Quantifying enhancement in aerosol radiative forcing during ‘extreme aerosol days’ in summer at Delhi National Capital Region, India

Sumant Kumar a, Sagnik Dey b,⁎, Arun Srivastava a

a School of Environmental Sciences, Jawaharlal Nehru University, New Delhi 110067, India
b Centre for Atmospheric Sciences, IIT Delhi, New Delhi 110016, India

HIGHLIGHTS
• Change in aerosol forcing in ‘extreme’ aerosol days at Delhi are quantified.
• Mean AOD increases > 107% during the ‘extreme’ aerosol days.
• Coarse-mode dust is the dominant (70.8%) contributor to AOD during ‘extreme’ days.
• Aerosol-induced heating rate increased by 75.1% from 1.7 K/day to 2.96 K/day.

GRAPHICAL ABSTRACT

Deviation (in %) of aerosol properties from ‘relatively clean’ days to ‘extreme’ aerosol days.

ABSTRACT

Changes in aerosol characteristics (spectral aerosol optical depth, AOD and composition) are examined during the transition from ‘relatively clean’ to ‘extreme’ aerosol days in the summer of 2012 at Delhi National Capital Region (NCR), India. AOD smaller than 0.54 (i.e. 12-year mean AOD − 1σ) represents ‘relatively clean’ days in Delhi during the summer. ‘Extreme’ days are defined by the condition when AOD0.5 exceeds 12-year mean AOD + 1 standard deviation (σ). Mean (±1σ) AOD increases to 1.2 ± 0.12 along with a decrease of Angstrom Exponent from 0.54 ± 0.09 to 0.22 ± 0.12 during the ‘extreme’ days. Aerosol composition is inferred by fixing the number concentrations of various individual species through iterative tweaking when simulated (following Mie theory) AOD spectrum matches with the measured one. Contribution of coarse mode dust to aerosol mass increased from 76.8% (relatively clean) to 96.8% (extreme events), while the corresponding contributions to AOD0.5 increased from 35.0% to 70.8%. Spectrally increasing single scattering albedo (SSA) and CALIPSO aerosol sub-type information support the dominant presence of dust during the ‘extreme’ aerosol days. Aerosol direct radiative forcing (ADRF) at the top-of-the-atmosphere increases from 21.2 W m−2 (relatively clean) to 56.6 W m−2 (extreme), while the corresponding change in surface ADRF is from −99.5 W m−2 to −153.5 W m−2. Coarse mode dust contributes 60.3% of the observed surface ADRF during the ‘extreme’ days. On the contrary, 0.4% mass fraction of black carbon (BC) translates into 13.1% contribution to AOD0.5 and 33.5% to surface ADRF during the ‘extreme’ days. The atmospheric heating rate increased by 75.1% from 1.7 K/day to 2.96 K/day during the ‘extreme’ days.

© 2016 Elsevier B.V. All rights reserved.

Keywords:
Aerosol radiative forcing
Extreme aerosol events
Delhi NCR

⁎ Corresponding author.

http://dx.doi.org/10.1016/j.scitotenv.2016.01.191
0048-9697/© 2016 Elsevier B.V. All rights reserved.
1. Introduction

Aerosols modify the Earth’s radiation budget by directly scattering and absorbing solar radiation and indirectly altering the cloud properties. However, quantification of aerosol direct radiative forcing (ADRF) continues to be uncertain due to heterogeneity in the spatial and temporal variability of their optical and microphysical properties (IPCC, Intergovernmental Panel on Climate Change, 2013). The Indo-Gangetic Basin (IGB) in northern India has been recognized as a major aerosol hotspot (Di Girolamo et al., 2004; Ramachandran and Cherian, 2008; Dey and Di Girolamo, 2010; Moorthy et al., 2013) in South Asia, where aerosol load continues to rise in the recent years (Dey and Di Girolamo, 2011). These studies have documented the seasonal and spatial variability of aerosol optical depth (AOD) over the IGB. Key features of these studies are as follows: (1) AOD is dominated by anthropogenic aerosols during the post-monsoon (Oct–Nov) and winter (Dec–Feb) season and shows a gradient from western (low) to eastern (high) IGB due to the influence of meteorology and topography; (2) AOD increases during the pre-monsoon (Mar–May) season due to the transport of dust from the west and central Asian and Indian dust sources; (3) AOD reduces due to washout by monsoon rain, but recovers quickly due to large emission sources and remains high during dry phase; (4) High AOD leads to large surface dimming and atmospheric heating.

Delhi National Capital Region (NCR), situated in the western IGB (Fig. 1) is the most polluted part of the IGB (Kumar et al., 2007; Lodhi et al., 2013). The source of pollution in this city is natural (dust from desert and nearby arid areas) as well as anthropogenic, like emission from industry, vehicles, biomass and biofuel (Tiwari et al., 2013; Srivastava et al., 2014). Many of these sources (e.g. dust and biomass burning) are seasonal in nature. During every summer, the region experiences several dust storms (Mishra et al., 2013a,b). These dust storms are classified as natural hazards as they disrupt daily life by degrading visibility and posing serious health risk to the exposed population. They add to aerosol load in this region, alter the aerosol optical properties (Dey et al., 2004) thereby change the radiation budget (Srivastava et al., 2011). Therefore, it is important to understand and examine the changes of aerosol characteristics and ADRF during such ‘extreme’
aerosol days. A ground based measurement of spectral AOD and black carbon (BC) was carried out during the summer of 2012 from 26th March to 10th June to examine the changes in aerosol properties in Delhi NCR as the summer progresses. This study reports the measured aerosol characteristics and examines the change in ADRF during the transition from ‘relatively clean’ to ‘extreme’ aerosol days.

2. Methodology

2.1. Site of study and meteorological condition

The site of study is shown in Fig. 1. AOD has been measured at five wavelengths — 380, 500, 675, 936 and 1020 nm at JNU campus (latitude 28.21° to 28.53° N and longitude 76.20° to 77.37° E). JNU is located at outskirts of southern part of Delhi. It is partly vegetative area and it is away from local industrial activity and heavy traffic. Therefore, it provides a site representative of regional aerosol characteristics, not influenced heavily by local pollution. Meteorological parameters are taken from weather underground (www.wunderground.com), which archives meteorological data measured by various national agencies across the world including India Meteorological Department. The data was collected every hour during the day. The temperature (T), relative humidity (RH) and wind speed (WS) during the campaign period varied from 29° to 39 °C, 14% to 58% and 1.9 to 13.4 km/h respectively (Fig. 2).

2.2. Analysis

AOD is measured by MICROTOPS sunphotometer (Solar light corporation, USA) at five different wavelengths. 380, 500, 675, 936, 1020 nm. The instrument uses a narrow band of interference filters. The sunphotometer is factory calibrated with an error margin of 0.002–0.021 in the visible to near infrared wavelength range (Singh et al., 2010). We have classified the aerosol days during the measurement period following the AOD climatology reported by Lodhi et al. (2013) for the period 2001–2012 at Delhi. These AOD data were also collected using the same instrument, thereby causing minimal instrumental bias in our analysis. When our measured daily AOD exceeds 12-year mean AOD + 1 standard deviation (σ) for the corresponding month reported by Lodhi et al. (2013), we categorize it as ‘extreme’ aerosol days. Days with AOD values smaller than 12-year mean AOD — 1σ are classified as ‘relatively clean’ days. Remaining days with AOD values between 12-year mean AOD — 1σ and 12-year mean AOD + 1σ are classified as ‘normal’ aerosol days. We classified 33.3%, 40.5% and 26.2% days as ‘relatively clean’, ‘normal’ and ‘extreme’ aerosol days during the observation period.

Aerosol composition is inferred from the measured AOD spectra and BC concentration using OPAC model (Hess et al., 1998). We choose five aerosol species to be externally mixed — BC, water-soluble, insoluble, dust in accumulation mode and dust in coarse mode. These species are chosen because previous studies (e.g. Singh et al., 2005, 2010) have identified them as the major aerosol species in Delhi during summer. Number concentration of BC is derived from the measured mass.

3. Results and discussions

3.1. Aerosol characteristics during the campaign period

Insoluble component contributes only 0.1% to total mass, while water-soluble and BC contributions remain confined within 1–2% during the measurement period (Fig. 3). Bulk of the aerosol mass is contributed by accumulation mode and coarse mode dust. Mass fraction of accumulation mode dust decreases from 19.5% during ‘relatively clean’ days to 1.5% to ‘extreme’ aerosol days. This decrease is compensated by coarse mode dust, whose mass fraction increases from 76.8% to 96.8%.

Box plot of AOD at 500 nm wavelength during the ‘relatively clean’, ‘normal’ and ‘extreme’ aerosol days is shown in Fig. 4. Median AOD during the ‘relatively clean’ days is observed to 0.51, which increases by ~4% to 0.74 during the normal days. During the ‘extreme’ pollution days, it increases further by 60.8% to reach a median value of 1.2. On
some given days during the ‘extreme’ events, AOD rises even above 1.8. This increase in AOD is accompanied by a consistent reduction in angstrom exponent, a first order indicator of aerosol size. Mean angstrom exponent of 0.54 ± 0.09 during the ‘relatively clean’ days implies dominance of large size particles, consistent with large mass fraction of coarse mode dust. Angstrom exponent further reduces to 0.35 ± 0.08 during ‘normal’ days, when mass fraction of coarse mode dust increases. During ‘extreme’ events when coarse mode dust contributes 96.8% to total aerosol mass, mean angstrom exponent drops to 0.22 ± 0.12.

The relative contributions of each aerosol species to AOD are displayed in Fig. 5. Relative contribution of insoluble component to AOD remains low (<0.1%), while that of water-soluble component remains in the range 12–14% (as compared to a mass fraction of 1–2% as shown in Fig. 3) during the measurement period. BC contributes 30.1% during the relatively ‘clean’ days, which decreases to 20.9% and 13.1% during the ‘normal’ and ‘extreme’ pollution days respectively. 20.5% (35.0%) relative contribution of accumulation mode dust (coarse mode dust) to AOD during the ‘relatively clean’ days decreases (increases) to 13.1% (52.8%) and further to 2.5% (70.8%) during the ‘normal’ and ‘extreme’ days respectively.

Mean SSA spectrum during the ‘relatively clean’, ‘normal’ and ‘extreme’ aerosol days as simulated from the reported aerosol composition is shown in Fig. 6. During the entire observation period, SSA shows an increasing trend with wavelength, implying the dominance of dust in the aerosol composition. Dust particles scatter large wavelength more efficiently and absorb wavelength closer to UV, thereby exhibiting an increasing SSA spectrum with wavelength (Dey et al., 2004). Overall, aerosols are highly absorbing during the summer. This is consistent with previous studies by Singh et al. (2005) at Delhi. Aerosol episodes are reflected by an increase in SSA at all the wavelengths, implying enhanced scattering of solar radiation. CALIOP aerosol retrieval during its pass over Delhi on 28th March 2012 is shown in Fig. 7. Based on the measurement, the day is characterized as an ‘extreme’ aerosol day. Aerosol layer can be easily noticed all the way up to 5 km over the Indo-Gangetic Basin. Aerosol sub-type retrieval indicates presence of ‘dust’ and ‘polluted dust’ in this region (bottom panel of Fig. 7). In the CALIOP algorithm, polluted dust is detected when natural mineral dust is transported from the source regions to urban areas and gets mixed with other species (Omar et al., 2009). CALIOP aerosol sub-type provides support to aerosol composition inferred from the measurements, suggesting the robustness of the hybrid approach as described earlier.

3.2. Aerosol direct radiative forcing (ADRF)

ADRF is defined as the change in net radiation due to aerosols. ADRF is calculated by Santa Barbara DISORT Atmospheric Radiative Transfer model (SBDART) developed by Ricchiazzi et al. (1998). We have used the aerosol spectral properties (SSA, asymmetry parameter and AOD) simulated by OPAC as input to SBDART. Other input parameters — total column ozone and surface albedo data have been taken from OMI Level 2G data products and water vapour has been taken from...
MODIS. Radiative fluxes at the TOA and surface have been calculated as function of solar zenith angle. Finally, diurnally averaged ADRF has been estimated following standard procedure demonstrated earlier by numerous researchers e.g. (Dey and Tripathi, 2008; Srivastava et al., 2011).

The mean TOA ADRF has been estimated to be 21.25 W m$^{-2}$ during the ‘relatively clean’ days, which increases to 27.29 W m$^{-2}$ and further to 56.56 W m$^{-2}$ during the ‘normal’ and ‘extreme’ aerosol days (Fig. 8). Large warming at the TOA is attributed to large aerosol load and highly absorbing aerosols present in the atmosphere of Delhi during the summer. The corresponding value for surface ADRF are $-99.47$, $-122.01$
and $-153.5 \text{ W m}^{-2}$ respectively. This results in a large atmospheric warming of $120.72, 149.3$ and $210.06 \text{ W m}^{-2}$ for the ‘relatively clean’, ‘normal’ and ‘extreme’ aerosol days, which translates into heating rates of 1.7, 2.1 and 2.96 K/day respectively. Results suggest that the aerosol induced heating is enhanced by ~75.1% for a corresponding increase of 133.3% increase in AOD from the ‘relatively clean’ to ‘extreme’ aerosol days. In terms of relative contribution of individual aerosol species, water-soluble components contribute to only 3–4% to estimated surface aerosol DRF (Fig. 9). BC contribution is highest (57.7%) for the ‘relatively clean’ days, followed by coarse (26.3%) and accumulation mode dust (11.9%). During the ‘normal’ aerosol days, contributions of BC and accumulation mode dust reduce to 45.3% and 8.6% respectively. They further reduce to 33.5% and 1.9% during the ‘extreme’ aerosol days. Relative contribution of coarse mode dust to surface DRF enhances to 42.1% and then to 60.3% during the ‘normal’ and ‘extreme’ aerosol days. The results presented here have implications for the regional climate. Aerosol loading and ADRF are very large in Delhi NCR. Mean AOD of 0.51 in the ‘relatively clean’ condition (based on 12-year climatology) suggests that the background aerosol concentration is enormous in Delhi NCR during the summer months. Aerosol direct radiative forcing efficiency (per unit AOD) at the TOA is estimated to be $41.7 \text{ W m}^{-2}$ and $47.1 \text{ W m}^{-2}$ for the ‘relatively clean’, ‘normal’ and ‘extreme’ aerosol days. Since mostly accumulation mode dust fraction to bulk aerosol reduces and that of coarse mode dust increases during the transition from ‘relatively clean’ to ‘extreme’ aerosol days; the TOA aerosol direct radiative forcing efficiency (related more to aerosol composition rather than total aerosol load) does not show drastic change. The corresponding values for the surface are $-195.0, -164.9$ and $-127.9 \text{ W m}^{-2}$ respectively. While TOA ADRF change from ‘relatively clean’ to ‘normal’ aerosol days is small ($-6 \text{ W m}^{-2}$) for a 45% increase in AOD, the enhancement is large ($29.3 \text{ W m}^{-2}$) for a 60.8% increase in AOD during the transition from ‘normal’ to ‘extreme’ aerosol days. The atmospheric heating is translated into 1.7 K/day during ‘relatively clean’, while it enhances by 75.1% to 2.96 K/day during the ‘extreme’ aerosol days.

Results presented above have several important climatic implications. Large surface dimming and atmospheric heating impact the regional circulation by modulating the wind pattern and temperature structure of the atmosphere (Das et al., 2014). We note that our estimates assume external mixing of aerosols. During the summertime, dusts transported from the arid regions to the IGB may mix with the anthropogenic species in a core-shell configuration, further enhancing the absorption (e.g. Dey et al., 2008). In that case, enhancement in ADRF would have been larger than the values reported here during the ‘extreme’ aerosol days. Another critical issue, not considered here, is the particle non-sphericity. Dust particles are non-spherical in shape and therefore, the ADRF may differ when the bulk aerosol composition is dominated by dust particles. Mishra et al. (2008) have shown that the deviation in TOA ADRF due to particle non-sphericity may be as large as 10–15% relative to spherical particle in the IGB. Hence the enhancement of AOD and subsequent ADRF presented here may be considered as conservative estimates.

4. Conclusions

We present a quantitative estimate of enhancement in AOD and aerosol direct radiative forcing during the transition from ‘relatively clean’ to ‘normal’ to ‘extreme’ aerosol days at Delhi National Capital region, India during the summer of 2012. The major conclusions of the study are as follows:

1. 33.3%, 40.5% and 26.2% days are categorized as ‘relatively clean’ (AOD smaller than 12-year mean $-1\sigma$), ‘normal’ (AOD between 12-year mean $-1\sigma$ and AOD $+1\sigma$) and ‘extreme’ (AOD exceeds 12-year mean AOD $+1\sigma$) aerosol days in Delhi NCR.
2. AOD increases from 0.51 to 1.2 during the ‘relatively clean’ to ‘extreme’ aerosol days along with a decrease of Angstrom Exponent from 0.54 ± 0.09 to 0.22 ± 0.12.
3. Relative contribution of coarse mode dust to aerosol mass increased from 76.8% (‘relatively clean’) to 96.8% (‘extreme’ events), while the corresponding contribution to AOD$_{0.5}$ increased from 35.0% to 70.8%. Spectrally increasing single scattering albedo and CALIPSO aerosol sub-type information support the dominant presence of dust during the ‘extreme’ aerosol days.
4. Aerosol direct radiative forcing (ADRF) at the top-of-the-atmosphere increases from 21.2 W m$^{-2}$ (‘relatively clean’) to 56.6 W m$^{-2}$ (‘extreme’), while the corresponding change in surface ADRF is from $-99.5 \text{ W m}^{-2}$ to $-153.5 \text{ W m}^{-2}$.
5. Coarse mode dust contributes 60.3% to the surface ADRF during the ‘extreme’ days. The atmospheric heating rate induced by aerosols is enhanced by 75.1% (from 1.7 K/day to 2.96 K/day) during the ‘extreme’ aerosol days.

Acknowledgements

SK acknowledges the University Grant Commission for providing by Basic Scientific Research (UGC BSR) fellowship. SK is also thankful to Parul Srivastava, IIT Delhi for helping in learning use of SBDART model. SD acknowledges support under FAST-Track scheme (SR/FTP/ES-191/2010).

We acknowledge the comments of anonymous reviewers that help improve the earlier version of the manuscript.

References

IPCC (Intergovernmental Panel on Climate Change), 2013. Stocker, T., Qin, D., Plattner, G., Tignor, M., Allen, S., Boschung, J., Nauels, A., Xia, Y., Bex, B., Midgley, B. Climate change 2013: the physical science basis. Contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change.


