

1 **Transported aerosols regulate the pre-monsoon rainfall over North-East India: a WRF-**
2 **Chem modelling study**

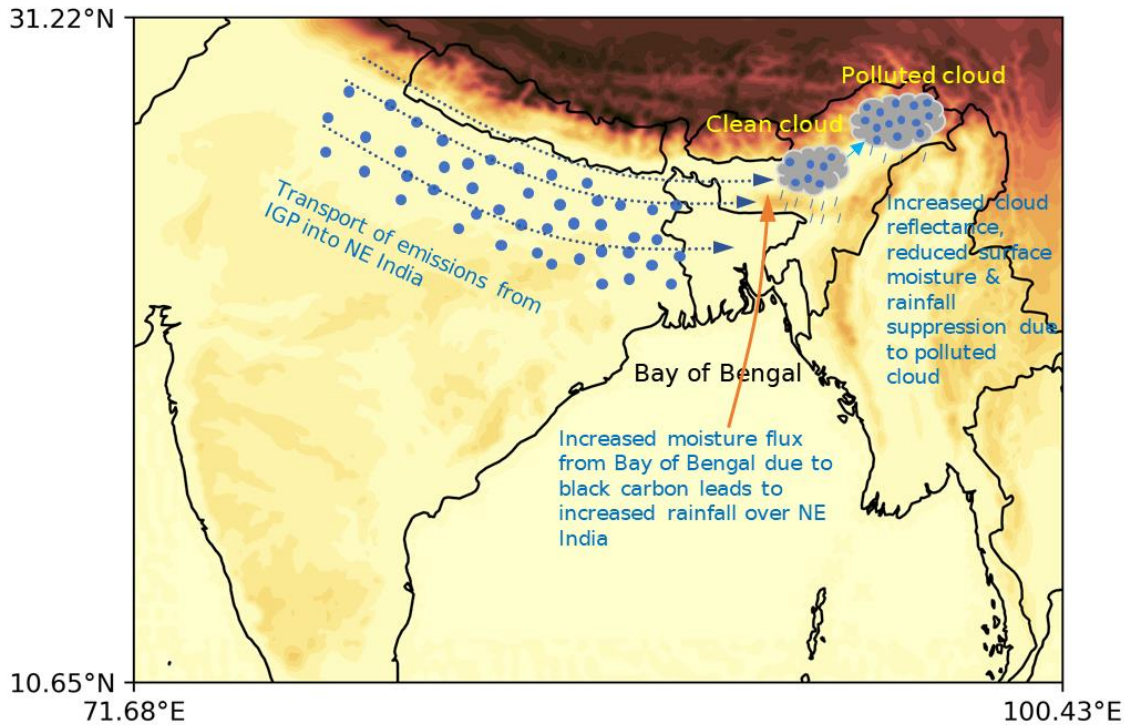
3 Neeldip Barman¹, Sharad Gokhale²

4 ¹Department of Civil Engineering, Indian Institute of Technology Guwahati, Guwahati, 781039, India

5 ²Department of Civil Engineering, Indian Institute of Technology Guwahati, Guwahati, 781039, India

6 *Correspondence to:* Sharad Gokhale (sharadbg@iitg.ac.in)

7 **Abstract.** The study differentiates and quantifies the impacts of aerosols emitted locally within North-East (NE)
8 India region and those transported from outside this region to ascertain whether local or transported aerosols are
9 more impactful in influencing this region's rainfall during the pre-monsoon season (March-April-May). Due to
10 the existence of a declining pre-monsoon rainfall trend in NE India, the study also quantified the role of different
11 aerosol effects on radiative forcing (RF) and rainfall. The study has been carried out using the WRF-Chem model
12 by comparing simulation scenarios where emissions were turned on and off within and outside the NE region.
13 The impact of all emissions as a whole and Black carbon (BC) specifically was studied. Results show that aerosols
14 transported primarily from the Indo-Gangetic Plain (IGP) were responsible for 93.98 % of the PM₁₀ mass over
15 NE India's atmosphere and 64.18 % of near-surface PM₁₀ concentration. Transported aerosols contributed >50 %
16 of BC, organic carbon, sulfate, nitrate, ammonium and dust aerosol concentration and hence a major contributor
17 to air pollution. Hence, the aerosol effects were much greater with transported aerosols. Indirect aerosol effect
18 was found to be the major effect and more impactful with transported aerosols that dominated both rainfall and
19 RF, and suppressed rainfall significantly than the direct and semi-direct effect. However, the increase in direct
20 radiative effects with an increase in transported BC counteracted the rainfall suppression caused by relevant
21 processes of other aerosol effects. Thus, this study shows atmospheric transport to be an important process for
22 this region as transported emissions, specifically from IGP were also found to have greater control over the
23 region's rainfall. Thus, emission control policies implemented in IGP will reduce air pollution as well as the
24 climatic impacts of aerosols over the NE India region.



25

26 **1 Introduction**

27 Aerosols regulate the Earth's energy budget and hydrological cycle through scattering and absorption of solar
 28 radiation and acting as sites for the formation of cloud droplets, which leads to its varied effects, viz. direct, semi-
 29 direct and indirect effects (Mitchell, 1971; Rosenfeld, 2012; Menon et al., 2002). The effects differ spatially
 30 depending on the constituents of aerosols, their physical and chemical properties as well as the quantity. Among
 31 these factors, atmospheric transport also plays an important role which extends the climatic impacts to the
 32 transported region from the source region (Lee et al., 2022). The IGP is a global hotspot of diverse aerosols (Ojha
 33 et al., 2020; Kumar et al., 2018) that impacts regional and global climate (Ramanathan et al., 2005; Tripathi et al.,
 34 2005; Sarangi et al., 2015). Air masses transport aerosols from the IGP to nearby regions, which also impact air
 35 quality (Bhat et al., 2022; Ojha et al., 2012). Bonasoni et al. (2010) showed that pollutants from the IGP follow
 36 the southern slope of the Himalayas as a path into the Bay of Bengal and NE India and similar observations were
 37 made by Gogoi et al. (2017). The condition becomes more critical in the pre-monsoon season when the westerlies
 38 directly transport air pollutants from the IGP to NE India. Among the aerosols, BC is a high climate-influencing
 39 aerosol component due to its strong absorption capability (Bond et al., 2013; Nenes et al., 2002; Koch and Del
 40 Genio, 2010) and IGP is the largest source region of it in India (Rana et al., 2019). Several studies (Guha et al.,
 41 2015; Sarkar et al., 2019; Chatterjee et al., 2010) found BC, among other aerosols measured at sites in NE India
 42 to be transported from the IGP. Moreover, in the NE India region, an increase in BC emissions was observed
 43 along with high BC concentrations near the surface level (Barman and Gokhale, 2019; Chaudhury et al., 2022;
 44 Singh and Gokhale, 2021). Tiwari et al. (2016) observed maximum BC concentration during this season in this
 45 region along with the highest surface RF. The region also observes the highest atmospheric heating and highest
 46 aerosol optical depth with an increasing trend during this period (Nair et al., 2017; Dahutia et al., 2018; Dahutia
 47 et al., 2019; Gogoi et al., 2017; Pathak et al., 2010; Pathak et al., 2016). The presence of high aerosol loading
 48 along with high atmospheric heating is likely to have varied aerosol effects over the region and may also have an

49 important role to play with the rainfall. Mondal et al. (2018) showed a decreasing trend of pre-monsoon rainfall
 50 in this biodiversity hotspot region. Few modelling studies (Kant et al., 2021; Kedia et al., 2016; Kedia et al., 2019)
 51 are available that studied the aerosol effect on rainfall over India. However, only Soni et al. (2017) and Barman
 52 and Gokhale (2022) studied the BC effect on pre-monsoon rainfall in this region but without the inclusion of
 53 aerosol indirect effect. Both studies found BC to increase total rainfall but Barman and Gokhale (2022) also found
 54 semi-direct effect to be a rainfall suppression mechanism by evaporating clouds between 1 to 2 km above ground
 55 level.

56 However, a few questions remained to be answered. How much is the contribution of transported aerosols
 57 to air pollution and climatic effects compared to those emitted within NE India region? What is the role of different
 58 aerosol effects on the rainfall mechanisms? Thus, this study was carried out with the following objectives (a)
 59 Compare the contributions of local and transported aerosols to air pollution and different climatic effects over NE
 60 India (b) Quantify the role of different aerosol effects on the climatic effects (c) Investigate the role of BC emitted
 61 within NE India and transported BC in such climatic effects. Here, transported aerosols include the transported
 62 primary aerosols emitted from outside NE India as well as the secondary aerosols formed from the transported
 63 emissions. Same goes for local emissions. Through qualitative and quantitative comparison of the impacts of local
 64 and transported aerosols, the study tries to find the source region of aerosols that has a greater impact on the
 65 atmosphere over NE India during the pre-monsoon season. Since observational studies cannot distinguish between
 66 the local and transported aerosols impacts, the study was carried out with numerical modelling. The effect of
 67 transported aerosols on different regions of the world has been studied (Krishnamohan et al., 2021; Wang et al.,
 68 2020; Bagtasa et al., 2019) but none of them covered the IGP and its impact on the nearby region.

69 2 Methods

70 The study used the WRF-Chem v4.2.1 model (Grell et al., 2005). The model configuration, modelling domain,
 71 model inputs and simulation period is similar to the one used in Barman and Gokhale (2022). Details regarding
 72 physical and chemical parametrization schemes and the emissions are provided in Table 1.

73 Table 1: Details of physical parametrizations, chemical parametrizations and emissions

Physical parametrizations	
Planetary boundary layer	MYNN3 (Nakanishi and Niino, 2006)
Radiation	RRTMG (Iacono et al., 2008)
Land surface model	NOAH (Tewari et al., 2004)
Cumulus scheme	Grell-Freitas (Grell and Freitas, 2014)
Microphysics	Morrison (Morrison et al., 2009)
Meteorology initial and boundary conditions	ERA5 (Hersbach et al., 2020)
Chemical parametrizations and emissions	
Chemistry scheme	MOZART (Emmons et al., 2010)
Aerosol scheme	MOSAIC (Zaveri et al., 2008)
Chemistry initial and boundary conditions	CAM-Chem (Lamarque et al., 2012)
Anthropogenic emissions	CAMS emission inventory (Granier et al., 2019)
Fire emissions	FINN (Wiedinmyer et al., 2010)

Dust emissions	Online model (Zhao et al., 2010)
Biogenic emissions	MEGAN v2.04 (Guenther et al., 2006)

74

75 The model was run at 10 km grid size for a duration of 13 days from 7-19 April 2018, out of which a 3-
76 day period from 7-9 April 2018 was discarded as spin-up and outputs from 10-19 April 2018 were used for
77 analysis. The period represents the mid of pre-monsoon season. Also, April 2018 was Indian Ocean Dipole and
78 ENSO neutral period and hence suitable for study of aerosol effects. The model domain is shown in Fig. 1(a)
79 which extends from 10.65° N to 31.22° N and 71.68° E to 100.43° E, and the NE India is the part of India within
80 the region bounded by the blue box. The region within the box is bounded by 22° N and 29° N latitudes and 89°
81 E and 97° E longitudes. The climatic situation during the study period was also described in Barman and Gokhale
82 (2022). The near surface wind flow was from the Bay of Bengal towards NE India, which gradually changed to
83 westerly wind flow carrying aerosols from IGP towards NE India. Hence the domain was selected by keeping the
84 NE India region near the upper-right corner of the domain. Descriptions of the simulations are provided in Table
85 2.

86 Table 2: Description of simulations

	Simulation name	Description of simulations
1.	NOR-I	Baseline simulation with all aerosol effects
2.	NOFEED-I	Same as NOR-I but with aerosol radiative effects turned off
3.	NOCHEM	Simulation with no atmospheric chemistry and aerosol effects
4.	No_EMISS_NE	Same as NOR-I but with emissions turned on only outside NE India
5.	Only_EMISS_NE	Same as NOR-I but with emissions turned on only within NE India
6.	No_EMISS_NE_4SO ₂	Same as No_EMISS_NE but with 4×SO ₂ emissions
7.	No_EMISS_NE_0.25SO ₂	Same as No_EMISS_NE but with 0.25×SO ₂ emissions
8.	No_EMISS_NE_NOFEED	Same as No_EMISS_NE but with aerosol radiative effects turned off
9.	Only_EMISS_NE_NOFEED	Same as Only_EMISS_NE but with aerosol radiative effects turned off
10.	No_NE_BCI	Same as NOR-I but with BC emissions turned on only outside NE India
11.	Only_NE_BCI	Same as NOR-I but with BC emissions turned on only within NE India
12.	4NOR-I	Same as NOR-I but with 4×BC emissions
13.	No_BC_ABS	Same as NOR-I but with BC absorption disabled
14.	NOR	Baseline simulation with only direct and semi-direct effect
15.	2NOR	Same as NOR but with 2×BC emissions
16.	No_NE_BC	Same as NOR but with BC emissions within NE India region turned off
17.	No_NE_2×BC	Same as No_NE_BC but with 2×BC emissions outside NE India
18.	Only_NE_BC	Same as NOR but with BC emissions turned off outside NE India
19.	Only_NE_2×BC	Same as Only_NE_BC but with 2×BC emissions inside NE India
20.	NOFEED	Same as NOR but with aerosol radiative effects off

87

88 All the simulations were conducted with the MOZART-MOSAIC scheme, except simulation 3, which was purely
89 a meteorology simulation and did not include any atmospheric chemistry and aerosol effects. Moreover,
90 simulations 1 to 13 (except 3), were conducted with the version of MOZART-MOSAIC scheme which also
91 supports indirect aerosol effect by coupling with the Morrison microphysics scheme along with direct and semi-
92 direct effect, while simulations 14 to 20 did not include indirect effect. The NOR simulation used in Barman and
93 Gokhale (2022), was also used in this study. NOR-I is also the baseline simulation run with the same baseline
94 emissions for the study period as NOR, but also includes indirect aerosol effect. No_EMITT_NE had all emissions
95 (biogenic, anthropogenic and dust) disabled within the region bounded by 22° N and 29° N latitudes and 89° E
96 and 97° E longitudes, shown by the blue box in Figure 1(a) while No_NE_BC and No_NE_BCI only had BC
97 emissions disabled within the same region. Only_EMITT_NE had all emissions disabled outside of the above
98 region along with boundary conditions for all chemical species modified to zero to nullify the transport of
99 emissions from outside the domain and similarly, Only_NE_BC and Only_NE_BCI had BC emissions disabled
100 outside the NE India region with boundary conditions for BC modified to zero. Remaining simulations can be
101 understood from Table 2 and their applications are understood from the results and discussion in Sect 3.

102 As per Ghan et al. (2012) and Bauer and Menon (2012), the total aerosol effect is the algebraic sum of
103 direct, indirect and semi-direct effects. Similar approaches were used by Yang et al. (2011). Thus,

104
$$\text{NOR-I} - \text{NOCHEM} = \text{Total aerosol effect} = \text{Direct} + \text{Semi-direct} + \text{Indirect}, \quad (1)$$

105 Both NOFEED-I and NOR-I includes indirect effect but NOFEED-I does not include aerosol radiative effects.
106 Thus,

107
$$\text{NOR-I} - \text{NOFEED-I} = \text{Direct} + \text{Semi-direct effect}, \quad (2)$$

108 Also, since NOFEED-I includes only indirect effect,

109
$$\text{NOFEED-I} - \text{NOCHEM} = \text{Indirect effect}, \quad (3)$$

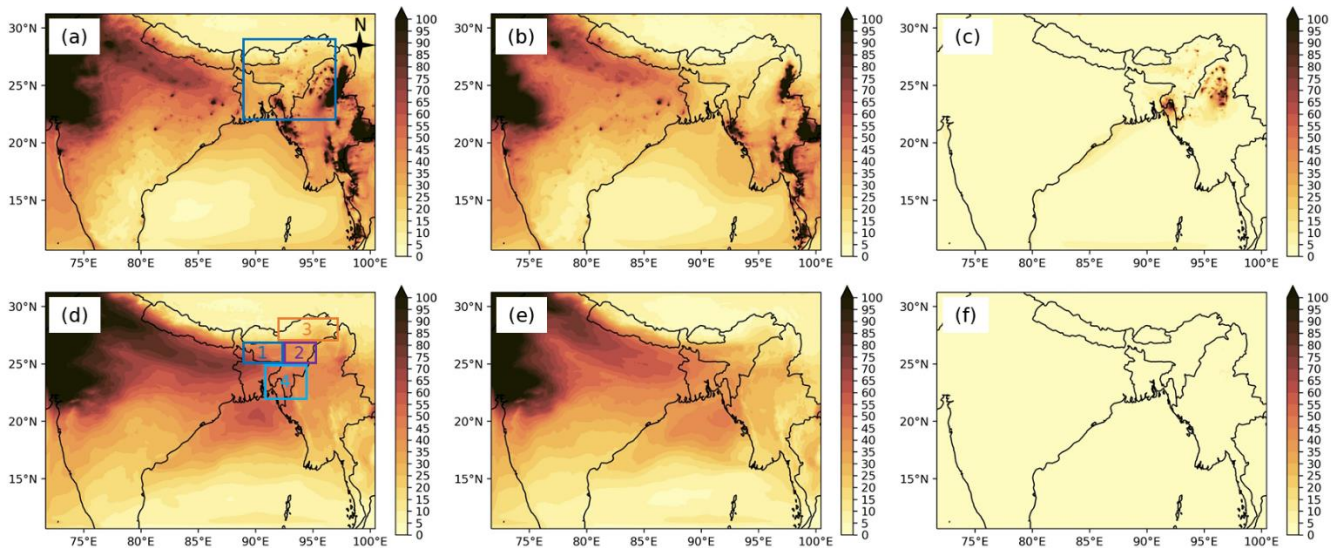
110 Similar approaches were used by Wang et al. (2015).

111 The NOR simulation utilised in this study was evaluated in Barman and Gokhale (2022). Moreover,
112 meteorological evaluation of NOR-I w.r.t wind direction, wind speed, temperature and humidity was carried out
113 against surface station datasets (<https://mesonet.agron.iastate.edu/sites/locate.php>) at Guwahati (26.10 °N, 91.58
114 °E), Kolkata (22.65 °N, 88.45 °E), Bangalore (13.20 °N, 77.70 °E), Patna (25.59 °N, 85.08 °E), Delhi (28.56 °N,
115 77.11 °E) and Mumbai (19.10 °N, 72.86 °E). Simulated rainfall was evaluated against the Indian Meteorological
116 Department (IMD) rainfall dataset of Pai et al. (2014)
117 (https://www.imdpune.gov.in/Clim_Pred_LRF_New/Grided_Data_Download.html). Index of agreement (IOA),
118 root mean square error (RMSE) and mean error (ME) were used as statistical parameters. As per the criteria of
119 Emery et al. (2001), the NOR-I simulation underpredicted temperature but showed good performance with wind
120 speed and wind direction but had large RMSE with wind direction, similar to the NOR simulation. Performance
121 statistics are provided in Table S1. Moreover, NOR and NOR-I simulated chemical species (BC, organic carbon,
122 dust and sulfate aerosol) were compared against the MERRA2 dataset
123 (https://disc.gsfc.nasa.gov/datasets/M2T1NXAER_5.12.4/summary) at the above locations. Performance
124 statistics are shown in Table S2. NOR gave a much better estimation of all the chemical species at all locations.

125 Moreover, the predicted chemical species of nitric oxide (NO), nitrogen dioxide (NO₂), sulfur dioxide (SO₂),
 126 PM_{2.5} and PM₁₀ were compared against in-situ observations at Delhi (28.56 °N, 77.11 °E), Kanpur (26.57 °N,
 127 80.32 °E), Patna (25.61 °N, 85.13 °E) and Siliguri (26.69 °N, 88.41 °E), obtained from Central Pollution Control
 128 Board, India (<https://app.cpcbcr.com/ccr/#/caaqm-dashboard-all/caaqm-landing/caaqm-data-availability>). These
 129 locations are located along the IGP. Performance statistics are given in Table S3. The performance statistics were
 130 better with both particulate matter than gaseous species. Comparatively the performance was better with
 131 MERRA2. The relatively lower performance with in-situ comparison may be due to the grid size as in-situ
 132 observations are affected by local emission sources as well the deficiencies in emission inventory. However, the
 133 inclusion of all aerosol effects greatly improved simulated rainfall performance with NE India regional average
 134 IOA: 0.52, ME: 3.72 mm day⁻¹, RMSE: 13.55 mm day⁻¹ compared to only considering direct + semi-direct effect
 135 (IOA: 0.40, ME: 9.22 mm day⁻¹, RMSE: 21.26 mm day⁻¹) in Barman and Gokhale (2022). The improvement in
 136 performance and decrease in ME show that indirect effect played a major role during this period in controlling
 137 and suppressing rainfall.

138 3 Results and discussion

139 3.1 PM₁₀ spatial and vertical distribution

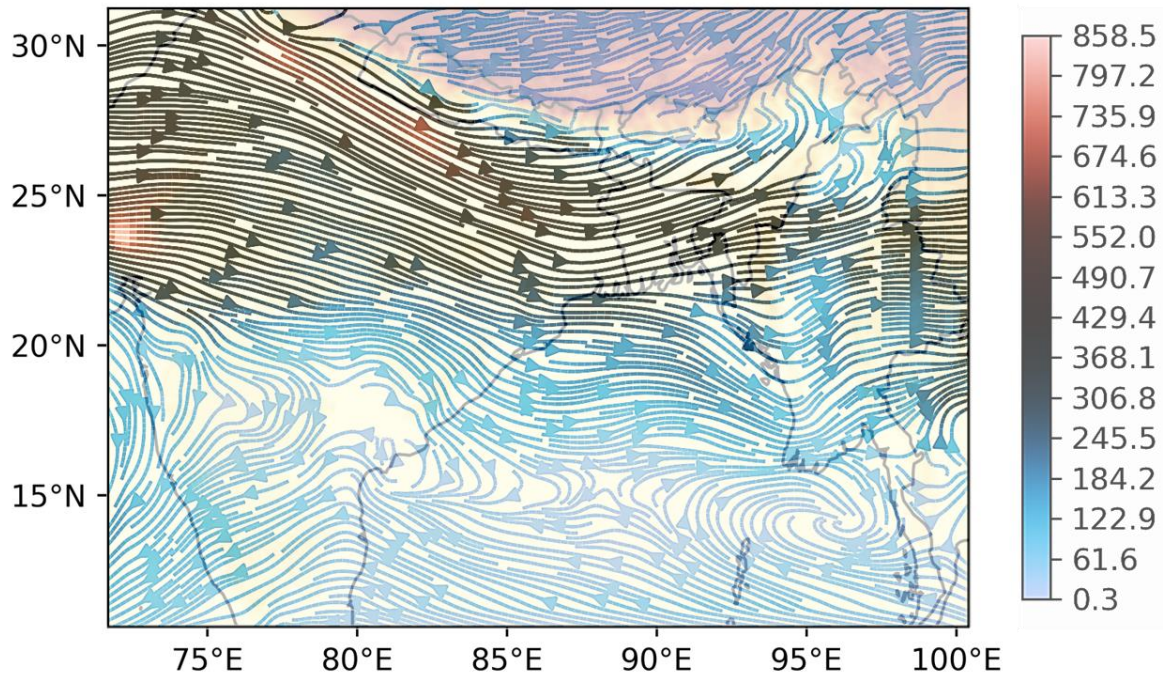


140

141 Figure 1: Spatial distribution of PM₁₀ concentration (µg m⁻³) in NOR-I, (a, d), No_EMISS_NE (b, e) and
 142 Only_EMISS_NE (c, f). Upper row shows distribution at model level 0 (near surface) and the lower row at model
 143 level 15

144 Figure 1 shows the time-averaged spatial distribution of PM₁₀ concentration. The NE India region was divided
 145 into four regions based on the proximity from the IGP, shown in Fig. 1(d). Region 1 and region 2 fall along the
 146 Brahmaputra River Valley, with region 1 being closest to IGP. Region 3 is mostly a mountainous region and 4 is
 147 the southern region closer to the Bay of Bengal. The spatial distribution of geopotential heights of model level 0
 148 and 15 are shown in Fig. S1, while region-wise (Fig. 1(d)) concentration values within NE India at the two
 149 atmospheric heights are shown in Table S4. PM₁₀ concentration contours shown in Fig. 1(a), 1(b), 1(d) and 1(e),
 150 emanating from IGP and spreading into NE India indicated the transport of aerosols from IGP into NE India. The
 151 similarity of these spatial distributions of No_EMISS_NE to the baseline scenario, NOR-I, especially within NE

152 India region inferred that most of the aerosol mass within NE India was contributed by transported aerosols, while
 153 PM₁₀ emitted or formed over NE India remained mainly confined within the region as shown in Fig. 1(c), possibly
 154 due to the mountainous terrain, as also described in Kundu et al. (2018). The transport of PM₁₀ can also be seen
 155 from Fig. 2, in which the streamline's arrow from IGP to NE India show the transport of air-mass and the colour
 156 of the streamlines show the PM₁₀ mass flux in $\mu\text{g m}^{-2} \text{s}^{-1}$. The flux was higher over IGP.



157
 158 Figure 2: Streamlines showing transport of air-mass from IGP to NE India and PM₁₀ mass flux ($\mu\text{g m}^{-2} \text{s}^{-1}$) at 1300
 159 m above terrain

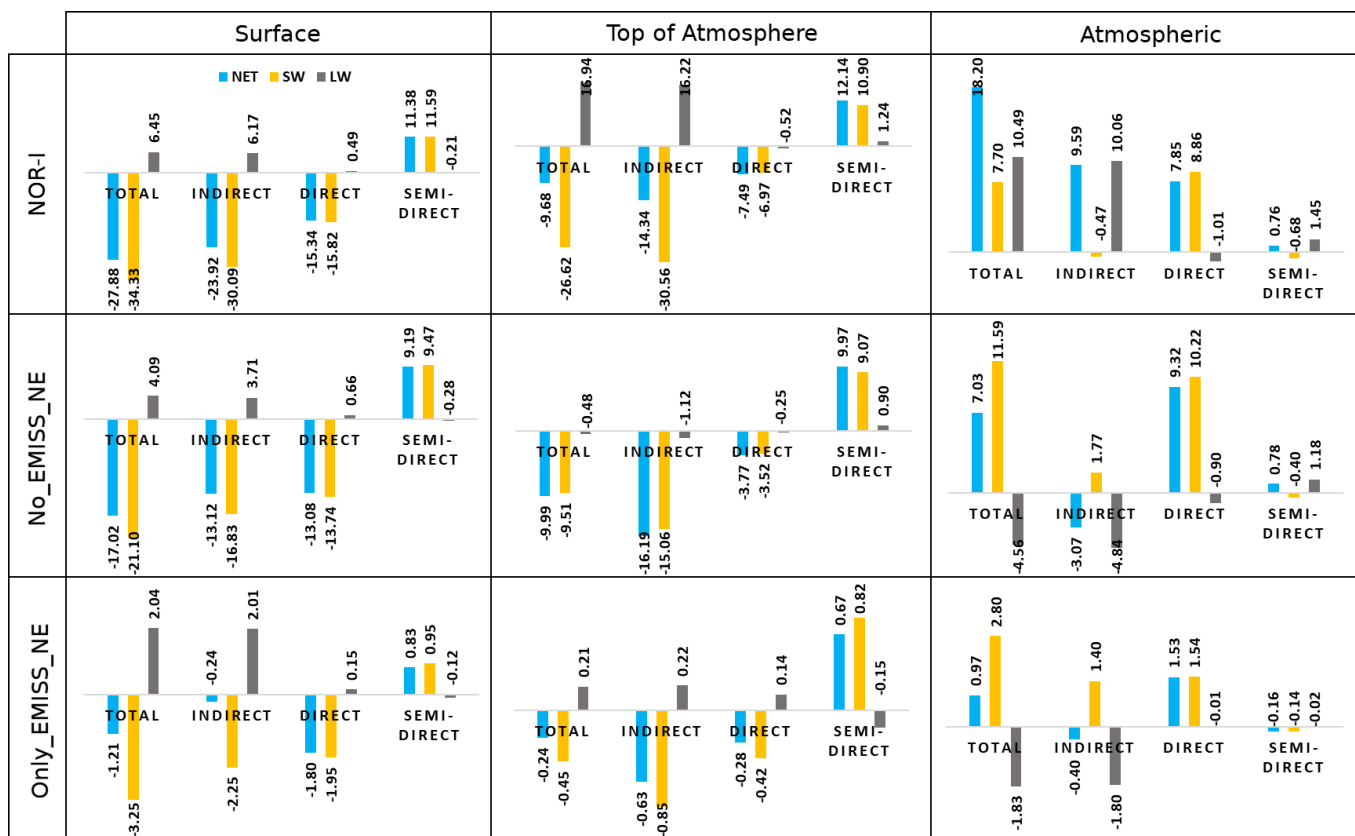
160 Both near the surface and at higher atmosphere (level 15), No_EMISS_NE showed a higher regional average
 161 concentration (surface: $14.46 \mu\text{g m}^{-3}$, higher atmosphere: $24.43 \mu\text{g m}^{-3}$) which was closer to the baseline scenario
 162 of NOR-I (surface: $27.43 \mu\text{g m}^{-3}$, higher atmosphere: $34.13 \mu\text{g m}^{-3}$) compared to the local emission scenario of
 163 Only_EMISS_NE (surface: $8.07 \mu\text{g m}^{-3}$, higher atmosphere: $0.98 \mu\text{g m}^{-3}$). Thus, transported aerosols contributed
 164 higher PM₁₀ concentration (64.18 %) than local emission and contribution from local emissions were negligible
 165 at higher atmosphere, as also seen in Fig. 1(f) and 96.14 % of it was contributed by transported aerosols. The
 166 higher concentration at higher atmosphere was due to transported aerosols developing an elevated PM₁₀ profile
 167 (Fig. S2) having maximum concentration near 2000 m and which shows much greater similarity with the baseline
 168 scenario. The long range transport and strong convective active during this season is responsible for the elevated
 169 profile (Pathak et al., 2016). Hence, transported aerosols contributed to bulk of the aerosols over NE India
 170 throughout the atmospheric column (93.98 %) indicated by the column integrated PM₁₀ mass of 313.97 g m^{-2}
 171 (No_EMISS_NE) and 20.08 g m^{-2} (Only_EMISS_NE). NOR-I had column integrated PM₁₀ mass of 466.63 g m^{-2} .
 172 Further analysis indicated that transported aerosols accounted for >50 % of BC, organic carbon, sulfate, nitrate,
 173 ammonium and dust aerosol mass over NE India's atmosphere as the column integrated mass for these species in
 174 No_EMISS_NE were 4.55, 19.59, 51.66, 2.20, 13.74 and 207.82 g m^{-2} , respectively, while it was 0.94, 6.51, 1.79,

175 0.12, 0.56 and 6.60 g m⁻², respectively in Only_EMISS_NE. The spatial distribution of column integrated mass
176 of these species can be seen in Figures S3, S4, S5, S6, S7 and S8. Regions 1, being in close proximity to IGP, as
177 seen in Fig. 1(c)), received maximum near surface aerosol mass (73.70 %) from transported aerosols, compared
178 to the other regions, followed by region 2 (66.86 %), 3 (60.48 %) and 4 (57.43 %). However, even though
179 No_EMISS_NE and Only_EMISS_NE is the bifurcation of NOR-I into two separate emission regions, the sum
180 of No_EMISS_NE and Only_EMISS_NE column integrated mass as well as concentrations didn't equate to NOR-
181 I values and is always less than it. This indicated formation of extra aerosol mass due to interaction of emissions
182 of the two regions.

183 **3.2 Aerosol effects of local and transported aerosols on radiative forcing**

184 RF due to different aerosol effects was estimated based on the methodology described in Sect. 2. Further details
185 regarding its estimation are provided in the supplementary.

186 The baseline scenario indicated that direct and indirect aerosol effects caused net (NET) surface and top
187 of the atmosphere (TOA) dimming while causing atmospheric heating, as seen in Fig. 3. This is due to the presence
188 of aerosols that scatter and absorb solar radiation, reducing it at the surface while increasing it at the top of the
189 atmosphere as well as causing atmospheric heating. Net direct surface, TOA and atmospheric RF were -15.34, -
190 7.49 and 7.85 Wm⁻² and was mainly contributed by short-wave (SW) radiation. Indirect effect had the same effect
191 on solar radiation as the direct effect and was due to the formation of numerous smaller cloud droplets which has
192 better reflectivity to solar radiation, also known as the 1st indirect effect or Twomey effect (Twomey, 1977).
193 However, positive atmospheric RF (18.20 W m⁻²) causing atmospheric heating (10.06 W m⁻²) was mainly caused
194 by long-wave (LW) radiation (16.22 W m⁻²) at the TOA contributed by indirect effect. This was due to greater
195 cloud cover (Fig. S9) at 8 – 10 km which is not seen in the other two scenarios. The indirect effect also caused
196 warming at the surface (6.17 W m⁻²), as its contributed to greater cloud cover (Nandan et al., 2022) and caused
197 heating of the surface through LW radiation. The total net surface RF was -27.88 W m⁻² out of which -23.92 W
198 m⁻² or 85.80% was contributed



199

200 Figure 3: NE India regional average RF ($W m^{-2}$) due to different aerosol effects at NET, SW and LW wavelengths
 201 in different emission scenarios

202 by indirect forcing. Indirect SW forcing ($-30.08 W m^{-2}$) was almost twice the direct SW forcing ($-15.82 W m^{-2}$),
 203 while semi-direct SW forcing ($+11.58 W m^{-2}$) was $\sim 75\%$ of the direct forcing. Semi-direct effect showed positive
 204 surface RF due to cloud cover reduction. Thus, atmospheric heating and the subsequent evaporation of clouds
 205 compensated to a large extent the reduction in solar radiation due to aerosols. The atmospheric RF ($0.76 W m^{-2}$)
 206 due to semi-direct effect was due to LW radiation, which may be due to increased solar radiation at the surface,
 207 which released the heat into the atmosphere in the form of LW radiation. However, this value was very small. The
 208 indirect RF contributed most to the total surface, TOA and atmospheric RF at both SW and LW wavelengths and
 209 hence was found to be the dominant aerosol effect affecting radiation over NE India.

210 Quantitatively, No_EMISS_NE provided RF values (surface: $-17.02 W m^{-2}$, TOA: $-9.99 W m^{-2}$ and
 211 atmospheric RF: $7.03 W m^{-2}$) that were much similar and closer to the baseline scenario (surface: $-27.88 W m^{-2}$,
 212 TOA: $-9.68 W m^{-2}$ and atmospheric RF: $18.20 W m^{-2}$) than Only_EMISS_NE (surface: $-1.21 W m^{-2}$, TOA: -0.24
 213 $W m^{-2}$ and atmospheric RF: $0.97 W m^{-2}$). Consequently, the No_EMISS_NE net indirect, direct and semi-direct
 214 surface RF values of -13.12 , -13.08 and $9.19 W m^{-2}$ were significantly larger than the corresponding
 215 Only_EMISS_NE RF values of -0.24 , -1.80 and $0.83 W m^{-2}$. A similar conclusion could be inferred at TOA also.
 216 Hence transported aerosols were primarily responsible for all the different aerosol effects on radiation over NE
 217 India as a greater amount of aerosol mass was contributed by it. Moreover, No_EMISS_NE net direct atmospheric
 218 RF ($9.32 W m^{-2}$) was found to be even higher than the baseline scenario ($7.85 W m^{-2}$). This indicated that the NE
 219 India region contained more scattering aerosols while transported aerosols contained more absorbing aerosols as

220 the difference in the direct atmospheric RF is mainly driven by changes in the TOA RF (-7.49 vs. -3.77 W m⁻²)
 221 than surface RF (-15.34 vs. -13.08 W m⁻²). Region 1 had the highest direct and semi-direct net surface RF of -
 222 20.41 W m⁻² and 19.20 W m⁻², respectively due to its close proximity to IGP.

223 3.3 Aerosol effects of local and transported aerosols on rainfall

224 Table 3: Changes in rainfall due to different aerosol effects in different scenarios (mm)

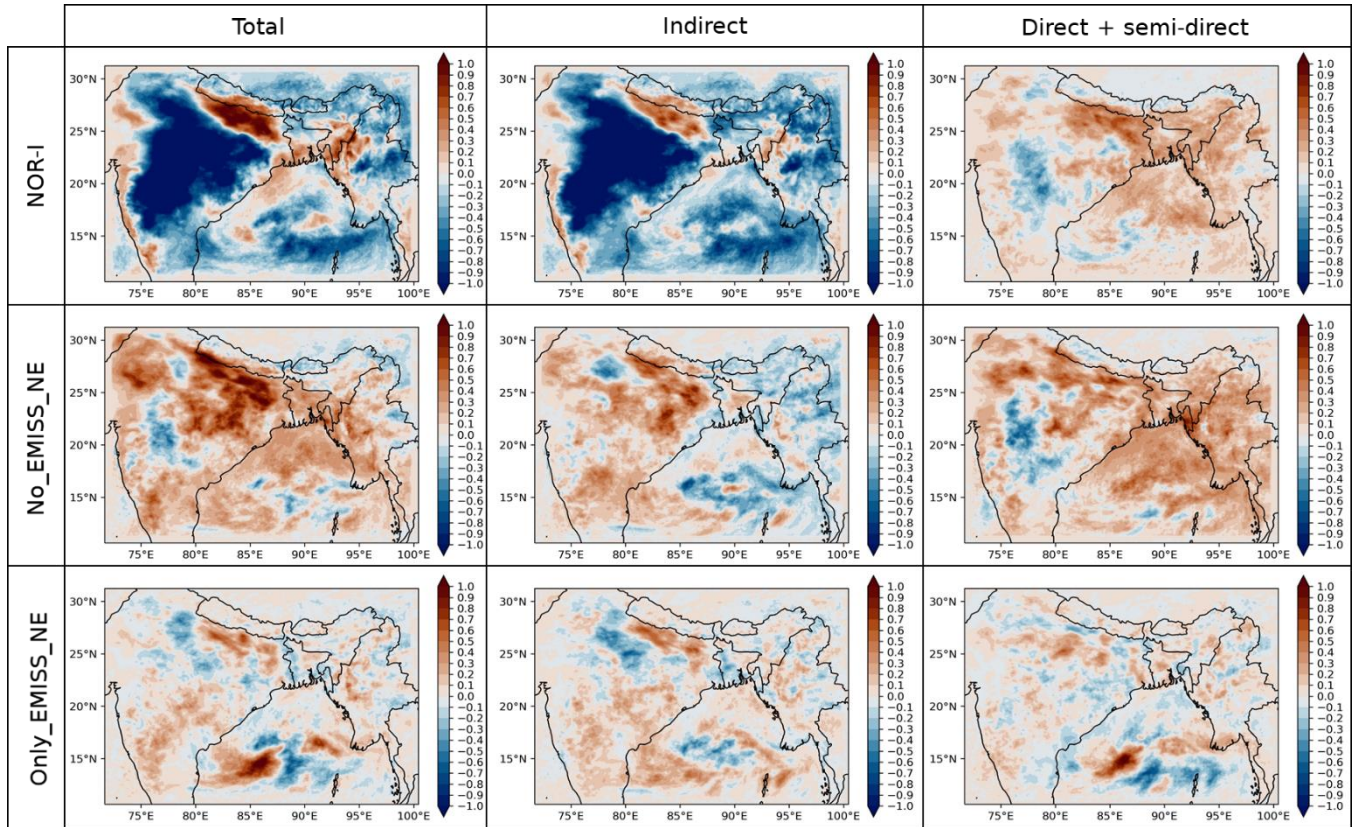
	Total aerosol effect	Direct + semi-direct	Indirect
NOR-I	-275.13	-17.04	-258.09
No_EMISS_NE	-73.06	-23.95	-49.11
Only_EMISS_NE	-24.45	-8.42	-16.04

225

226 The quantitative changes in regional average rainfall amounts over NE India due to the different aerosol effects
 227 induced by the aerosols in different scenarios are provided in Table 3. Region-wise values can be read from Table
 228 S5. Rainfall from region 4 was not considered due to large errors being associated with it (Fig. S10). In the baseline
 229 scenario (NOR-I), the total aerosol effect caused rainfall suppression in all three regions, with a regional total of
 230 -275.13 mm, shown in Table 3. Reductions in rainfall due to the total aerosol effect was contributed by
 231 suppressions due to both direct + semi-direct and indirect effect and was observed in all the considered regions.
 232 The highest suppression was observed in region 3 (-102.60 mm), followed by region 1 (-100.60 mm). The role of
 233 direct + semi-direct effect was observed to be minimal with a total regional suppression of -17.04 mm while the
 234 indirect effect (-258.09 mm) was responsible for almost the whole of the suppression of -275.13 mm. Region 1
 235 observed the highest suppression of -13.21 mm due to direct + semi-direct effect as this region's radiation was
 236 highest impacted by these effects.

237 Direct effect could suppress rainfall by reducing surface evaporation and convection through surface
 238 dimming while semi-direct by evaporation of clouds (Talukdar et al., 2019; Lohmann and Feichter, 2001; Habib
 239 et al., 2006; Bollasina et al., 2011; Koch and Del Genio, 2010b). However, the surface dimming by indirect effect
 240 (-23.92 W m⁻²) with NOR-I was much larger than the combined direct + semi-direct effect (-3.96 W m⁻²). Hence
 241 the reduction in surface moisture flux due to indirect effect (-6.45×10^{-6} kg m⁻² s⁻¹) was much greater than due to
 242 combined direct + semi-direct effect (-1.1×10^{-6} kg m⁻² s⁻¹) and much similar to the reduction due to total aerosol
 243 effect (-7.56×10^{-6} kg m⁻² s⁻¹). This was also observed in the case of No_EMISS_NE. The greater surface dimming
 244 of -17.02 W m⁻² in No_EMISS_NE caused a much higher negative surface moisture flux change of -3.82×10^{-6} kg
 245 m⁻² s⁻¹ due to total aerosol effect, mostly contributed by indirect effect (-2.79×10^{-6} kg m⁻² s⁻¹) compared to direct
 246 + semi-direct effect (-1.03×10^{-6} kg m⁻² s⁻¹). Hence, indirect effect in NOR-I and No_EMISS_NE dominated
 247 moisture reduction through reduction in surface moisture flux over most areas of NE India at both low and high-
 248 terrain regions, as seen in Fig. 4.

249 However, direct + semi-direct effect caused an increase of moisture in NOR-I and No_EMISS_NE over
 250 most of NE India in spite of a negative surface moisture flux not observed in Only_EMISS_NE. This indicated
 251 that direct + semi-direct caused an increase in the transport of moisture from another region, in this case from Bay
 252 of Bengal. The equivalent potential temperature (EPT) profiles in Fig. 5 compared the atmospheric stability due
 253 to

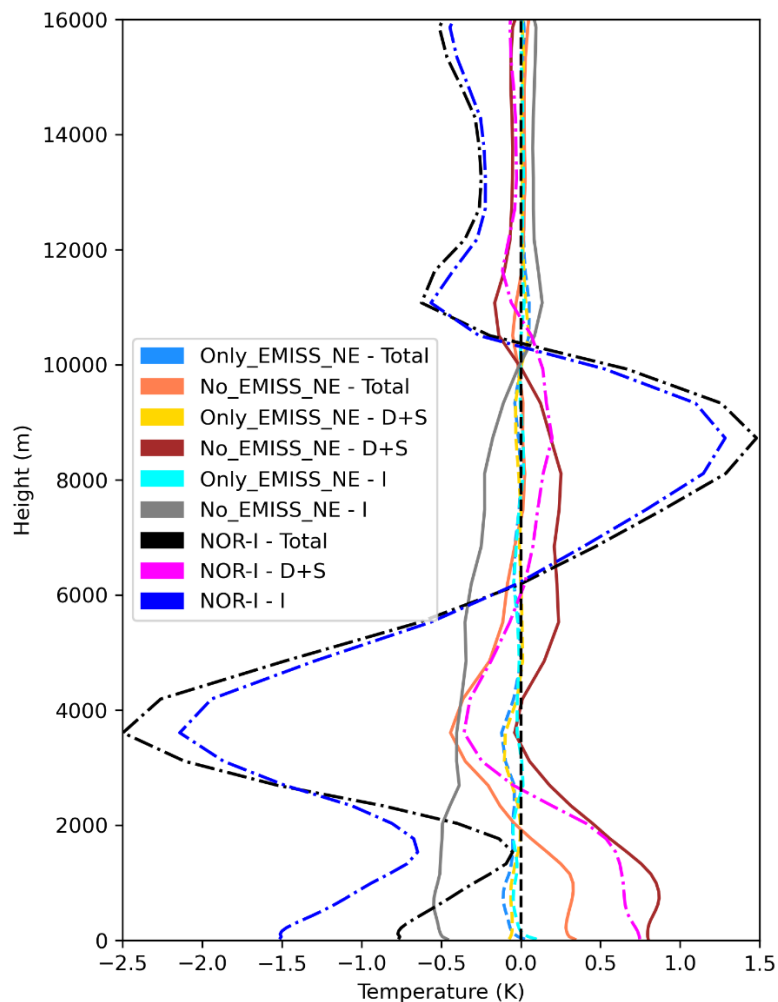


254

255 Figure 4: Spatial distribution of change in near-surface water vapor mixing ratio (g kg^{-1}) due to total aerosol effect,
 256 direct + semi-direct effect and indirect effect

257 different aerosol effects. The greater surface dimming due to the indirect effect in No_EMISS_NE caused not
 258 only negative surface moisture flux but also a significant increase in atmospheric stability (indicated by increasing
 259 value of indirect effect EPT profile with height), reducing convection, which possibly also contributed reduction
 260 to rainfall suppression. However, although the direct + semi-direct EPT profile showed increased atmospheric
 261 stability below 1 km, but created an overall unstable atmosphere in the lower atmosphere. This instability,
 262 primarily caused due to atmospheric heating of BC, created an unstable region over NE India which facilitated
 263 the increased transport of moisture from the Bay of Bengal (discussed later). Hence, though the direct effect
 264 reduces rainfall by reducing surface moisture flux and convection but also possibly enhances it by transporting
 265 moisture. This transported moisture possibly compensated to some extent the rainfall reduction due to a decrease
 266 in surface moisture flux, convection and cloud evaporation caused by direct and semi-direct effects. Hence, the
 267 rainfall reduction due to direct + semi-direct effect (-17.04 mm) was possibly significantly less than the indirect
 268 effect (-258.09 mm). Thus, the effect of direct and indirect effects on dynamics was distinctly different. The EPT
 269 profile of the total aerosol effect in No_EMISS_NE showed an unstable lower atmosphere, supporting moisture
 270 transport. Similar explanation could be given for moisture increase due to direct + semi-direct in NOR-I but the
 271 increase in atmospheric stability and moisture reduction due to greater surface dimming by its indirect effect was
 272 significantly larger, which created an overall stable atmosphere due to total aerosol effect in NOR-I. The EPT
 273 profiles of Only_EMISS_NE showed almost zero perturbation throughout the atmosphere and hence was unable
 274 to affect atmospheric stability and cause moisture transport. Thus, the direct + semi-direct effect in
 275 Only_EMISS_NE did not show significant moisture change in Fig. 4. Moreover, the significantly smaller surface
 276 dimming (-1.21 W m^{-2}) in Only_EMISS_NE caused very small but positive change of $8.15 \times 10^{-8} \text{ kg m}^{-2} \text{ s}^{-1}$ due to

277 the total aerosol effect and hence similar moisture change is observed in Fig. 4. Hence aerosols emitted solely
 278 from NE India had negligible capability in affecting moisture through different aerosol effects. Moisture reduction
 279 over NE India was much greater due to the indirect effect in No_EMISS_NE compared to Only_EMISS_NE,
 280 while moisture increase was much greater in No_EMISS_NE compared to Only_EMISS_NE due to a higher
 281 direct + semi-direct effect.

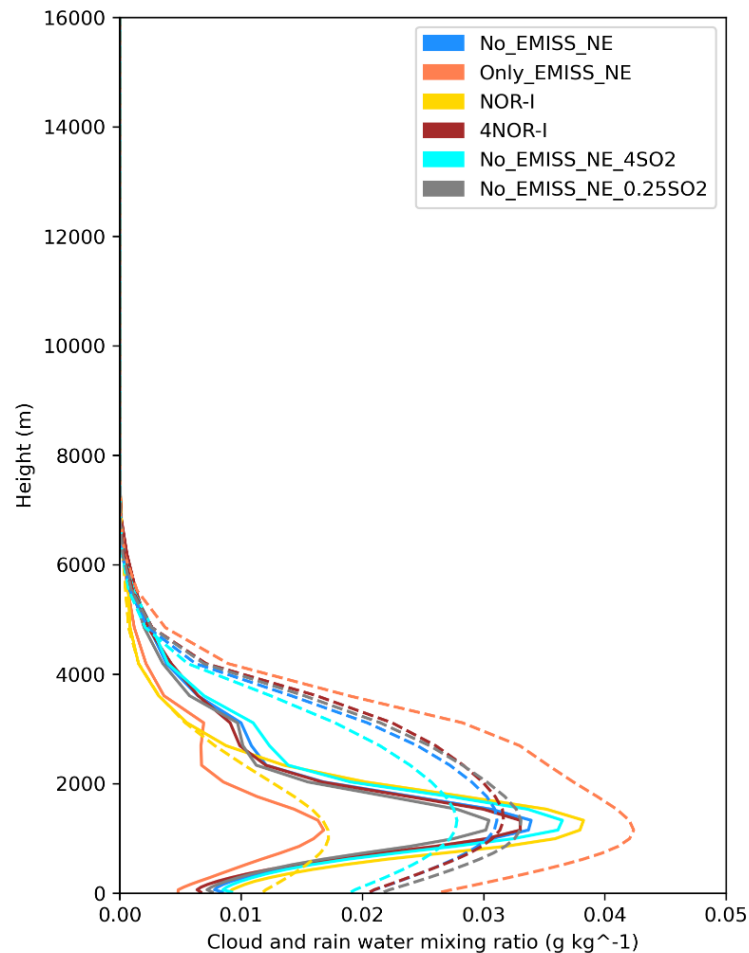


282

283 Figure 5: Perturbation of EPT (K) due to total aerosol effect (Total), direct + semi-direct (D+S) and indirect (I) aerosol effect
 284 in No_EMISS_NE (non-dashed), Only_EMISS_NE (dashed) and NOR-I (dashdot)

285 Moreover, the positive NE India regional average difference of column integrated cloud condensation
 286 nuclei (CCN) number ($4.38 \times 10^{10} \text{ m}^{-2}$), cloud droplet number ($4.42 \times 10^{13} \text{ m}^{-2}$) and cloudwater (27.93 g m^{-2}), and
 287 estimated from No_EMISS_NE – Only_EMISS_NE indicated that transported aerosols had a greater impact
 288 through aerosol indirect effect (Zhang et al., 2010). The presence of larger aerosol amounts in the form of CCN
 289 affects the cloud lifetime by affecting the conversion from cloudwater to rainwater, thus, to rainfall, thereby
 290 suppressing rainfall, also known as the 2nd indirect effect (Shiogama et al., 2010; Cherian et al., 2017). The
 291 presence of a large amount of CCN facilitates condensation of water vapor on numerous CCN particles, producing
 292 numerous cloud droplets with smaller radii. This restricts small cloud droplets to grow in size due to reduction in
 293 interaction with other cloud droplets which affect its conversion to rain droplet, and thus to rainfall. Due to more

294 aerosol mass over NE India (Sect. 3.1), NOR-I and No_EMISS_NE had significantly higher cloudwater compared
295 to



296

297 Figure 6: NE India regional average vertical profiles of cloudwater mixing ratio (non-dashed) and rainwater mixing ratio
298 (dashed) in different scenarios (g kg^{-1})

299 Only_EMISS_NE, as seen in Fig. 6. Consequently, NOR-I and No_EMISS_NE had a significantly lower
300 rainwater mixing ratio than Only_EMISS_NE. Thus, rainfall suppression due to indirect effect was highest in
301 NOR-I, followed by No_EMISS_NE and Only_EMISS_NE. Hence, the combined effect of reduction in moisture,
302 instability and rainfall formation contributed to the reduction in rainfall through indirect and total aerosol effects.
303 This could be a possible key mechanism associated with the decreasing rainfall trend in the region. Reduction of
304 moisture due to the direct effect of aerosols and evaporation of clouds by BC were found to be possible
305 mechanisms by Barman and Gokhale (2022). However, this study shows that the contribution of direct and semi-
306 direct effects was very small compared to the indirect effect. The indirect effect has been found to be the dominant
307 aerosol effect in many studies (Wang et al., 2015; Liu et al., 2016) and was found to suppress monsoon rainfall
308 over India (Manoj et al., 2012). Aerosol indirect effect is mainly dictated by the warm clouds (Christensen et al.,
309 2016). Thus, the higher cloud cover associated with NOR-I and No_EMISS_NE in lower atmosphere which
310 affected SW radiation more in Sect. 3.2, was due to a greater amount of cloudwater in lower atmosphere.

311 Moreover, No_EMISS_NE and Only_EMISS_NE simulations were evaluated against the IMD rainfall
312 dataset and NOR-I simulation to check whether the local or transported aerosols had greater control over the

313 rainfall in NE India. No_EMISS_NE showed better regional average rainfall statistics than Only_EMISS_NE
 314 with higher IOA (0.48 vs. 0.47), lower RMSE (18.85 vs. 20.37 mm day⁻¹), and lower ME (6.94 vs. 8.22 mm day⁻¹)
 315 ¹) on comparing with the IMD rainfall dataset. Also, the simulated rainfall of No_EMISS_NE showed higher
 316 rainfall similarity with NOR-I than Only_EMISS_NE with higher IOA (0.65 vs. 0.63), lower RMSE (56.32 vs.
 317 61.92 mm day⁻¹) and lower ME (39.30 vs. 39.81 mm day⁻¹). Hence, No_EMISS_NE showed more similarity with
 318 the baseline scenario as well as observed data and had greater control over the region's rainfall.

319 **3.4 Role of local and transported BC**

320 In section 3.3, the direct effect showed to increase moisture over NE India through an increase in atmospheric
 321 instability, caused mainly due to atmospheric heating of BC (Barman and Gokhale (2022)) Hence, to negate the
 322 effects of the indirect effect on atmospheric dynamics, scenarios in Table 1 containing only direct and semi-direct
 323 effects were used in this analysis. Moreover, NOR gave a much better performance with BC concentration
 324 estimation (Table S2) than when the indirect effect was included (NOR-I). The results from No_EMISS_NE,
 325 Only_EMISS_NE, No_NE_BCI and Only_NE_BCI scenarios were compared and related.

326 **3.4.1 Radiative heating**

327 The regional average vertical profiles of NOR, 2NOR, No_NE_BC, No_NE_2×BC, Only_NE_BC and
 328 Only_NE_2×BC can be seen from Fig. S11, in which the transported BC and local BC profiles resemble the
 329 No_EMISS_NE and Only_EMISS_NE PM₁₀ profiles, respectively. IGP was the dominant source of transported
 330 BC (Fig. S12). In transported BC scenarios, BC was available up to much higher atmospheric height and profiles
 331 showed elevated concentration at around 1500 m indicating stronger BC transport at that height. In Only_NE_BC
 332 and Only_NE_2×BC, BC was confined near the surface, which decreased continuously. The atmospheric heating
 333 rate (HR) was estimated as per Liou (1980).

$$334 \quad HR = \frac{g}{C_p} \cdot \frac{\Delta F}{\Delta P}, \quad (4)$$

335 where g is the acceleration due to gravity (9.81 m s⁻²), C_p is the specific heat capacity of air at constant pressure
 336 (1.005 kJ K⁻¹ kg⁻¹), ΔF the atmospheric RF and ΔP is the atmospheric pressure (300 hPa) difference between
 337 surface and 3 km altitude as most of the BC was present below this height. Moreover, in order to compare the
 338 effectiveness of heating by local and transported BC, two parameters, heating efficiency (HE) and heating slope
 339 (HS), were defined by equations 5 and 6.

$$340 \quad HE = \frac{HR}{\text{Column sum of BC concentration within 3 km (CC)}}, \quad (5)$$

$$341 \quad HS = \frac{\Delta HR}{\Delta CC}, \quad (6)$$

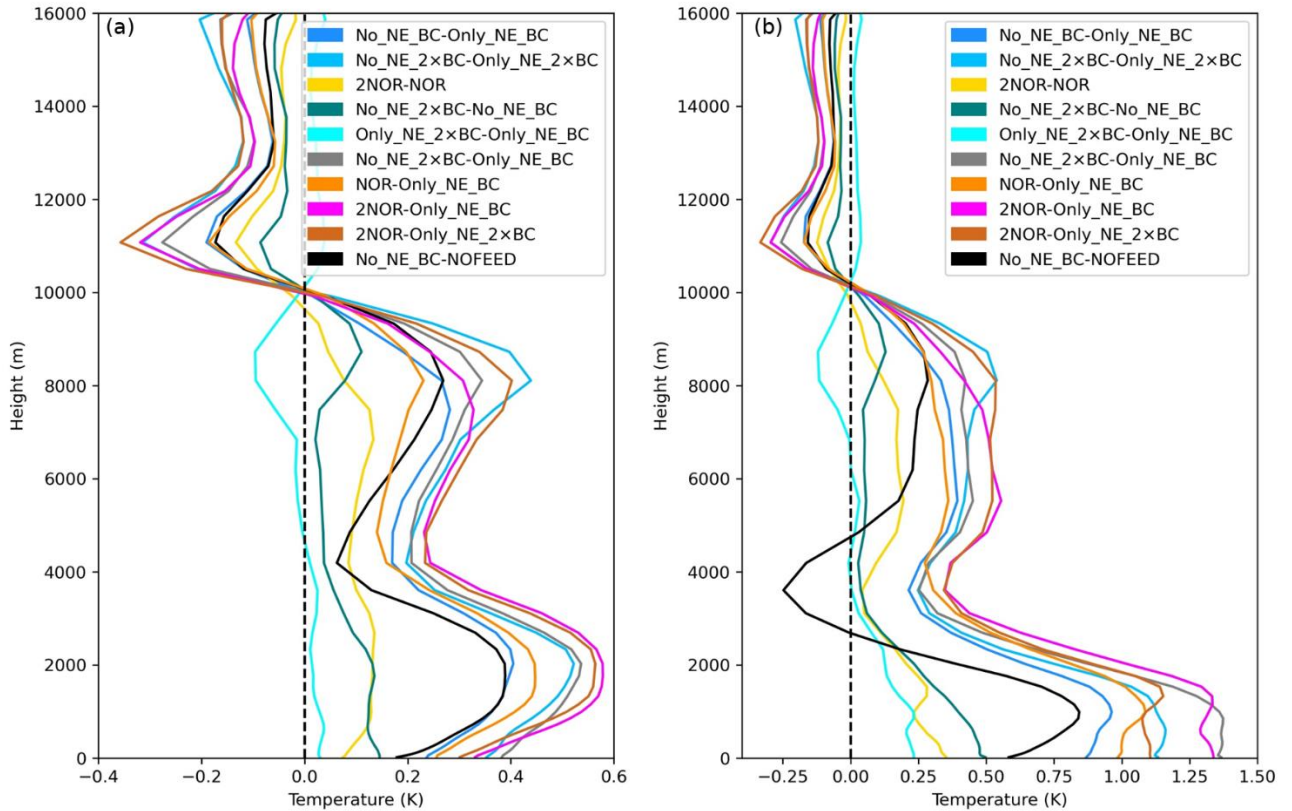
342 HE has units of K day⁻¹ μg⁻¹ m³, thus measuring the heating contributed by per unit concentration of BC below 3
 343 km. HE was used to assess the effect of BC vertical distribution on atmospheric heating while HS was used to
 344 assess the response of atmospheric heating rate to BC concentration changes and has similar units as HE. CC has
 345 units of μg m⁻³.

346 Table 4: NE India region average values of columnar BC concentration ($\mu\text{g m}^{-3}$) and atmospheric heating
 347 parameters in different scenarios

	No_NE_BC	No_NE_2×BC	Only_NE_BC	Only_NE_2×BC
HR	0.460	0.597	0.123	0.178
CC	12.458	18.391	3.905	7.563
HE	0.037	0.032	0.032	0.024
ΔHE	-0.004		-0.008	
HS	0.023		0.015	

348 The quantitative values of the parameters are provided in Table 4. Only_NE_BC had a regional net average HR
 349 of 0.123 K day^{-1} compared to 0.460 K day^{-1} of No_NE_BC. This indicated a 3.73 times higher atmospheric heating
 350 rate by transported BC. An increase in local emissions from Only_NE_BC to Only_NE_2×BC caused a small
 351 increase in heating rate of 0.055 K day^{-1} compared to the increase of 0.137 K day^{-1} from No_NE_BC to
 352 No_NE_2×BC. As per the definition, HE was inversely proportional to CC and this was exactly followed in all
 353 regions across all scenarios (Fig. S13 and S14). However, HE was higher in the case of transported BC compared
 354 to local BC with values of $0.037 \text{ K day}^{-1} \mu\text{g}^{-1} \text{ m}^3$ (No_NE_BC) vs. $0.032 \text{ K day}^{-1} \mu\text{g}^{-1} \text{ m}^3$ (Only_NE_BC) and
 355 $0.032 \text{ K day}^{-1} \mu\text{g}^{-1} \text{ m}^3$ (No_NE_2×BC) vs. $0.024 \text{ K day}^{-1} \mu\text{g}^{-1} \text{ m}^3$ (Only_NE_2×BC), even if CC was higher in the
 356 case of transported BC. The reason might be that transported BC might have undergone a higher amount of
 357 chemical transformation due to higher atmospheric time, leading to a higher lensing effect on the BC core,
 358 resulting in enhanced absorption (Liu et al., 2015). Also, it was observed that on increasing emissions, the decrease
 359 in HE was smaller in the case of transported BC ($-0.004 \text{ K day}^{-1} \mu\text{g}^{-1} \text{ m}^3$) than local BC ($-0.008 \text{ K day}^{-1} \mu\text{g}^{-1} \text{ m}^3$).
 360 Hence, with the increase in BC emissions, HE decreased more when BC was more concentrated near the surface
 361 than in the atmosphere. HS indicated that atmospheric heating increased at a higher rate of $0.023 \text{ K day}^{-1} \mu\text{g}^{-1} \text{ m}^3$
 362 with increasing transported BC compared to $0.015 \text{ K day}^{-1} \mu\text{g}^{-1} \text{ m}^3$. Thus, the increase in transported BC emissions
 363 had more impact on atmospheric heating over NE India than when present near the surface with local emissions.

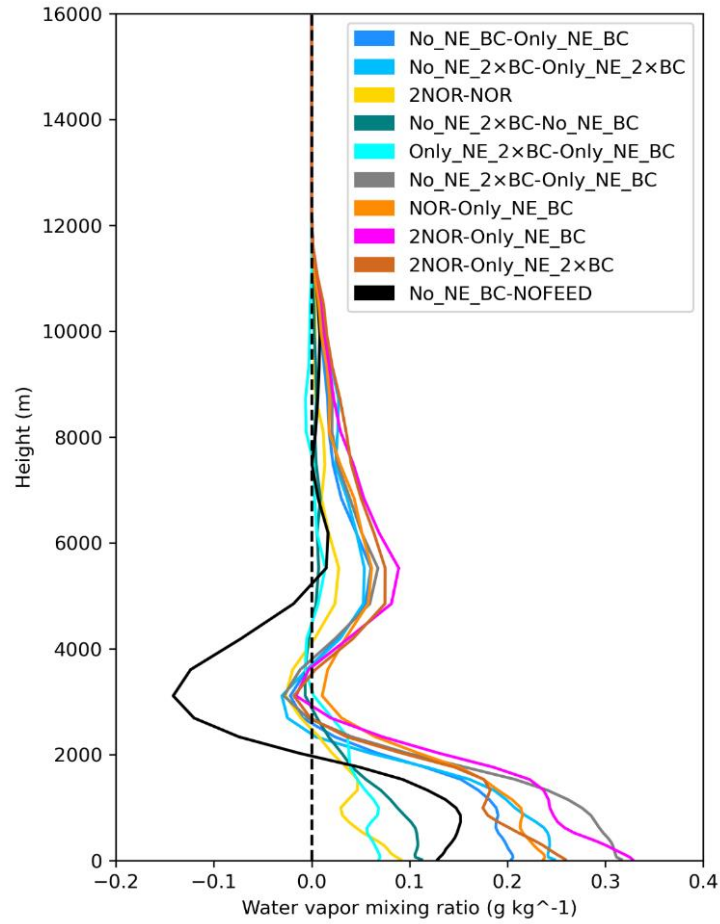
364 3.4.2 Atmospheric stability and moisture



365

366 Figure 7: Regionally averaged vertical profiles showing perturbations in a) potential temperature (K) b)
 367 equivalent potential temperature (K)

368 Barman and Gokhale (2022), as well as Soni et al. (2017), showed an increased influx of moisture into the region
 369 during pre-monsoon due to BC. In order to compare and separate the effects of local and transported BC on
 370 atmospheric stability through temperature and moisture, potential temperature (PT) and EPT were estimated. PT
 371 estimates atmospheric stability based on temperature, while EPT accounts for both temperature and moisture and
 372 is a more realistic parameter. In most of the profiles in both parameters in Fig. 7(a) and 7(b), positive perturbation
 373 was observed approximately below 10 km and negative above it which indicated an increase in atmospheric
 374 instability and vice-versa for an increase in atmospheric stability(Zhao et al., 2011). The positive perturbations
 375 below 10 km varied with height and were most profound in the profiles No_NE_BC – Only_NE_BC,
 376 No_NE_2xBC – Only_NE_2xBC and No_NE_2xBC – Only_NE_BC, each of which was estimated from the
 377 difference between a transported BC scenario and local BC scenario. These profiles showed similarity with the
 378 corresponding profiles of NOR – Only_NE_BC, 2NOR – Only_NE_2xBC and 2NOR – Only_NE_BC in both
 379 the parameters, indicating that they were closer to the normal atmospheric scenario. The positive perturbations
 380 were, however, comparatively smaller with 2NOR – NOR, No_NE_2xBC – No_NE_BC and Only_NE_2xBC –
 381 Only_NE_BC in both the parameters, each pair being the same scenario with only a difference in emission rates.
 382 This shows that BC atmospheric distribution played an important role on instability. The Only_NE_2xBC –
 383 Only_NE_BC profile not only showed a smaller increase in instability than No_NE_2xBC – No_NE_BC profile
 384 but also contributed to the smallest increase in instability in both the parameters. Thus, transported BC and an
 385 increase in transported BC emissions led to higher atmospheric instability than local BC.



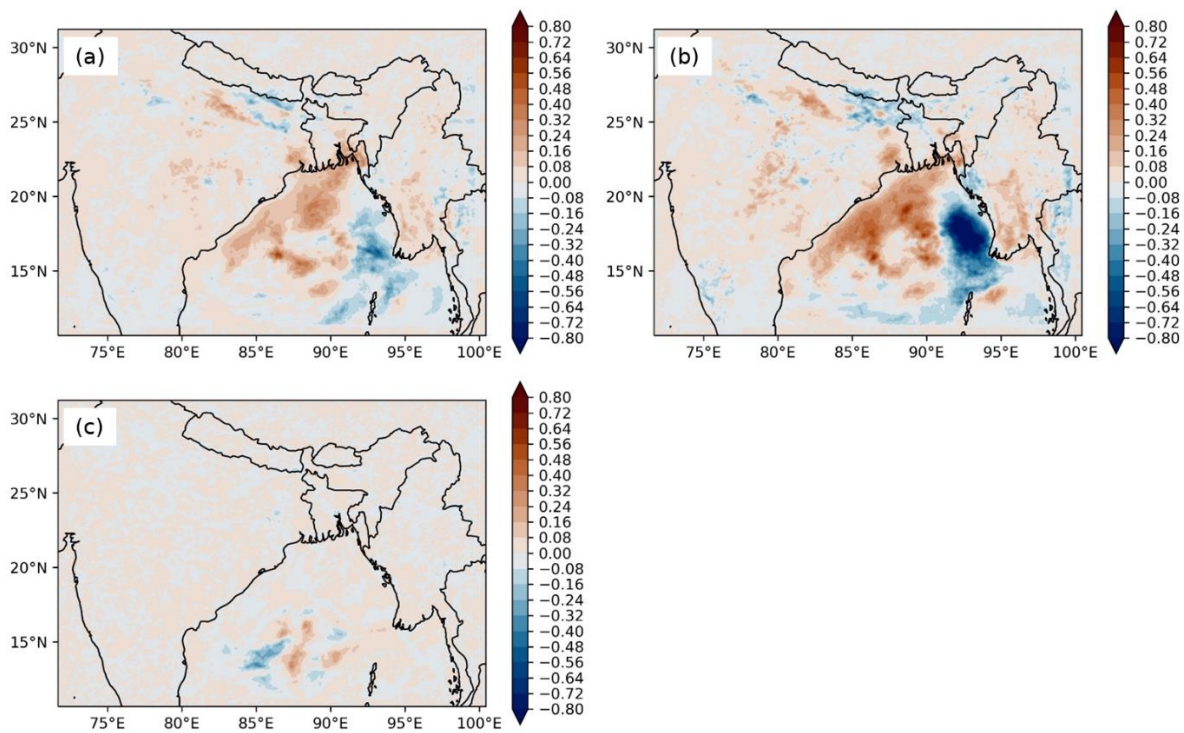
386

387 Figure 8: Regionally averaged vertical profiles showing perturbations in water vapor mixing ratio (g kg^{-1})

388 Moreover, EPT profiles showed higher positive perturbations and hence higher instability compared to
 389 the corresponding PT profiles with values exceeding 1.25 K. The positive difference or additional instability
 390 between the corresponding profiles of Fig. 7(a) and 7(b) was due to moisture. The difference also indicated that
 391 moisture contributed even more to the instability than BC. The peaks for EPT existed closer to the surface due to
 392 most of the moisture also remaining near the surface, as shown in Fig. 8. However, there occurred a region of
 393 increased stability from the ground surface to the first peak of transported BC profiles at approximately 1000 m,
 394 indicated by increasing temperature with height. Thus, transported BC may also be responsible for air quality
 395 scenarios over NE India by creating a stable boundary layer. The close qualitative and quantitative similarity
 396 between No_NE_BC – NOFEED, No_NE_BC – Only_NE_BC and NOR – Only_NE_BC profiles in Fig. 7(a)
 397 showed that aerosol radiative effect due to transported BC was intricately linked with the PT profile and the
 398 positive perturbations in each of these profiles were also closely linked with BC. This was also seen in Fig. 7(b),
 399 but since it also included the effect of moisture, larger differences were seen.

400 BC, whether transported or emitted locally, caused a positive perturbation in moisture at least below 2
 401 km altitude, as seen in Fig. 8. The perturbation was much larger in profiles that had a combination of transported
 402 and local BC scenarios and which had higher transported BC emissions and followed the pattern similar to PT
 403 and EPT. This links BC, instability and moisture in the region, i.e., higher transported BC caused higher instability
 404 which brought a higher amount of moisture which would possibly again cause higher instability. It was the same

405 for scenarios that included indirect effect, as can be observed from the similarities of the No_NE_BCI –
 406 Only_NE_BCI (Fig. S15) and No_NE_BC – Only_NE_BC profile in Fig.8. Furthermore, the similarity of
 407 No_EMISS_NE – Only_EMISS_NE profile with No_NE_BCI – Only_NE_BCI (Fig. S15) inferred that direct
 408 radiative effect of transported BC was responsible for the moisture increase in Fig. 4. The higher moisture with
 409 transported BC scenarios was due to higher moisture flux caused by it over Bay of Bengal compared to local BC
 410 and can be verified from Fig. 9. Quantitatively, No_NE_BC (33.95 kg m^{-2}) and No_NE_2×BC (34.15 kg m^{-2}) had
 411 higher region average precipitable water vapor than Only_NE_BC (33.49 kg m^{-2}) and Only_NE_2×BC (33.64 kg m^{-2}).
 412 Hence transported BC in Sect. 3.3 was primarily responsible for transporting moisture from the Bay of
 413 Bengal by affecting the atmospheric dynamics. The mechanism is similar to the "heat pump" model by Lau et al.
 414 (2006).

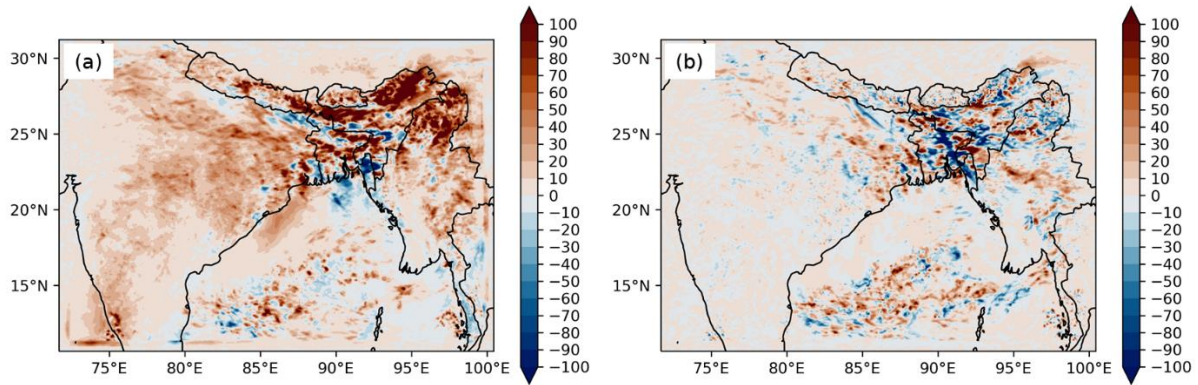


415
 416 Figure 9: Spatial distributions of change in moisture flux ($\text{g s}^{-1} \text{ m}^{-2}$) in a) No_NE_BC – Only_NE_BC b)
 417 No_NE_2×BC – Only_NE_BC c) Only_NE_2×BC – Only_NE_BC near surface

418 3.5 Rainfall response to emissions

419 Similar to NOR-I – NOCHEM, No_BC_ABS – NOCHEM gave the rainfall change due to total aerosol effect,
 420 but without BC absorption. The higher negative rainfall change of -275.13 mm with NOR-I – NOCHEM
 421 compared to -266.78 mm with No_BC_ABS – NOCHEM showed BC absorption to reduce rainfall. The higher
 422 reduction with NOR-I – NOCHEM was mainly due to higher rainfall reduction in region 1, where the direct and
 423 semi-direct effect was maximum. This shows BC initially suppressed rainfall even though moisture increased due
 424 to it. However, with the increase in BC emissions, rainfall increased and the rainfall suppression due to the total
 425 aerosol effect reduced substantially to -64.44 mm with 4NOR-I – NOCHEM compared to -275.13 mm with NOR-
 426 I – NOCHEM and similarly, rainfall due to direct and semi-direct with 4NOR-I – NOFEED-I showed a positive
 427 rainfall change of 193.64 mm compared to -17.04 mm with NOR-I – NOFEED. Similarly, 4NOR-I – NOR-I gave

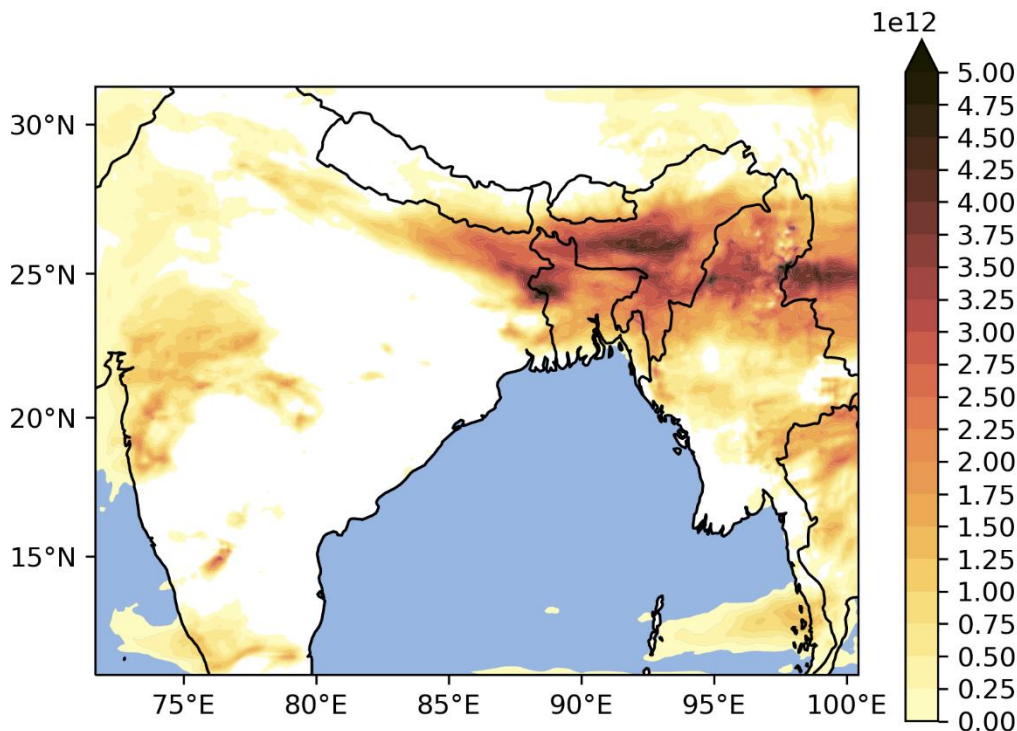
428 a rainfall enhancement of 225.24 mm. Spatial distribution of change in rainfall is shown in Fig. 10(a) which show
 429 rainfall change primarily occurring over NE India and along the valley.



430

431 Figure 10: Spatial distributions of change in rainfall (mm) in a) 4NOR-I – NOR-I b) No_EMISS_NE_4SO₂ –
 432 No_EMISS_NE_0.25SO₂

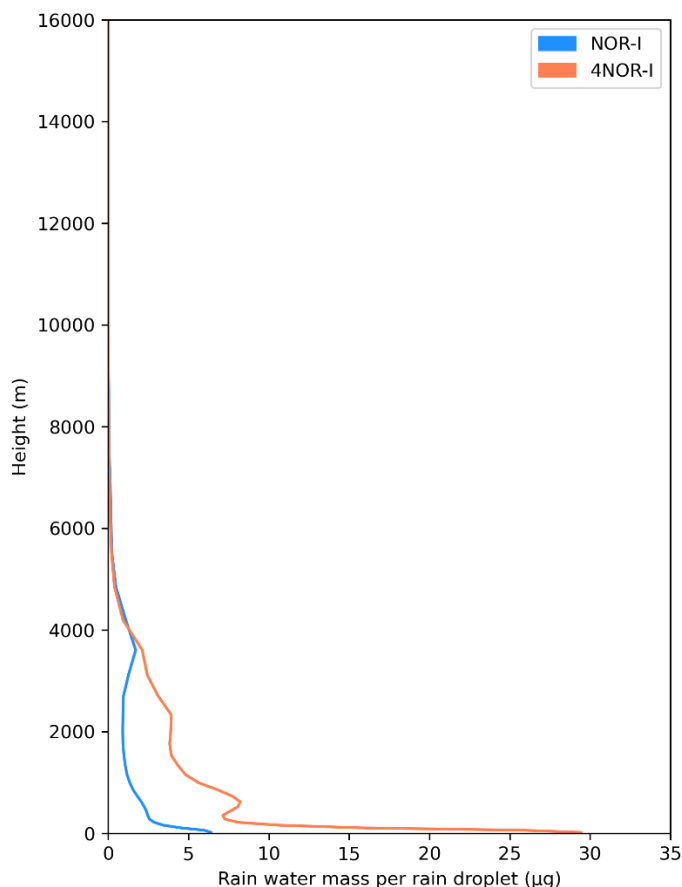
433 Aged BC also contributes as CCN (Lambe et al., 2015). The enhancement in BC emission did increase the column
 434 average CCN concentration to 2252 m⁻³ (4NOR-I) from 2024 m⁻³ (NOR-I), but the increase was largely
 435 disproportionate to the 4 times BC emission increase. The enhancement over NE India can also be seen from the
 436 spatial distribution of column integrated CCN in Fig. 11.



437

438 Figure 11: Spatial distribution of column integrated CCN number (m⁻²), estimated from 4NOR-I – NOR-I
 439 Enhancement of CCN number concentration generally leads to enhancement of indirect aerosol effect (Yu et al.,
 440 2013) and also seen later in case of sulfate aerosol. However, in spite of the increase in CCN, cloudwater mixing
 441 ratio was lower in 4NOR-I than NOR-I, as seen in Fig. 6 and 4NOR-I caused significantly more rainfall formation

442 than NOR-I, as can be seen from the rainwater mixing ratio profiles. This may be related to the suppression of
 443 CCN activation due to BC, as observed over Central India (Nair Jayachandran et al., 2020). Also BC contributes
 444 marginally to indirect effect (Kristjánsson, 2002). Thus, the increased moisture (Fig. S15) did not remain stored
 445 as cloudwater even though there was an increase in CCN, but it got converted to rainwater. The large increase in
 446 moisture, caused by the increase in atmospheric instability possibly condensed on relatively a smaller number of
 447 CCN particles promoting larger cloud droplets which enhanced rainfall. Moreover, the ratio of rainwater mixing
 448 ratio to rain droplet number concentration gave the amount of rain water per rain droplet, or indirectly the rain
 449 droplet size. The vertical profile of this ratio is shown in Figure 12, which shows higher values for 4NOR-I.



450

451 Figure 12: NE India region averaged vertical profiles of rain water mass per rain droplet

452 Collision is the primary mechanism of rain development in warm clouds (Lamb and Verlinde, 2011). Since rain
 453 droplets are formed from the gathering of cloud droplets, the higher value for 4NOR-I indicated larger rain droplet
 454 formation, possibly through better collisions among the cloud droplets, besides higher moisture availability. This
 455 indicated that the increase in BC emissions didn't contribute to rainfall suppression through indirect aerosol effect
 456 though there was an increase in CCN concentration, but rather counteracted the suppression of rainfall due to the
 457 indirect effect of other aerosol species. The rainfall enhancement was due to an increase in moisture, contributed
 458 by the transported fraction of BC, as explained in Sect. 3.4.2. Moreover, rainfall suppression was also more due
 459 to transported aerosols, mainly contributed by indirect effect (Table 3). Also, among the non-absorbing aerosols,
 460 sulfate aerosol is an important contributor to CCN and indirect effect (Kristjánsson, 2002). Its concentration was
 461 found to be the highest among non-absorbing aerosols and most of its mass over NE India was found to be

462 transported (Sect. 3.1). Concentration profiles can be seen from Fig. S16. Hence, the response of rainfall over NE
463 India was checked by increasing (No_EMISS_NE_4SO₂) and decreasing (No_EMISS_NE_0.25SO₂) SO₂
464 emissions outside NE India and compared against the baseline transported scenario (No_EMISS_NE) since sulfate
465 is mainly formed within the atmosphere by oxidation of SO₂ (Wang et al., 2021). Similar to the increase in BC
466 emissions, No_EMISS_NE_4SO₂ caused an increase in column average CCN concentration to 3524 m⁻³ compared
467 to 1753 m⁻³ in No_EMISS_NE, while No_EMISS_NE_0.25SO₂ showed a decrease (1390 m⁻³). However, contrary
468 to the BC, an increase in SO₂ emissions with No_EMISS_NE_4SO₂ caused an increase in cloudwater mixing ratio
469 compared to No_EMISS_NE, as seen in Fig. 6, while its decrease also caused a decrease. Thus,
470 No_EMISS_NE_4SO₂ and No_EMISS_NE_0.25SO₂ had lower and higher rainwater mixing ratio, respectively,
471 compared to No_EMISS_NE. Consequently, No_EMISS_NE_4SO₂ had higher rainfall suppression and gave
472 lesser rainfall (-22.23 mm) compared to No_EMISS_NE_0.25SO₂. Spatial distribution is shown in Fig. 10(b)
473 which show mainly negative change over the region. Thus, an increase in non-absorbing aerosol caused rainfall
474 suppression through indirect effect. The indirect effect was observed to be the dominant aerosol effect for
475 suppressing rainfall. However, with an increase in BC, suppression of rainfall due to direct and semi-direct effects
476 through surface processes (surface moisture flux, convection) and cloud evaporation as well as due to indirect
477 aerosol effect (atmospheric stability, surface moisture flux and cloud to rainwater conversion) becomes
478 comparatively weaker mechanisms than the direct effect of radiative heating by BC, enhancing rainfall through
479 the transport of moisture. However, the increase in transported SO₂ emissions also caused further suppression of
480 rainfall. Hence, an increase in transported aerosols of an absorbing aerosol (BC) and a non-absorbing aerosol
481 (sulfate), both being a contributor to CCN, exerted different impacts to indirect effect parameters and thus to
482 rainfall and hence most likely controls the enhancement and suppression of pre-monsoon rainfall over NE India,
483 thus counteracting each other. However, since decreasing rainfall trend has been observed, the impacts of the
484 indirect aerosol effect could be dominant. Here, the response of only one non-absorbing aerosol (sulfate) was
485 checked and possibly has contributions from other similar species also. Other non-absorbing aerosol species like
486 nitrate also contribute to indirect aerosol effect (Wang et al., 2010; Zaveri et al., 2021) which may contribute to
487 rainfall suppression as sulfate.

488 Moreover, the percentage of the simulation time different aerosol effects and BC emissions increased
489 (inc) or suppressed (dec) rainfall under different rainfall intensities (low: 0-5, medium: 5-10, high: > 10 mm day⁻¹;
490 defined as per (Raju et al., 2015)) and the rainfall amount under those intensities was estimated. Regional
491 average values are provided in Table S6 and S7. All aerosol effects caused a higher decrease across all rainfall
492 intensities, except the indirect effect, which indicated a higher increase in low-intensity rainfall (6.52 mm vs. -
493 6.48 mm; 21.44 % vs. 20.58 %). High-intensity rain was primarily responsible for rainfall changes across all the
494 scenarios and effects. The indirect effect decreased high-intensity rainfall duration (18.85 vs. 12.38 %) and amount
495 (-399.41 mm vs. 141.62 mm) and was primarily responsible for the rainfall suppression in total aerosol effect (-
496 411.34 mm). The total aerosol effect with enhanced BC emissions (4NOR-I – NOCHEM) showed a significantly
497 higher increase (275.47 mm vs. 137.16 mm) as well as a significantly lower decrease (-337.23 vs. -411.34) in
498 high-intensity rainfall compared to total aerosol effect with baseline BC emissions (NOR-I – NOCHEM). Similar
499 results in time and rainfall amount between BC increase and direct + semi-direct effect with BC increase scenarios
500 inferred that enhanced radiative effects due to BC increase were mainly responsible for higher high-intensity
501 rainfall duration and rainfall amount, while the indirect aerosol effect was mainly involved in its suppression,

502 possibly due to the increased atmospheric stability associated with it. Barman and Gokhale (2022) also showed
503 similar results with BC emissions increase, but this study verifies the role of direct radiative effects of BC in it.
504 Thus, BC increased rainfall over NE India but in the form of high-intensity rainfall. Hence, relative fractions of
505 BC and the other aerosols contributing to indirect effect possibly decide the amount of rainfall and its intensity
506 over the region. However, indirect effect also caused high-intensity rainfall but with lesser amount than its
507 suppression and may be involved in catastrophic flood events at local scales (Wang et al., 2022).

508 **4 Conclusions**

509 Transported aerosols, primarily from IGP, were found to be responsible for the bulk of the aerosol mass (93.98
510 %) over NE India while contributing 64.18 % of near-surface PM₁₀ concentration, thus primarily responsible for
511 air pollution as climatic impacts over the region during pre-monsoon season. The climatic impacts, both w.r.t. RF
512 as well as rainfall, were dominated by the indirect aerosol effect. The impacts of the indirect aerosol effects of
513 transported aerosols were much higher in affecting radiation (-13.12 W m⁻² vs. -0.24 W m⁻² at the surface, 7.30 W
514 m⁻² vs. 0.97 W m⁻² in the atmosphere) as well as suppressing rainfall (-49.11 mm vs. -16.04 mm) compared to
515 local emissions. The greater surface dimming by transported aerosols caused a higher negative change in surface
516 moisture flux (-3.82×10⁻⁶ kg m⁻² s⁻¹ vs. 8.15×10⁻⁸ kg m⁻² s⁻¹) as well as higher aerosol mass reduced cloudwater to
517 rainwater conversion, both of which contributed to higher rainfall suppression. Transported aerosols caused
518 4.42×10¹³ m² higher cloud droplets than local emissions. The atmospheric instability due to the direct + semi-
519 direct effect and indirect effect of transported aerosols were found to be contradictory and caused an increase and
520 decrease, respectively. The direct effect of transported aerosols, though also caused negative surface moisture flux
521 over NE India (-1.03×10⁻⁶ kg m⁻² s⁻¹), however, increased moisture over NE India, increasing moisture flux over
522 the Bay of Bengal. Further analysis showed that transported BC was more efficient in atmospheric heating over
523 NE India and together with the higher transported BC mass, an increase in its emissions caused higher atmospheric
524 instability over the region, which brought more moisture from the Bay of Bengal. The increased moisture further
525 contributed to higher instability. Hence, the rainfall suppression caused through the different atmospheric
526 processes by direct, semi-direct and indirect effects was reduced and nullified with the increase in BC emissions,
527 but the rainfall increase was mainly in the form of high-intensity rainfall. The increase in BC did not show a
528 positive change in cloudwater, though it contributed to CCN. The direct effect of BC thus overpowered the other
529 rainfall-suppressing processes. Indirect aerosol effect and radiative heating were the main rainfall-controlling
530 factors. Hence, changes in emissions of aerosols or chemical species contributing to these processes will possibly
531 contribute to rainfall suppression and enhancement over NE India. Moreover, rainfall simulated with transported
532 aerosols were found to be more similar to the IMD observation datasets as well as the baseline emission scenario,
533 indicating its possible greater influence in the real-world scenario.

534 The study shows that the atmospheric transport of emissions from IGP to NE India has a significant
535 impact on NE India's rainfall during pre-monsoon and the impacts are even greater than the emissions within the
536 NE India region.

537 **Data availability.** Model outputs are available upon request.

538 **Author contributions.** NB - conceptualization, methodology, model simulation, visualisation, manuscript
539 writing, SG - conceptualization, methodology and supervision, manuscript review and editing.

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