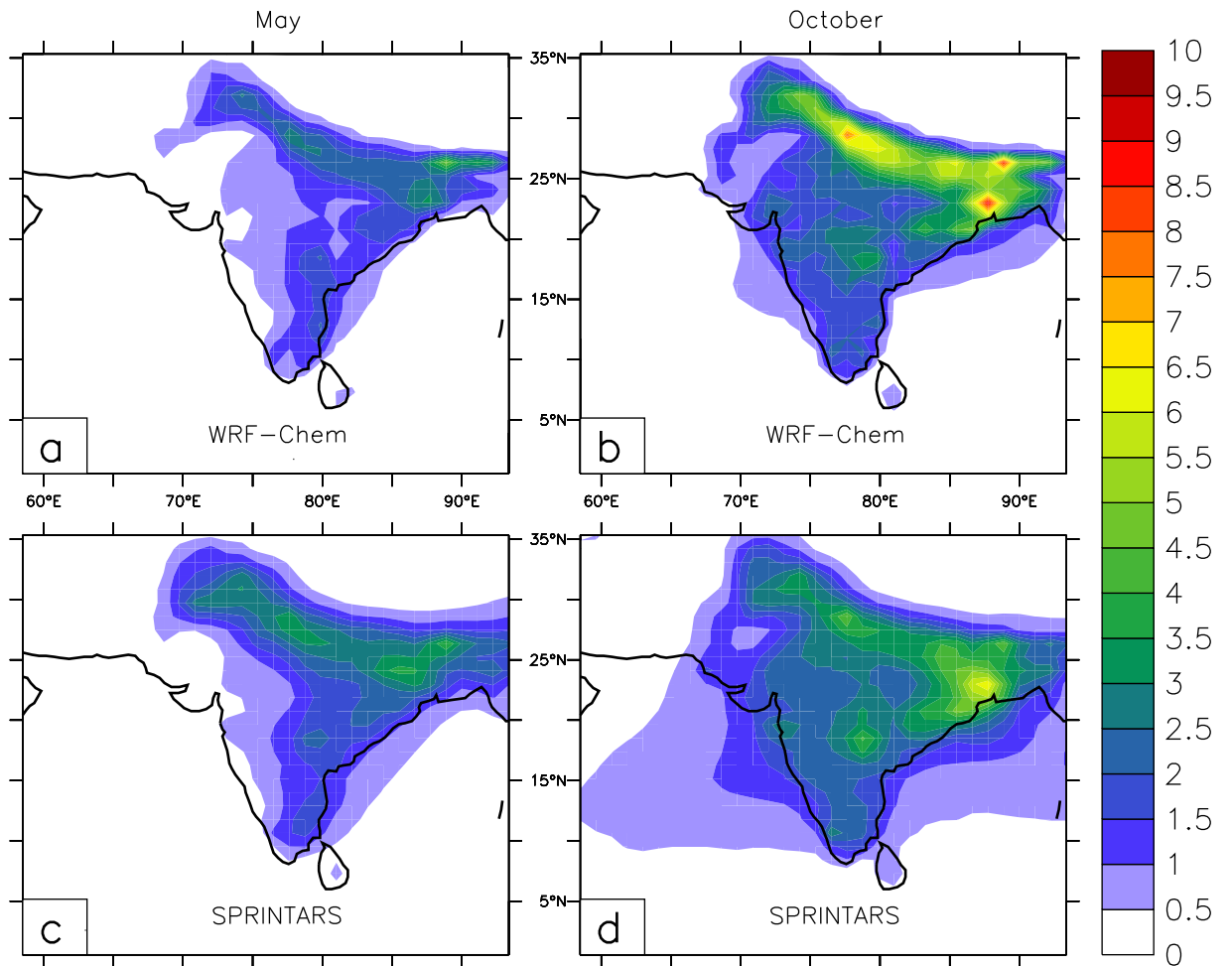


Figure 3: Comparison of model-simulated near-surface BC mass concentration ($\mu\text{g m}^{-3}$) over Indian region for both the months

262 During the post-monsoon month of October 2011, the near-surface BC
 263 mass concentration is in-general higher vis-a-vis May mainly due to convec-
 264 tively stable atmospheric conditions, which inhibit the vertical transport of
 265 surface BC (which was active during May) and confine the particulates to
 266 the lower atmospheric levels (Fig.6, Govardhan et al. (2015)). In WRF-Chem
 267 simulations (Fig.3b), this increases the BC loading over regional hot-spots
 268 like IGP to reach an average value of 6-7 $\mu\text{g m}^{-3}$ (from 3 $\mu\text{g m}^{-3}$ in pre-
 269 monsoon season), with local maxima (Delhi, Bengal coast and some spots
 270 in north-east) higher than 8.5-9 $\mu\text{g m}^{-3}$ (Fig.3b). The central Indian re-
 271 gion shows a BC near-surface loading of around 4 $\mu\text{g m}^{-3}$, while that over
 272 Southern India is seen to be around 1-2 $\mu\text{g m}^{-3}$. The east-west gradient
 273 in surface BC concentrations during the pre-monsoon season (Fig.3a and
 274 Fig.3c) decreases significantly during the post-monsoon season due to the
 275 reversed direction of low levels winds. The oceanic bodies also experience in-
 276 crements in BC loading (Fig.3b). Post monsoon, the regions adjacent to the
 277 western coast line of India (BC upto 1 $\mu\text{g m}^{-3}$) face an outflow of BC from
 278 western India due to the prevailing winds from the interior. Such an outflow
 279 of BC is also seen over the northern part of the BoB (BC : 1-1.5 $\mu\text{g m}^{-3}$),
 280 which happens to receive it from IGP. SPRINTARS simulations for the same
 281 month (Fig.3d) also show an increase in near-surface BC values. A familiar
 282 BC hot-spot over GP is seen with values reaching upto 4-5 $\mu\text{g m}^{-3}$, with a
 283 maxima over coastal regions of Bengal (BC : 6 $\mu\text{g m}^{-3}$), in SPRINTARS. The
 284 rest of the landmass (CI: 3.5-4 $\mu\text{g m}^{-3}$, SI: 2-3 $\mu\text{g m}^{-3}$) is seen to be under
 285 relatively lower BC surface loading. Similar to WRF-Chem, SPRINTARS
 286 also depicts the transport of the continental BC mass into the surrounding
 287 water bodies around India during the post-monsoon season. The AS has a
 288 BC loading of 0.5 to 1.5 $\mu\text{g m}^{-3}$ with maxima occurring near the coast and
 289 the transport of BC extending up to 60^oE. The BoB also has a higher BC
 290 loading vis-a-vis May (Fig.3c) with maxima around the northern part of the
 291 Bay, with values ranging from 0.5 to 1.5 $\mu\text{g m}^{-3}$. For both the water basins,
 292 SPRINTARS shows a farther transport of BC vis-a-vis WRF-Chem owing
 293 to its coarser horizontal resolution. As in May, during October too we found
 294 that the simulated patterns were similar with a spatial correlation of 0.91.

295 4.1.3. Quantification of differences (Model-Model)

296 In this section we quantify the similarities/differences in model simula-
 297 tions of NSBC over the complete domain. Scaling Factor-BC (SF-BC, which
 298 is the ratio of magnitude of the simulated BC in SPRINTARS to that in

299 WRF-Chem) is used to quantify the similarities and differences. So, the
 300 higher the value of SF-BC, the higher is the underestimation of BC in WRF-
 301 Chem vis-a-vis SPRINTARS. Such SF-BCs are averaged over 9 specific re-
 302 gional boxes (Fig.4) spanning the entire domain, for both the months (Table
 303 1).

304 The boxes are so chosen that all of them individually capture regions
 305 with some peculiar features, while together, they cover the main critical re-
 306 gions. It may be noted that for May (Table 1, column 2), WRF-Chem shows
 307 better comparison with SPRINTARS over the land (grid boxes like NIGP,
 308 Bengal, CI, SI and LNAS) as compared to that over oceans. Over most the
 309 land regions, SF-BC values are closer to 1, showing a relatively lesser under-
 310 estimation in WRF-Chem vis-a-vis SPRINTARS. Similarly during October
 311 2011 (Table 1, column 3), the land regions show better agreement between
 312 WRF-Chem and SPRINTARS vis-a-vis that over oceans. In-general, with a
 313 few exceptions, models show a satisfactory agreement in simulated BC mass
 314 concentrations over the Indian land-mass:-the home of BC emission sources.
 315 Thus, interestingly though the models SPRINTARS and WRF-Chem differ
 316 in many aspects, they show some kind of an agreement in the simulated
 317 BC mass concentrations. To understand this further, we next examine the
 318 emission scenarios used in these models.

Table 1: Ratio of model simulated BC (SPRINTARS BC/WRF-Chem BC) during May and October 2011

Region	May	Oct
NIGP	1.5	0.85
Bengal	1.19	0.77
CI	1.39	1.18
SI	1.11	1.2
NW	2.04	1.41
LNAS	1.39	1.75
BoB	1.45	4
HBoB	1.56	1.64
AS	1.67	2.5
NAS	1.92	2.63

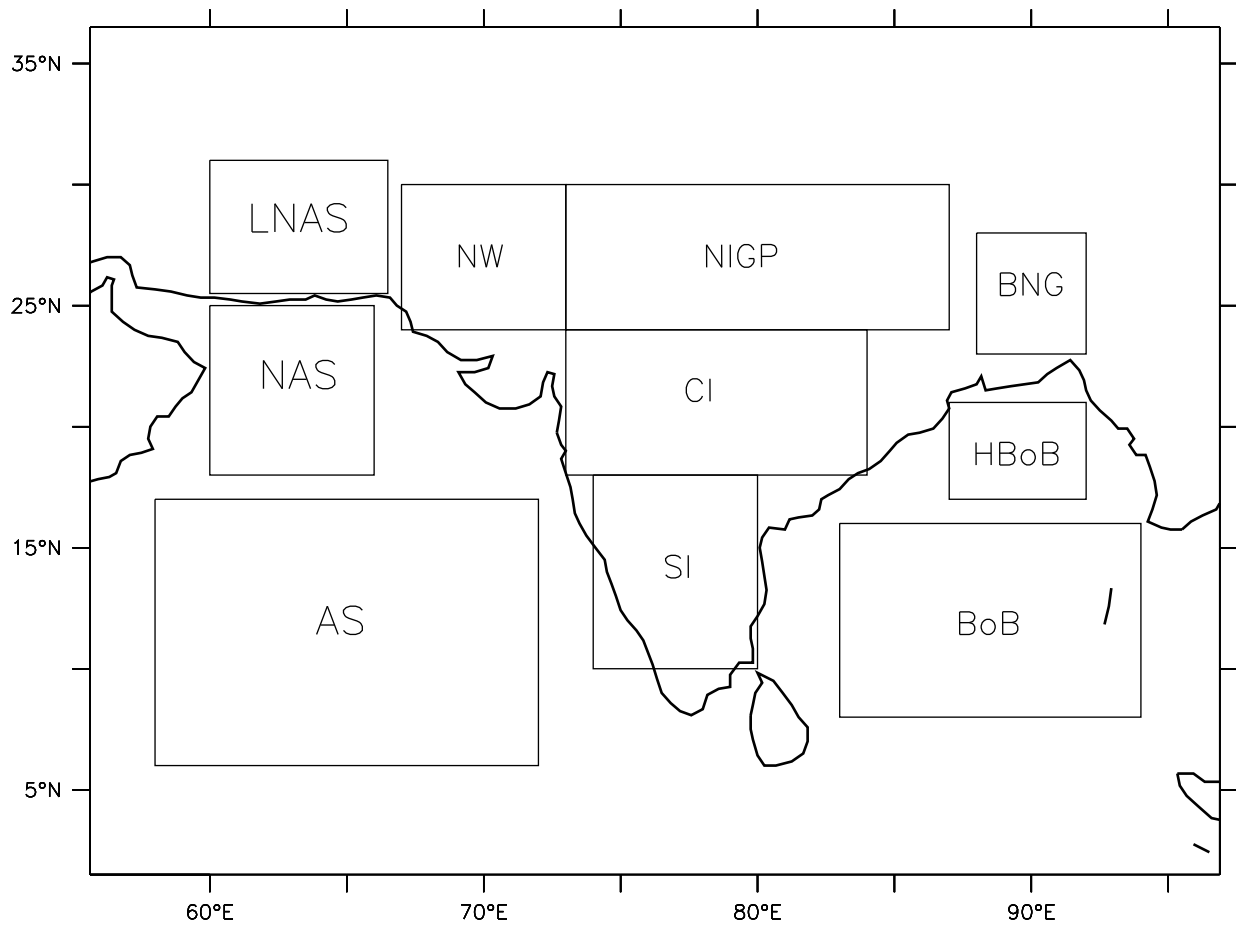


Figure 4: Regional boxes used in calculation of AF for different species. Box BNG is only used in AF for BC analysis. LNAS- Landmass North of Arabian Sea, NW- North-Western part of Indian subcontinent, NIGP- Northern India along with Gangetic Plains, BNG- Bengal, CI- Central India, SI- Southern India, NAS- Northern part of Arabian Sea, AS- Arabian Sea, HBoB- Head part of Bay of Bengal, BoB- Bay of Bengal

Table 2: Comparison between GOCART and RCP8.5 emissions scenarios, and WRF-Chem and SPRINTARS BC over the observational stations viz. BLR-Bengaluru, CHN-Chennai, TVM-Trivandrum, ANT-Anantpur, HYD-Hyderabad, DEL-Delhi, VNS-Varanasi and RNCH- Ranchi.

Location	GOCART/ RCP8.5 emissions	WRF-Chem BC/ SPRINTARS BC	Observed BC / WRF-Chem BC	Observed BC/ SPRINTARS BC
BLR	1.53	0.87	1.59	1.38
CHN	2.03	1.15	2.88	3.3
TVM	1.25	1.25	3.71	4.64
HYD	1.55	0.81	1.88	1.52
ANT	1.62	0.69	3.71	2.55
DEL	1.39	0.82	2.44	1.98
VNS	1.67	1.08	2.56	2.75
RNCH	1.58	1.08	1.13	1.21

319 *4.1.4. BC emissions inventories: WRF-Chem vs SPRINTARS:*

320 To understand the reasons behind agreements in simulated BC by WRF-
321 Chem and SPRINTARS, we in this section, compare the anthropogenic BC
322 emissions used in these 2 models. BC emissions used in WRF-Chem are
323 from GOCART database. The emissions are prepared using a standard WRF-
324 Chem utility *prep_chem_sources*. SPRINTARS on the other hand uses BC
325 emissions from RCP 8.5 (Riahi et al., 2007) concentrations scenario. The
326 emission inventories exhibit a spatial correlation coefficient of 0.97. This in-
327 dicates that, both the models (WRF-Chem and SPRINTARS) employ sim-
328 ilar spatial map of BC emissions. This could be one of the reasons behind
329 very high spatial correlation between model simulated BC. We next compare
330 the emission magnitudes in these 2 emission inventories over the observation
331 locations, in table 2.

332 Along with emission comparisons, we have also shown the comparisons
333 between simulated BC and measured BC over the station. It may be noted
334 from table 2 that, though the models differ in magnitudes of prescribed BC
335 emissions (Column 2, table 2), the differences in the simulated BC mass
336 concentrations (Column 2, table 2) are always within $\pm 20\%$ (except for
337 Anantpur). Also, both the models underestimate the observations largely
338 by similar margins (Column 4 and Column 5, table 2). Nevertheless, it
339 is seen that, over the urban stations like Bengaluru, Hyderabad, Chennai,

Table 3: Comparison between 850 hpa wind speeds in WRF-Chem, SPRINTARS and MERRA over the 10 regional grid boxes

Region	WRF-Chem/ MERRA		SPRINTARS/MERRA	
	May	October	May	October
LNAS	1.15	1.22	1.17	0.99
NW	1.28	1.53	1.17	1.03
NIGP	1.1	1.52	0.98	0.86
BNG	1.29	1.86	1.03	0.82
CI	1.39	1.17	0.98	0.95
SI	1.52	1.6	0.7	1.1
NAS	0.95	0.92	0.78	0.62
AS	1.15	1.55	1.16	0.9
HBoB	1.03	0.88	0.99	0.64
BoB	1.52	0.88	0.9	0.97

340 Varanasi, Ranchi and Delhi, as well as over the semi-urban station Anantpur
341 the emissions in GOCART are higher than that in RCP 8.5 (Column 2, ta-
342 ble 2), while the simulated concentrations are lower in WRF-Chem compared
343 to SPRINTARS (Column 3, especially for Bengaluru, Hyderabad, Anantpur
344 and Delhi). Even for other stations like Chennai, Varanasi and Ranchi, the
345 WRF simulated concentrations are not significantly higher (as could be ex-
346 pected from the corresponding higher emission) than those by SPRINTARS.
347 We next examine the possible role of meteorological parameters in governing
348 this scenario.

349 4.1.5. Simulated meteorological parameters: WRF-Chem vs SPRINTARS

350 In the light of such agreements in the simulated near-surface BC mass-
351 concentrations in-spite of the different emissions magnitudes, we compare the
352 simulated meteorological variables in the models. Simulations of low levels
353 winds and boundary layer heights have a significant effect on simulations
354 of near-surface BC mass concentrations (Govardhan et al., 2015). We here
355 compare low-level (850 hpa) wind speeds in WRF-Chem with the reanalysis
356 dataset MERRA. Same comparisons are done between SPRINTARS and
357 MERRA as well (Table 3).

358 Such comparisons are done over the 10 regional boxes within the model
359 domain (Fig.4). It can be seen that, WRF in-general overestimates 850 hpa
360 wind-speeds vis-a-vis MERRA. Such overestimations of wind are higher in

361 October vis-a-vis May over the land-only regions (LNAS to SI). SPRINTARS,
362 on the other hand shows a good comparison with MERRA, with magnitude
363 ratios being closer to 1 over most of the regional boxes within the domain,
364 during May 2011. During October 2011, SPRINTARS underestimate wind
365 speeds vis-a vis MERRA over a few regional boxes. Thus, WRF-Chem looks
366 to overestimate winds speeds over the Indian region, while SPRINTARS
367 does a slight underestimation. Since SPRINTARS is nudged with observa-
368 tions for the simulations meteorological parameters, we would thus expect
369 such agreement between SPRINTARS simulated meteorological parameters
370 and the observations. It is important to note that, near-surface wind affect
371 the horizontal transport of pollutants within lower atmosphere. Thus higher
372 the wind speeds, higher the transport of pollutants from their sources. This
373 could reduce the concentrations of pollutants in the source regions. Low-level
374 wind speeds and boundary layer heights affect simulations of BC over Indian
375 region (Govardhan et al., 2015). We would have liked to compare boundary
376 layer height simulations in WRF-Chem with SPRINTARS, but boundary
377 layer height is not stored as an output variable in SPRINTARS simulations.
378 Nevertheless, in a previous study Govardhan et al. (2015), we found that,
379 WRF-Chem over estimates the boundary layer height over the region vis-
380 a-vis MERRA. Thus it appears that, WRF-Chem overestimates meteoro-
381 logical parameters like low-level winds and boundary layer heights vis-a-vis
382 MERRA and SPRINTARS simulations. The higher the low-level wind-speed
383 and boundary layer heights the lower will be the near-surface concentration
384 of BC (Govardhan et al., 2015). Thus, given that WRF-Chem prescribes
385 higher BC emissions over the Indian region as compared to SPRINTARS, we
386 would in-general expect that the simulated BC concentrations in WRF-Chem
387 would also be higher. But due to overestimated meteorological parameters in
388 WRF-Chem, it does an enhanced ventilation of surface BC mass and hence
389 surface BC mass concentration reduces. Thus, the models WRF-Chem and
390 SPRINTARS do not differ significantly in simulated BC mass concentrations
391 due to the balance between overestimated meteorological variables in WRF-
392 Chem and lower emissions of BC in SPRINTARS. Interestingly, we noticed
393 that, even SPRINTARS, a model which employs realistic meteorology in its
394 formulation, underestimates NSBC. In presence of realistic meteorological
395 parameters, we would expect that, such an underestimation would be caused
396 primarily due to an an underestimation in prescribed emissions of BC over
397 the region. Thus, it seems that there is a possible low bias in the BC emis-
398 sions inventory used by SPRINTARS model, which could give rise to such

Table 4: Spatial correlation coefficient between GOCART, RCP8.5 and other inventories

Inventory	CC with MACCity	CC with REAS	CC with SAFAR_India
GOCART	0.87	0.91	0.76
RCP 8.5	0.9	0.93	0.78

399 underestimations of NSBC. In the next section, we attempt to quantify this
 400 low bias in BC emissions used in SPRINTARS.

401 *4.1.6. Comparison of BC emissions used in SPRINTARS and WRF-Chem*
 402 *with other emission inventories:*

403 In this section, we have compared RCP8.5 (BC emissions inventory used
 404 in SPRINTARS) and GOCART (BC emissions inventory used in WRF-
 405 Chem) with the globally widely used emission inventories viz. MACCity(Lamarque
 406 et al., 2010, Granier et al., 2011, Diehl et al., 2012), REAS(Ohara et al., 2007)
 407 and SAFAR_India (Sahu et al., 2008). Such comparisons are done for the pe-
 408 riod of May and October 2011, for which we have the model simulations.
 409 We have first examined the spatial match between the BC emission invento-
 410 rories used in the 2 models and the globally widely used emission inventories.
 411 The spatial correlation coefficient between the anthropogenic BC emissions
 412 in RCP 8.5, GOCART and that from the other inventories are listed in table
 413 4.

414 It can be seen that, GOCART and RCP 8.5 have significantly high cor-
 415 relations ($P < 0.0001$) with MACCity and REAS; but comparatively lesser
 416 correlation with SAFAR_India. Thus the spatial variation of all the 4 inven-
 417 tories are quite similar to each other, but they deviate somewhat from that
 418 of SAFAR_India. The SAFAR_India inventory has data only over India as it
 419 is generated from activity information about BC sources pertaining to India
 420 only. But, the other inventories have data over neighboring countries as well,
 421 and thus due to the lack of data in SAFAR_India inventory we get lesser
 422 spatial correlation of the other inventories with SAFAR_India. We then have
 423 done a magnitude-wise comparison of GOCART and RCP 8.5 with those
 424 inventories. Here we have taken ratios of area averaged BC emissions in GO-
 425 CART and RCP8.5 inventories with that from the other emission inventories
 426 and the same are listed in table 5. The spatial grid boxes chosen for this
 427 comparison are basically same as the ones used for BC spatial comparison
 428 between the models (Fig.4). We have selected only the land-only boxes out
 429 of those, for this comparison.

Table 5: Ratio of BC emissions in GOCART and RCP8.5 inventories with the other inventories over 4 regional grid boxes

Region	Lon,Lat	GOCART/ MACCity	GOCART/ REAS	GOCART/ SAFAR_India	RCP8.5/ MACCity	RCP8.5/ REAS	RCP8.5/ SAFAR_India
NIGP	73 ⁰ E:87 ⁰ E, 24 ⁰ N:30 ⁰ N	1.43	1.13	0.83	0.91	0.72	0.53
Bengal	88 ⁰ E:92 ⁰ E, 23 ⁰ N:28 ⁰ N	2.07	0.96	1.85	1.3	0.6	1.15
CI	73 ⁰ E:84 ⁰ E, 18 ⁰ N:24 ⁰ N	1.96	1.17	1.02	1.18	0.71	0.62
SI	74 ⁰ E:80 ⁰ E, 10 ⁰ N:18 ⁰ N	1.94	1.22	1.07	1.17	0.74	0.65

430 From Column 3-5, table 5 it can be seen that, GOCART inventory overes-
431 timates MACCity everywhere but is much closer to REAS and SAFAR_India
432 inventories. In almost all cases, GOCART emissions inventory has higher
433 emissions than the widely used inventories. On the other hand, the RCP8.5
434 scenario agrees fairly well with MACCity (Column 6-8, Table 5); but strongly
435 underestimates REAS and SAFAR_India over entire region. Thus overall,
436 it can be said that, the BC emissions used in WRF-Chem i.e. GOCART
437 emissions are comparable with the other widely use emission inventories,
438 but that in SPRINTARS (RCP8.5) looks to underestimate vis-a-vis 2 of the
439 3 widely used inventories considered in this study. But, all the available
440 emission inventories essentially provide estimates of the emissions and they
441 have some uncertainties associated with them. So, it is possible that the
442 widely used emission inventories like MACCity, REAS and SAFAR_India
443 could have some uncertainties associated with them. So comparing RCP8.5
444 with the other widely used inventories will not give us a clear idea about a
445 positive/negative bias in RCP8.5 with respect to reality. Nevertheless, in the
446 next section we try to quantify this uncertainties in RCP 8.5 BC emissions
447 as well as other emission inventories used in this study.

448 *4.1.7. Quantification of emission underestimations in RCP 8.5, GOCART*
449 *and other inventories:*

450 In general, the NSBC at a station bears a direct relationship with BC
451 emissions occurring over the station. Thus we can write,

452

$$NSBC \propto E_BC \quad (1)$$

453

Where,

454

NSBC = Near-surface BC mass concentrations over a station

456

E_BC = BC emissions over the station

458

459

Hence,

$$NSBC = K(Met) \times E_BC \quad (2)$$

460

where,

461

K(Met) = A constant which represents the effect of local meteorological conditions (low level wind speeds, boundary layer mixing, deposition etc.) on NSBC.

463

464

Extending (2) we can write, for station measurements of BC,

$$NSBC_{obs} = K(Met)_{obs} \times E_BC_{real} \quad (3)$$

465

466

and for SPRINTARS model simulations of NSBC

$$NSBC_{SPI} = K(Met)_{SPI} \times E_BC_{SPI} \quad (4)$$

467

where,

468

NSBC_{obs} = Observed Near-surface BC mass concentration

469

NSBC_{SPI} = Simulated Near-surface BC mass concentration by SPRINTARS

470

K(Met)_{obs} = K(Met) for observations

471

K(Met)_{SPI} = K(Met) for SPRINTARS

472

E_BC_{real} = Real-life BC emissions over the observational station

473

E_BC_{SPI} = BC emissions in SPRINTARS simulation over the station

474

475

Taking ratios of (3) and (4),

$$\frac{NSBC_{Obs}}{NSBC_{SPI}} = \frac{K(Met)_{Obs} \times (E_BC)_{real}}{K(Met)_{SPI} \times (E_BC)_{SPI}} \quad (5)$$

476

477

Now, for SPRINTARS, the meteorological parameters are nudged with observations at every 6 hours. Hence, we could expect that, the simulated meteorological parameters in SPRINTARS would be realistic.

478

Table 6: Multiplication factors by which the BC emissions in 5 different emission inventories are needed to be multiplied to match the BC emissions in reality, across the different observational stations viz. BLR-Bengaluru, CHN-Chennai, TVM-Trivandrum, ANT-Anantpur, HYD-Hyderabad, DEL-Delhi, VNS-Varanasi and RNCH- Ranchi.

Station	MF_RCP8.5	MF_MACC	MF_REAS	MF_SAF	MF_GOC
BLR	1.5	1.59	1.7	0.85	0.98
CHN	3.47	4.27	2.22	1.84	1.71
TVM	5.87	5.88	5.15	3.34	4.69
HYD	1.77	2.85	1.56	1.43	1.14
ANT	2.69	3.6	2.37	1.69	1.66
DEL	1.98	1.43	1.69	0.63	1.42
VNS	2.5	1.63	1.59	1.3	1.5
RNCH	1.18	1.99	0.96	1.42	0.75
Mean	2.62	2.91	2.15	1.56	1.73

479 Hence,

$$K(Met)_{obs} = K(Met)_{SPI} \quad (6)$$

480 Substituting (6) in (5) we get,

$$E_{BC_{real}} = \frac{NSBC_{obs}}{NSBC_{SPI}} \times E_{BC_{SPI}} \quad (7)$$

481 i.e.

$$E_{BC_{real}} = MF_{RCP8.5} \times E_{BC_{SPI}} \quad (8)$$

482 where,

483 $MF_{RCP8.5} = NSBC_{obs}/NSBC_{SPI}$ = Multiplication Factor for RCP8.5, a
 484 factor by which BC emissions in RCP8.5 emissions inventory are needed to
 485 be multiplied to get close to the BC emissions in reality.

486 Thus, from (8) it can be seen that, to get the realistic BC emissions
 487 in SPRINTARS model (i.e. in RCP8.5 emissions inventory), we need to
 488 multiply the original emission magnitudes by a factor, $MF_{RCP8.5}$. Such
 489 $MF_{RCP8.5}$ values are computed for all the available observational stations
 490 in Table 6. Thus with the help of such $MF_{RCP8.5}$ values we attempt to
 491 quantify the discrepancy between the BC emissions in reality vis-a-vis BC
 492 emissions in RCP8.5 emission scenario. From column 2, Table 6, it can be
 493 that, in-general, $MF_{RCP8.5}$ values vary from station to station. It may be
 494 noted that, $MF_{RCP8.5}$ shows higher values over the coastal stations like

495 Chennai (3.47) and Trivandrum (5.87). The mean values of MF_RCP8.5
496 comes out to be 2.62, while that without considering the coastal stations
497 comes out to be 1.94.

498

499 Thus, if we multiply the RCP8.5 BC emissions by the station specific
500 MF_RCP8.5, we would get the most realistic BC emissions at the particu-
501 lar station. Then the ratio of those most realistic BC emissions with BC
502 emissions in the other inventories (e.g MACCcity, REAS, SAFAR_India and
503 GOCART) would give the quantification of the underestimation in BC emis-
504 sions for those particular stations in India in these emission inventories.
505 MF_MACC, MF_REAS, MF_SAF and MF_GOC respectively represent these
506 underestimations for the various emission inventories under consideration.
507 Thus, for a station, MF_MACC= (Most realistic BC emissions / BC emis-
508 sions in MACCcity inventory) similarly, we can define MF_REAS, MF_SAF
509 and MF_GOC. Largely, it can be seen that, all the emission inventories un-
510 derestimate BC over coastal stations (Chennai and Trivandrum) by a large
511 margin. The inventory SAFAR_India shows minimum MF values. MACCcity
512 inventory appears to be the one which needs a large multiplication factor to
513 match the real-life BC emissions compared to the other inventories. Over
514 most of the stations SAFAR_India and GOCART show good agreements.
515 Thus, with this analysis, we have attempted to find out the underestimation
516 for BC emissions in currently available, widely used emission inventories. In
517 case of WRF-Chem, we notice that, the emission inventory for BC needs
518 to be modified by factors which range from 0.75 to 4.69 depending upon
519 the location. Also, such a quantification of BC emissions underestimation
520 in WRF-Chem helps us understand role of other factors (simulations of me-
521 teorological parameters like low-levels winds and boundary layer heights)
522 in affecting the BC underestimation in WRF-Chem. A similar comparison
523 of multiple emissions inventories for anthropogenic NO_x emissions has also
524 been carried out recently in Jena et al. (2015). They found that all those in-
525 ventories overestimate the anthropogenic NO_x emissions over Indian region,
526 mainly due to the errors in activity related data.

527

528 *4.2. Aerosol Optical Depth (AOD)*

529 AOD is one of the most important parameters used in the assessment of
530 climate impacts of aerosols. It represents the fraction of incident light (of any
531 specific wavelength) scattered and absorbed by total column aerosol loading.
532 Owing to its larger spatial coverage in comparison with station observations,
533 we have compared model simulated AOD (550 nm) with that from MODIS
534 and MISR satellite products, in-spite of known uncertainties (Kahn et al.,
535 2007, Levy et al., 2010, Remer et al., 2005, Zhang et al., 2005).

536 *4.2.1. Uncertainties in satellite retrievals of AOD:*

537 In-general, MODIS instrument on-board Terra and Aqua satellites, quote
538 the following pre-launch uncertainties in the AOD values (King et al., 1999,
539 Remer et al., 2005).

540 $\Delta\tau = \pm 0.05 \pm 0.15 \times \tau$ over land

541 $\Delta\tau = \pm 0.03 \pm 0.05 \times \tau$ over ocean where, $\tau = AOD$

542 There are many studies (Jethva et al., 2007b,a, Choudhry et al., 2012,
543 More et al., 2013, Bibi et al., 2015, Misra et al., 2015) which have evalu-
544 ated the MODIS and MISR retrieved AOD over location across India with
545 the corresponding ground based measurements from AERONET network.
546 All these studies highlight the limitations of satellite retrievals of AOD. The
547 AOD retrieved using DeepBlue algorithm from MODIS instrument, in gen-
548 eral underestimates stations measurements of AOD over different stations
549 across India (More et al., 2013, Bibi et al., 2015, Misra et al., 2015). Never-
550 theless, satellites provide a regional picture of aerosol loading. Hence, despite
551 certain issues with satellite estimates of AOD, we still use them to compare
552 the models' performances; mainly due to their spatial coverage.

553 *4.2.2. Intercomparison and evaluation for May 2011*

554 An examination of satellite AOD products, namely, MODIS (Fig.5a) and
555 MISR (Fig.5c) for the pre-monsoon month (May) over the simulation do-
556 main, reveals the following features a) a regional AOD hot-spot over the
557 IGP and Sindh region (MODIS : 0.7-0.8, MISR : 0.6-0.7) b) higher AOD
558 (upto 0.55) over the Eastern coast of India. c) moderate AOD (upto 0.60)
559 over the rest of the Indian landmass d) an AOD east-west gradient across
560 the Indian land mass with a higher AOD (upto 0.7) over the BoB and a
561 relatively lower aerosol burden over AS (AOD : 0.45). e) the moderately
562 loaded west coast of India with AOD that reaches upto 0.5. It can also be
563 noted that, though the satellite products agree with each other over most

564 parts of the study domain, they differ significantly over AOD hot-spots like
565 IGP and Sindh. MODIS overestimates AOD over those regions vis-a-vis the
566 MISR product.

567

568 For the same month, WRF-Chem (Fig.5e) shows an AOD hot-spot over
569 the Gangetic plain (GP) region with AOD values ranging upto 0.4-0.45. Such
570 a hot spot is a combined result of the abundance of high concentrations of
571 anthropogenic aerosols (BC) over the densely populated belt (Fig.3a) and the
572 seasonal transport of natural aerosols (mineral dust) due to the prevailing
573 direction of the winds (Lau et al., 2006). Higher values of AOD (0.4-0.45)
574 are also seen over the eastern coast of India, which could also be associated
575 with anthropogenic aerosol mass over the belt (Fig.3a). Over the rest of the
576 land mass of India, WRF-Chem shows a relatively lower aerosol burden with
577 AOD ranging upto 0.2-0.3. Over the BoB, WRF-Chem exhibits a higher
578 AOD (upto 0.4), which could be mainly attributed to the transport of the
579 continental aerosol mass from the eastern-coast of India into the Bay, due
580 to the direction of prevailing wind fields over the region during this season.
581 WRF-Chem also simulates a higher AOD (upto 0.3) over the northern part
582 of AS, which could be related to the transport of mineral dust from the re-
583 gion north-west of it. The rest of the AS has aerosol loading with an average
584 AOD of about 0.15. Of the 2 basins, the BoB is seen to have a larger burden
585 of aerosol mass during the month of May, and this could be due to the ad-
586 vection of BC from the densely populated GP into the Bay. So, WRF-Chem
587 successfully simulates the east-west gradient in AOD over the basins lying
588 on either side of India.

589

590 For the same month, SPRINTARS (Fig.5g) exhibits a very high AOD over
591 the semi arid region around Sindh with values reaching upto 1.3. The main
592 cause for such a high AOD could be the presence of mineral dust aerosols over
593 the region, which arise due to the drier conditions of the underlying surface.
594 SPRINTARS also shows a secondary AOD hot-spot over the GP regions
595 (AOD : 0.7), the possible causes for which could be an anthropogenic aerosol
596 mass and/or mineral dust loading. The AOD over the rest of the Indian land
597 mass in SPRINTARS is about 0.4-0.45. Over the BoB, SPRINTARS shows
598 a lower AOD vis-a-vis WRF-Chem, with a mean value around 0.3, and a
599 spatial pattern similar to that of WRF-Chem (Fig.5e). Over the northern
600 part of AS, SPRINTARS exhibits a higher AOD (average value of 0.55) the
601 cause of which could be the heavy dust loading over the northern part of it.

602 The AOD decreases away from the coast in the interior AS. In general the
603 models are in better agreement in the simulation of AOD over the oceanic
604 regions vis-a-vis land.

605 *4.2.3. Intercomparison and evaluation for October 2011*

606 A reduction in AOD during October 2011 was seen in both the satellite
607 AOD products, MODIS (Fig.5b) and MISR (Fig.5d). Nevertheless, both the
608 products exhibited a regional AOD hot-spot over GP with AOD values reach-
609 ing upto 0.8 in MODIS and 0.7 in MISR. Over the rest of Indian land mass,
610 also, satellite products show a reduction in AOD with a high AOD (0.5) over
611 the southern peninsula, seen clearly in MODIS (Fig.5b). Over the BoB, both
612 the products show moderate AOD values reaching upto 0.5, still lower than
613 May. A peculiar spatial pattern of AOD is seen in both the products over
614 AS with maxima (AOD : 0.45) occurring near the western coast of India.
615 During October 2011, in general, the aerosol burden over the entire domain
616 decreases. Mineral dust dominates this aerosol blanket over the Indian region
617 (on the basis of total mass) during pre-monsoonal months. Such a reduction
618 in aerosol loading during October mainly occurs because of reduction in the
619 mineral dust mass due to a) scavenging of the particulates by the monsoonal
620 rainfall b) reduced dust emissions from the source region due to wetter sur-
621 face conditions and c) absence of the assisting winds which cause dust influx
622 into the Indian region during pre-monsoonal months. This reduction in AOD
623 is well captured by both the models (WRF-Chem Fig.5f and SPRINTARS
624 Fig.5h). WRF-Chem (Fig.5f) still exhibited an AOD hot-spot over IGP with
625 values reaching upto 0.4. The rest of the land mass was seen to be under
626 a lower aerosol burden with the AOD around 0.2. The higher AOD over
627 the BoB which prevailed during May (Fig.5e), decreases to a relatively lower
628 value around 0.1. Such a large reduction in AOD was not seen over AS (ex-
629 cept the northern part). The AOD over the central (0.15) and eastern parts
630 (0.2) of AS was still comparable to that in May (Fig.5e), while a reduction
631 in AOD was seen over the northern part of AS. During the same month,
632 SPRINTARS simulations (Fig.5h) exhibited a very strong reduction in AOD
633 vis-a-vis that in May (Fig.5g). The AOD over the entire Indian land mass
634 (except southern India and some parts of the eastern coast) decreased to very
635 low values (0.1-0.2). The southern part of India was seen with a moderately
636 high AOD upto 0.3 while that over the eastern-coast was seen to be around
637 0.25. Aerosol loading over the BoB completely decreased with an average
638 value not exceeding 0.1. Nevertheless, such a large reduction in AOD was

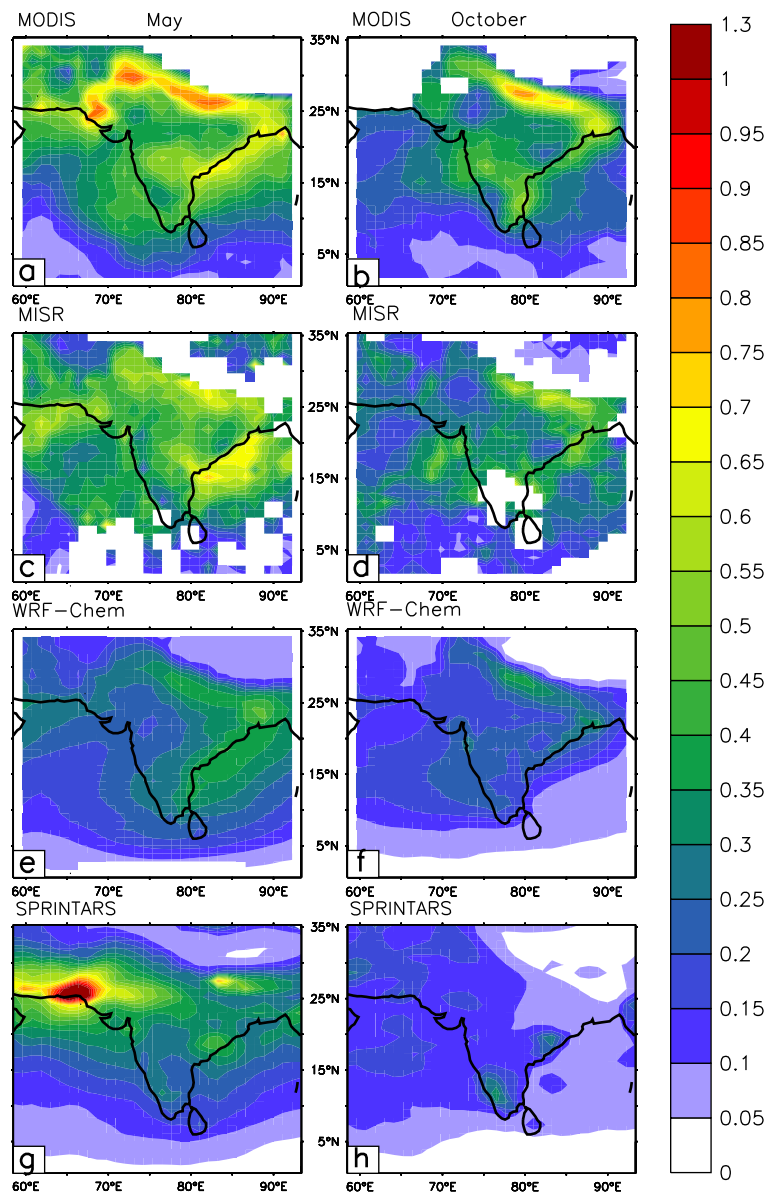


Figure 5: Monthly mean AOD (550nm) over Indian region for a)MODIS,May 2011 b) MODIS, October 2011, c) MISR, May 2011 d) MISR, October 2011 e) WRF-Chem May 2011 f) WRF-Chem, October 2011 g) SPRINTARS, May 2011 h) SPRINTARS, October 2011

639 not seen over AS (except the northern part), the average AOD over the entire
640 AS attained an average AOD value upto 0.15. The AOD gradient switched
641 its polarity vis-a-vis May, in both the model simulations (WRF-Chem Fig.5f
642 and SPRINTARS Fig.5h) which was not that clearly seen in the satellite
643 products (MODIS Fig.5b and MISR Fig.5d)

644 4.2.4. Quantification of the AOD differences

645 It can be clearly seen that both the models (WRF-Chem Fig.5e, Fig.5f
646 and SPRINTARS Fig.5g, Fig.5h) underestimate the AOD vis-a-vis satellite
647 products (MODIS Fig.5a, Fig.5b and MISR Fig.5c, Fig.5d) over the entire
648 domain with a few exceptions. To quantify the AOD underestimation, we
649 define a parameter ‘Adjustment Factor AOD’ (AF-AOD). AF-AOD is the
650 ratio of the satellite AOD and the model AOD. We computed AF-AOD
651 for 9 different regional boxes (Fig.4) of our complete domain and they are
652 listed in Table 7 (for May 2011) and Table 8 (for October 2011). From the
653 AF-AOD analysis for May (Table 7) it can be seen that, the WRF-Chem
654 (Column 2 and 3) shows higher AF-AOD values vis-a-vis SPRINTARS, over
655 the entire Indian landmass except SI for both the satellite products. WRF-
656 Chem does maximum underestimation of the AOD over the LNAS, NW
657 and NIGP regions (Avg. AF-AOD LNAS= 1.84, NW=2.1, NIGP=1.84);
658 which have large anthropogenic and natural aerosol loading. Over rest of
659 the landmass it shows an average AF-AOD of 1.57. Over the oceans, WRF-
660 Chem shows lower AF-AOD vis-a-vis SPRINTARS, with 1.66 over the AS
661 and 1.55 over the BoB. On the other hand, SPRINTARS over LNAS and
662 NW actually overestimates the AOD with an AF-AOD less than 1. Even
663 over rest of the land mass, SPRINTARS shows a lower AF-AOD value (1.5)
664 vis-a-vis WRF-Chem. Over the oceans, SPRINTARS underestimates AOD
665 over most of the water-mass except NAS.

666 For October (Table 8), unlike for May, WRF-Chem (Column 2 and 3)
667 has better simulations of AOD (with lower AF- AOD with both the satellite
668 products), over the entire domain (land and oceans) vis-a-vis SPRINTARS
669 (column 4 and 5). Over land, both the models show a higher AF-AOD in Oc-
670 tober vis-a-vis May. WRF-Chem shows a large AF-AOD over AOD hotspots
671 like LNAS (2.03), NW (1.47) and NIGP (1.76), while over the rest of the
672 landmass, it exhibits relatively lower AF-AOD (1.62). Over the BoB, both
673 the models show a larger AF-AODs during October vis-a-vis May. WRF-
674 Chem presents a mean AF-AOD of 2.78 over the entire BoB, and an AF-AOD
675 of 1.46 over AS. For the same month, SPRINTARS (column 4 and 5), shows

Table 7: Adjustment factor between models and satellite data, for **May, 2011**. AF= Satellite AOD / Model AOD

Region	WRF-Chem		SPRINTARS	
	MODIS	MISR	MODIS	MISR
Land				
LNAS	1.93	1.75	0.68	0.62
NW	2.38	1.824	1	0.77
NIGP	1.9	1.79	1.524	1.44
CI	1.64	1.61	1.3	1.29
SI	1.63	1.41	1.88	1.63
Ocean				
NAS	1.67	1.8	0.97	1.07
AS	1.55	1.64	1.84	1.94
HBoB	1.63	1.61	1.97	1.94
BoB	1.34	1.47	1.95	2.14

676 a very high AF-AOD over NIGP (3.85), relatively lower AFs over LNAS,
677 NW and an average AF around 2.35 over the rest of the landmass. Over the
678 oceans, SPRINTARS does a large underestimation with an AF-AOD of 4.1
679 over the BoB, but does simulate AOD better over AS with a relatively lower
680 AF-AOD of 1.68. So, while both the models underestimate AOD vis-a-vis
681 satellite observations, SPRINTARS has a lower AF-AOD during May and
682 a very large AF-AODs during October. WRF-Chem has a comparable AF-
683 AOD during both the months and a lower AF-AOD vis-a-vis SPRINTARS
684 during October.

685

686 Additionally, WRF-Chem shows a higher spatial correlation with the
687 MODIS and MISR AOD spatial pattern than SPRINTARS (Table 9), for
688 both months. It can also be seen that there are larger differences in mod-
689 els' spatial correlation coefficients with satellites for the month of October.
690 SPRINTARS shows a very low correlation with satellite AOD products dur-
691 ing October, while WRF-Chem replicates the pattern of satellite AOD during
692 both months with significantly high correlation coefficients. So, WRF-Chem,
693 while underestimating satellite AOD, mimics the AOD pattern significantly
694 well. So, both models while underestimating AOD vis-a-vis satellite obser-

Table 8: Adjustment factor between models and satellite data, for **October, 2011**. AF= Satellite AOD / Model AOD

Region	WRFChem		SPRINTARS	
Land	MODIS	MISR	MODIS	MISR
Sindh	NA	2.03	NA	1.53
NW	NA	1.47	NA	1.69
NIGP	1.81	1.71	3.96	3.75
SI	1.87	NA	2.34	2.17
CI	1.52	1.46	2.49	2.4
Ocean				
NAS	1.46	1.47	1.45	1.54
AS	1.3	1.62	1.66	2.08
HboB	2.19	2.07	5.13	4.86
BoB	3.18	3.47	3.04	3.31

Table 9: Spatial correlation between the AOD simulated by the models and retrieved by the satellite products

Model	Model and MISR		Model and MODIS	
	May	Oct	May	Oct
SPRINTARS	0.56	0.27	0.63	0.33
WRF-Chem	0.78	0.71	0.82	0.81

695 vations, significantly differ in simulating AOD over the Indian region. So,
696 the similarities seen in model simulated patterns of BC completely disappear
697 when model simulated AOD patterns are compared.

698

699 The possible cause for this difference between models could be related
700 to the simulation of various aerosol species. We next examined the concen-
701 tration of various aerosol species in order to determine the cause of these
702 differences. Here, we have examined major natural aerosols present over the
703 Indian region viz. dust and sea-salt. We additionally, re-examined the role of
704 BC simulations from models in causing such differences in AOD distributions.

705 *4.3. Mineral Dust*

706 We examined simulations of near-surface mineral dust mass concentra-
707 tions over the entire study domain for May and October, using both the
708 models (Fig.6). Near surface dust concentration is controlled by emission
709 (related to wind speed, erodability and soil moisture), advection and depo-
710 sition.

711 *4.3.1. Comparison of Dust simulations, during pre-monsoon*

712 During the pre-monsoonal month (May), WRF-Chem (Fig.6a) shows an
713 in-flux of a large dust mass into the Indian region due to the action of pre-
714 vailing surface winds in the dry and semi-arid regions of Arabia and the
715 Sindh and Thar deserts. The main dust source region appears to be around
716 the Thar desert with mass concentrations reaching as high as $400 \mu\text{g m}^{-3}$.
717 Additionally, secondary sources are also seen over the deserts of Sindh and
718 Arabia with average dust mass concentrations around $200 \mu\text{g m}^{-3}$. The en-
719 tire Indian landmass (except North-east India) is seen to be affected by the
720 dust inflow from these source regions. The regions closer to the dust sources
721 (the Central and North-western part of India) show near-surface dust mass
722 concentrations as high as $120 \mu\text{g m}^{-3}$, while those farther away from the
723 sources show concentrations more than $30 \mu\text{g m}^{-3}$. Such a large dust mass
724 is also seen over the oceanic part also, especially over the AS. The northern
725 part of the sea bears a heavy dust cover with a dust mass concentration
726 higher than $90 \mu\text{g m}^{-3}$, which is comparable to inland regions like the Cen-
727 tral and North-western part of India. The central part of the AS experiences
728 a relatively clearer atmosphere with lower dust mass concentrations (up to
729 $30 \mu\text{g m}^{-3}$), as it is relatively distant from the dust source regions. For the
730 same month, SPRINTARS (Fig.6c) also simulates heavy dust loading over
731 most parts of the study domain. The main dust source in SPRINTARS sim-
732 ulations is seen to be around the Sindh region with a dust loading of more
733 than $600\text{-}800 \mu\text{g m}^{-3}$, which is twice that over the WRF-Chem dust source.
734 Such a heavy dust cover is seen to be spread across an area of 7° long \times
735 3° lat. Additionally, secondary dust sources are seen to be over the Thar
736 desert and GP with dust mass concentrations as high as $350\text{-}500 \mu\text{g m}^{-3}$.
737 Dust production from these source regions affects the entire northern part of
738 India due to transport by the prevailing wind fields, resulting in dust mass
739 concentrations around $90 \mu\text{g m}^{-3}$ near the source regions and $30\text{-}60 \mu\text{g m}^{-3}$
740 away from the source regions. The southern part of India in SPRINTARS
741 simulations is seen to be relatively free from such a dust blanket. Most part

742 of the oceanic regions of the study domain have a lower dust loading (less
 743 than $30 \mu\text{g m}^{-3}$) in SPRINTARS simulations, except the northern part of
 744 AS. This region, being close to the dust source region, is loaded with near-
 745 surface dust with mass concentrations ranging from $30\text{-}90 \mu\text{g m}^{-3}$. Thus,
 746 during the pre-monsoonal month (May), though both the models simulate
 747 dust production over the desert regions and its advection into the rest of the
 748 Indian region, they differ in the details. While WRF-Chem (Fig.6a) shows
 749 a dust source around the Thar desert, SPRINTARS shows a more intense
 750 and larger dust source around the Sindh region. Also, the secondary dust
 751 source over GP in SPRINTARS is not seen in WRF-Chem simulations. Ad-
 752 ditionally, the magnitudes of dust mass concentrations over the dust sources
 753 in SPRINTARS are significantly higher vis-a-vis WRF-Chem. In addition
 754 to the differences in dust sources, the models also differ in simulating the
 755 transport of dust over the domain. WRF-Chem (Fig.6a) shows larger in-
 756 land transport, with dust covering almost the entire Indian land-mass; in
 757 contrast, the transport is limited to central India in SPRINTARS (Fig.6c).
 758 Oceans are seen to be almost free from the dust blanket (except northern
 759 AS) in SPRINTARS (Fig.6c), while WRF-Chem exhibits a moderate dust
 760 loading over the central to northern part of AS.

761 4.3.2. Comparison of Dust simulations, during post-monsoon

762 For the post-monsoonal month (October), WRF-Chem (Fig.6b) and SPRINT-
 763 ARS (Fig.6d) simulate a reduction in the overall dust burden over the re-
 764 gion, mainly attributed to the reduced production of dust aerosols due to
 765 damp surface conditions and the removal of atmospheric dust loading due
 766 to the monsoonal rainfall. Nevertheless, WRF-Chem (Fig.6b) still exhibits a
 767 dust source over the same regions (Thar, Sindh and Arabia) as seen in May
 768 (Fig.6a), though the magnitudes have almost decreased to $(1/4)^{th}$ vis-a-vis
 769 May. The inland transport of dust also decreases with the magnitudes going
 770 down to $(1/3)^{rd}$ vis-a-vis May due to weaker and reversed wind fields during
 771 the post monsoon period compared to the pre monsoon. A similar reduction
 772 in dust is also seen over AS with concentrations going down to $(1/4)^{th}$ that in
 773 May. For the same month, SPRINTARS (Fig.6d) shows a reduction in dust
 774 mass concentrations over the source regions (mainly Sindh and Arabia) by a
 775 factor close to 4-5 as compared to May (Fig.6c). The dust source over GP,
 776 which was active during May in SPRINTARS does not appear in the October
 777 simulations. SPRINTARS also simulates a reduction in the inland transport
 778 of dust into the Indian land-mass. The dust cover over AS also gets thinner

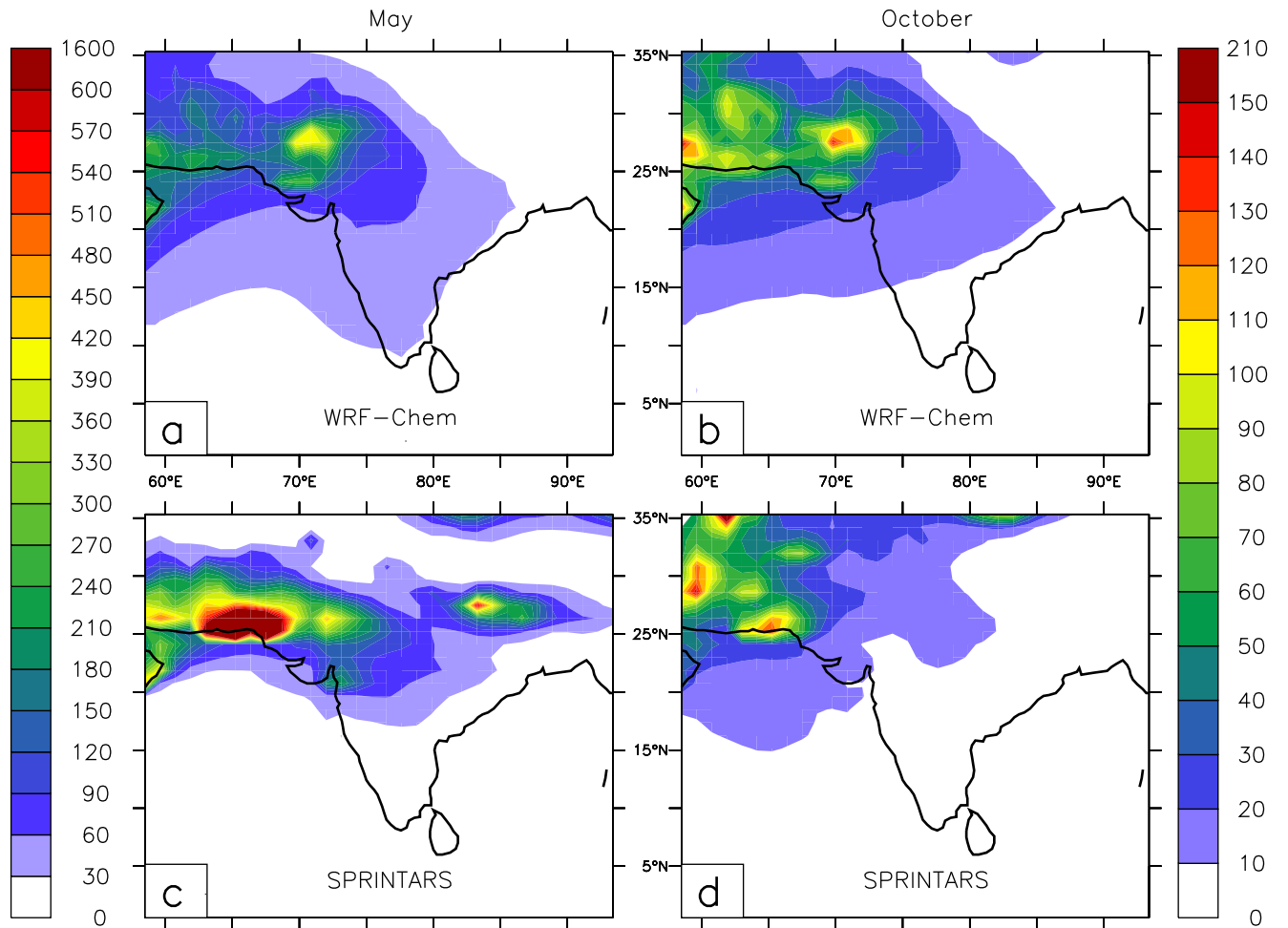


Figure 6: Comparison of model-simulated near-surface dust concentration ($\mu\text{ g m}^{-3}$) over Indian region for May and October 2011. Left colorbar should be read for May and right one should be read for October

779 in October in SPRINTARS vis-a-vis May by a factor of 3 over the Northern
780 AS. As in May, the models differ in simulating dust during October, also.
781 The main differences in the model simulations of dust in October appear to
782 be with the location of dust sources and the inland transport of dust.

783 *4.3.3. Quantification of dust differences*

784 To quantify such differences between model simulated near-surface dust
785 mass concentrations, we computed the Scaling Factors (SF-Dust). Here,
786 SF-Dust is the ratio of dust mass concentration in WRF-Chem to that in
787 SPRINTARS. Please note the difference between SF-Dust used in this section
788 and SF-BC used previously. SF-Dust for different locations is calculated for
789 the same 9 regional boxes (Fig.4) and is presented in Table 10. It may be
790 noted that for the month of heavy dust loading (May, column 2, Table 10),
791 SPRINTARS exhibits a higher dust mass vis-a-vis WRF-Chem over most of
792 the landmass (except SI) and over the northern part of AS, resulting in a
793 SF-Dust lower than 1. Large differences are especially seen over regions of
794 LNAS (SF=0.39) and NIGP (SF=0.56). The differences in modelled dust
795 concentrations near the dust source locations (regions like LNAS, NW and
796 NAS) could be related to the differences in the production of dust aerosols
797 within the models, while that over the regions farther away (SI, BoB, HBoB
798 and AS) could be related to the differences in the transport of dust aerosols
799 within the models. For May, the models exhibit a SF-Dust lower than 1 over
800 LNAS, NW and NAS. This points towards a possible higher production of
801 dust aerosols in SPRINTARS vis-a-vis WRF-Chem. For the same month,
802 the models exhibit SF-Dust greater than 1 over regions like AS, BOB, HBoB
803 and SI, which hints at a weaker transport of dust in SPRINTARS vis-a-vis
804 WRF-chem. So, it may be noted that, while SPRINTARS exhibits higher
805 production of dust aerosol for May, it mimics a weaker transport of those
806 aerosols away from the source region vis-a-vis WRF-Chem. For the month of
807 October (Column 3, Table 10), WRF-Chem overestimates dust concentration
808 over most of the domain (except LNAS). The models are in better agreement
809 over the surrounding oceanic regions such as NAS, AS and BoB vis-a-vis
810 the Indian landmass. Large differences are particularly noticeable over NW
811 (SF-Dust=2.62), due to a mismatch between model simulated dust source
812 locations and CI (SF-Dust= 2.14), SI (SF-Dust=2.59), due to a subsequent
813 lower inland transport of dust.

Table 10: Scaling factor between models for dust during May and October 2011 SF-Dust=
WRF Dust/SPRINTARS Dust

Region	SF May	SF October
Sindh	0.39	0.97
NIGP	0.56	1.84
NW	0.69	2.62
CI	0.85	2.14
SI	1.9	2.59
NAS	0.66	1.31
AS	2.4	1.23
BoB	2.19	1.53
HBoB	2.58	3.34

814 *4.3.4. Potential Causes of Discrepancy*

815 To understand the causes behind such differences between the dust sim-
816 ulations in the models, we first examined the formulation of the production
817 of dust aerosols in both the models. Takemura et al. (2000, 2009) discuss
818 the emission methodology employed in SPRINTARS, while WRF-Chem em-
819 ploys the dust emission model discussed in Ginoux et al. (2001). In general,
820 dust particles become airborne due to the action of near-surface winds on
821 the underlying surface. The particles get lifted up from the surface beyond
822 a particular value of wind-speed, termed as the threshold wind speed. The
823 rate of production of dust aerosols is directly related to wind magnitudes
824 beyond the threshold wind-speed. Also, such a lifting of mineral dust parti-
825 cles from the surface is controlled by soil moisture. The higher Soil Moisture
826 (SM) level, the lower is the lifting of particles from the surface and hence the
827 lower is the production of dust aerosols. Both the models (WRF-Chem and
828 SPRINTARS) propose these 2 criteria for dust emissions, but they differ in
829 the details.

830

831 The models employ different threshold wind speeds (U_t) for the lifting
832 of dust aerosols from the surface. SPRINTARS assumes a value of 6.5 m
833 s^{-1} as the threshold wind speed (U_t), while WRF-Chem calculates U_t as a
834 function of dust particle diameter, density and SM levels within the grid box.
835 To understand the differences in these 2 dust emission models, we plotted
836 the sensitivity of dust emissions in both the models to 10m wind speeds
837 (U_{10}) (Fig.7). In Fig.7 dust emission/production rates in the 2 models are

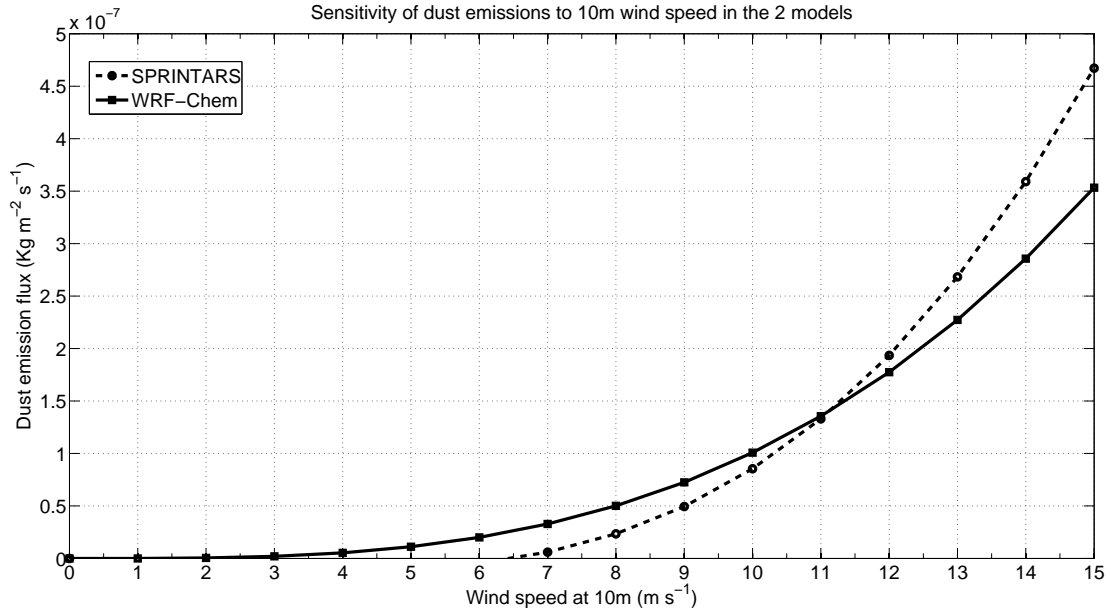


Figure 7: Sensitivity of model-simulated dust emissions to U_{10} . Here, dust emissions are calculated by considering particle diameter to be $1.4 \mu\text{m}$, particle density to be 2500 kg m^{-3} $SM=0.001 \text{ m m}^{-1}$

838 plotted as a function of U_{10} , for the particles with diameter $1.4 \mu\text{m}$, density
 839 2500 kg m^{-3} and with a fixed SM value of 0.001 m m^{-1} . It may be noted
 840 that for both the models WRF-Chem and SPRINTARS, dust emissions are
 841 directly proportional to 10m wind speeds beyond a threshold value. But, the
 842 threshold values for the models differ by a large margin. For WRF-chem, it
 843 happens to be around 2 m s^{-1} , for the specific values of the aforementioned
 844 parameters, while for SPRINTARS it is 6.5 m s^{-1} . Also, it could be noted
 845 that, for lower U_{10} values WRF-Chem has higher dust emissions vis-a-vis
 846 SPRINTARS, while at higher U_{10} values ($U_{10} > 11 \text{ m s}^{-1}$), SPRINTARS
 847 show higher dust emissions vis-a-vis WRF-Chem.

848 During pre-monsoon month, in-general U_{10} values could go higher than
 849 11 m s^{-1} over the dust source region in these 2 models, which could give
 850 rise to higher dust emissions in SPRINTARS vis-a-vis WRF-Chem. While,
 851 during Post-monsoon, the U_{10} magnitudes would in-general be low ($U_{10} <$
 852 11 m s^{-1}) and thus could cause higher dust emissions in WRF-Chem vis-

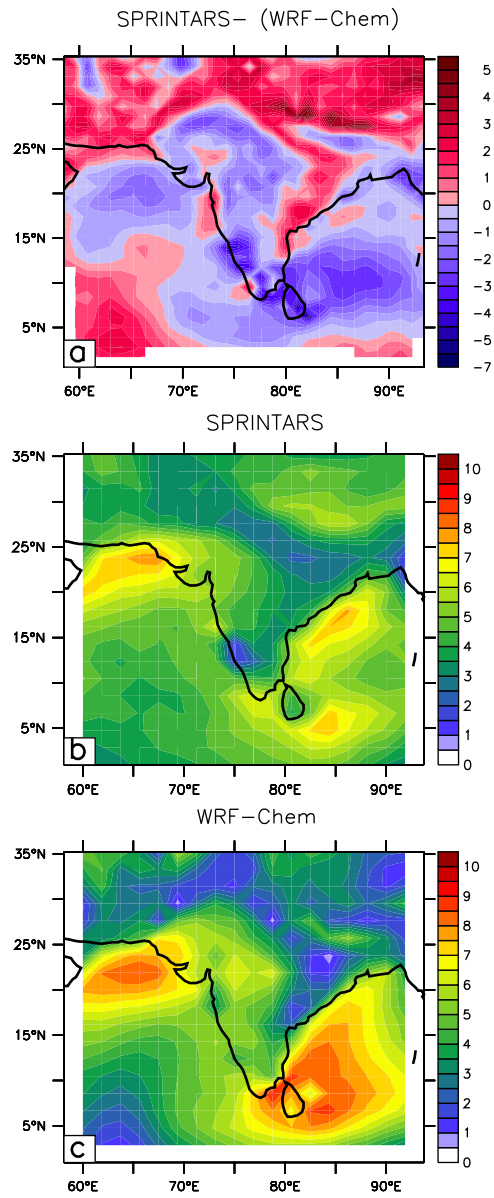


Figure 8: Wind at 10m height from surface during May 2011 a) SPRINTARS-(WRF-Chem) b) SPRINTARS c) WRF-chem. All values are in m s^{-1}

853 a-vis SPRINTARS. This to a certain extent explains the differences in dust
854 mass concentration in these models during pre-monsoon and post-monsoon.
855 To understand this further, we have compared the near-surface wind fields in
856 these 2 model simulations for the month of heavy dust loading (i.e. May) and
857 they are presented in Fig.8. It can be seen that (Fig.8a), in general, WRF-
858 Chem overestimates surface wind speeds over the entire domain. However,
859 SPRINTARS shows higher wind speeds over the semi-arid regions of Sindh
860 and Arabia. These higher winds along with inherent differences in dust
861 production rates in these 2 models, give rise to higher emission fluxes and
862 consequently higher near-surface mass concentrations in SPRINTARS. Also,
863 higher wind speeds over the GP region (Fig.8a) in SPRINTARS simulations
864 vis-a-vis WRF-chem, generates higher dust emissions and thus builds higher
865 dust concentrations. Such a dust source over GP is virtually absent in WRF-
866 Chem simulations. Similar comparisons were done for SM simulations in both
867 the models (Figure not shown), but the models do not differ much in the
868 simulations of SM. Thus, differences in meteorology (U_{10}) and dust emission
869 strategies cause the models to differ in the simulations of dust aerosols. This
870 difference in dust concentrations affects AOD patterns in the two models.

871 4.3.5. Role of Refractive Index

872 It may be noted that, SPRINTARS (Fig.5g, Fig.5h) fails to simulate the
873 AOD hot spot over GP as seen in satellite products MODIS (Fig.5a, Fig.5b),
874 MISR (Fig.5c, Fig.5d) and in WRF-Chem (Fig.5e, Fig.5f). We examine whether
875 this is related to NSBC. As seen in Fig.3, the spatial pattern of near-surface
876 mass concentrations of BC aerosols in these 2 model simulations are similar.
877 Both the models capture the BC hot-spot over the GP region during both
878 months. Also, the BC magnitudes over Indian landmass in the 2 model sim-
879 ulations satisfactorily agree with each other with a few exceptions. In spite
880 of such similarities, SPRINTARS does not capture the the expected AOD
881 hot-spot over the GP region. An examination of the optical properties of BC
882 used in these 2 models reveals that WRF-Chem uses $RI_{BC} = (1.85 - 0.71 \times i)$
883 while SPRINTARS employs $RI_{BC} = (1.75 - 0.44 \times i)$ which is in agreement with
884 OPAC database. It can be seen that, the imaginary part of the RI for BC
885 in SPRINTARS is 60% of that in WRF-Chem. So, BC in WRF-Chem has
886 higher absorbing characteristics than in SPRINTARS and OPAC. Thus, due
887 to such differences in RI values, BC in WRF-Chem affects model simulated
888 AOD more significantly than that in SPRINTARS in spite of having lower
889 mass concentrations (Table 1, Fig.3). Hence, the model simulated AOD dif-

890 fers over GP region.

891

892 Thus, differences in the AOD simulated by the two models over land
893 appear to be linked with the differences associated with model simulations
894 dust aerosol and the optical properties of BC aerosols within the models.

895 4.4. *Sea-Salt*

896 Additionally, we have also studied model simulated sea-salt aerosol dis-
897 tributions (Fig.9). It is to be noted that both the models use identical strate-
898 gies for the emissions/productions of sea-salt aerosols. Here, as we examine
899 the model simulated near-surface concentrations of sea-salt, we can expect
900 emissions to play a major role in governing these distributions at least over
901 oceans. So, given all these factors, the differences in model-simulated sea-
902 salt distributions would mainly arise due to the corresponding differences
903 in the models' meteorology. This would highlight the effect of using purely
904 model-generated meteorology on the aerosol scenario over the oceanic parts
905 of the Indian region. It is seen that during May 2011 both the models (WRF-
906 Chem, Fig.9a and SPRINTARS, Fig.9c), depict a similar pattern of sea-salt
907 concentrations over the oceanic regions, however WRF-Chem underestimates
908 the magnitudes vis-a-vis SPRINTARS. The regional maxima occur over the
909 northern part of AS and over the western part of the BoB. Over these re-
910 gions, WRF-chem depicts a mean concentrations around $6-8 \mu\text{g m}^{-3}$, while
911 SPRINTARS exhibits magnitudes exceeding $10 \mu\text{g m}^{-3}$. The production of
912 sea-salt over oceans is mainly governed by the interaction of near-surface
913 winds with the water surface. Both the models employ identical formula-
914 tion for the production of sea-salt aerosols. Thus, to understand the rea-
915 sons behind such differences in model-simulated sea-salt concentrations, we
916 have again examined simulations of 10m winds over the oceanic regions for
917 WRF-Chem (Fig.8c) and SPRINTARS (Fig.8b), for May 2011. We note that
918 regions with higher winds have higher sea-salt concentrations (Fig.8b and
919 Fig.8c). The higher sea-salt magnitudes over the eastern coast in SPRINT-
920 ARS (vis-a-vis WRF-Chem) appear to be related to higher wind magnitudes
921 over that region in SPRINTARS. Also, the differences in sea-salt concentra-
922 tions over AS in both the models appear to be loosely linked with the higher
923 winds in SPRINTARS over the northern part of the AS. Additionally, both
924 the models show inland transport of sea-salt mainly due to prevailing winds
925 (ocean to land) during the pre-monsoon months. The landward incursion of
926 sea-salt is higher in WRF-Chem (Fig.9a) than in SPRINTARS (Fig.9c), and

927 this could be attributed to the higher wind speeds in WRF-Chem over land
928 during May (Fig.8a).

929

930 For October 2011, both the models (WRF-Chem, Fig.9b and SPRINT-
931 ARS, Fig.9d) exhibit a reduction in sea-salt mass concentrations (similar to
932 dust) which could be mainly attributed to the reduced wind speeds in Octo-
933 ber vis-a-vis May. The mean concentrations over the source regions are seen
934 to be around 2-3 $\mu\text{g m}^{-3}$ for WRF-Chem and 3-4 $\mu\text{g m}^{-3}$ for SPRINTARS.
935 Also, the inland-transport of sea-salt which was seen to be very high during
936 May, decreases due to the reversal of the direction of the winds in October
937 (land to ocean). The simulated pattern of sea-salt concentration is similar
938 in the two models the exception being over the Eastern Coast. However,
939 WRF-Chem underestimates the magnitudes of sea-salt mass concentrations
940 vis-a-vis SPRINTARS. The online model overestimates the wind speeds over
941 land and thus transports sea-salts farther into the land, while underestim-
942 ating the sea-salt production over the oceanic regions.

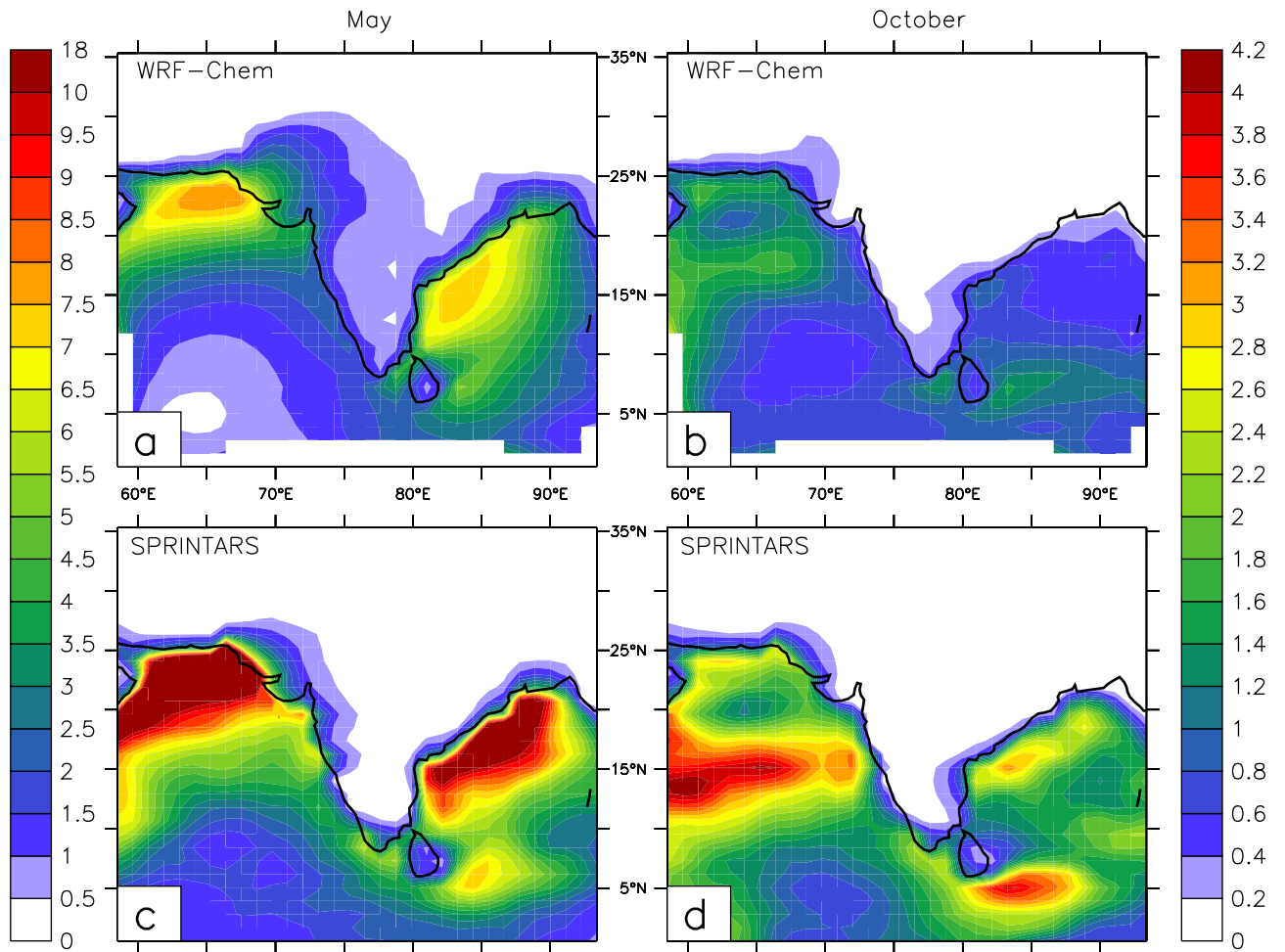


Figure 9: Comparison of model-simulated near-surface sea-salt mass concentration ($\mu\text{g m}^{-3}$) over Indian region during May and October 2011. Left colorbar should be read for May and right one should be read for October

943 **5. Conclusions**

944 The aerosol burden over the south Asian region assumes importance due
945 to its potential impact on Monsoonal rainfall (Lau et al., 2006, Ramanathan
946 et al., 2005), which could affect this densely populated part of the world. In
947 this regard it is imperative to understand some characteristics of this aerosol
948 loading, namely the aerosol species involved, the causes for the production of
949 such species, the lifetime of these short-lived particulates, spatial variation,
950 seasonality, vertical distribution, etc. There are many different models which
951 can simulate such aerosol species. There have been many studies which have
952 simulated the effects of such an aerosol loading on monsoonal rainfall over
953 the region. Nevertheless, it would be worthwhile to validate such models for
954 their representation of the aerosols over India. This validation can be done
955 by comparing the model simulated parameters (mass concentrations, optical
956 depths etc.) with corresponding measurements executed at different obser-
957 vatories spanning the country.

958
959 In this study, we compared simulations of aerosols over the Indian region
960 by WRF-Chem and SPRINTARS, with satellite and surface based observa-
961 tional data, for May and October 2011. These models essentially differ in
962 may aspects including a). horizontal resolution b). physical domain c). for-
963 mulation of meteorological component. This comparison between the models
964 of different kinds and simultaneous evaluation with observations, helped us
965 understand the probable cause for the incorrect simulations of aerosols over
966 the Indian region. We find that both the models underestimate near-surface
967 BC mass concentrations vis-a-vis measurements done at observatories over 8
968 stations across India, with the AF ranging from 1.1 to 5.8. Largely, both the
969 models underestimate the observations by similar margins. Both the models
970 have a similar spatial pattern of NSBC with a correlation of 0.92. Keep-
971 ing in mind such an agreement between BC simulations by two such models
972 which differ in many aspects, we inspected the BC emissions scenarios used
973 in these models. The BC emission scenarios used in these 2 models had a
974 very good spatial match with spatial correlation coefficient of 0.97. Such
975 a high spatial CC in prescribed BC emissions, could have resulted in high
976 spatial CC in simulated BC concentrations. Additionally, it is important to
977 note that the underlying BC emission inventories used in these two mod-
978 els have a horizontal grid spacing of $0.5^0 \times 0.5^0$ (SPRINTARS) and $1^0 \times 1^0$
979 (WRF-Chem). Since the horizontal grid spacing of the underlying BC emis-

980 sion inventories are comparable, they represent all the point sources of BC
981 emissions over Indian region roughly by similar magnitudes. Hence in addi-
982 tion to the similar spatial pattern of BC emissions the underlying inventories
983 get a match in the magnitudes of BC emissions as well. This factor plays
984 an important role in giving the similarities in the model simulated NSBC in-
985 spite of the differences in the horizontal resolution of the models. We further
986 noted that, WRF-Chem prescribes higher magnitudes of BC emissions vis-
987 a-vis SPRINTARS over all the observational locations. But, we found that
988 the meteorological parameters in WRF-Chem (low-level winds and bound-
989 ary layer height) are overestimated vis-a-vis SPRINTARS and the re-analysis
990 dataset MERRA. Thus, even though the models (WRF-Chem and SPRINT-
991 ARS) differ in many aspects the model simulated NSBC largely agree with
992 each other mainly due the balance between the overestimated meteorolog-
993 ical variables in WRF-Chem and lower emissions of BC in SPRINTARS.
994 We further compared the BC emissions inventory used in these two models
995 (GOCART in WRF-Chem and RCP8.5 in SPRINTARS) with three other
996 widely used emission inventories (MACCcity, REAS and SAFAR_India). Both
997 GOCART and RCP 8.5 showed a good spatial match with the other global
998 emission inventories. But, our analysis shows that, all the five BC emission
999 inventories considered in this study underestimate the actual BC emissions
1000 over Indian region, by a factor which ranges from 1.5 to 2.9. Such a quan-
1001 tification of underestimations in BC emissions inventory used in WRF-Chem
1002 (GOCART), also helped us understand the role played by underestimated
1003 emissions and overestimated meteorology in underestimation of NSBC in
1004 WRF-Chem.

1005

1006 While BC simulations from the two models are in good agreement, there
1007 are significant differences in model simulations of AOD. Both the models un-
1008 derestimate AOD over the region vis-a-vis satellite observational data from
1009 MODIS and MISR instruments, for both the months of the model simu-
1010 lations. SPRINTARS AOD values are closer to observational AOD magni-
1011 tudes, than in WRF-Chem. Nevertheless, WRF-Chem significantly replicates
1012 the pattern of satellite AOD with a spatial correlation coefficient around 0.8.
1013 Dust also is a significant contributor to aerosol loading over the Indian region.
1014 An examination of dust simulations by the models reveals that the models
1015 differ significantly in simulating dust over the Indian region. The main differ-
1016 ences occur due to differences in meteorology and dust emission formulations
1017 within the models. Dust aerosol looks to dominate the composite AOD in

1018 SPRINTARS simulations. These differences in dust concentrations appears
1019 to be the major cause of differences in model simulated AOD especially over
1020 Sindh and parts of GP. Models also differ in prescribing the optical prop-
1021 erties for BC aerosol. WRF-Chem employs BC absorption 1.6 times higher
1022 vis-a-vis SPRINTARS. This results in differences in AOD mainly over BC
1023 hot-spots like GP. Thus while the models agree with each other in simulating
1024 near-surface BC concentrations, they differ in simulating AOD over the re-
1025 gion, the main causes of which could be the differences in meteorology, dust
1026 emissions strategy and RI values for BC.

1027

1028 Additionally, we also examined model simulated concentrations of sea-salt
1029 aerosols. While both the models show a similar spatial pattern of sea-salt
1030 concentration, WRF-Chem underestimated the magnitude of sea-salt vis-
1031 a-vis SPRINTARS. Also, it showed a greater inland transport of sea-salt
1032 vis-a-vis SPRINTARS. As the models employ the same strategy for sea-salt
1033 emissions, the differences in sea-salt aerosols are related to the differences in
1034 near-surface winds over the region.

1035

1036 This model inter-comparison and evaluation study augments the current
1037 knowledge regarding the state of aerosol simulating models over Indian re-
1038 gion. While there are a few studies (Nair et al., 2012, Moorthy et al., 2013,
1039 Pan et al., 2015, Govardhan et al., 2015) available regarding the performances
1040 of such models, this study inter-compares and evaluates the 2 different kinds
1041 of models, for an identical period, which does not overlap with that in previ-
1042 ous studies. Such an exercise highlights the role played by meteorology and
1043 emissions' inventories in affecting the aerosol simulations in such models. It
1044 indicates that emission scenarios for anthropogenic aerosols and emission for-
1045 mulation for natural aerosol need to be re-examined and improved. However,
1046 more such intercomparisons (and subsequent modifications in the models) are
1047 also needed to be done for longer periods and over different regions to reduce
1048 the uncertainties in the model predictions.

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