



1 **Tropospheric warming over the North Indian Ocean caused by the South Asian**
2 **anthropogenic aerosols: possible implications**

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17 **Abstract**

18 Atmospheric concentrations of South Asian anthropogenic aerosols and their transport play a
19 key role in the regional hydrological cycle. Here, we use the ECHAM6-HAMMOZ
20 chemistry-climate model to show the structure and implications of the transport pathways of
21 these aerosols during **spring**. Our simulations indicate that large amounts of anthropogenic
22 aerosols are transported from South Asia to **the North Indian Ocean (the Arabian Sea and**
23 **North Bay of Bengal)**. These aerosols are then lifted into the upper troposphere and lower
24 stratosphere (UTLS) by **the** convection over the Arabian Sea and Bay of Bengal. In the
25 **UTLS, they are further transported to the southern hemisphere (30-40°S) and downward into**
26 **the troposphere** by the **secondary circulation induced by the aerosol changes**. The
27 carbonaceous aerosols are also transported to the Arctic and Antarctic producing local
28 heating (0.002 – 0.05 K d⁻¹).



29 The presence of anthropogenic aerosols causes negative radiative forcing (RF) at the TOA (-
30 $0.90 \pm 0.089 \text{ W m}^{-2}$) and surface ($-5.87 \pm 0.31 \text{ W m}^{-2}$) and atmospheric warming ($+4.96 \pm 0.24 \text{ W}$
31 m^{-2}) over South Asia ($60^\circ \text{ E} - 90^\circ \text{ E}$, $8^\circ \text{ N} - 23^\circ \text{ N}$), except over the Indo-Gangetic plain
32 ($75^\circ \text{ E} - 83^\circ \text{ E}$, $23^\circ \text{ N} - 30^\circ \text{ N}$) where RF at the TOA is positive ($+1.27 \pm 0.16 \text{ W m}^{-2}$) due to
33 large concentrations of absorbing aerosols. The carbonaceous aerosols produced in-
34 atmospheric heating along the aerosol column extending from the boundary layer to the
35 UTLS (0.01 to 0.3 K d^{-1}) and in the stratosphere globally (0.002 to 0.012 K d^{-1}). The heating
36 of the troposphere increases water vapor concentrations, which are then transported from
37 highly convective region (i.e. the Arabian Sea) to the UTLS (increasing water vapor by $0.02 -$
38 0.06 ppmv).

39 Keywords: South Asian Anthropogenic aerosols; warming over the Arabian Sea; transport of
40 aerosols and water vapor to the UTLS in spring.

41



42 1. Introduction

43 Understanding the variability of anthropogenic aerosol loading over the North Indian
44 Ocean is of utmost importance since (1) it regulates the Asian hydrological cycle via
45 modulating atmospheric convection, heating rates, and moisture transport (Ramanathan et al.
46 2005; Corrigan et al., 2008; Budhavant et al., 2018, Meehl et al., 2008), and (2) it leads to
47 adverse impacts on marine ecosystems (Mahowald et al., 2018; Collins et al., 2019). Several
48 observations indicate that ~~the~~ aerosol loading over the North Indian Ocean during the spring
49 season is strongly influenced by South Asian aerosols. Aircraft measurements during the
50 Indian Ocean Experiment (INDOEX) (February–March 1999) showed the presence of a thick
51 layer (surface to 3.2 km) of anthropogenic aerosols (BC~14 %, sulfate 34 %, ammonium 11
52 %) over the North Indian Ocean (Dickerson et al., 2002; Mayol-Bracero et al., 2002) with
53 sources over South Asia. Several other in situ observations, e.g. over the Maldives during
54 November 2014 – March 2015, show that air masses arising from the Indo-Gangetic Plain
55 contain very high amounts (97 %) of ~~the~~ elemental carbon in the fine mode. Other
56 anthropogenic species such as organic carbon, non-sea-salt, potassium, and ammonium (70–
57 95%) were also observed in the fine mode (Bhuhvant et al., 2018). Observations from the
58 Geosphere-Biosphere Programme over the Bay of Bengal during spring (March 2016) also
59 show abundant anthropogenic aerosols (sulfate and nitrate) having sources over the Indo-
60 Gangetic plain (Nair et al., 2017).

61 The aerosol loading over South Asia has been increasing at an alarming rate (rate of
62 increase in AOD 0.004 per year during 1988 – 2013) (Babu et al., 2013). For the last two
63 decades, ~~the~~ AOD increase (by 12 %) over south Asia has been attributed to the strong
64 increase in anthropogenic aerosols (sulfate, black carbon, and organic carbon), while natural
65 aerosol remained unchanged (Ramachandran et al., 2020a). The major sources of



66 anthropogenic aerosols are the combustion of domestic fuels, industrial emissions,
67 transportation, and open burning (Paliwal et al., 2016). The growth of the economy of India
68 led to a 41 % increase in BC and 35 % in OC from 2000 to 2010 (Lu et al., 2011). The
69 emissions of sulfur dioxide (SO₂) which leads to the production of sulfate aerosols have
70 doubled during 2006 – 2017 (Fadnavis et al., 2019). Figure 1 a-c shows the annual mean
71 emission of BC, OC, and sulfate aerosols over South Asia in 2016, with high emissions over
72 the Indo-Gangetic Plain (BC 7E-12 – 17E-12 Kg m⁻² S⁻¹, OC: 25E-12 - 70E-12 Kg m⁻² S⁻¹,
73 sulfate: 2E-12 - 5E-12 Kg m⁻² S⁻¹). Higher amounts of aerosols over the Indo-Gangetic Plain
74 are associated with densely populated regions and industrial and vehicular emissions
75 (Karambelas et al., 2018, Fadnavis et al., 2019). Past studies also show substantially higher
76 amounts of aerosols over North India compared to rest of the Indian region (Ramachandran et
77 al., 2020b, Fadnavis et al., 2013, 2017a, 2017b). Over the Indo-Gangetic plain, these
78 emissions show a peak in spring (Fig. 1d), with increases for BC of 0 – 3 %, OC 0 – 8.7 %,
79 and sulfate 0 – 0.2 %, compared to annual means. This peak in emissions in spring is to a
80 large extent driven by springtime agricultural crop burning and biomass burning activity
81 (Chavan et al., 2021).

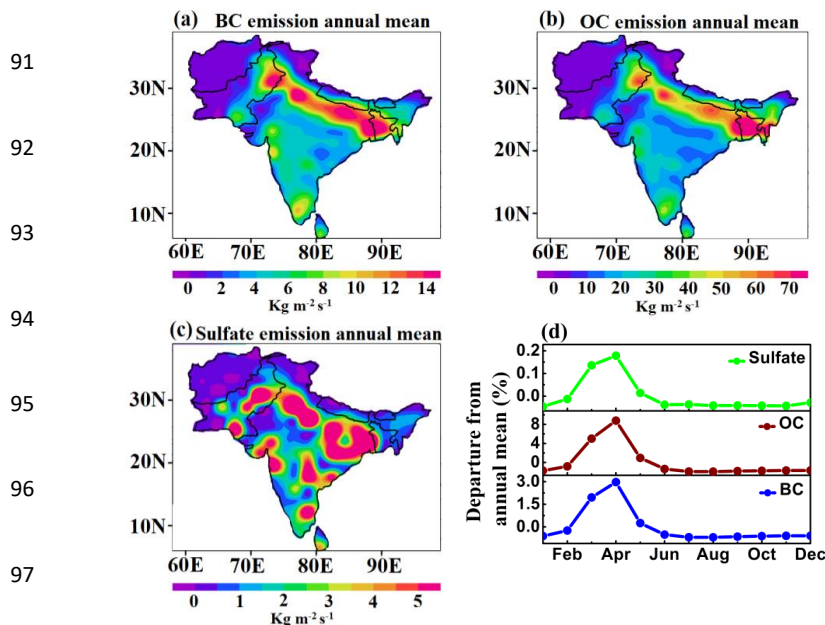
82 While the presence of sulfate aerosols lead to a cooling of the atmosphere below due to
83 their strong scattering properties, carbonaceous aerosols produce atmospheric warming via
84 absorption of solar radiation (Fadnavis et al., 2019, Penner et al., 1998). Previous studies
85 showed that the doubling of carbonaceous aerosols loading over Asia (10° S – 50° N, 65° E–
86 155° E) led to significant atmospheric warming (in-atmospheric RF 5.11W m⁻², Fadnavis et al.
87 2017b).

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98 Figure 1: Spatial distribution of year 2016 annual mean total emission ($\text{kg m}^{-2} \text{S}^{-1}$) of (a)
99 BC, (b) OC, (c) Sulfate aerosols, (d) time series of monthly departure from annual mean
100 total emissions (%) of BC, OC, and Sulfate aerosols averaged over Indo-Gangetic plain
101 ($23^\circ - 30^\circ \text{N}$, $78 - 90^\circ \text{E}$).

102 During spring, the prevailing convective instability over the Bay of Bengal and the
103 Arabian Sea transports aerosol from the boundary layer to the upper troposphere
104 (Romatschke and Houze, 2011). Airborne observations during winter and spring, e.g. the
105 Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container
106 (CARIBIC) in March 1999 and January 2001 (Papaspriopoulos et al., 2002), and the Indian
107 Ocean Experiment (INDOEX) in February-March 1999 show elevated aerosol amounts near
108 8 – 12 km over the Indian Ocean and South Asia (De Reus et al., 2001). Recently, using a set
109 of model simulations, Chavan et al., (2021) reported the transport of biomass burning
110 aerosols to the upper troposphere by the convection in spring 2013.



111 Here, we investigate the source of the very large aerosol loading over the Arabian Sea
112 during spring. These aerosols produce atmospheric warming leading to enhanced water vapor
113 that is transported to the UTLS. Once in the lower stratosphere, the water vapour is
114 transported globally, which has implications for tropospheric temperatures and possibly
115 stratospheric ozone. For this purpose, we performed a series of five simulations using the
116 ECHAM6-HAMMOZ model for changes in anthropogenic aerosol over South Asia.

117 **2. Model simulations and satellite data**

118 **2.1 ECHAM6-HAMMOZ experimental set-up**

119 We used the state of art ECHAM6–HAM aerosol–chemistry–climate model. It
120 comprises of the general circulation module ECHAM6, coupled to the aerosol and cloud
121 microphysics module **HAM** (Stier et al., 2005; Tegen et al., 2019). HAM predicts the
122 nucleation, growth, evolution, and sinks of sulfate, black carbon (BC), **particulate organic**
123 **matter** (POM), sea salt (SS), and mineral dust (DU) aerosols. The size distribution of the
124 aerosol population is described by seven log-normal modes with prescribed variance as in
125 the M7 aerosol module (Stier et al., 2005; Zhang et al., 2012). Moreover, HAM explicitly
126 simulates the impact of aerosol species on cloud droplet and ice crystal formation. Aerosol
127 particles **can act as cloud condensation nuclei or ice-nucleating particles**. Other relevant
128 cloud microphysical processes such as evaporation of cloud droplets, sublimation of ice
129 crystals, ice crystal sedimentation, and detrainment of ice crystals from convective cloud
130 tops are simulated interactively (Neubauer et al., 2014). The anthropogenic and fire
131 emissions of sulfate, black carbon (BC), and organic carbon (OC) are based on the
132 AEROCOM-ACCMIP-II emission inventory. Other details of the model and emissions are
133 reported by Fadnavis et al. (2017a, 2019, 2021a, b).



134 The model simulations are performed at the T63 spectral resolution corresponding to
135 $1.875^\circ \times 1.875^\circ$ in the horizontal dimension, while the vertical resolution is described by 47
136 hybrid σ -p levels from the surface up to 0.01 hPa (approx. 80 km). The simulations have
137 been carried out at a time step of 20 min. Monthly varying Atmospheric Model Inter-
138 comparison Project (AMIP) sea surface temperature (SST) and sea ice cover (SIC) (Taylor et
139 al., 2000) were used as lower boundary conditions.

140 We performed five model experiments: (1) a control (CTL) simulation where all aerosol
141 emissions are included and four perturbed experiments where (2) all anthropogenic aerosol
142 emissions (black carbon, organic carbon, and sulfate) are switched off over South Asia (75° –
143 100° E, 8° – 40° N, see Fig. 1) during the study period (2001 – 2016) (referred to as Aerooff),
144 (3) only anthropogenic black carbon emissions (BC) switched off during the study period,
145 (BCoff), (4) only anthropogenic organic carbon (OC) emissions switched off (OCoff) during
146 the study period, and (5) only anthropogenic sulfate aerosol emissions switched off (Suloff)
147 during the study period (see Table 1). All simulations were performed from 1 January 2001 to
148 December 2016 from stabilized initial fields created after a model integration for one year.
149 Dust emission parameterization is the same in all ~~the~~ simulations and is based on Tegen et al.
150 (2002). The analysis is performed for spring (March – May) averaged for the period 2001 –
151 2016. We compare the CTL with Aerooff, BCoff, OCoff, and Suloff simulations to
152 understand the impact of south Asian anthropogenic aerosols over the Indian region and
153 surrounding ocean.

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157 Table -1: Details of eECHAM6-HAMMOZ model simulations performed in this study.

Experiment name	Duration	Aerosol species on/off	Boundary conditions
CTL	2001 – 2016	All aerosols species globally, as per AEROCOM-ACCMIP-II emission inventory.	AMIP Sea surface temperature and sea ice concentration.
Aerooff	2001 – 2016	Anthropogenic BC, OC, and sulfate aerosols switch off over South Asia during 2001 – 2016.	AMIP Sea surface temperature and sea ice concentration.
BCoff	2001 – 2016	Anthropogenic BC aerosols switch off over South Asia during 2001 – 2016.	AMIP Sea surface temperature and sea ice concentration.
OCoff	2001 – 2016	Anthropogenic OC aerosols switch off over South Asia during 2001 – 2016.	AMIP Sea surface temperature and sea ice concentration.
Suloff	2001 – 2016	Anthropogenic sulfate aerosols switch off over South Asia during 2001 – 2016.	AMIP Sea surface temperature and sea ice concentration.

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159 2.2 AOD satellite observations

160 In this study we use the last fifteen years (2001 – 2016) of aerosol optical depth at 0.55 μm
161 (AOD) obtained from the Moderate Resolution Imaging Spectroradiometer
162 (MODIS) instrument onboard the NASA EOS Terra satellite. The MODIS instrument
163 measure radiance in 36 spectral channels at spatial resolution ranging from 250 m to 1 km
164 with a 2300 km wide swath, allowing for almost daily global coverage. Terra MODIS
165 (MOD08_M3 V6.1) AOD aerosol products are retrieved using the Deep Blue (DB) algorithm
166 (Mhawish et al., 2019). The algorithm calculates the column aerosol loading at 0.55 μm over
167 land and ocean. The **AOD data from MODIS Terra** can be downloaded from
168 <https://ladsweb.modaps.eosdis.nasa.gov/archive/allData/61/MODATML2/>



169 AOD data from the Multi-angle Imaging Spectro-Radiometer (MISR) for the same period as
170 MODIS (2001 – 2016) is also used for model evaluation. The MISR sensor onboard the Terra
171 satellite has been operational since 1999. It makes measurements at four spectral bands
172 centered at 443 nm, 555 nm, 670 nm, and 865 nm (Diner et al., 2008). In this study we used
173 level 3 (MIL3MAE_v4) monthly mean aerosol optical depth at 555 nm wavelength at spatial
174 resolution $0.5^\circ \times 0.5^\circ$. The MISR AOD data is available for download at
175 <https://misr.jpl.nasa.gov/getData/accessData/>.

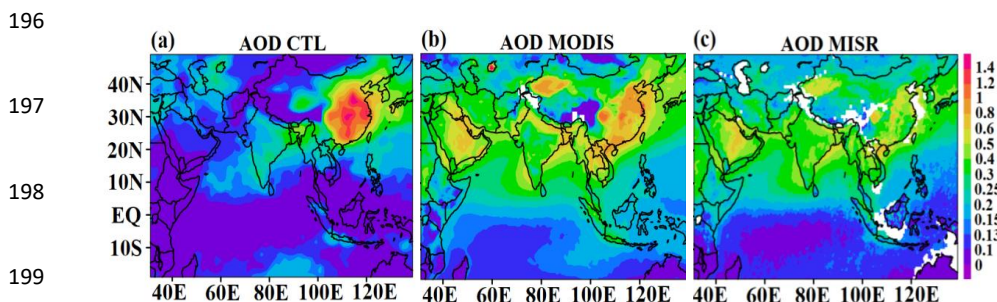
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177 2.3 Model evaluation

178 We evaluate model performance by comparing simulated AOD (from CTL simulations)
179 with MISR and MODIS data for the spring season. The model simulations show high
180 amounts of AOD over the Indo-Gangetic plain, Myanmar, and East Asia, consistent with
181 MODIS and MISR observations, despite quantitative differences (Fig. 2). Compared to
182 observations, the model underestimates AOD over the Indo-Gangetic plain (model: 0.15 to
183 0.4, MODIS: 0.4 to 0.8, MISR: 0.3 to 0.5) and overestimates AOD over East Asia (model:
184 0.6 to 1.4, MODIS: 0.4 to 1.2, MISR: 0.2 to 0.5). Over the Myanmar region, the model
185 underestimates AOD in comparison to MODIS, but overestimates it in comparison to MISR
186 (model: 0.15 to 0.5, MODIS: 0.3 to 0.8 MISR: 0.15 to 0.3). There are differences among
187 satellite observations and between the model and observations. The differences are due to
188 uncertainties in the model due to model transport processes, emission inventory, and
189 parametrisations (Fadnavis et al. 2014, 2015, 2018, 2019) and there are uncertainties in
190 satellite measurements (Bibi et al., 2015). With model biases present in both the CTL and
191 the perturbed simulations, investigating anomalies removes some of the model bias. In the
192 past Fadnavis et al. (2018, 2019, 2020, 2021a,b) reported model evaluations for AOD,
193 absorbing aerosol index, precipitation, mixing ratio of black carbon aerosol and cloud ice



194 with various measurements that show the fair performance of the ECHAM6-HAMMOZ
195 model.



200 **Figure 2:** Spatial distribution of AOD average for the spring season during 2001 – 2016, from
201 (a) ECHAM6-HAMMOZ CTL simulations, (b) MODIS measurements average for the spring
202 season during 2001 – 2016, (c) MISR measurement average for the spring season during
203 2001 – 2016.

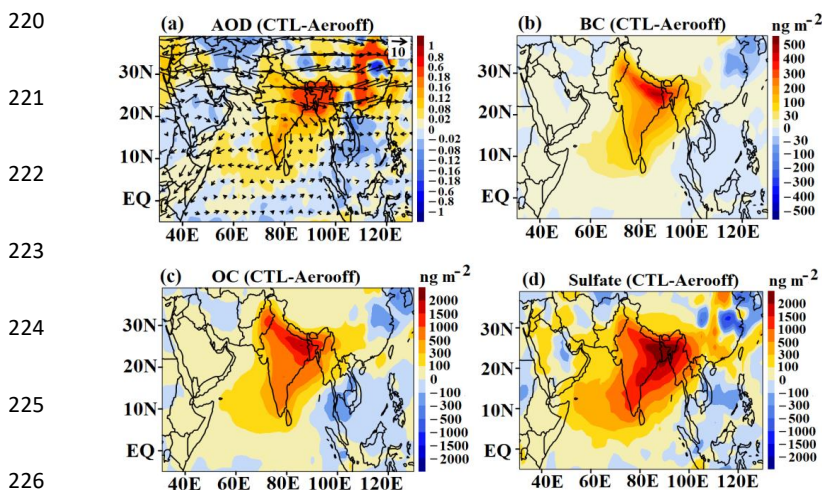
204 4. Results and discussions

205 4.1 Transport of South Asian aerosols to the North Indian Ocean

206 The spatial distribution of AOD anomalies from the Aerooff simulation shows positive
207 anomalies of AOD extending from South Asia to the Arabian Sea and the North Bay of
208 Bengal (10 – 20° N) (Fig. 3a). The wind vectors indicate that these are transported from the
209 Indo-Gangetic plain to the Arabian Sea and the Bay of Bengal. The transported aerosols
210 enhanced the AOD by 0.18 – 0.8 (30 – 80 %) over the North Bay of Bengal and by 0.02 – 0.12
211 (20 – 60 %) over the Arabian Sea. This is consistent with previous studies where 50 – 60 %
212 enhancements in the AOD over the tropical Indian Ocean due to anthropogenic aerosols have
213 been reported (Satheesh et al. 1999; Jose et al. 2020). Chemical analysis of aerosols observed
214 over the south-eastern coastal Arabian Sea also shows the dominance of anthropogenic
215 aerosols having sources over the Indian region (73 %) (Aswini et al., 2020). Analysis of
216 MODIS satellite observations (2003 – 2017) likewise shows that anthropogenic sources
217 contributed ~60 – 70% to the aerosol loading over the east coast and west coast of India (Jose



218 et al. 2020). Measurements over the Equatorial Indian Ocean further show a substantial
219 increase in AOD (~80 %) due to anthropogenic aerosols (Gogoi et al., 2019).



227 Figure 3: Spatial distribution of (a) AOD anomalies averaged for spring during 2001 –
228 2016 (CTL - Aerooff), and anomalies of tropospheric column of (b) BC, (c) OC, and (d)
229 sulfate aerosols (ng m^{-2}) (CTL-Aerooff). The vectors in Fig.1a indicate winds (m s^{-1}) at
230 850 hPa.

231 The distribution of anomalies in the tropospheric column of BC, OC, and sulfate aerosols also
232 indicates that these aerosols are transported from South Asia to the Bay of Bengal and the
233 Arabian Sea (Fig. 3b-d). Enhancement of sulfate and OC aerosol ($100 - 2000 \text{ ng m}^{-2}$) is
234 higher than BC ($100 - 500 \text{ ng m}^{-2}$) over the South Asian region (Fig. 3b-d). The total
235 carbonaceous aerosol (BC and OC together) dominates over the sulfate aerosols. These
236 anthropogenic aerosols over the tropical Indian Ocean affect the radiation budget and cloud
237 cover over the Indian Ocean (Satheesh et al. 1999; McFarquhar and Wang 2006).

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240 3.2. Radiative forcing

241 The anthropogenic aerosols over the tropical Indian Ocean affect the radiation budget and
242 cloud cover (McFarquhar and Wang 2006). Here, we discuss the impact of south Asian
243 anthropogenic aerosols on radiative forcing (RF). Figures 4a-c show anomalies in net
244 radiative forcing (RF) at the TOA, surface, and in-atmosphere (TOA - surface) for Aerooff
245 simulations (CTL - Aerooff). The anthropogenic aerosols have produced a cooling at the
246 TOA (except over the Indo-Gangetic plain) and surface (see Fig. 4a-b). The simulated RF
247 values over the Arabian Sea (55 – 70° E, 8 – 20° N), Bay of Bengal (88 – 92° E, 12 – 20° N),
248 and Indo-Gangetic Plain (75 – 83° E, 26 – 30° N) are tabulated in Table-S1. The RF
249 estimates show that aerosols have produced significant cooling at the TOA and surface over
250 the Arabian Sea (TOA: $-0.72 \pm 0.14 \text{ W m}^{-2}$, surface: $-3.0 \pm 0.28 \text{ W m}^{-2}$), Bay of Bengal (TOA:-
251 $1.24 \pm 0.15 \text{ W m}^{-2}$, surface: $-5.14 \pm 0.44 \text{ W m}^{-2}$), and in-atmospheric warming over the above
252 regions (Arabian Sea $+2.27 \pm 0.19 \text{ W m}^{-2}$; Bay of Bengal: $+3.89 \pm 0.30 \text{ W m}^{-2}$) (Fig. 4 c). The
253 Indo Gangetic Plain shows positive anomalies of RF at the TOA ($+1.27 \pm 0.16 \text{ W m}^{-2}$),
254 negative at the surface ($-11.16 \pm 0.50 \text{ W m}^{-2}$), and atmospheric warming of $+12.44 \pm 0.42 \text{ W m}^{-2}$.
255 In agreement with our results, several previous studies have reported negative RF at the
256 surface and TOA, and atmospheric warming over the north Indian Ocean caused by enhanced
257 anthropogenic aerosol. For example, Pathak et al. (2020) reported negative aerosol RF at the
258 TOA (-2 to -4 W m^{-2}) over the Bay of Bengal and the Arabian Sea during spring 2009 - 2013.
259 Reddy et al., (2004) estimated positive in-atmosphere RF over the North Indian Ocean ($+25$
260 W m^{-2}). The aerosol radiative forcing estimated from satellite measurements (January to
261 March 1999) over the north Indian ocean is also negative at TOA (-4 and -14 W m^{-2}) and
262 surface (-12 to -42 W m^{-2}) (Satheesh and Ramanathan 2000; Rajeev and Ramanathan et al,
263 2001). The clear sky aerosol direct radiative forcing estimated from measurements during the
264 INDOEX experiment (January to March in 1999) over the north Indian Ocean also show



265 similar results (TOA: -7 W m^{-2} , surface: -23 W m^{-2} , and in-atmosphere: $+16 \text{ W m}^{-2}$)
266 (Ramanathan et al. 2001). These studies attribute positive in-atmospheric radiative forcing to
267 absorbing aerosols (especially black carbon) that lead to heating of the atmosphere (Rajeev
268 and Ramanathan 2001; Satheesh et al 2002).

269 Analysis of the perturbed model experiments indicates that anthropogenic BC
270 aerosols (Fig. 4d-f) have produced a warming at the TOA (Arabian Sea: $1.24 \pm 0.13 \text{ W m}^{-2}$,
271 Bay of Bengal: $1.54 \pm 0.26 \text{ W m}^{-2}$, Indo-Gangetic Plain: $4.33 \pm 0.17 \text{ W m}^{-2}$) and cooling at the
272 surface (Arabian Sea: $-2.56 \pm 0.25 \text{ W m}^{-2}$, Bay of Bengal: $-3.70 \pm 0.49 \text{ W m}^{-2}$, Indo-Gangetic
273 Plain: $-9.27 \pm 0.37 \text{ W m}^{-2}$). OC (Fig. 4g-i) and sulfate (Fig. 4j-l) aerosols have produced
274 significant cooling at the TOA (OC: -0.21 ± 0.13 to $-0.44 \pm 0.15 \text{ W m}^{-2}$; Sulfate: -1.55 ± 0.16 to -
275 $2.14 \pm 0.17 \text{ W m}^{-2}$) and surface (OC: -0.49 ± 0.31 to $-2.56 \pm 0.45 \text{ W m}^{-2}$, Sulfate: -1.19 ± 0.24 to -
276 $2.67 \pm 0.36 \text{ W m}^{-2}$) over the above regions (listed in Table-S1). Figures 4d, 4g, and Fig. 4j
277 further confirm our finding that the positive anomalies of radiative forcing in the Indo-
278 Gangetic plain are due to BC aerosols because of its absorbing property. All the aerosols
279 produce in-atmospheric warming over the Indian region and the north Indian Ocean. The
280 atmospheric warming over the Arabian Sea and Bay of Bengal is due to BC and OC aerosols
281 with larger contributions by the BC aerosols.

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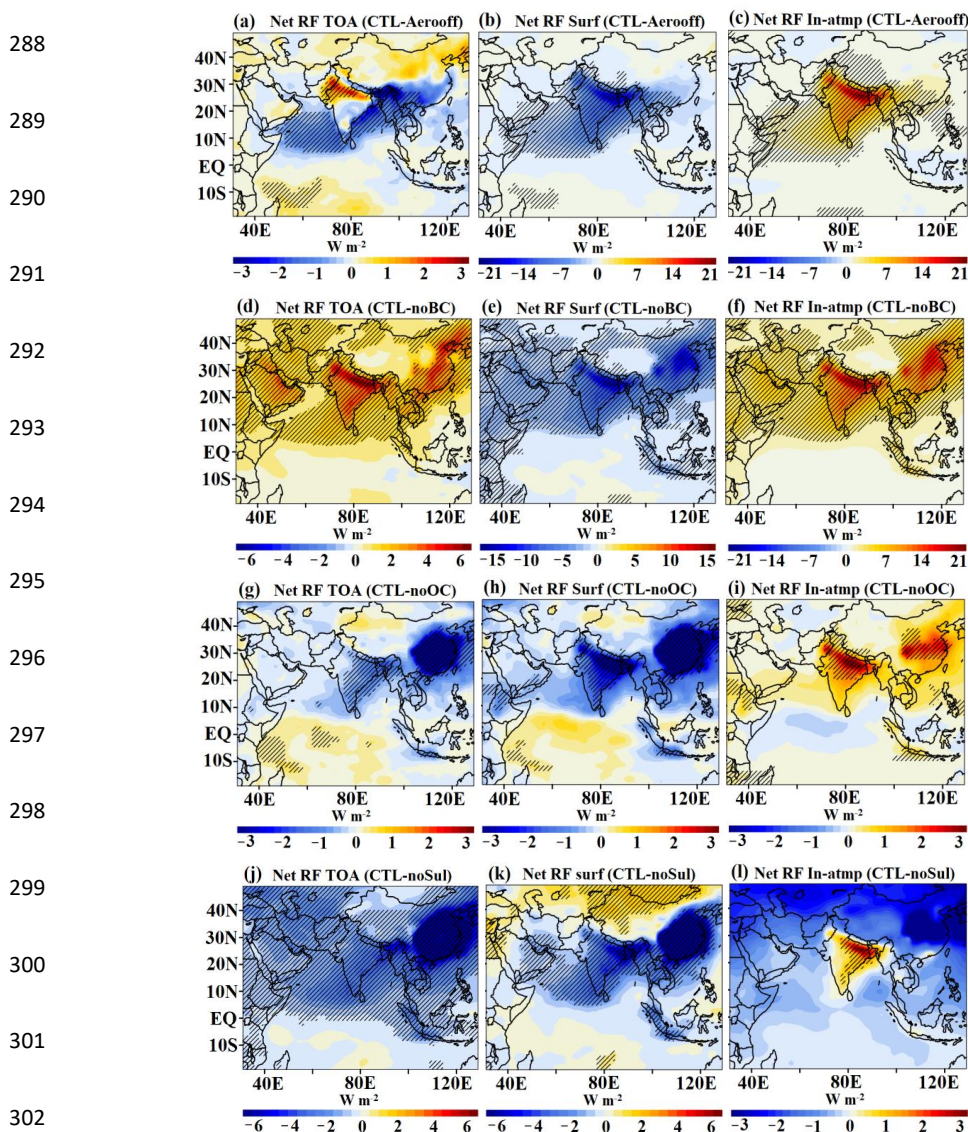
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303 Figure 4: Spatial distribution of net aerosol radiative forcing (CTL - Aerooff) (W m^{-2})
 304 averaged for spring during 2001 – 2016 (a) TOA, (b) same as (a) but for surface, (c) same
 305 as (a) but for in-atmosphere (TOA - surface), (d) spatial distribution of radiative forcing at
 306 the TOA (CTL - BCoff) averaged for spring during 2001 – 2016, (e) same as (d) but for
 307 surface, (f) same as (d) but for in-atmosphere (TOA - surface), (g) spatial distribution of
 308 radiative forcing at the TOA (CTL - OCoff) averaged for spring during 2001 – 2016, (h)
 309 same as (g) but for surface, (i) same as (h) but for in-atmosphere (TOA - surface), (j)
 310 spatial distribution of radiative forcing at the TOA (CTL - Suloff) averaged for spring
 311 during 2001 – 2016, (k) same as (j) but for surface, (l) same as (k) but for in-atmosphere
 312 (TOA - surface). The hatched lines in figure a-l indicate 99% confidence level for the
 313 mean differences.

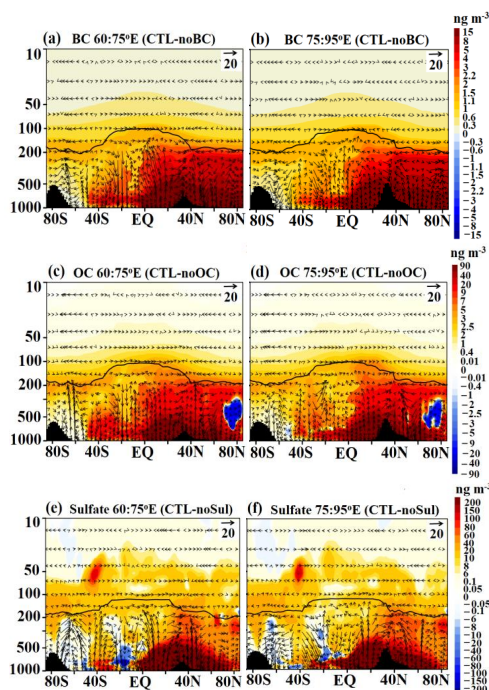


314 3.3. Transport of Asian anthropogenic aerosols into the UTLS

315 Further, we investigate the vertical distribution of aerosols that are transported to the north
316 Indian Ocean. Meridional sections over the Arabian Sea (60 – 75° E) and Bay-of-Bengal (75
317 – 95 °E) for BC, OC, and sulfate aerosol anomalies indicate that these aerosols are
318 transported from the boundary layer of both regions and north India into the UTLS (Figure 5
319 and Fig. S1). The spring convection occurring over the Arabian Sea and Bay-of-Bengal
320 which is shown by the combined distribution of CDNC and ICNC (Fig. S2a) plays an
321 important role in the vertical transport. The prevailing spring convection is further
322 invigorated over the Arabian Sea by the transported aerosols there which is not the case for
323 the Bay of Bengal region (Fig. S2b). The aerosol loading over the North-Indian region forms
324 clouds and elicit convection there (Fig. S2c-d). The distribution of wind resolved circulation
325 shows a strong ascent over the Arabian Sea, and the Bay-of-Bengal regions, while the steep
326 orography of the Himalayas over North India also plays an important role in the vertical
327 transport to the upper troposphere (Fig. 5 and Fig. S1). Figure 5 also shows that aerosols
328 induce a secondary circulation, ascending winds over 10 – 30° N and descent over 30 – 40°
329 S. BC, OC, and sulfate aerosols are transported to the UTLS, moving southward and
330 downward ~30 – 40° S (Fig. 5a-f, and Fig. S1) due to this secondary circulation. The aerosol
331 enhancement in the lower troposphere (1000 – 500 hPa) over 30 – 40° S is therefore due to
332 the combined impact of horizontal transport and downward transport from the UTLS due to
333 this secondary circulation. Further, in the UTLS these aerosols are transported globally.
334 There is enhancement in the Arctic (BC: 0.6 to 1.5 ng m⁻³, OC: 0.4 to 7 ng m⁻³, Sulfate: 0.1 to
335 20 ng m⁻³) and Antarctic (BC: 0.6 to 3 ng m⁻³, OC: 1 to 5 ng m⁻³, Sulfate: 6 to 40 ng m⁻³) in
336 the lower stratosphere (180 – 90 hPa) (see Figure 5). Our analysis shows that transport to the
337 Arctic and Antarctic occurs every year in the UTLS which causes heating in the lower
338 stratosphere (see Section 3.4).



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350 Figure 5: Meridional cross-section over Arabian Sea (averaged 60 – 75° E) and Bay-of-
351 Bengal (75 – 95° E) and for the spring season during 2001 – 2016 of anomalies (ng m^{-3}) of
352 (a–b) BC aerosols (CTL-BCoff), (c–d) OC aerosols (CTL-OCoff), (e–f) sulfate aerosols
353 (CTL-Suloff). Vectors in Figs. a–f indicate anomalies of winds (m s^{-1}) (the vertical velocity
354 field has been scaled by 300 and the units are m s^{-1}).

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356 3.4. Impacts on the heating rate and water vapor

357 Carbonaceous aerosols absorb solar radiation, leading to atmospheric heating, while
358 predominately scattering aerosols such as sulfate reflect and scatter back solar radiation,
359 therefore cooling the atmosphere below (Fadnavis et al., 2019). The vertical distribution of
360 heating rate anomalies induced by all the anthropogenic Asian aerosols (CTL - Aerooff) over
361 the North Indian Ocean (Arabian Sea and North Bay of Bengal, 50 – 100° E) indicates a
362 significant increase in heating rates in the region of elevated anthropogenic aerosols in the
363 troposphere (0.05 K d^{-1}) and stratosphere (0.002 K d^{-1}) (Fig. 6a–d, Fig. 5, and Fig. S1).

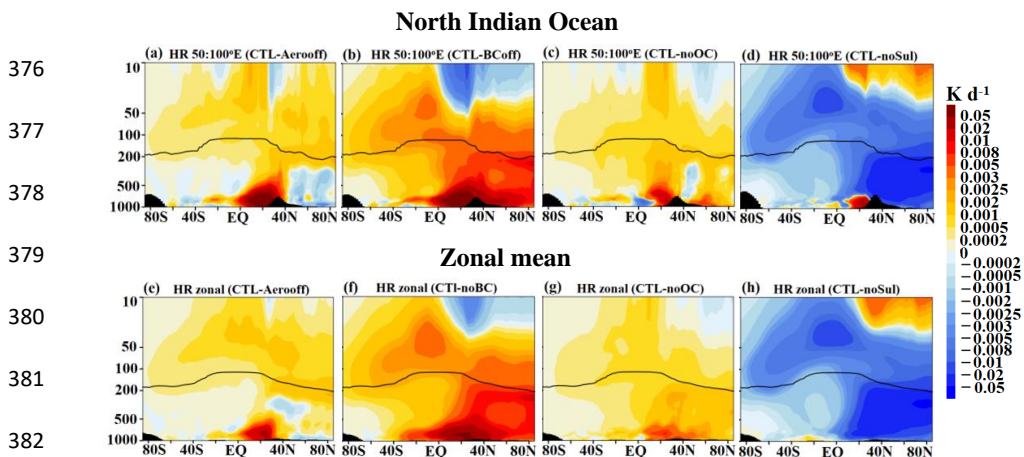
364 Heating rate anomalies estimated over the North Indian Ocean from BC (CTL - BCoff), OC



365 (CTL - OCoff), and Sulfate (CTL - Suloff) show that BC and OC aerosols produce heating in
366 the troposphere (10 – 40° N) (BC: 0.001 to 0.05 K d⁻¹, OC: 0.0002 to 0.02 K d⁻¹) and
367 stratosphere (100 – 50 hPa, 90°S – 90°N) (BC: 0.001 to 0.008 K d⁻¹, OC: 0.0002 to 0.002 K
368 d⁻¹), while sulfate aerosols produce atmospheric cooling in the troposphere -0.001 to -0.05
369 (500 – tropopause) and stratosphere -0.001 to -0.008 K d⁻¹ (tropopause – 50 hPa) (Fig. 6a-d).
370 There is anomalous heating in the tropical stratosphere (20° S – 20° N) (0.001 to 0.002 K d⁻¹)
371 seen in CTL-Aerooff simulations (Fig. 6a), mainly due to carbonaceous aerosols (Fig. 6b-c).

372 The zonal mean distribution of heating rates (Fig 6 e-h) shows that the South Asian
373 carbonaceous aerosols lead to 0.002 – 0.01 K d⁻¹ heating in the lower stratosphere globally
374 (100 – 50 hPa) (Fig. 6f-g), larger than the cooling induced by sulfate aerosols (Fig. 6h).

375



383 Figure 6: Meridional cross-section over the North Indian Ocean (averaged over the
384 Arabian Sea and Bay of Bengal region, 50 – 100° E) of anomalies of heating rates (K d⁻¹)
385 averaged for the spring season during 2001 – 2016 (a) from CTL - Aerooff simulation, (b)
386 same (a) but from CTL - BCoff simulation (c) same (a) but from CTL - OCoff simulation,
387 (d) same (a) but from CTL - Suloff simulation. (e) Zonal mean (0 – 360°) anomalies in
388 heating rate for CTL - Aerooff simulation, (f) same as (e) but from CTL - BCoff
389 simulation, (g) same as (e) but from CTL - OCoff simulation, (h) same as (e) but from
390 CTL - Suloff simulation.



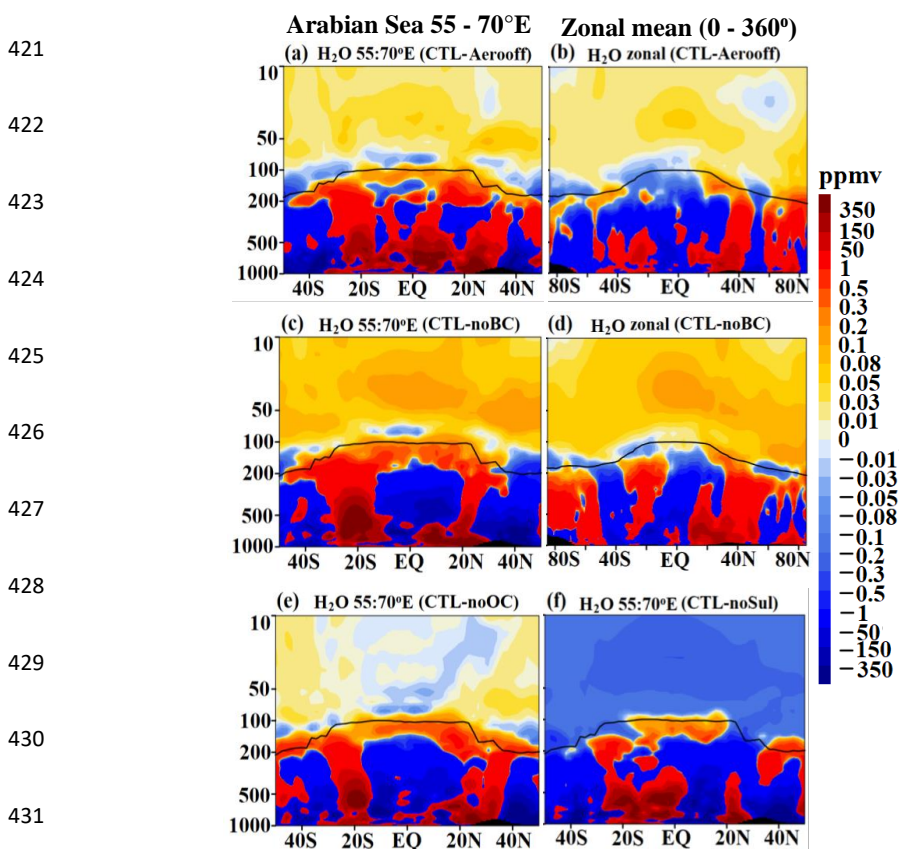
391 In general, these aerosols increase heating in the troposphere over the South Asian region
392 (Fig. 6a) and northern Arabian Sea and Bay of Bengal (10 – 30° N). This **enhanced heating**
393 **invigorates the convection process**, which results in an increase in cloud cover (Fig. S2c) and
394 deepening of the OLR (Fig. S2d). The invigorated convection provides positive feedback on
395 the vertical ascent into the free troposphere that **extends to above the tropopause** into the
396 lower stratosphere over the Arabian Sea and Bay-of-Bengal-North-India region (Fig. S3a-b)
397 ([Fadnavis et al., 2013](#); [Randel et al., 2010](#)).

398 The vertical distribution of water vapor over the Indian Ocean (CTL - Aerooff) **shows that**
399 **water vapor (0 – 0.3 ppmv) is transported to the UTLS from the Arabian Sea** (55 – 70° E, 0 –
400 30° N) (Fig. 7a) along the path of elevated aerosols (Fig. 5). Interestingly, there is an
401 enhancement in water vapor over the southern Indian Ocean (20 – 30° S, 55 – 70° E) along
402 the path of the descending branch of aerosols (BC, OC, and sulfate). This is due to the
403 **significant heating caused by carbonaceous aerosols** (Fig. 6b-c) which leads to enhancement
404 of tropospheric water vapor (Fig. 7a) over the Arabian Sea. The zonal mean (averaged for 0 –
405 360°) anomalies of water vapor (Fig. 7b) show an enhancement by 0.03-0.08 ppmv (0 – 4 %)
406 in the global stratosphere (Fig. 7b). There is an enhancement in **the lower stratosphere** in the
407 **Antarctic (60 – 90° S) by 0.01 to 0.03 ppmv and in the Arctic (80 – 90° N) by 0.01 - 0.1 ppmv**
408 caused by anthropogenic aerosols (CTL-Aerooff).

409 The impact of BC (CTL - BCoff), OC (CTL - OCoff), and Sulfate (CTL - Suloff) on water
410 vapor distribution (Figs. 7 c-f) shows that BC aerosols play a major role in water vapor
411 enhancement in the stratosphere (Fig. 7 c), (100 – 10 hPa). Water vapor enhancement by BC
412 aerosols over the Arabian Sea region is **~0.03 – 0.3 ppmv (Fig. 7c) and 0.01 – 0.2 ppmv** in
413 the global stratosphere (Fig. 7d). There is significant enhancement of water vapor due to BC
414 aerosols in the Arctic (0.01 – 0.2 ppmv) and Antarctic (0.01 – 0.1 ppmv) (Fig. 7d). The water
415 vapor enhancement by OC aerosols is negligible in the tropical stratosphere and there is no



416 contribution of sulfate aerosols (Fig. 7 e-f). The sulfate aerosols cause negligible heating by
417 **abortion of infra-red radiations** over the Arabian Sea that leads to water vapor enhancement
418 from the boundary layer to the mid-troposphere (500 hPa), near the tropopause, and in the
419 path of descending branch of secondary circulation over the South Indian Ocean (~20° S)
420 (Fig. 7f).

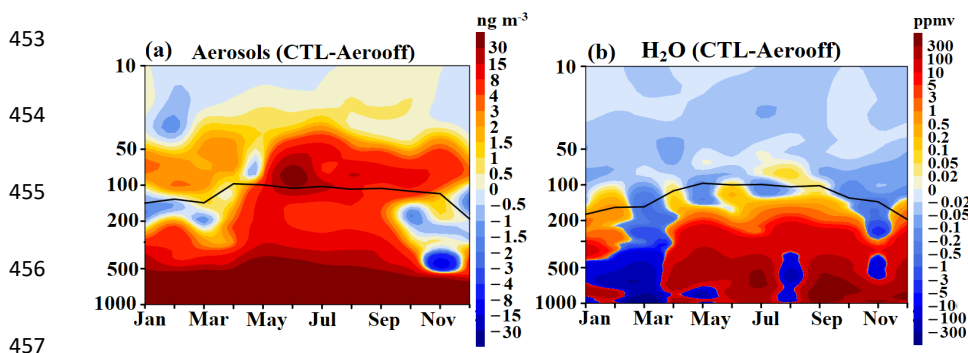


432 Figure 7: (a) Meridional cross-section over Arabian Sea region (averaged 55 – 70° E) of
433 **anomalies of water vapour (ppmv)** (CTL - Aerooff) the for spring season during 2001 – 2016,
434 (b) same as (a) but zonal mean (average for longitudes 0 – 360°), (c) same as (a) but from
435 CTL - BCoff simulations, (d) same as (c) but zonal mean (average for longitudes 0 –360°),
436 **(e) same as (a) but from CTL - OCoff simulations, (f) same as (a) but from CTL - Suloff**
437 **simulations.**

438 Although the focus of the manuscript is on transport of aerosols during the spring season, ~~it~~
439 ~~should be noted that the~~ anthropogenic South Asian aerosols are also transported to the UTLS



440 during the monsoon season (Fadnavis et al., 2013, 2017, 2019). Annual distribution
441 anomalies of aerosols (average of BC, OC and sulfate) show transport of aerosols into the
442 UTLS during spring and monsoon season (April to September) from the Arabian Sea, Bay-
443 of-Bengal-North-India region (Fig. 8a). These aerosols enhance tropospheric heating thereby
444 transporting elevated water vapour into the lower stratosphere (Fig. 8b). Injection of aerosols
445 into the lower stratosphere occurs every year however there is interannual variability. We
446 show the vertical distribution of aerosols for two normal years when there was no large scale
447 ocean-atmosphere coupling phenomenon like El Niño southern oscillation or Indian Ocean
448 Dipole (2008 and 2016) in Fig. S4a-b. It also shows transport of aerosols into the lower
449 stratosphere during spring and monsoon seasons (March-September). The aerosol induced
450 enhanced water vapour also shows enhancement in the lower stratosphere during the same
451 time (Figs. S4c-d). In the lower stratosphere, these aerosols persist for a few months (Fig. 8a)
452 thus their effect will be seen for an extended time.

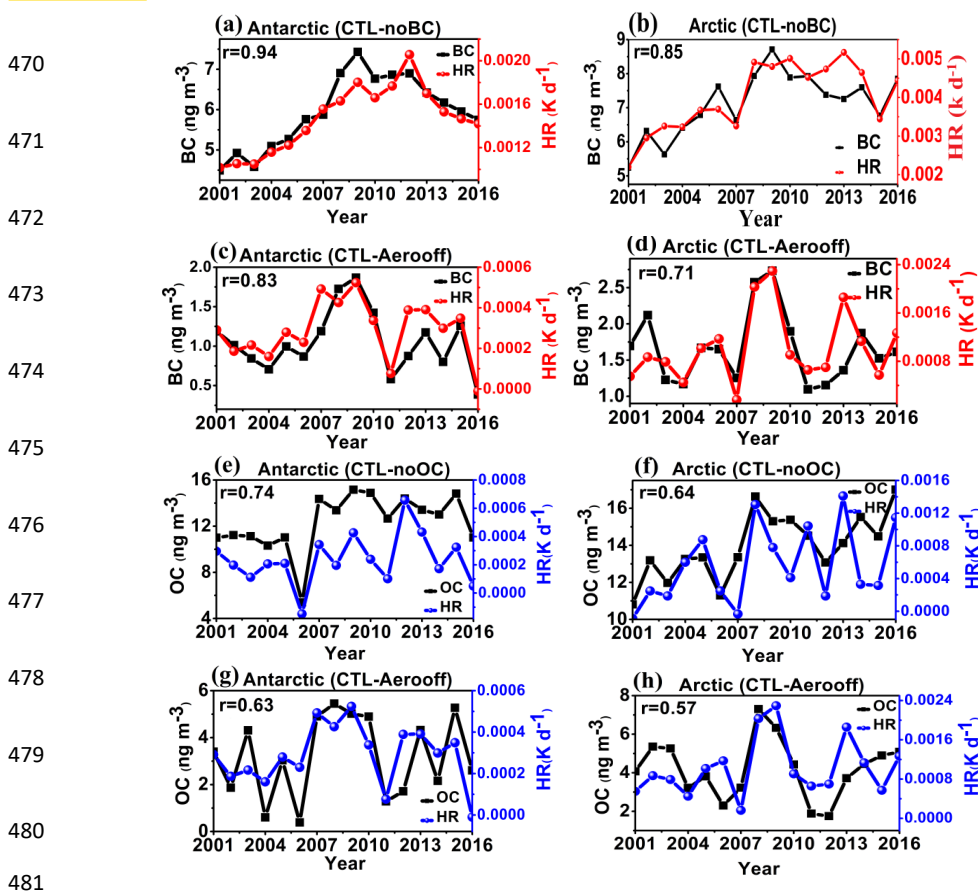


458 Figure 8: (a) Annual distribution of anomalies of aerosols (CTL - Aerooff) (averaged of BC,
459 OC and sulfate aerosols) (ng m^{-3}) averaged for Arabian Sea, Bay-of-Bengal-North-India
460 region ($55 - 110^\circ \text{E}$, $10 - 30^\circ \text{N}$), (b) same as (a) but for water vapour (ppmv).

461 Further, we analyze the correlation between heating rates and carbonaceous aerosol amounts
462 in the UTLS (180 – 70 hPa) in the Arctic and Antarctic during 2001 – 2016 (spring mean)
463 (Fig. 9) from Aerooff, BCoff, and OCOff in comparison with CTL simulations. The



464 carbonaceous aerosols show a positive correlation (correlation coefficient r : 0.57 to 0.94)
 465 with the UTLS heating rates indicating that transported carbonaceous aerosols **enhance UTLS**
 466 **heating** in the Arctic and Antarctic. It should be noted that transport of aerosols to the Arctic
 467 and Antarctic also occurs during the monsoon season (Fadnavis et al., 2017a, 2017b, 2019)
 468 which **may affect the dynamics and aerosol amounts in the spring of the next year in the**
 469 **UTLS.**



482 Figure 9: (a) Time series of BC aerosols and heating rates averaged for spring and UTLS
 483 (180 – 70 hPa) at the Antarctic (60 – 90 °S, 0 – 360 °)(from CTL - BCoff, (b) same as (a) but
 484 in the Arctic (65 – 85° N, 0 – 360°), (c) same as (a) but from CTL - Aerooff, (d) same as (b)
 485 but from CTL - Aerooff, (e) same as (a) but for OC (CTL - OCoff), (f) same as (b) but for
 486 OC (CTL - OCoff), (g) same as (c) but for OC, (h) same as (d) but for OC. The correlation
 487 coefficient (r) between anomalies of BC/OC aerosols and heating rates is indicated in panels
 488 a-h.



489 Importantly, South Asian aerosols enhance water vapor in the lower stratosphere, globally.
490 Water vapor being a greenhouse gas further **enhances the heating of the troposphere** leading
491 to a positive feedback. The increase in water vapor in the stratosphere also warms the Earth's
492 surface (Shindell, 2001; Solomon et al., 2010). Solomon et al. (2010) estimated that an
493 increase in the stratospheric water vapor by 1 ppmv accounts for 0.24 W m^{-2} radiative
494 **forcing**. The SABER and MLS observations showed an increase in stratospheric water vapor
495 by 0.45 ppmv globally during 2003 – 2017 (Yue et al., 2019). Thus the radiative forcing due
496 to water vapor increase (0.02 – 0.14 ppmv) in response to South Asian anthropogenic
497 aerosols is **not negligible** for surface warming globally. Further, increasing stratospheric
498 water vapour could also lead to ozone depletion (e.g., Shindell, 2001, Robrecht et al., 2019).

499 **4. Conclusions**

500 A series of ECHAM6-HAMMOZ chemistry-climate simulations for South Asian
501 anthropogenic aerosols were used to understand the transport pathways of **Asian** aerosols and
502 their associated impacts. The analysis is performed for the spring season, when emissions of
503 anthropogenic aerosols (BC, OC, and sulfate) over **south** Asia peak. The model simulations
504 show that large amounts of South Asian aerosols are transported during spring to the Arabian
505 Sea (increases in AOD by: 0.02 – 0.12 from CTL - **AeroOff**) and Bay of Bengal (increases in
506 AOD by: 0.16 to 0.8 from CTL - Aerooff). The anthropogenic aerosols are further lifted **up**
507 into the UTLS from the Arabian Sea and Bay-of-Bengal-North-India. In the UTLS, they are
508 also transported **to** the southern hemisphere (30 – 40 °S) ~~and downward in the troposphere~~ by
509 the secondary circulation induced by the aerosol changes. In the UTLS, these aerosols (BC,
510 OC, and Sulfate) are transported globally.

511 The anthropogenic aerosol produces significant radiative impacts over the Indo-Gangetic
512 Plain (RF anomalies estimated from CTL-Aerooff simulations, TOA: $+1.27 \pm 0.16 \text{ W m}^{-2}$,



513 Surface: $-11.16 \pm 0.50 \text{ W m}^{-2}$, In-atmosphere: $+12.44 \pm 0.42 \text{ W m}^{-2}$) and the Arabian Sea (RF at
514 the TOA: $-0.72 \pm 0.14 \text{ W m}^{-2}$, surface: $-3.00 \pm 0.28 \text{ W m}^{-2}$, In-atmosphere: $+2.27 \pm 0.19 \text{ W m}^{-2}$).
515 Interestingly, RF at the TOA over the Indo-Gangetic Plain is positive $+4.33 \pm 0.17 \text{ W m}^{-2}$ due
516 to the emission of BC aerosols. The anthropogenic aerosols enhance heating in the
517 troposphere (estimated from CTL-Aerooff) by 0.002 to 0.05 K d^{-1} and UTLS by 0.001 to
518 0.002 K d^{-1} leading to more cloud formation (cloud cover anomalies enhanced by 2 to 12 %)
519 and intensification of convection (OLR anomalies -0.5 to -10 W m^{-2}). This invigorated
520 convection provides a positive feedback on the vertical updraft of aerosols into the free
521 troposphere and above the tropopause into the lower stratosphere (Fadnavis et al., 2013;
522 Randel et al., 2010). The tropospheric heating/cooling caused by the anthropogenic aerosols
523 over South Asia and the North Indian Ocean during spring has implications on the Indian
524 summer monsoon rainfall a few months later (Fadnavis et al., 2017a; Fadnavis and
525 Chattopadhyay, 2017).

526

527 The heating of the troposphere by the carbonaceous aerosol increases evaporation and
528 thereby tropospheric water vapor amounts over the North Indian Ocean and adjoining
529 regions. The elevated water vapor is transported from highly convective regions (i.e. Arabian
530 Sea) to the UTLS, from where it is then transported globally. BC aerosols play a major role in
531 water vapor enhancement in the lower stratosphere, globally (increased water vapor) by 0.01
532 to 0.1 ppmv . Water vapor being a greenhouse gas further enhances the heating of the
533 troposphere leading to a positive feedback. The increase in water vapor in the stratosphere
534 also warms the Earth's surface (Shindell, 2001; Solomon et al., 2010).

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537 **Data availability:** The data used in this study are generated from ECHAM6-HAMMOZ
538 model simulations at the High-performance computing system in the Indian Institute of
539 Tropical Meteorology, Pune, India. The AOD data from MODIS Terra used here can be
540 downloaded from <https://ladsweb.modaps.eosdis.nasa.gov/archive/allData/61/MODATML2/>,
541 and MISR from <https://misr.jpl.nasa.gov/getData/accessData/>.

542

543

544 **Author contributions:** S. F. initiated the idea. A. J., S. S., A. A., performed model analysis.
545 R. M., and A. R. contributed to analysis and study design. All authors contributed to the
546 writing and discussions of the manuscript.

547

548 **Competing Interests:** Some authors are members of the editorial board of Atmospheric
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550 the authors have also no other competing interests to declare.

551



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