1	AIR POLLUTION OVER THE GANGES BASIN AND						
2	NORTH-WEST BAY OF BENGAL IN THE EARLY POST-						
3	MONSOON SEASON BASED ON NASA MERRAERO						
4	DATA						
5							
6	Pavel Kishcha,						
7	Department of Geophysical, Atmospheric and Planetary Sciences, Tel-Aviv University,						
8	69978 Tel-Aviv, Israel						
9							
10	Arlindo M. da Silva						
11	Global Modeling and Assimilation Office, NASA/GSFC, Greenbelt, Maryland USA.						
12							
13	Boris Starobinets						
14	Department of Geophysical, Atmospheric and Planetary Sciences, Tel-Aviv University,						
15	69978 Tel-Aviv, Israel						
16							
17	Pinhas Alpert						
18	Department of Geophysical, Atmospheric and Planetary Sciences, Tel-Aviv University,						
19	69978 Tel-Aviv, Israel						
20							
21	Journal of Geophysical Research – Atmospheres,						
22	Accepted for publication on January 13, 2014						
23							
24							

#### 25 Abstract

26 The MERRA Aerosol Reanalysis (MERRAero) has been recently developed at NASA's 27 Global Modeling Assimilation Office (GMAO). This reanalysis is based on a version of 28 the GEOS-5 model radiatively coupled with GOCART aerosols, and it includes 29 assimilation of bias-corrected Aerosol Optical Thickness (AOT) from the MODIS 30 sensor on both Terra and Aqua satellites. In October over the period 2002-2009, 31 MERRAero showed that AOT was lower over the east of the Ganges basin than over 32 the north-west of the Ganges basin: this was despite the fact that the east of the Ganges 33 basin should have produced higher anthropogenic aerosol emissions because of higher 34 population density, increased industrial output and transportation. This is evidence that 35 higher aerosol emissions do not always correspond to higher AOT over the areas where 36 the effects of meteorological factors on AOT dominate those of aerosol emissions. 37 MODIS AOT assimilation was essential for correcting modeled AOT mainly over the 38 north-west of the Ganges basin, where AOT increments were maximal. Over the east of 39 the Ganges basin and north-west BoB, AOT increments were low and MODIS AOT 40 assimilation did not contribute significantly to modeled AOT. Our analysis showed that 41 increasing AOT trends over north-west BoB (exceeding those over the east of the 42 Ganges basin) were reproduced by GEOS-5, not because of MODIS AOT assimilation, 43 but mainly because of the model capability of reproducing meteorological factors 44 contributing to AOT trends. Moreover, vertically integrated aerosol mass flux was 45 sensitive to wind convergence causing aerosol accumulation over north-west BoB.

46

47

48

#### 50 1. Introduction

51 The Indian subcontinent (and the Ganges basin in particular) is characterized by a 52 significant population growth accompanied by developing industry, agriculture, and 53 increasing transportation. This has resulted in declining air quality [Di Girolamo et al. 54 2004, Ramanathan and Ramana, 2005, Tripathi et al., 2005, Prasad and Singh, 2007, 55 Kaskaoutis et al., 2011a, Dey and Di Girolamo, 2011, Krishna Moorthy et al., 2013]. 56 With respect to air pollution, one could suggest some relationship between population 57 figures and anthropogenic aerosol emissions. Indeed, Kishcha et al. [2011] showed that, 58 over extensive areas with differing population densities in the Indian subcontinent, the 59 higher the averaged population density – the larger the averaged AOT. In addition, the 60 larger the population growth - the stronger the increasing AOT trends.

61

62 In accordance with Di Girolamo et al. [2004], Prasad and Singh [2007], Kumar et al. 63 [2010], prevailing winds blowing along the Ganges basin in the post-monsoon and 64 winter months transport anthropogenic aerosol particles into the Bay of Bengal. A 65 number of sea expeditions to BoB were conducted to investigate the resulting increased 66 levels of air pollution over BoB [Ramachandran and Jayaraman, 2003; Vinoj et al., 2004; Ganguly et al., 2005, Moorthy et al., 2008, Kumar et al., 2010, Kaskaoutis et al., 67 68 2011b]. Moreover, long-term AOT trends over South Asia, including BoB, were 69 examined, using different satellite AOT data sets, by Mishchenko and Geogdzhayev 70 [2007], Zhao et al. [2008], Zhang and Reid [2010], Kaskaoutis et al. [2011a], Dey and 71 Di Girolamo [2011], and Hsu et al. [2012]. Based on AVHRR satellite data, 72 Mishchenko and Geogdzhayev [2007] compared over-water AOT averaged over two 73 separate periods, 1988–1991 and 2002–2005, and found significant changes. Zhao et al. 74 [2008] studied AOT trends over the whole area of BoB for spring, summer, autumn, and 75 winter during the 25-year period 1981 - 2005, using AVHRR data. Using MODIS-76 Terra Level 2 AOT data, Zhang and Reid [2010] analyzed AOT trends over the whole 77 area of BoB for all months during the 10-year period 2000 - 2009. The spatial 78 distribution of decadal (2000 - 2009) MODIS Level-3 AOT trends over South Asia, 79 including BoB, in different months was obtained by Kaskaoutis et al. [2011a]. Using MISR aerosol data, decadal (2000 - 2009) AOT trends over the Indian subcontinent and 80 81 surrounding sea areas were also estimated by Dey and Di Girolamo [2011]. Hsu et al.

[2012] created maps of SeaWiFS AOT trends over the period 1998 – 2010 for each of
the four seasons.

84

85 The early post-monsoon season over the study region is characterized by aerosol 86 transport from the Ganges basin to north-west BoB by prevailing winds; and still 87 significant rainfall of over 150 mm/month over the east of the Ganges basin and north-88 west BoB. It would be reasonable to consider that AOT trends over sea areas in BoB 89 were created by changes in aerosol sources on the land in the Indian subcontinent. In 90 our previous study [Kishcha et al., 2012], we found that it was not always the case. 91 Specifically, we found that, in October, MODIS showed strong increasing aerosol 92 optical thickness (AOT) trends over north-west Bay of Bengal (BoB) in the absence of 93 AOT trends over the east of the Indian subcontinent. This was unexpected, because 94 sources of anthropogenic pollution were located over the Indian subcontinent, mainly in 95 the Ganges basin, and aerosol transport from the Indian subcontinent to north-west BoB 96 was carried out by prevailing winds.

97

98 It was our purpose to determine whether existing state-of-the-art aerosol data-99 assimilated systems were capable of reproducing the aforementioned AOT trends over 100 north-west BoB in the early post-monsoon season, in the presence of significant rainfall. 101 For the model, it would be a challenge just to obtain correct space-time distribution of 102 rainfall, which is of importance for estimating aerosol wet removal. The NASA 103 Goddard Earth Observing System (GEOS-5) was used to extend the NASA Modern 104 Era-Retrospective Analysis for Research and Applications (MERRA) reanalysis by 105 adding five atmospheric aerosol components (sulfates, organic carbon, black carbon, 106 desert dust, and sea-salt). In the current study, the obtained eight-year (2002 - 2009)107 assimilated aerosol dataset (so-called MERRAero) was applied to examine aerosol 108 trends over the Ganges basin and north-west Bay of Bengal (BoB) in the post-monsoon 109 season. In addition, using air pollution modeling allowed us to look at the situation over 110 the study region from the point of view of air quality. This was carried out by estimating 111 the contribution of various aerosol species to AOT and its trends.

112 Note that AOT assimilation was effective only for two short periods of MODIS's 113 appearance over the study area. All other times (18 hours per day) the GEOS-5 model 114 worked independently of MODIS.

115

## 116 2. GEOS-5 and the MERRA Aerosol Reanalysis (MERRAero)

117

## 118 2.1 GEOS-5 Earth Modeling System

119 GEOS-5 is the latest version of the NASA Global Modeling and Assimilation Office 120 (GMAO) Earth system model. GEOS-5 contains components for atmospheric 121 circulation and composition (including atmospheric data assimilation), ocean circulation 122 and biogeochemistry, and land surface processes. Components and individual 123 parameterizations within components are coupled under the Earth System Modeling 124 Framework (ESMF) [Hill et al., 2004]. In addition to traditional meteorological 125 parameters (winds, temperatures, etc. [Rienecker et al., 2008]), GEOS-5 includes 126 modules representing the atmospheric composition, most notably aerosols [Colarco et 127 al., 2010], and tropospheric/stratospheric chemical constituents [Pawson et al., 2008], 128 and the impact of these constituents on the radiative processes of the atmosphere.

129

## 130 2.2 Aerosols in GEOS-5

131 GEOS-5 includes modules representing atmospheric composition, including aerosols 132 [Colarco et al., 2010] and tropospheric and stratospheric chemical constituents [Pawson 133 et al., 2008]. The current generation aerosol module is based on a version of the 134 Goddard Chemistry, Aerosol, Radiation, and Transport (GOCART) model [Chin et al., 135 2002]. GOCART treats the sources, sinks, and chemistry of dust, sulfate, sea salt, and 136 black and organic carbon aerosols. Aerosol species are assumed to be external mixtures. 137 Both dust and sea salt have wind-speed dependent emission functions [Colarco et al., 138 2010], while sulfate and carbonaceous species have emissions principally from fossil 139 fuel combustion, biomass burning, and bio-fuel consumption, with additional biogenic 140 sources of organic carbon. Sulfate has additional chemical production from oxidation of 141 SO2 and dimethylsulfide (DMS), as well as a database of volcanic SO2 emissions and 142 injection heights. Aerosol emissions for sulfate and carbonaceous species are based on

the AeroCom version 2 hindcast inventories [Dr. Thomas Diehl, personal communication, and http://aerocom.met.no/emissions.html). Daily biomass burning emissions are from the Quick Fire Emission Dataset (QFED) and are derived from MODIS fire radiative power retrievals [Darmenov and da Silva, 2013]. Total mass of sulfate and carbonaceous aerosols are tracked, while for dust and sea salt the particle size distribution is explicitly resolved across five non-interacting size bins for each.

149

150 For all aerosol species, optical properties are primarily from the commonly used Optical 151 Properties of Aerosols and Clouds (OPAC) data set [Hess et al., 1998]. OPAC provides 152 the spectrally varying refractive index and a humidification factor for each aerosol 153 species which, together with assumptions about the particle size distribution of each species, are used to construct spectrally varying lookup tables of aerosol optical 154 155 properties such as the mass extinction efficiency, single scattering albedo, and 156 asymmetry parameter, inputs required by our radiative transfer codes (details are in 157 Colarco et al. [2010], and references therein).

158

## 159 2.3 GEOS-5 Data Assimilation

GEOS-5 has a mature atmospheric data assimilation system that builds upon the Gridpoint Statistical Interpolation (GSI) algorithm, jointly developed with NCEP [Wu et al. 2002, Derber et al. 2003, Rienecker et al. 2008]. The GSI solver was originally developed at NCEP as an unified 3D-Var analysis system for supporting global and regional models. GSI includes all the in-situ and remotely sensed data used for operational weather prediction at NCEP.

GEOS-5 also includes assimilation of AOT observations from the MODIS sensor on both Terra and Aqua satellites. Based on the work of Zhang and Reid [2006] and Lary [2009], a back-propagation neural network has been developed to correct observational biases related to cloud contamination, surface parameterization, aerosol microphysics, etc. On-line quality control is performed with the adaptive buddy check of Dee et al. [2001], with observation and background errors estimated using the maximum likelihood approach of Dee and da Silva [1999].

#### 174 2.4 MERRA Aerosol (MERRAero) Reanalysis

175 MERRA is a NASA reanalysis for the satellite era using a major new version of the 176 Goddard Earth Observing System Data Assimilation System Version 5 (GEOS-5) 177 [Rienecker et al., 2011]. The Project focuses on historical analyses of the hydrological 178 cycle from the NASA EOS suite of observations in a climate context, on a broad range 179 of weather and climate time scales and places. The MERRA time period covers the 180 modern era of remotely sensed data, from 1979 through the present, and the special 181 focus of the atmospheric assimilation is the hydrological cycle. Like other similar 182 reanalysis, MERRA provides meteorological parameters (winds, temperature, 183 humidity), along with a number of other diagnostics such as surface and top of the 184 atmosphere fluxes, diabatic terms and the observational corrections imposed by the data 185 assimilation procedure.

186 As a step toward an Integrated Earth System Analysis (IESA), the GMAO is producing 187 several parallel re-analyses of other components of the earth system such as ocean, land 188 and atmospheric composition. Of particular relevance for this paper the MERA Aerosol 189 Reanalysis (MERRAero), where MODIS AOT observations are assimilated providing a 190 companion aerosol gridded datasets that can be used to study the impact of aerosols on 191 the atmospheric circulation and on air quality in general. Table 1 summarizes the main 192 attributes of MERRAero. Notice that MERRAero only covers the later years of 193 MERRA, capitalizing on the improved aerosol measurements from NASA's EOS 194 platforms.

195

## **3. Method**

197 Following our previous study [Kishcha et al., 2012], we analyzed long-term variations of AOT over seven zones, each 3° x 3°, located in the Ganges basin and north-west BoB 198 199 (Fig. 1). As mentioned, in the post-monsoon period, prevailing winds blow along the 200 Ganges basin. The specified zones in the Ganges basin provide us with an opportunity 201 for analyzing air pollution trends produced by local sources and aerosol transport. Fig. 202 la shows the spatial distribution of eight-year mean MERRAero AOT over the region 203 under consideration in October, together with the location of zones 3° x 3° in the Indian 204 subcontinent (zone 1 to zone 5) and in the Bay of Bengal (zones 6 and 7). MERRAero 205 monthly AOT data are available from the year 2002. To analyze AOT and its trends

over the Indian subcontinent and north-west BoB, we used monthly MERRAero AOT
data with horizontal resolution of approximately 50 km, during the eight-year period
2002 – 2009. To estimate the NASA GEOS-5 model performance, 3-hour MERRAero
AOT data together with AOT increments were used.

210

211 A linear fit was used to determine the resulting trend of aerosol optical thickness during 212 the study period (2002 - 2009) over each of the aforementioned zones. The obtained 213 AOT trend values correspond to the slope of the linear fit. To ensure that the linear fit 214 produced normally distributed residuals, they were required to pass the Shapiro–Wilk 215 normality test [Shapiro and Wilk, 1965, Razali and Wah, 2011]. If the residuals were 216 normally distributed, they could be used in a t-test, in order to estimate the statistical 217 significance of a linear fit. The statistical significance of the AOT trend was checked by 218 applying the significance level (p) value, i.e. p < 0.05 for statistically significant AOT 219 trends at the 95 % confidence level.

220

#### **4. Results**

## 222 4.1. Total MERRAero AOT and its trends in October

In accordance with spatial distribution of eight-year mean AOT in the early postmonsoon season (October), MERRAero showed high AOT values over the Ganges basin with a maximum over the north-west part of the Ganges basin (Fig. 1a). Therefore, MERRAero data were able to reproduce the main structure of aerosol distribution over the Ganges basin. The Ganges basin is the most polluted part of the Indian subcontinent, where highly-populated areas and main industrial centers are located.

230

We analyzed zone-to-zone variations of MERRAero AOT averaged over the specified zones. In the early post-monsoon season (October), MERRAero showed mainly decreasing AOT variations from zone 3 to zone 5 (Fig. 2a and Table 2). Note that this decrease in AOT from north-west to east of the Ganges basin does not correspond to the distribution of population density: population density is higher in the east of the Ganges basin (zones 4 and 5) than in the north-west of the Ganges basin (zone 1) (Fig. 3). At first glance, this is contradictory to our previous findings on the relationship between AOT and population density in the Indian subcontinent [Kishcha et al., 2011]. It should be mentioned, however, that, in our previous study, we used averaging over significant areas of the Indian subcontinent with differing population densities.

241

242 The most probable reason for the decrease in AOT over the east of the Ganges basin, 243 where population density is the highest in the entire Ganges basin, is wet removal 244 processes after significant rainfall in October. Monthly accumulated Tropical Rainfall 245 Measuring Mission (TRMM) rainfall data from the 3B42V6 archive, on a  $0.25^{\circ} \times 0.25^{\circ}$ 246 latitude-longitude grid [Huffman et al., 2007], were used to estimate zone-to-zone 247 variations of eight-year (2002 – 2009) mean TRMM rainfall over the specified zones in 248 October (Fig. 4a). High rainfall values of over 150 mm can be seen in October over the 249 east of the Ganges basin (zone 5) and north-west BoB (zone 6). Moreover, rainfall data 250 showed that, over the east of the Ganges basin, the accumulated rainfall in October in 251 the first four-year period 2002 - 2005 was essentially higher than in the second four-252 year period 2006 – 2009 (Fig. 4a). As a result, higher values of MERRAero AOT over 253 the east of the Ganges basin (zones 4 and 5) were observed in the second four-year 254 period 2006 – 2009 than in the first four-year period 2002 – 2005 (Fig. 2b). Moreover, 255 the aforementioned decrease in the eight year (2002 - 2009) mean AOT over the east of 256 the Ganges basin and north-west BoB in October corresponds to the increase in model-257 simulated aerosol wet deposition over the specified zones towards north-west BoB, in 258 accordance with the spatial distribution shown in Fig. 4b.

259

260 Therefore, we have arrived at the interesting point, which is as follows. The east of the 261 Ganges basin should have produced higher anthropogenic aerosol emissions than the 262 north-west of the Ganges basin because of higher population density, increased 263 industrial output and transportation. Nevertheless, MERRAero showed that AOT over 264 the east of the Ganges basin was lower than that over the north-west of the Ganges 265 basin. This is evidence that higher aerosol emissions do not always correspond to higher 266 AOT. This can take place over the areas where the effects of meteorological factors on 267 AOT (in this case, precipitation) dominate those of aerosol emissions.

268

Spatial distributions of MERRAero AOT trends during the eight-year (2002 – 2009)
study period showed strong increasing AOT trends over north-west BoB exceeding
those over the Ganges basin (Fig. 1b). This indicates that MERRAero is capable of

reproducing the main features of the phenomenon of strong increasing AOT trends over
north-west BoB in the early post-monsoon season, in line with our previous study
[Kishcha et al., 2012].

275

# 276 **4.2. Effects of rainfall on MERRAero AOT**

277 As mentioned, in the early post-monsoon season (October), intense rainfall can be 278 frequently observed over the east of the Ganges basin. These severe precipitation events 279 could strongly affect AOT over the east of the Ganges basin due to aerosol wet removal processes. To understand the rain effects on AOT over the east of the Ganges basin 280 281 (zone 5), we compared year-to-year variations of assimilated MERRAero AOT and 282 TRMM accumulated rainfall, over zone 5 in each October during the study period (not 283 shown). Rainfall data showed that the accumulated rainfall in October in the first four-284 year period 2002-2005 was higher than in the second four-year period 2006-2009. A 285 strong inverse relationship (with a high negative correlation of over -0.8) between 286 changes in assimilated MERRAero AOT and rainfall is clearly seen: each increase in 287 rainfall was accompanied by a decrease in assimilated AOT. The aforementioned 288 decrease in rainfall over zone 5 in October during the study period can explain some 289 increasing trend in MERRAero AOT observed over that area in October (Table 2). 290 There was some dissimilarity in the rainfall amount between the east of the Ganges 291 basin (zone 5) and north-west BoB (zone 6): north-west BoB does not show as clear 292 decreasing trends in rainfall amount as the east of the Ganges basin does.

293

# 4.3. AOT of different aerosol species and their trends in the early post-monsoonseason

296 As known, satellite remote sensing data can not distinguish between various aerosol 297 species. MERRAero provides us with an opportunity to look at the situation over the 298 study region from the point of view of air quality. This was carried out by estimating the 299 contribution of various aerosol species to AOT and its trends. Based on MERRAero 300 model data, Fig. 5a represents zone-to-zone variations of eight-year (2002 – 2009) mean 301 AOT of several aerosol components (desert dust; organic and black carbon; and 302 sulfates) averaged over specified zones in October, and their trends. One can see that 303 over the west of the Ganges basin (zones 1 - 3), where precipitation was minimal (Fig. 304 4a), each aerosol component changes from zone to zone in a different way, in 305 accordance with its own emissions, transport, and gravitational settling (Fig. 5a). By 306 contrast, over the east of the Ganges basin (zones 4 - 5), a general decrease in AOT can 307 be observed due to increasing aerosol wet removal towards north-west BoB (Fig. 4b). 308 Over zone 1, there is a considerable amount of carbon aerosols (as a result of crop waste 309 burning [Sharma et al., 2010]), dust particles, and sulfate aerosols (Fig. 5a). This 310 explains the AOT maximum over the north-west of the Ganges basin in October. Over 311 sea areas (zones 6 - 7), aerosols are dominated by anthropogenic air pollution, such as 312 sulfates and carbon aerosols (Fig. 5a). This predominance can affect cloud formation, 313 atmospheric dynamics, and even marine life in this region.

314

315 By contrast to sulfates and carbonates, dust aerosol particles have no sources along the 316 Ganges basin. Therefore, dust distribution along the Ganges basin is determined by 317 aerosol transport (by the action of prevailing winds blowing along the Ganges basin) 318 and by deposition processes. One can see that the eight-year mean dust AOT values 319 noticeably decreased along the Ganges basin and over north-west BoB. This resulted in 320 the decrease in dust contribution to the total AOT from approximately 30% over zone 1 321 to 8% over zones from 5 to 7 (Table 3). Furthermore, dust AOT trends did not change in 322 transition from land to sea: approximately the same slightly increasing dust AOT trends 323 of ~0.004 yr-1 were obtained along the east of the Ganges basin and over north-west 324 BoB (Figs. 5b and 6c). We found that these AOT trends over zones from 5 to 7 were 325 statistically significant (Table 3). The same dust AOT trends along the Ganges basin 326 and over north-west BoB suggest an increasing trend in some external source of dust 327 emissions, outside the Ganges basin. It should be kept in mind that MERRAero only 328 assimilates total AOT and that the trend in aerosol speciation may depend on the trend 329 (or lack thereof) of the specified emissions.

330

331 The distribution of sulfate AOT along the Ganges basin is determined by sulfate aerosol 332 emissions, together with aerosol transport (by the action of prevailing winds) and 333 deposition processes (Fig. 5a). The sulfate contribution to the total AOT increased along 334 the Ganges basin from approximately 30 % over zone 1 to ~56% over zone 5 (Table 3). 335 Over north-west BoB (zones 6 and 7), the sulfate contribution to the total AOT was 336 over 50% (Table 3). Thus, according to MERRAero AOT data, sulfates were the major 337 atmospheric aerosol component over the east of the Ganges basin and over north-west 338 BoB. Moreover, MERRAero data showed that sulfate AOT trends changed in transition 339 from land to sea: strong statistically-significant increasing sulfate AOT trends (of 0.008

343 With respect to organic and black carbon aerosols, their distribution of eight-year mean 344 AOT values along the Ganges showed a wide maximum from the north-west to the 345 center of the Ganges basin (zones from 1 to 3) (Fig. 5a). As mentioned, this area of 346 maximum carbon AOT is known for crop waste burning aerosols [Sharma et al., 2010. 347 Venkataraman et al., 2006]. AOT values of carbon aerosols decrease to the east from 348 zone 3 (Fig. 5a). As discussed in Section 4.1, the reason for the decrease in AOT over 349 the east of the Ganges basin in October is significant rainfall accompanied by aerosol 350 wet removal processes. The joint contribution of organic and black carbon aerosols to 351 the total AOT is  $\sim 38\%$  over the north-west of the Ganges basin (zones from 1 to 3); 352 ~35% over the east of the Ganges basin (zone 5), and approximately 27% over north-353 west BoB (zones 6 and 7) (Table 3). Similar to AOT trends of sulfate aerosols, 354 MERRAero showed that AOT trends of carbon aerosols changed in transition from land 355 to sea: increasing AOT trends in organic and black carbon AOT over the sea (zones 6 356 and 7) exceeded those over zone 5 in the land (Fig. 5b and 6b, and Table 3).

357

Based on MERRAero data, we found that, in October, the contribution of sea-salt aerosols to the total AOT over the east of the Ganges basin was even lower than that of desert dust. Over north-west BoB, desert dust and sea-salt aerosols equally contributed to the total AOT (Fig. 7a). Our analysis showed that sea-salt aerosols did not contribute to the increasing AOT trends over north-west BoB: no sea-salt AOT trend was observed in year-to-year variations during the study period (Fig. 7a). This was supported by yearto-year variations of MERRA surface winds over north-west BoB (Fig. 7b).

365

## 366 4.4. Factors contributing to AOT trends over north-west BoB

MERRAero showed increasing AOT trends over north-west BoB in October exceeding AOT trends over the east of the Ganges basin (Fig. 1b). This was despite the fact that sources of air pollution are located on the land, mainly in the Ganges basin. There could be several factors contributing to the increasing AOT trends over north-west BoB. First, there were changes in the atmospheric circulation over north-west BoB in October during the eight-year study period (Fig. 8). Mean wind vectors of the 700-850 hPa layer in each October during the 8-year period under consideration were analyzed (Fig. 8). 374 The 700-850 hPa layer is considered as indicative of wind in the lower troposphere, 375 where aerosol transport mainly occurs [Dunion and Velden, 2004]. During the second 376 4-year period (2006 - 2009), prevailing winds blowing mainly from land to sea (Fig. 8, 377 e - h) resulted in a drier environment and less precipitation over the east of the Ganges 378 basin and north-west BoB (Fig. 4a) than during the first 4-year period (2002 – 2005) 379 (Fig. 8, a - d). This caused less wet removal of air pollution in the second 4-year period 380 than in the first 4-year period. Second, our analysis showed that, during the eight-year 381 study period, there was an increasing number of days (Np, in percentage form) in each 382 October when prevailing winds blew from land to sea (Fig. 9). This suggests some 383 increasing trends in the transport of anthropogenic air pollution from their sources in the 384 east of the Ganges basin to north-west BoB. Third, for Octobers when Np > 50%, wind 385 convergence was observed over north-west BoB causing the accumulation of aerosol 386 particles over that region (Fig. 10), in line with our previous study [Kishcha et al., 387 2012]. All the three factors contributed to the increasing AOT trend over north-west 388 BoB in the early post-monsoon season.

389

390 Fig. 11 represents maps of the absolute value of the magnitude of monthly vertically 391 integrated mass flux of aerosols including sulfates, carbonates, and desert dust over the 392 study region. These maps were obtained for each October between 2002 and 2009. One 393 can see that, over north-west BoB (zone 6), the aerosol flux was higher during the 394 second 4-year period (2006 - 2009) (when prevailing winds blew mainly from land to 395 sea) than during the first 4-year period (2002 - 2005) (when prevailing winds blew 396 frequently from sea to land). In October 2009, monthly mean wind over zone 6 was 397 minimal (less than 1 m/s) (Fig. 8h). As the aerosol flux is proportional to wind, then one 398 could expect that the aerosol flux over zone 6 should be also minimal. However, as 399 shown in Fig. 11h, in October 2009, the aerosol flux over zone 6 was maximal during 400 the study period. The maximal flux under the minimal wind indicates accumulating 401 aerosol particles over zone 6. This is supported by the analysis of wind convergence: the 402 wind convergence over zone 6 was also maximal in October 2009 (Fig. 10e). Therefore, 403 we can conclude that vertically integrated aerosol mass flux is sensitive to wind 404 convergence causing aerosol accumulation. Moreover, the aerosol flux is sensitive to 405 precipitation: its magnitude is maximal over the north-west of the Ganges basin, and 406 decreases towards north-west BoB over the specified zones in the east of the Ganges 407 basin, due to wet removal processes after significant rainfall (Fig. 11). Therefore, the

analysis of aerosol fluxes supports our findings on the effects of wind convergence andprecipitation on AOT.

410

## 411 5. Analysis of AOT increments

412 MODIS crosses the study area twice a day. Specifically, MODIS-Terra crosses the 413 study area at approximately 04:30 UT (10:30 LT), while MODIS-Aqua at 414 approximately 08:30 UT (14:30 LT). MODIS-Terra AOT retrievals were used for 415 updating MERRAero AOT at 6 UT, while MODIS-Aqua retrievals were used for updating MERRAero AOT at 9 UT. It is clear that, after the two consecutive updates, 416 417 MERRAero AOT at 9 UT corresponds in the best way to available MODIS 418 measurements over the study area. During all other times (18 hours per day), the model 419 simulates air pollution over the study area independently of MODIS, using available 420 meteorology and predefined aerosol emissions. Taking into account the uncertainty of 421 aerosol emissions over such a complex study area, which includes the highly-populated 422 and polluted Ganges basin, one could expect some accumulation of model errors in 423 AOT simulations during the 18-hour period without data assimilation.

424

425 We estimated the NASA GEOS-5 model performance over the study area analyzing 426 model AOT increments. These AOT increments are the field differences between 427 MODIS AOT and modeled AOT. AOT increments include a complex combination of 428 all model errors in AOT simulations over each specific location. To analyze AOT 429 increments, 3-hour MERRAero AOT data were used. These 3-hour data allowed us to 430 distinguish between 3-hour periods with and without MODIS AOT assimilation. Fig. 431 12a represents a spatial distribution of the eight year (2002 - 2009) mean AOT 432 increments at 6 UT. One can see maximal AOT increments over the north-west of the 433 Ganges basin (zones 1-3), where precipitation was minimal as shown in Fig. 4a. In the 434 absence of precipitation, the deficiency in anthropogenic aerosol emissions could be the 435 main contributor to the high AOT increments. By contrast, over the east of the Ganges 436 basin and north-west BoB (zones 4 - 7), AOT increments were low, indicating that 437 MODIS AOT assimilation did not contribute significantly to modeled AOT there. This 438 was despite the fact that, in the east of the Ganges basin (zones 4 - 5), the population 439 density is the highest in the entire Ganges basin (Fig. 3), which means the highest 440 industrial and transportation emissions. The most probable reason why AOT increments 441 were lower over zones 4 - 5 than over zones 1 - 3, is the aforementioned significant

increase in rainfall (accompanied by aerosol wet removal processes) towards north-west
BoB. Over the east of the Ganges basin and north-west BoB, the NASA GEOS-5 model
was capable of reproducing the effect of significant rainfall on MERRAero AOT which,
in turn, dominates the effect of aerosol emissions there.

446

447 Fig. 12b represents a spatial distribution of the eight year (2002 – 2009) mean AOT 448 increments at 9 UT. One can see that, in spite of the MODIS AOT assimilation at 6 UT, 449 the AOT increments at 9 UT are quite significant over the north-west of the Ganges 450 basin (zones 1 - 2). Therefore, MODIS AOT assimilations at 6 UT and 9 UT were 451 essential for correcting modeled AOT mainly over the north-west of the Ganges basin, 452 where AOT increments were maximal. Over the east of the Ganges basin and north-453 west BoB where AOT increments were low, MODIS AOT assimilation did not 454 contribute significantly to modeled AOT.

455

456 Our findings are further illustrated by Fig. 13 which represents a comparison between 457 spatial distributions of eight-year (2002 - 2009) AOT trends at 6 UT with and without 458 AOT assimilation. Here, the modeled AOT without MODIS assimilation was obtained 459 as the field difference between MODIS AOT and AOT increments at 6 UT. One can see 460 some similarity in the two distributions of AOT trends, namely, AOT trends over north-461 west BoB exceed those over the Ganges basin (Fig. 13). This is evidence that the 462 increasing AOT trends over north-west BoB were reproduced by the model not only 463 because MODIS AOT assimilation provided us with an opportunity to correct the 464 uncertainty in aerosol emissions, but mainly because the model was capable of 465 reproducing changes in meteorological factors contributing to the AOT trends.

466

#### 467 **6.** Conclusions

The recently developed eight-year (2002 – 2009) MERRAero assimilated aerosol data set was applied to the study of AOT and its trends over the Ganges basin and north-west BoB in the early post-monsoon season. MERRAero showed that AOT was lower over the east of the Ganges basin than over the north-west of the Ganges basin: this was despite the fact that the east of the Ganges basin should have produced higher anthropogenic aerosol emissions due to higher population density, increased industrial output and transportation. This is evidence that higher aerosol emissions do not always

475 correspond to higher AOT over the areas where the effects of meteorological factors on476 AOT dominate those of aerosol emissions.

477

478 Based on the analysis of model AOT increments, we have arrived at the important point 479 of the current study which is as follows: the decrease in AOT over the east of the 480 Ganges basin towards north-west BoB was reproduced by the model not only because of MODIS AOT assimilation, but mainly because of the model capability of 481 482 reproducing meteorological factors contributing to AOT. MODIS AOT assimilation 483 was essential over the north-west of the Ganges basin, where AOT increments were 484 maximal, indicating the deficiency in anthropogenic aerosol emissions used in the 485 model. Over the east of the Ganges basin and north-west BoB, AOT increments were 486 low and AOT assimilation did not contribute significantly to modeled AOT there.

487

In October, in the absence of aerosol sources in north-west BoB, MERRAero showed increasing AOT trends over north-west BoB exceeding those over the east of the Ganges basin. Similar AOT trends were obtained using MERRAero AOT with and without MODIS AOT assimilation. Various aerosol components showed strong increasing AOT trends over north-west BoB. Therefore, using MERRAero AOT, we obtained similar results with respect to AOT trends over north-west BoB, as in our previous study based on MODIS data [Kishcha et al., 2012].

495

496 There were a number of meteorological factors contributing to the increasing AOT497 trends over north-west BoB:

498 499 • an increasing number of days in each October when prevailing winds blew from land to sea, resulting in an increase in air pollution over north-west BoB;

during the second 4-year period (2006 – 2009), prevailing winds blowing mainly
 from land to sea were responsible for a drier environment with less precipitation
 causing less wet removal of air pollution than in the first 4-year period (2002 –
 2005);

in October 2009, the vertically integrated aerosol mass flux over north-west BoB
 (zone 6) was maximal, while monthly mean wind was minimal. This indicates
 accumulating aerosol particles over north-west BoB. The wind convergence over
 north-west BoB was also maximal in October 2009. Therefore, vertically

integrated aerosol mass flux is sensitive to wind convergence causing aerosol accumulation.

510

511 The MERRAero AOT data set allowed us to determine aerosol species responsible for 512 the AOT maximum over the north-west of the Ganges basin, which are both natural 513 aerosols (dust particles) and anthropogenic aerosols (carbon aerosols from bio-mass 514 burning and sulfates). In accordance with MERRAero data, most of dust particles fell 515 out due to gravitational settling and wet deposition during dust transport from the north-516 west to the east of the Ganges basin. As a result, over the east of the Ganges basin and 517 north-west BoB in the early post-monsoon season, aerosols were dominated by 518 anthropogenic air pollution, such as sulfates and carbon aerosols. Our analysis showed 519 that sea-salt aerosols did not contribute to the increasing AOT trends over north-west 520 BoB: no sea-salt AOT trend was observed in year-to-year variations during the study 521 period.

522

523 We used MERRAero aerosol reanalysis over the Ganges basin and north-west BoB, but 524 this global NASA product can be used over other places. Our findings illustrate that, 525 while analyzing MERRAero AOT over other places, model AOT increments could be 526 helpful in determining (a) areas where effects of aerosol emissions on AOT dominate 527 those of meteorology, and MODIS AOT assimilation would be essential for correcting 528 modeled AOT, and (b) areas where effects of meteorological factors on AOT dominate 529 those of aerosol emissions, and MODIS AOT assimilation would not contribute 530 significantly to modeled AOT.

#### 532 **References**

- 533 Chin, M., P. Ginoux, S. Kinne, O. Torres, B. Holben, B.N. Duncan, R.V. Martin, J.
- Logan, A. Higurashi, T. Nakajima (2002), Tropospheric aerosol optical
  thickness from the GOCART model and comparisons with satellite and sun
  photometer measurements. *J. Atmos. Phys.* 59, 461–483, doi:
  http://dx.doi.org/10.1175/1520-0469(2002)059.
- Colarco, P., A. da Silva, M. Chin, and T. Diehl (2010), Online simulations of global
  aerosol distributions in the NASA GEOS-4 model and comparisons to
  datellite and ground-based aerosol optical depth. *J. Geophys. Res.*, 115,
  D14207, doi:10.1029/2009JD012820.
- 542 Darmenov, A., and A.M. da Silva (2013), The quick fire emissions dataset (QFED)
  543 Documentation of versions 2.1, 2.2 and 2.4. NASA Technical Report Series
  544 on Global Modeling and Data Assimilation. NASA TM-2013-104606, 32,
  545 183p.
- 546 Dee, D.P., L. Rukhovets, R. Todling, A.M. da Silva, and J.W. Larson (2001), An
  547 Adaptive Buddy Check for Observational Quality Control. *Quarterly Journal*548 *of the Royal Meteorological Society*, 127(577), 2451–2471,
  549 doi:10.1002/qj.49712757714.
- Dee, D.P., and A.M. da Silva (1999), Maximum-Likelihood Estimation of Forecast
  and Observation Error Covariance Parameters. Part I: Methodology. *Mon. Weather Rev.*, 127(8), 1822–1834, doi:http://dx.doi.org/10.1175/1520-0493.
- 553 Derber, J. C., R.J. Purser, W.-S. Wu, R. Treadon, M. Pondeca, D. Parrish, and D.
- 554 Kleist (2003), Flow-dependent Jb in a global grid-point 3D-Var. Proc.
- 555 ECMWF annual seminar on recent developments in data assimilation for
- atmosphere and ocean. Reading, UK, 8-12 Sept. 2003.

- 557 Dey, S., and L. Di Girolamo (2011), A decade of change in aerosol properties over
  558 the Indian subcontinent, *Geophys. Res. Lett.*, 38, L14811,
  559 doi:10.1029/2011GL048153.
- 560 DI Girolamo, L., T.C. Bond, D. Bramer, D.J. Diner, F. Fettinger, R.A. Kahn, J.V.
- 561 Matronchik, M.V. Ramanathan, and P.J. Rash (2004), Analysis of Multi-angle
- 562Imaging SpectroRadiometer (MISR) aerosol optical depths over greater India563during winter 2001 2004. Geophys. Res. Lett., 31, L23115,
- 564 doi:10.1029/2004GL021273.
- Dunion, J. P., and C. S. Velden (2004), The impact of the Saharan air layer on
  Atlantic tropical cyclone activity, *Bull. Am. Meteorol. Soc.*, 85(3), 353–365,
  doi:10.1175/BAMS-85-3-353.
- Ganguly, D., A. Jayaraman, and H. Gadhavi (2005), In situ ship cruise
  measurements of mass concentration and size distribution of aerosols over
  Bay of Bengal and their radiative impacts. *J. Geophys. Res.*, 110, D06205,
  doi:10.1029/2004JD005325.
- Hess, M, P Koepke, and I Schult (1998), Optical Properties of Aerosols and
  Clouds: the Software Package OPAC. *Bulletin of the American Meteorological Society*, 79 (5), 831–844, DOI:10.1175/1520-0477(1998)079.
- 575 Hill, C, C. DeLuca, Balaji, M. Suarez, and A. da Silva (2004): The architecture of
  576 the Earth System Modeling Framework. *Computing in Science and*577 *Engineering*, 6(1), 18–28.
- Hsu, N.C., R. Gautam, A.M. Sayer, C. Bettenhausen, C. Li, M.J. Jeong, S.C. Tsay,
  and B.N. Holben (2012), Global and regional trends of aerosol optical depth
  over land and ocean using SeaWiFS measurements from 1997 to 2010.

- 581
   Atmos. Chem. Phys. Discuss., 12, 8465-8501, doi:10.5194/acpd-12-8465 

   582
   2012.
- Huffman, G.J., R.F. Adler, D.T. Bolvin, G. Gu, E.J. Nelkin, K.P. Bowman, Y.
  Hong, E.F. Stocker, D.B. Wolff (2007), The TRMM multisatellite
  precipitation analysis (TMPA): quasi-global, multiyear, combined-sensor
  precipitation estimates at fine scales. *J. Hydrometeorology*, 8, 38 55, doi:
  10.1175/JHM560.1.
- Kaskaoutis, D.G., S.K. Kharol, P.R. Sinha, R.P. Singh, K.V.S. Badarinath, W.
  Mehdi, and M. Sharma (2011a). Contrasting aerosol trends over South Asia
  during the last decade based on MODIS observations. *Atmos. Meas. Tech. Discuss.*, 4, 5275–5323, doi:10.5194/amtd-4-5275-2011.
- Kaskaoutis, D.G., S. Kumar Kharol, P.R. Sinha, R.P. Singh, H.D. Kambezidis, A.
  Rani Sharma, and K.V.S. Badarinath (2011b), Extremely large anthropogenic
  aerosol contribution to total aerosol load over the Bay of Bengal during winter
  season. *Atmos. Chem. Phys.*, 11, 7097–10 7117, doi:10.5194/acp-11-70972011.
- Kishcha, P., B. Starobinets, O. Kalashnikova, and P. Alpert (2011), Aerosol optical
  thickness trends and population growth in the Indian subcontinent. *Int. J. Remote. Sens.*, 32, 9137-9149, doi:10.1080/01431161.2010.550333.
- Kishcha, P., B. Starobinets, C.N. Long, P. Alpert (2012), Unexpected increasing
  AOT trends over north-west Bay of Bengal in the early post-monsoon season.
- 602 J. Geophys. Res., 117, D23208, doi 10.1029/2012JD018726.
- Krishna Moorthy, K., S. Suresh Babu, M.R. Manoj, and S.K. Satheesh (2013),
  Buildup of aerosols over the Indian region. *Geophys. Res. Lett.*,
  doi:10.1002/GRL.50165.

- Kumar, A., M.M. Sarin, and B. Srinivas (2010), Aerosol iron solubility over Bay of
  Bengal: Role of anthropogenic sources and chemical processing, *Marine Chemistry*, 121, 167–175.
- 609 Lary, D.J., L.A. Remer, D. MacNeill, B. Roscoe, and S Paradise (2009), Machine
- Learning and Bias Correction of MODIS Aerosol Optical Depth. *Geoscience and Remote Sensing Letters*, IEEE, 6(4), 694–698, doi:
  10.1109/LGRS.2009.2023605.
- Mishchenko, M. I., and I. V. Geogdzhayev (2007), Satellite remote sensing reveals
  regional tropospheric aerosol trends, Opt. Express, 15, 7423 7438,
  doi:10.1364/OE.15.007423.
- Moorthy, K.K., S.K. Satheesh, S.S. Babu, and C.B.S. Dutt (2008), Integrated
  campaign for aerosols, gases and radiation budget (ICARB): An overview. *J. Earth Syst. Sci.*, 117, 243–262, doi:10.1007/s12040-008-0029-7.
- 619 Pawson, S., R.S Stolarski, A.R. Douglass, P.A. Newman, J.E.c Nielsen, S.M. Frith, 620 and M.L Gupta (2008), Goddard Earth Observing System Chemistry-Climate 621 Model Simulations of Stratospheric Ozone-Temperature Coupling Between 622 1950 and 2005. J. Geophys. 113(D12), Res., D12103, 623 doi:10.1029/2007JD009511.
- Prasad, A.K. and R.P. Singh (2007), Comparison of MISR-MODIS aerosol optical
  depth over the Indo-Gangetic basin during the winter and summer seasons
  (2000-2005). *Remote Sensing of Environment*, 107, 109 119,
  doi:10.1016/j.rse.2006.09.026.
- Ramachandran, S. and A. Jayaraman (2003), Spectral aerosol optical depths over
  Bay of Bengal and Chennai: II Sources, anthropogenic influence and model
  estimates. *Atmos. Env.*, 37, 1951–1962, doi:10.1016/S1352-2310(03)00082-7.

631	Ramanathan, V., and M. Ramana (2005), Persistent, widespread and strongly
632	absorbing haze over the Himalayan foothills and the Indo-Gangetic plains.
633	Pure and Applied Geophysics, 162, 1609 - 1626, doi:10.1007/s00024-005-
634	2685-8.

- Razali, N.M., and Y.B. Wah (2011), Power comparisons of Shapiro-Wilks,
  Kolmogorov-Smirnov, Lilliefors and Anderson-Darling tests. *J. Statistical Modeling and Analytics*, 2, 21-33.
- Rienecker, MM, MJ Suarez, R Todling, J Bacmeister, L Takacs, H-C Liu, W Gu,
  M. Sienkiewicz, R.D. Koster, R. Gelaro, I. Stajner, and E. Nielsen (2008),
- 640 The GEOS-5 Data Assimilation System--Documentation of Version 5.0.1,

641 5.1.0, and 5.2.0. NASA/TM-2007-104606, 27, 1-118.

- 642 Rienecker, M.M., M.J. Suarez, R. Gelaro, R. Todling, J. Bacmeister, E. Liu, M.G.
- 643 Bosilovich, S.D. Schubert, L. Takacs, G.-K. Kim, S. Bloom, J. Chen, D.
- 644 Collins, A. Conaty, A. da Silva, et al. (2011), MERRA: NASA's Modern-Era
- 645 Retrospective Analysis for Research and Applications. J. Climate, 24, 3624-
- 646 3648, doi: 10.1175/JCLI-D-11-00015.1.
- Shapiro, S.S., and M.B. Wilk (1965), An analysis of variance test for normality
  (complete samples). Biometrika, 52, 591 611, doi:10.1093/biomet/52.34.591.
- Sharma, A.R., S.K. Kharol, K.V.S. Badarinath, and D. Singh (2010), Impact of
  agriculture crop residue burning on atmospheric aerosol loading a study
  over Punjab State. India, *Ann. Geophys.*, 28, 367–379, doi:10.5194/angeo-28367-2010.
- Tripathi, S.N., S. Day, A. Chandel, S. Srivastava, R.P. Singh, and B. Holben
  (2005), Comparison of MODIS and AERONET derived aerosol optical depth

656 over the Ganga basin, India. Ann. Geophys., 23, 1093-1101,
657 doi:10.5194/angeo-23-1093-2005.

- 658 Venkataraman, C., G. Habib, D. Kadamba, M. Shrivastava, J.-F., Leon, B. 659 Crouzille, O. Boucher, D. Streets (2006), Emissions from open biomass 660 burning in India: Integrating the inventory approach with high-resolution 661 Moderate Resolution Imaging Spectroradiometer (MODIS) active-fire and 662 cover land data. Global Biogeochem. Cycles, 20, GB2013, 663 doi:10.1029/2005GB002547.
- 664 Vinoj, V., S.S. Babu, S.K. Satheesh, K.K. Moorthy, and Y.J. Kaufman (2004), Radiative forcing by aerosols over the Bay of Bengal region derived from 665 shipborne, island-based, and satellite (Moderate-Resolution 666 Imaging 667 Spectroradiometer) observations. J. Geophys. Res. 109. D05203. 668 doi:10.1029/2003JD004329.
- Wu, W.S., R.J. Purser, and D.F. Parrish (2002): Three-dimensional variational
  analysis with spatially inhomogeneous covariances. *Mon. Weather. Rev.*, 130,

671 2905–2916, doi:<u>http://dx.doi.org/10.1175/1520-0493</u>.

- Zhang, J. and J.S. Reid (2006), MODIS Aerosol Product Analysis for Data
  Assimilation: Assessment of Over-Ocean Level 2 Aerosol Optical Thickness
  Retrievals. J. Geophys. Res., 111(D22), D22207. doi:10.1029/2005JD006898.
- 675 Zhang, J.L. and J.S. Reid (2010), A decadal regional and global trend analysis of
- the aerosol optical depth using a data-assimilation grade over-water MODIS
- and Level 2 MISR aerosol products. Atmos. Chem. Phys., 10, 10949-10963,
- 678 doi:10.5194/acp-10-10949-2010.
- Zhao, T. X.-P., I. Laszlo, W. Guo, A. Heidinger, C. Cao, A. Jelenak, D. Tarpley,
  and J. Sullivan (2008), Study of long-term trend in aerosol optical thickness

- 681 observed from operational AVHRR satellite instrument, J. Geophys. Res.,
- 682 113, D07201, doi:10.1029/2007JD009061.

6	8	4
---	---	---

Acknowledgements. This study was made with support from and in cooperation with
the international Virtual Institute DESERVE (Dead Sea Research Venue), funded by the
German Helmholtz Association. We also acknowledge the GES-DISC Interactive
Online Visualization and Analysis Infrastructure (Giovanni) for providing us with
TRMM data.

- 692 Table 1. The overview of main attributes of NASA MERRAero assimilated aerosol
- 693 data.

Feature	Description
Model	GEOS-5 Earth Modeling System (with GOCART aerosol components); Constrained by MERRA Meteorology (Replay) Land sees obs. precipitation (like MERRA <i>Land</i> ) Driven by QFED daily Biomass Emissions
Aerosol data assimilation	Local Displacement Ensembles (LDE) MODIS reflectances AERONET Calibrated AOT's (Neural Net) Stringent cloud screening
Period	mid 2002-present (Aqua + Terra)
Resolution	Horizontal: nominally 50 km Vertical: 72 layers, top ~85 km
Aerosol Species	Dust, sea-salt, sulfates, organic & black carbon

- 698 Table 2. Eight-year (2002-2009) mean AOT ( $\tau$ ), standard deviation (sd), and AOT slope
- 699 (a) of MERRAero AOT averaged over the specified zones in October<sup>v</sup>.

Area	Zone #	Geographic	τ	sd	α	S - W	
		Coordinates			(per year)	test	р
IS	1	28.5N - 31.5N	0.53	0.07	-0.005	Normal	Not significant
		72.7E – 75.7E					-
	2	27N - 30N	0.50	0.08	-0.004	Normal	Not significant
		75.7E – 78.7E					-
	3	25N - 28N	0.51	0.08	0.008	Normal	Not significant
		78.7E-81.7E					-
	4	24N - 27N	0.43	0.05	0.012	Normal	Not significant
		82.5E-85.5E					-
	5	22N – 25N	0.35	0.05	0.010	Normal	Not significant
		86E – 89E					-
BoB	6	18N – 21N	0.22	0.05	0.015	Normal	0.043
		87E – 90E					
	7	15N – 18N	0.20	0.05	0.020	Normal	0.006
		84E – 87E					

<sup>v</sup>The decision based on the Shapiro – Wilk normality test for residuals (S-W test) and
the significance level (p) are also displayed. If the p value was too high as compared
with the 0.05 significance level, the obtained linear fit was considered as statistically
insignificant.

Table 3. The eight-year mean AOT ( $\tau$ ), standard deviation (sd), and AOT slope ( $\alpha$ ) for long-term changes of MERRAero AOT for different aerosol species (desert dust; organic and black carbon; and sulfates) averaged over specified zones in October. F corresponds to the fraction of aerosol component AOT (in percentages) from the total MERRAero AOT.

712

Area	Zone #	F	τ	sd	α	S - W	
		%			(per year)	test	р
					Sulfates		
IS	1	31.7	0.17	0.04	0.001	Normal	Not significant
	2	36.7	0.18	0.04	-0.001	Normal	Not significant
	3	43.7	0.22	0.05	0.004	Normal	Not significant
	4	49.7	0.22	0.04	0.006	Normal	Not significant
	5	55.9	0.20	0.03	0.004	Normal	Not significant
BoB	6	52.9	0.12	0.03	0.008	Normal	0.050
	7	50.7	0.10	0.03	0.011	Normal	0.004
					Organic and black carbon		
IS	1	37.7	0.20	0.05	-0.011	Normal	Not significant
	2	39.2	0.20	0.03	-0.009	Normal	Not significant
	3	37.7	0.19	0.02	-0.001	Normal	Not significant
	4	37.5	0.16	0.02	0.002	Normal	Not significant
	5	34.9	0.12	0.02	0.003	Normal	Not significant
BoB	6	28.8	0.06	0.02	0.004	Normal	Not significant
	7	25.9	0.05	0.02	0.005	Normal	0.026
					Desert dust		
IS	1	29.5	0.16	0.04	0.006	Normal	Not significant
	2	23.2	0.12	0.03	0.006	Normal	Not significant
	3	17.8	0.09	0.03	0.005	Normal	Not significant
	4	12.2	0.05	0.02	0.004	Normal	Not significant
	5	8.0	0.03	0.01	0.004	Normal	0.035
BoB	6	8.3	0.02	0.01	0.004	Normal	0.012
	7	8.1	0.02	0.01	0.004	Normal	0.008

713

714





Figure 1. Spatial distributions of (a) the eight year (2002 – 2009) mean MERRAero AOT and (b) its trends (characterized by AOT slopes) in October. The AOT trend values correspond to the slope of the linear regression analysis. The squares show the locations of zones 1 to 7 within the study region.



Figure 2. a - zone-to-zone variations of eight-year (2002-2009) mean MERRAero AOT averaged over the specified zones. b - zone-to-zone variations of MERRAero AOT averaged over the first four-year (2002-2005) period and over the second four-year (2006-2009) period. The error bars show the standard error of mean AOT.





Figure 3. a – population density (persons km<sup>-2</sup>) distributions over the Indian subcontinent. b – zone-to-zone variations of population density averaged over the specified zones. The GPW-v3 gridded population density data for the year 2005 were used (http://sedac.ciesin.columbia.edu/data/collection/gpw-v3).



Figure 4. a - zone-to-zone variations of TRMM accumulated rainfall over the specified
zones in October averaged over the eight-year study period (2002 – 2009), over the first
4-year period (2002 – 2005), and over the second 4-year period (2006 – 2009). The
error bars show the standard error of mean accumulated rainfall. TRMM data from the
3B42V6 archive were used.

743 b – the spatial distribution of the eight year (2002 – 2009) mean model-simulated 744 aerosol wet deposition rate (kg m<sup>-2</sup> s<sup>-1</sup>) in October.



Figure 5. Zone-to-zone variations of (a) eight-year (2002-2009) mean MERRA AOT of various aerosol species (sulfates (SU), organic and black carbon (OC & BC), and desert dust) averaged over the specified zones in October, and (b) their AOT trends (characterized by the slope of the linear regression analysis). The error bars show the standard error of mean AOT.



Figure 6. Year-to-year variations of AOT for various aerosol species (such as sulfates, organic carbon, and desert dust) over zones 6 and 7 in north-west BoB in October. The straight lines (dashed for zone 7 and solid for zone 6) designate linear fits.

- 757
- 758



Figure 7. Year-to-year variations of (a) MERRAero AOT of sea-salt aerosols and (b)
MERRA surface wind over north-west BoB (zones 6 and 7) in October.



Figure 8. Spatial distributions of mean MERRA wind vectors of the 700-850 hPa layer in each October during the study period. 766



Figure 9. Numbers of days (in percentage form) in each October during the study period
when prevailing wind (transporting air pollution) blew from the east of the Ganges
basin (zone 5) to north-west BoB (zone 6).



Figure 10. Spatial distributions of mean wind convergence (10<sup>-6</sup> s<sup>-1</sup>) of the 700-850 hPa
layer for each October, when the number of days with prevailing winds, blowing from
the east of the Ganges basin (zone 5) to north-west BoB (zone 6), exceeded 50%.
MERRA wind reanalysis data were used.



Figure 11. Spatial distributions of the absolute value of the magnitude of monthly integrated mass flux (kg m<sup>-1</sup> month<sup>-1</sup>) of aerosols (including sulfates, carbonates, and desert dust) in each October during the study period. The absolute value of the flux magnitude was obtained by the following expression:  $((Fu)^2 + (Fv)^2)^{1/2}$ , where Fu is the vertically integrated zonal mass flux and Fv is the vertically integrated meridional mass flux.





Figure 12. Spatial distributions of the eight year (2002 – 2009) mean AOT increments

in October at (a) 6 UT and (b) 9 UT.





12N

9N

6N

3N

795

12N ·

9N ·

6N

3N

0.01

0.005

-0.005