On the Contribution of Particulate Matter (PM_{2.5}) to Direct Radiative Forcing over Two Urban Environments in India

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ABSTRACT

Radiative forcing by particulate matter (PM_{2.5}) has been estimated for a period of one year (January–December 2015) over Delhi and Pune (polluted urban metro cities in India). In situ observations of PM_{2.5} and black carbon (BC) over both the cities were obtained from the ground-based System of Air Quality Forecasting and Research (SAFAR) network of stations. Observations have shown that PM concentrations over Pune had a strong diurnal cycle as compared to Delhi in all the seasons. Also, comparisons of the mode values and seasonal frequency distributions (FDs) over Pune and Delhi showed that pollution levels over Delhi were consistently above National Ambient Air Quality Standards (NAAQS). The mean monthly PM_{2.5} values ranged from 61.5 to 162.9 over Delhi and from 17.4 to 74.05 over Pune. The BC mass contribution to PM_{2.5} was found to be 10% to 25% over Pune. However, the contribution of BC to PM_{2.5} was up to 35% over Delhi. Radiative forcing due to PM_{2.5} (PRF) over both the sites was estimated using the Optical Properties of Aerosols and Clouds (OPAC) model along with the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model. The PRF in the atmosphere was between +7.73 Wm⁻² and +14.51 Wm⁻² over Delhi and between +3.12 Wm⁻² and +12.15 Wm⁻² over Pune. Sensitivity experiments showed that the impact of the increase in the hygroscopicity of the aerosols on the PRF was overshadowed by the net changes in albedo.

Keywords: PM_{2.5}; AOD; Albedo; Radiative forcing.

INTRODUCTION

Atmospheric aerosols controls Earth's climate by influencing the radiation budget at regional and global scales. Aerosols (particulate matter (PM)) can be classified as PM₁₀, PM_{2.5}, and PM₁ based on their aerodynamic diameter. They directly influence the climate by scattering and absorbing the incoming solar radiation (Charlson and Schwartz, 1992; Tiwari *et al.*, 2016). Aerosols also influence the climate indirectly by modifying the cloud microphysics (Twomey, 1974; Panicker *et al.*, 2016). Direct aerosol radiative forcing generally is estimated in the shortwave region (0.3–3 μm) and is calculated at the surface and at the Top of the Atmosphere (TOA) to find the net effect in the atmosphere. While the aerosol forcing at the surface always shows a reduction in radiation, the forcing at TOA depends on aerosol species (positive for absorbing aerosols and negative for scattering species). The difference between TOA and surface forcing provides the net gain of radiation due to aerosols in the atmosphere, known as atmospheric aerosol forcing (Panicker and Lee, 2012;

Panicker *et al.*, 2018). Black carbon (BC) is one of the most important absorbing aerosol species in the atmosphere (which is a part of PM_{2.5}), generated mainly from incomplete combustion of fossil fuels and biomass burning (Koelmans *et al.*, 2006; Penner *et al.*, 1993; Cooke and Wilson, 1996). Approximately 20% of BC is reported to be emitted from burning of biofuels, 40% from fossil fuels, and 40% from open biomass burning on a global basis (Ramanathan and Carmichael, 2008). To quantify the climatic effects of PM, it is necessary to determine their local dynamics. Since PM can have high spatial and temporal variability, it is necessary to characterize the PM properties at multiple sites (Lagudu *et al.*, 2011; Kumar *et al.*, 2012). Over South Asia, the extensive study of aerosol direct RF has been made using the single column as well as 3D models and field experiments to understand the climate implications (Ramanathan *et al.*, 2005; Lau *et al.*, 2006; Moorthy *et al.*, 2009; Lawrence and Lelieveld, 2010; Bollasina *et al.*, 2011; Vinoj *et al.*, 2014). Experiments such as Indian Ocean Experiment (INDOEX) (Jayaraman *et al.*, 1998; Satheesh *et al.*, 1999) reported clear-sky aerosol surface forcing of about −29 Wm⁻² over the tropical Indian Ocean. Several studies have been carried out on the radiative forcing of aerosols in regional scale over India (Pandithurai *et al.*, 2004; Panicker *et al.*, 2008; Babu *et al.*, 2002; Tiwari *et al.*, 2013b; Srikanth *et al.*, 2007; Bibi *et al.*, 2017a; etc.). However, most of the studies reported

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were based on a single point observation representing the whole area. SAFAR is a program intended to measure the air pollutants at micro-environmental level over important metro cities. Hence in this study, we are incorporating $PM_{2.5}$ observations from rural/semi-urban and urban locations under SAFAR and the average forcing due to these different environments, which are a true representative of Pune and Delhi and are presented in this paper. The increase in emissions from vehicular and energy sectors are the main sources of growing ambient concentration levels of $PM_{2.5}$ (Gupta *et al.*, 2007). The major sources of PM_{10} and $PM_{2.5}$ in Delhi were reported as windblown dust from unpaved roads, fuel combustion, biomass burning and long-range transport (Tiware *et al.*, 2009). PM concentrations over Pune have been sparsely reported by the Central Pollution Control Board (CPCB, 2010) of India. Long-term exposure to ambient $PM_{2.5}$ has potential health risks including premature death (Brauer *et al.*, 2012; Pope *et al.*, 2002). From the above viewpoint, our present study is an attempt to utilize the $PM_{2.5}$ and BC observed data over Delhi and Pune to estimate the PRF. A brief description of the study area and methodology are provided in Section 2. Detailed results on PRF are discussed in Section 3. Results are summarized in Section 4.

Site Description and Data

The sampling of PM was carried out in different SAFAR locations over Delhi (28.37°N, 77.12°E; altitude: 300 m) and Pune (18.5°N, 73.8°E; altitude: 550 m). Delhi is the National Capital Region of India with 25 million population,

making it the second most populous city in India, and also it is one of the most polluted urban environments over the globe (Tiware *et al.*, 2013a). According to Lodhi *et al.* (2013), main sources of the particulate pollution over Delhi is contributed by anthropogenic emissions from the power plants, vehicles, industrial sector, seasonal agricultural biomass burning and dust storm events over the region. Delhi is characterized with a semi-arid type of climate with high summer-monsoon (July–September) rainfall and dry period during the pre-monsoon (March–June) and post-monsoon (October–November) seasons, while winter (December–February) is chilly and foggy (Tiware *et al.*, 2010; Srivastava *et al.*, 2014b). Wind speeds in general are low ($2\text{--}5\text{ m s}^{-1}$), blowing from southeast directions during monsoon to northwest during the rest of the year, influencing aerosol sources, types and transport pathways (Lodhi *et al.*, 2013). Pune is a rapidly expanding metropolitan city with a population over 4 million. Pune receives annual rainfall at an average of 76 cm. Almost 80% of the annual rainfall takes place during monsoon (June–September). During post-monsoon/winter, winds are normally from east-northeast crossing across the northeast and central India, and dispersion of particulate pollutants are comparatively less due to the low ventilation coefficients (Raju *et al.*, 2014). The location map of measurement sites are depicted in Fig. 1.

The data used in this study are obtained from the SAFAR network for a period of one year (January–December 2015). SAFAR network consists of Air Quality Monitoring Stations (AQMS) and Automatic Weather Stations (AWS) set up across Delhi and Pune in different environments, which

Fig. 1. Observations site of Delhi and Pune.

continuously monitors air pollutants and meteorological parameters. More details of the SAFAR network, air quality and weather monitoring setups used in this study are available elsewhere (<http://safar.tropmet.res.in/>). PM was continuously monitored using Beta Attenuation Monitor (BAM-1020; Met One Instruments, Inc., USA). The instrument allows the particles of concentration with a cut-off aerodynamic diameter of $2.5\ \mu\text{m}$ and less and hence this set of equipment measures $\text{PM}_{2.5}$ particulates only with a resolution of $0.1\ \mu\text{g m}^{-3}$ (Ali et al., 2013; Panicker et al., 2016). The present study uses the measurements of $\text{PM}_{2.5}$ over eight different environments over Delhi and Pune. The main monitoring stations are placed at strategically selected locations by including conditions with vehicular transport, industrial pollution, biomass burning, residential burning etc. BC data was also obtained from the SAFAR network measured by an Aethalometer (Surendran et al., 2013). The calibration and correction methodology of Aethalometer are described elsewhere (Allen et al., 1999; Babich et al., 2000; Weingartner et al., 2003; Arnott et al., 2005; Corrigan et al., 2006). Apart from these visibility and relative humidity data obtained from India Meteorological Department (IMD), water vapor data sets from the Moderate Resolution Imaging Spectroradiometer (MODIS), ozone from Ozone Monitoring Instrument (OMI) satellites, Albedo data from MODIS-BRDF (Bidirectional Reflectance Distribution Function) multispectral wavelength (<https://modis-land.gsfc.nasa.gov/brdf.html>) over both the sites also were used in this study.

Methodology

To estimate the PRF, the PM mass concentration has been bifurcated into water-soluble (WS) and water-insoluble (WIS) components according to prevailing mass concentration conditions as suggested by Hess et al. (1998) and Panicker et al. (2010). The WS and WIS mass concentrations were converted to number density and were used in the OPAC model to obtain aerosol optical properties such as Aerosol Optical Depth (AOD), single scattering albedo (SSA) and asymmetric parameter (ASP) as explained in Hess et al.

(1998).

OPAC model simulates the aerosol optical properties, such as AOD, SSA, and ASP at 60 different wavelengths, at 8 different relative humidity conditions for a given aerosol chemical composition. In this study, we have modeled the aerosol optical properties (AOD, SSA, and ASP) at different RH levels corresponding to the ambient RH values obtained from the India Meteorological Department (IMD) observations for the entire year 2015 for individual cities. Also, the scale heights, which were derived by taking the ratio between AOD and extinction coefficient (σ) as explained in Hayasaka et al. (2007), were adjusted in OPAC model. σ was derived using the visibility observations from IMD as suggested by Panicker et al. (2010). The WS and WIS combinations in OPAC were adjusted to tune OPAC derived AOD to match with the AOD obtained from authentic sun photometer (at 550 nm) observations (from Aerosol Robotic Network) over the sites (Fig. 2). It was adjusted with a $\pm 5\%$ error and hence the resultant aerosol optical properties which are calculated from water-soluble and insoluble PM are reasonable to simulate radiative fluxes and further to estimate radiative forcing.

SBDART model is facilitated with vertically inhomogeneous, non isothermal plane-parallel media, and is shown to be computationally efficient in reliably resolving the radiative transfer equation. The OPAC derived AOD, SSA, and ASP were used in the SBDART model (Ricchiazzi et al., 1998) to obtain the fluxes at the surface and TOA. Fluxes also were derived for “no aerosol” condition and difference in aerosol and no aerosol cases were taken to get PRF. We also used the MODIS-BRDF albedo, water vapor and ozone (from OMI) data over both the sites along with aerosol data sets in SBDART. The default tropical aerosol profile and tropical model profile of temperature and humidity (McClatchey et al., 1972) also were used for estimating PRF. The atmospheric heating rate is calculated for each month using the following equation (Pandithurai et al., 2004; Panicker et al., 2014; Srivastava et al., 2014a; Bibi et al., 2017b):

Fig. 2. Comparison of measured AOD/SSA (AERONET) and OPAC derived AOD/SSA over (a) Delhi and (b) Pune region.

$$\frac{\partial T}{\partial t} = \frac{g}{c_p} \frac{\Delta F}{\Delta P} \quad (1)$$

where $\partial T/\partial t$ is the atmospheric heating rate in K day^{-1} , g is the acceleration due to gravity (9.8 m s^{-2}), c_p is the specific heat capacity of air at constant pressure (i.e., $1006 \text{ J kg}^{-1} \text{ K}^{-1}$) and ΔP (300 hPa) is the atmospheric pressure difference between top and bottom layers of the atmosphere where most aerosols are present which contribute to local heating (Pathak *et al.*, 2010).

RESULTS AND DISCUSSION

Temporal Variability of $\text{PM}_{2.5}$

The monthly variations of $\text{PM}_{2.5}$ have been analyzed using the in-situ measurements from SAFAR network as explained in Section 2. Fig. 3 depicts the monthly variations of $\text{PM}_{2.5}$ during 2015. The mean $\text{PM}_{2.5}$ concentrations were calculated by averaging the observations of eight monitoring stations over Delhi and Pune. The $\text{PM}_{2.5}$ concentrations had ranged between $61.5 \mu\text{g m}^{-3}$ to $162.95 \mu\text{g m}^{-3}$ and showed minimum during monsoon ($67.25 \pm 39.85 \mu\text{g m}^{-3}$) and maximum during winter ($145.8 \pm 50.1 \mu\text{g m}^{-3}$) over Delhi. The respective post-monsoon and pre-monsoon concentrations were observed to be $133.86 \pm 40.02 \mu\text{g m}^{-3}$ and $115.7 \pm 68.8 \mu\text{g m}^{-3}$ over Delhi. The $\text{PM}_{2.5}$ concentrations observed here were found to be lower than that reported in the previous study over Delhi (Tiwari *et al.*, 2013b). Tiwari *et al.* (2013b) identified that concentration of $\text{PM}_{2.5}$ is high during the post-monsoon period attributed to enhanced biomass burning over the region. However, in present study, we found higher $\text{PM}_{2.5}$ values in winter, could be due to low-level inversions leading to trapping of pollutants within the boundary layer. The lower concentrations of $\text{PM}_{2.5}$ in monsoon season may be due to rain and washout mechanisms. The enhancement in PM during post-monsoon is associated with the transport of aerosols due to crop burning in the surrounding regions of Haryana and Punjab

during the season (Tiwari *et al.*, 2013b). We further analyzed the diurnal cycle of $\text{PM}_{2.5}$ throughout the year during different seasons. The pattern showed a higher variation (in terms of their standard deviation) during pre-monsoon and post-monsoon seasons over Delhi (Fig. 4(a)). $\text{PM}_{2.5}$ concentrations showed three major peaks over Delhi. Morning peak observed at around 5–7 IST is associated with fumigating effect due to the breaking of boundary layer as explained in Safai *et al.* (2007). The peak at 10 IST could be due to large traffic during office hours and the evening peak (around 22 IST) could be associated with the nocturnal cooling and associated lowering of boundary layer, leading to trapping of pollutants (Fig. 4(a)).

The annual mean mass concentrations of $\text{PM}_{2.5}$ over Pune during the study period were observed to be $41.04 \pm 18.1 \mu\text{g m}^{-3}$ (ranging between $17.40 \mu\text{g m}^{-3}$ to $74.03 \mu\text{g m}^{-3}$ in different months). The minimum concentration was during the monsoon season ($22.06 \mu\text{g m}^{-3}$), while maximum values were observed during winter ($64.29 \mu\text{g m}^{-3}$). The seasonal variability was as low as $10 \mu\text{g m}^{-3}$ in terms of standard deviations from their daily concentrations over Pune region (it was on average $\sim 45 \mu\text{g m}^{-3}$ over Delhi). It may be noted that the PM concentrations over Delhi were 1.7–4-fold higher compared to Pune in different months. This mainly is attributed to the larger dust loading over northern parts of India during pre-monsoon season and also due to enhanced vehicular transport and lower ventilation of pollutants (Pandithurai *et al.*, 2008) compared to Pune. Diurnal variability of $\text{PM}_{2.5}$ over Pune in different months showed a peak in the morning around 9–10 IST attributed to the high traffic during the office hours. The second peak observed in the night (around 21–23 IST) could be associated with the settling of pollutants at lower levels due to inversions (Fig. 4(b)).

BC is a highly absorbing aerosol species and is a major component of $\text{PM}_{2.5}$. We also analyzed the contribution of BC to $\text{PM}_{2.5}$ concentrations over both the sites. The monthly cycle of BC from SAFAR network over both Delhi and Pune were found to be matching well with the previous

Fig. 3. Monthly variations of $\text{PM}_{2.5}$ concentrations over Delhi and Pune during 2015.

Fig. 4. Diurnal Variability of $PM_{2.5}$ concentrations over Delhi, Pune and their difference during 2015.

results (Safai *et al.*, 2007; Tiwari *et al.*, 2013a). Fig. 5 monthly variation showed the minimum concentration of $10.8 \mu\text{g m}^{-3}$ and maximum of $32.1 \mu\text{g m}^{-3}$ respectively during August and May over Delhi. However seasonal variation showed higher concentration in winter ($25.2 \mu\text{g m}^{-3}$) and minimal concentration in monsoon ($16.1 \mu\text{g m}^{-3}$) seasons. Many previous studies (Tripathi *et al.*, 2005; Tiwari *et al.*, 2013; Bibi *et al.*, 2017b, c) also reported the similar feature of higher values during the winter season over adjacent regions. BC mass concentration ranged between $1.59 \mu\text{g m}^{-3}$ (June) to $9.06 \mu\text{g m}^{-3}$ (December) over Pune. The seasonal mean BC mass concentration was high in the winter season ($7.58 \mu\text{g m}^{-3}$) and was low during monsoon season ($2.2 \mu\text{g m}^{-3}$). BC mass contribution to $PM_{2.5}$ found to be 6–20% over Pune. However, the contribution of BC to $PM_{2.5}$ was up to 30% in Delhi (Fig. 6). The study by Islam *et al.* (2014) over Bangladesh reported a 5–35% of BC concentration in $PM_{2.5}$. Liu *et al.* (2016) reported that BC to $PM_{2.5}$ ratio is 4.6% over Beijing. Marinoni *et al.* (2010) reported that contribution of BC to fine aerosol mass at Nepal was found 10.8%. Hyvarinen *et al.* (2009) showed a ~4% average BC to $PM_{2.5}$ ratio at Mukteshwar, India. While Putaud *et al.* (2004) found a contribution in the range of 5–10% BC to $PM_{2.5}$ at European sites.

Frequency Distribution of $PM_{2.5}$

Seasonal frequency distribution (FD) of hourly $PM_{2.5}$ concentrations with a bin width of $20 \mu\text{g m}^{-3}$ over Pune and Delhi is shown in Fig. 7. It showed a right side skewed distribution with different mode values over different

seasons. The peak distribution of $PM_{2.5}$ concentrations observed at Pune was within limits of National Ambient Air Quality Standards (NAAQS), i.e., $60 \mu\text{g m}^{-3}$. The mode of the $PM_{2.5}$ concentrations slightly found to cross the NAAQS in winter (62.7) over Pune. The mode of FD in pre-monsoon season was found to be 47.1 . Similarly, it was 37.5 in post-monsoon and 14.4 during monsoon seasons over Pune. FD of $PM_{2.5}$ over Delhi showed a Gaussian pattern in different seasons and the concentration levels in all the seasons were always above the NAAQS (except during the monsoon season). This illustrates the prevalence of poor quality of air during pre-monsoon, post-monsoon and winter seasons. Apart from the local uplifting of dry and loose soil by convection, the maximum concentration level in the pre-monsoon season over Delhi is mainly contributed by wind-blown dust (Trivedi *et al.*, 2014). The enhancement in PM during the post-monsoon season could be due to crop residue burning from the northern part of India. The wintertime high values of PM are attributed to low ventilation due to temperature inversions. The dominance of lower concentration levels during monsoon is due to washout effect of rain mainly on coarse-mode particles.

Direct Radiative Forcing

The major aerosol optical parameters such as AOD, SSA, and ASP were derived using OPAC model as explained in Section 2.1. Fig. 2 depicts the comparison of monthly averaged OPAC-derived AOD and SSA with AERONET observations. The OPAC-derived values of AOD and SSA found to be matching well with the observations.

Fig. 5. Monthly variations of BC concentration over Delhi and Pune for the year 2015.

Fig. 6. Monthly variations of BC to PM_{2.5} mass fraction ratio over Delhi and Pune for the year 2015.

AOD showed consistently high values over Delhi compared to Pune. SSA values showed variation from 0.89 to 0.99 over Delhi and 0.81 to 0.95 over Pune during the observational period. Many previous studies reported the radiative forcing estimates over Delhi and Pune using the in-situ measurements of aerosols over single location representing the city (e.g., Panicker *et al.*, 2010; Srivastav *et al.*, 2012; Tiwari *et al.*, 2013). However, the present study extends the estimates of PRF by using average data sets of PM_{2.5} from different environments of SAFAR network located in both the cities.

Fig. 7 depicts the PRF estimates based on the monthly values of PM_{2.5} (segregated as WS and WIS components as explained in Section 2). The PRF found to be ranging between -20.10 Wm^{-2} to -35.61 Wm^{-2} at the surface, -11.90 Wm^{-2} to -22.41 Wm^{-2} at the TOA and $+7.73 \text{ Wm}^{-2}$

to $+14.51 \text{ Wm}^{-2}$ in the atmosphere over Delhi. Monthly values of PRF found to be higher in November (-35.61 Wm^{-2} at the surface, -22.41 Wm^{-2} at TOA and $+13.23 \text{ Wm}^{-2}$ at atmosphere) over Delhi. The high aerosol loading from crop burning during post-monsoon (October–November) could be the reason for high PM values and hence high forcing during November (Bisht *et al.*, 2015). Over Pune, the PRF values found to be ranging between -14.17 Wm^{-2} to -39.11 Wm^{-2} at the surface, -8.38 Wm^{-2} to -20.91 Wm^{-2} at TOA and $+3.12 \text{ Wm}^{-2}$ to $+19.20 \text{ Wm}^{-2}$ in the atmosphere (Fig. 8). The highest atmospheric forcing over Pune was found in January (19.20 Wm^{-2}) and low during July (3.12 Wm^{-2}). The radiative forcing reported over similar environments in Pakistan (Iftikhar *et al.*, 2018) and Iran (Gharibzadeh *et al.*, 2017) showed higher forcing compared to the present study.

Fig. 7. Frequency Distribution of $PM_{2.5}$ over Delhi and Pune with different seasons and their mode values.

Fig. 8. PRF at the surface, TOA, and the atmosphere over (a) Delhi and (b) Pune along with their corresponding atmospheric heating rate.

To unravel the changes in forcing in $PM_{2.5}$ due to different chemical composition and albedo conditions, we carried out sensitivity analysis over both the sites and depicted in Table 1. The changes in albedo (vegetative and sand albedo) found to induce a change of ~3–15% in PRF at the surface and ~6–26% at TOA over Delhi. However, the change in PRF due to albedo sensitivity was ~11–23% at the surface and ~20–48% at TOA over Pune. It is also observed that the changes in forcing to sand and vegetation albedo showed high variability over Pune as compared to Delhi. A 10% change in WS mass of $PM_{2.5}$ found to induce a change of 10% in AOD over Delhi and Pune. Further, the

same change (change of 10% in WS component) found to induce a change of ~5–7% at the surface and ~6–9% at TOA in PRF over Delhi. The same change however induced ~1–7% at the surface and ~5–9% at TOA in PRF over Pune (Table 2). The above sensitivity experiments illustrate that the impact of hygroscopicity on PRF is less dominant compared to changes in albedo.

The aerosol heating rates also were estimated over Delhi and Pune as explained in Section 2. Atmospheric heating rates over Delhi ranged between 0.47 to 0.92 K day⁻¹, whereas over Pune it ranged between 0.17 to 0.75 K day⁻¹ during the observational period.

Table 1. Sensitivity analysis by varying the WS and WIS mass concentration by 10%.

Station	Month	Original - AOD	Change in AOD by decreased 10% (WS)	Change in AOD by increased 10% (WS)	Original			Forcing due to 10% decrease in WS.			Forcing due to 10% increase in WS		
					PM_FORCING								
					SUF	TOA	TOA	SUF	TOA	TOA	SUF	TOA	TOA
Delhi	Mar	0.50	0.45	0.55	-20.09	-11.90	-18.58	-10.86	-21.56	-12.95	-38.21	-24.33	-12.95
	Nov	1.04	0.96	1.16	-35.61	-22.41	-33.90	-21.00	-38.21	-24.33	-19.82	-12.64	-12.64
Pune	Mar	0.40	0.36	0.43	-18.52	-11.60	-17.21	-10.54	-28.69	-16.88	-27.87	-15.32	-15.32
	Nov	0.48	0.46	0.49	-28.28	-16.12	-27.87	-15.32	-28.69	-16.88	-27.87	-15.32	-15.32

Table 2. Sensitivity analysis by varying the Albedo parameters with sand, vegetation, and MODIS BRDF.

Station	Month	Original forcing (with MODIS BRDF ALBEDO)			SBDART_MODEL SAND ALBEDO			SBDART_MODEL VEGETATIVE ALBEDO			SBDART_MODEL 50% VEGE AND 50% SAND ALBEDO			SBDART_MODEL 70% VEGE AND 70% SAND ALBEDO			SBDART_MODEL 30% VEGE AND 70% SAND ALBEDO		
		SUF	TOA	TOA	SUF	TOA	TOA	SUF	TOA	TOA	SUF	TOA	TOA	SUF	TOA	TOA	SUF	TOA	TOA
		SUF	TOA	TOA	SUF	TOA	TOA	SUF	TOA	TOA	SUF	TOA	TOA	SUF	TOA	TOA	SUF	TOA	TOA
Delhi	Mar	-20.10	-11.90	-19.49	-11.23	-17.96	-9.34	-18.65	-10.22	-18.35	-9.85	-18.97	-10.61	-32.15	-18.59	-18.59	-32.15	-18.59	-18.59
	Nov	-35.61	-22.41	-32.99	-19.55	-30.42	-16.53	-31.62	-17.97	-31.12	-17.38	-32.15	-18.59	-32.15	-18.59	-18.59	-32.15	-18.59	-18.59
Pune	Mar	-18.53	-11.61	-16.47	-9.31	-15.17	-7.68	-15.74	-8.42	-15.49	-8.11	-16.02	-8.76	-16.02	-8.76	-8.76	-16.02	-8.76	-8.76
	Nov	-28.28	-16.13	-24.19	-11.45	-21.68	-8.35	-22.85	-9.82	-22.36	-9.22	-23.37	-10.46	-23.37	-10.46	-10.46	-23.37	-10.46	-10.46

SUMMARY

1. Radiative forcing by PM_{2.5} (PRF) was estimated over two Indian metro cities (Delhi and Pune) using the in-situ measurements of PM_{2.5} from the SAFAR network.
2. The results show that the monthly variations of PM_{2.5} (and BC) were consistently high over Delhi as compared to Pune. This difference may be due to the long-range transport of crop residue, lower ventilation of pollutants because of temperature inversions, and enhanced anthropogenic activities as well as higher dust loading over Delhi than Pune.
3. The PRF was estimated at the surface, in the atmosphere, and at the TOA over both the regions using the OPAC model in conjunction with the SBDART model.
4. The estimates from the SBDART model simulations indicated that the PRF ranged from -20.09 Wm^{-2} to -35.61 Wm^{-2} at the surface and from -11.90 Wm^{-2} to -22.41 Wm^{-2} at the TOA, inducing an atmospheric forcing of $7.732\text{--}14.51 \text{ Wm}^{-2}$ over Delhi.
5. The PRF over Pune ranged from -14.17 Wm^{-2} to -39.11 Wm^{-2} at the surface and from -8.38 Wm^{-2} to -20.91 Wm^{-2} at the TOA. The aerosol atmospheric absorption over Pune ranged from $+3.12 \text{ Wm}^{-2}$ to $+19.20 \text{ Wm}^{-2}$ in different seasons.
6. The sensitivity of the albedo (for vegetation and sand) induced a change of $\sim 3\text{--}15\%$ at the surface and $\sim 6\text{--}26\%$ at the TOA in the PRF over Delhi. The same changes induced a greater change in the PRF ($\sim 11\text{--}23\%$ at the surface and $\sim 20\text{--}48\%$ at the TOA) over Pune.
7. A change of 10% in the WS component of the PM_{2.5} induced a change of $\sim 5\text{--}7\%$ at the surface and $\sim 6\text{--}9\%$ at the TOA in the PRF over Delhi. The change in the PRF over Pune due to changes in hygroscopicity (changes in WS content) ranged $\sim 1\text{--}7\%$ at the surface and $\sim 5\text{--}9\%$ at the TOA. Hence, we conclude that the impact of the increase in the hygroscopicity of the aerosols on the PRF was overshadowed by the net changes in albedo.
8. Atmospheric heating rates over Delhi were in the range of $0.47\text{--}0.92 \text{ K day}^{-1}$ in different months, whereas the variation over Pune was $0.17\text{--}0.75 \text{ K day}^{-1}$ during the observational period.

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