Importance of aerosol non-sphericity in estimating aerosol radiative forcing in Indo-Gangetic Basin

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HIGHLIGHTS

• Two years of aerosol composition at Delhi located in the Indo-Gangetic Basin were analyzed.
• Dust optical properties calculated for three non-spherical shapes - spheroid, cylinder and chebyshev.
• Non-spherical dust enhances at mid-visible SSA by 0.04.
• Treatment of dust as ‘spherical’ biases TOA forcing to warming at Delhi.
• Non-sphericity needs to be accounted, but choice of shape may not be important.

Abstract

Aerosols are usually presumed spherical in shape while estimating the direct radiative forcing (DRF) using observations or in the models. In the Indo-Gangetic Basin (IGB), a regional aerosol hotspot where dust is a major aerosol species and has been observed to be non-spherical in shape, it is important to test the validity of this assumption. We address this issue using measured chemical composition at megacity Delhi, a representative site of the western IGB. Based on the observation, we choose three non-spherical shapes - spheroid, cylinder and chebyshev, and compute their optical properties. Non-spherical dust enhances aerosol extinction coefficient (βext) and single scattering albedo (SSA) at visible wavelengths by ~0.05 km−1 and ~0.04 respectively, while it decreases asymmetry parameter (g) by ~0.1. Accounting non-sphericity leads top-of-the-atmosphere (TOA) dust DRF to more cooling due to enhanced backscattering and increases surface dimming due to enhanced βext.

Outgoing shortwave flux at TOA increases by up to 3.3% for composite aerosols with non-spherical dust externally mixed with other spherical species. Our results show that while non-sphericity needs to be accounted for, choice of shape may not be important in estimating aerosol DRF in the IGB.

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Keywords:
Aerosols
Non-spherical particles
Indo Gangetic Basin
Aerosol direct radiative forcing

1. Introduction

Aerosols perturb the Earth’s radiation budget directly by scattering and/or absorbing solar radiation (Haywood and Boucher, 2000) and indirectly by altering cloud characteristics (Twomey, 1974, 1977; Albrecht, 1989; Lohmann and Feichter, 2005). Quantification of aerosol induced change in radiation budget, represented as direct radiative forcing (DRF) (Chung et al., 2005) has improved at global scale due to multinational efforts in form of coordinated ground-based, aircraft and shipborne field campaigns (Singh, 2005; Dumka et al., 2014; More, 2013), spread of surface observational network, advent of new generation
sensors providing global coverage of aerosol parameters over the last few decades. Yet, uncertainty in aerosol DRF continues to be large, especially at regional scale, due to lack of information about aerosol composition, vertical distribution, mixing state and particle shape at required space and time scale (Boucher et al., 2013). The problem is critical in the Indian monsoon region characterized by large aerosol loading (Dey and Di Girolamo, 2010) that is increasing in recent years (Dey and Di Girolamo, 2011). Several studies (e.g. Chung and Ramanathan, 2006; Lau and Kim, 2006 and many more later on) have shown significant changes in atmospheric circulation and precipitation in response to total aerosol DRF in this region, implying that our interpretation of aerosol-climate interaction depends on the accuracy of aerosol DRF.

Most of the climate models underestimate aerosol optical depth (AOD) in the Indian monsoon region (Sanap et al., 2015). Therefore, estimation of aerosol DRF, using information obtained from aerosol parameters measured directly, needs to be accurate to understand the fidelity of the models. Despite commendable effort by Indian Space Research Organization in developing and maintaining a network of aerosol observations (Moorthy et al., 2013) and efforts from other organizations (e.g. AERONET under NASA, Holben et al., 1998) and individuals (as summarized in the ‘Introduction’ section of Dey and Di Girolamo, 2010), continuous direct measurements are mostly limited to columnar AOD spectrum and black carbon (BC) mass concentration. In absence of direct information, aerosol composition is typically inferred by iteratively tuning the number concentrations of individual species to match AOD spectrum simulated by Mie theory (therefore assuming spherical particle) with measured spectrum constrained by other measurements (viz. BC mass, spectral SSA etc.) (Satheesh and Srinivasan, 2006; Dey and Tripathi, 2008). Aerosol optical properties derived by this approach are then used to estimate aerosol DRF. Other key assumptions frequently involved are mixing state and representative vertical distribution. Each of these factors contributes to uncertainty in estimated aerosol DRF. Previously, we examined the sensitivity of aerosol properties to choice of mixing state (Dey et al., 2008) and sensitivity of dust optical properties to non-sphericity (Mishra et al., 2008) using AERONET-retrieved aerosol spectral optical properties at Kanpur, an urban site in the polluted Indo-Gangetic Basin (IGB) in northern India. In absence of direct chemical measurements, aerosol composition was inferred. Hence separating uncertainty in DRF due to particle shape from uncertainty due to aerosol composition was difficult. Moreover, it is not tested whether any change in optical properties of non-spherical dust (relative to spherical dust) will reflect in optical properties of composite aerosols (where non-spherical dust is a major component) in the IGB, because of lack of robust chemical composition data. Therefore, it is important to understand the implication of particle shape on composite optical properties using external mixing state (which is most common in the climate models and aerosol retrievals) from measured chemical data from the IGB where dust is a major aerosol species (Dey and Di Girolamo, 2010).

We consider three most common non-spherical shapes - spheroid, cylinder and chelyshev, as identified in the analysis of SEM (scanning electron microscope) images of aerosols containing dust in India (Negi et al., 1996). Similar shapes were observed in the IGB (Fig. 8 of Kumar et al., 2014). First, deviations of spectral extinction coefficient ($f_{tot}$), single scattering albedo (SSA) and asymmetry parameter ($g$) relative to spherical particle are estimated using T-matrix code (Mishchenko and Travis, 1998). T-matrix code only deals with the accumulation mode dust. Then aerosol DRF for external mixing was estimated by replacing optical properties of spherical dust by those of volume equivalent non-spherical particles in the composite aerosol mixture. Implications of such modulations of aerosol DRF are discussed. Along with T-matrix code, we have used two versions of database OPAC (Optical Properties of Aerosols and Clouds) in this study. OPAC (Hess and Schult, 1998) has been used to estimate optical properties of spherical dust (both coarse and accumulation). We note that in OPAC 4.0, only spheroid dust properties were added, while those of other spherical species remain unchanged.

2. Aerosol data and methodology

We analyze aerosol composition data for the period 2007–2008. Aerosol samples were collected at megacity Delhi using a PM$_{10}$ sampler and concentrations of various elements and ions were measured using ED-XRF and ICP-MS (Perrino et al., 2011). BC was measured directly by an Aethalometer (model AE-21, Magee Scientific, USA) using mass flow rate of 2 L min$^{-1}$ at 5 min time interval. First, mass of dust is quantified using measured ‘$A$’ mass and its proportion in the Earth’s crust following Tare et al. (2006). Total dust is divided into ‘accumulation’ (Dust$_{acc}$ in Fig. 1) and ‘coarse’ (Dust$_{c}$) mode in a way that the ratios of optical depths of these two modes match with optical depths of medium (equivalent to accumulation mode) and large (equivalent to coarse mode) particles in MISR-retrieved optical depths. MISR assumes dust to be present in these two modes in its aerosol retrieval algorithm (Kahn et al., 2010). Next, mass of insoluble species is determined by subtracting the mass of ions from their elements counterparts. Remaining mass (difference between PM$_{10}$ mass and sum of masses of all the ions, insoluble species, dust and BC) is attributed to organic carbon (OC). 35% of OC is water-soluble (Ram and Sarin, 2010), therefore total mass of water-soluble components is estimated by summing total ions and 35% of OC mass; while that of insoluble components is determined by summing insoluble mass and remaining 65% of OC mass.

Monthly variation of mass fractions of individual aerosol species at Delhi is shown in Fig. 1. Dust is ubiquitous throughout the year with higher mass fraction (>20%) during April to September. Even in the winter months (December–February), dust mass fraction varies within 9–17% in Delhi, which is unusual in terms of typical urban setting across the world. High dust load in Delhi was reported earlier by many researchers (Singh et al., 2010; Srivastava et al., 2011, 2014), where the source was attributed primarily to long range transport from the arid regions in the west during the pre-monsoon and monsoon months in addition to local sources (re-suspension of road dust and construction activities) prevailing throughout the year. Large (>10%) BC fraction from November to February can be attributed to shallow boundary layer that does not allow aerosols to disperse easily. Mass fractions of water-soluble and insoluble species are more or less uniform throughout the year.

Three distinct shapes of dust particles - spheroid, cylinder and chelyshev, were identified from the SEM image of aerosols collected at Delhi (Kumar et al., 2014) by comparing with the SEM image in Negi et al. (1996). We model spheroid and cylinder shaped particles as both ‘oblate’ and ‘prolate’ and chelyshev shaped particles as function of various degree of deformation. Based on literature (e.g. Mishra et al., 2012 and Mishra et al., 2015), we fix the ranges of aspect ratio of

![Fig. 1. Monthly variation of mass fractions (in %) of individual aerosol species at Delhi.](image-url)
spheroid and cylinder shapes to be in the range 1/3 to 3. Chebyshev particles are modeled as:

\[ r(\theta, \phi) = r_0 [1 + \varepsilon T_n(\cos\theta)] \]

where, \( n \) is polynomial and \( \varepsilon \) is degree of deformation. We calculate optical properties for deformation of \(-0.2\) and \(0.2\), and \(-0.15\) and \(0.15\) respectively for 2nd and 4th order polynomial. In T-matrix framework, only accumulation mode dust is considered to be non-spherical because T-matrix code cannot converge for coarse particles (Mishchenko and Travis, 1998). To account for non-sphericity for coarse-mode dust, we utilize OPAC 4.0 (Koepke and Hess, 2015), where only spheroid shape is considered due to the limitation of the code.

Refractive index (RI) of dust used in the calculation is taken from Mishra et al. (2015). We calculate scattering and absorption cross-section and \( g \) of non-spherical dust for the shortwave region (0.25 to 4 \( \mu \)m). SSA is simply the ratio of scattering and extinction (sum of scattering and absorption) efficiency, which are determined as extinction cross-section per unit surface area of the respective shape. Extinction and scattering cross-sections are then converted to \( \beta_{ext/sca} \) using the number size distribution derived from the measured mass for these non-spherical shapes.

\[ \beta_{ext/sca} = N \times C_{ext/sca} \]

where \( N \) is the number concentration of non-spherical dust particles per unit volume of air and \( C \) is the cross-section of particle. We consider the log-normal size distribution parameters (mode radius = 0.39 \( \mu \)m and \( \sigma = 2.0 \)) for accumulation mode dust (Hess and Schult, 1998). Our calculations from T-matrix (Fig. 2) show that extinction and scattering cross section and SSA for a specific shape are not significantly different for various aspect ratios and degree of deformation. Only asymmetry parameter reduces in visible wavelength for smaller aspect ratios in oblate and prolate ‘spheroid’ and ‘cylinder’. Moreover, the differences in SSA and extinction cross-sections are negligible across the individual shapes. Integration over the entire shortwave region results in negligible differences in optical properties across the three shapes relative to the properties for spherical dust. Since we do not have the information about the relative proportions of these three individual non-spherical shapes and it is not possible to obtain such information unless a large number of samples is examined under scanning electron microscope, we calculate the optical properties for non-spherical dusts assuming equal proportions of these three shapes.

We finally compute the spectral optical properties for the composite aerosols in an external mixing as follows:

\[ \beta_{ext/sca} = \sum_{i=1}^{5} \beta_{ext/sca,i} \]

\[ SSA = \frac{\sum_{i=1}^{5} \beta_{ext,i} \times SSA_i}{\sum_{i=1}^{5} \beta_{ext,i}} \]

\[ g = \frac{\sum_{i=1}^{5} \beta_{sca,i} \times g_i}{\sum_{i=1}^{5} \beta_{sca,i}} \]

In the above Eqs. (3)–(5), \( i \) refer to individual aerosol species (five of them i.e., BC, WS, WINS, coarse and accumulation dust), where optical

Fig. 2. Spectral optical properties for (top panel) spheroid shaped dust, (middle panel) cylinder shaped dust and (bottom panel) chebyshev shaped dust particles for various ranges of aspect ratios and degree of deformation as calculated by T-matrix.
properties of volume equivalent non-spherical shapes replace optical properties of those spherical accumulation dust particles. We also compute optical properties of externally mixed aerosols using the same composition assuming entire dust to be spherical. Aerosol scale height and thickness of aerosol layer are considered from vertical profiles of aerosols extracted from CALIOP over Delhi from our earlier study (Srivastava et al., 2014). These spectral optical properties are used as input in SBDART radiative transfer code (Ricchiazzi et al., 1998) along with other non-aerosol inputs such as surface albedo and ozone concentration from OMI and water vapor from MODIS. Net fluxes at the top-of-the-atmosphere (TOA) and surface with and without aerosols are computed to estimate TOA and surface aerosol DRF (similar to the previous studies reported, e.g. Dey and Tripathi, 2008; Singh et al., 2010). In case of OPAC 4.0, $\beta_{\text{ext}}$, SSA and $g$ are calculated for the spheroid dust in both accumulation and coarse mode. The entire methodology starting from chemical speciation followed by estimation of dust optical properties using T-matrix code (accumulation mode non-spherical dust) and OPAC 4.0 (for total non-spherical dust) to simulation of DRF using SBDART is summarized in a flow chart (Fig. 3).

3. Results and discussion

Fig. 4 shows the comparison between spectral $\beta_{\text{ext}}$, SSA and $g$ for non-spherical (average over three shapes) and spherical accumulation mode dust in the shortwave. Dust $\beta_{\text{ext}}$ is observed to be highest during the months with large dust mass fraction. When dust particle is considered to be non-spherical, $\beta_{\text{ext}}$ is enhanced by $>0.05$ km$^{-1}$ at visible and NIR wavelengths. $\beta_{\text{ext}}$ depends on extinction cross-section. Therefore, enhancement in $\beta_{\text{ext}}$ can be attributed to either a larger scattering cross-section for non-spherical dust or change in extinction efficiency or both. The monthly variation in enhancement of $\beta_{\text{ext}}$ depends on the mass fractions (Fig. 1). Unlike $\beta_{\text{ext}}$, changes in other two important aerosol optical properties do not exhibit monthly variation, because they are not directly dependent on mass. Relative to spherical dust, SSA of non-spherical dust increases in the shortwave, while $g$ (asymmetry parameter) reduces. In another study, Koepke and Hess (2015) reported 4% to 6% change in the optical properties of spherical to non-spherical “desert aerosol” in the shortwave. However, we point out that in Koepke and Hess (2015), desert aerosol was composed of dust and water-soluble particles. In order to examine, the change in spectral optical properties total (accumulation and coarse) spherical to total (accumulation and coarse) non-spherical using the same OPAC 4.0 model for our case, we use total dust as spheroid and calculate the mean.

Fig. 3. Flowchart explaining the method from aerosol speciation to radiative forcing estimation.

Fig. 4. Comparison between (top panel) spectral $\beta_{\text{ext}}$ (in km$^{-1}$) in each month and (bottom panel) spectral SSA and $g$ (both unit-less) for the non-spherical (T-matrix) (averaged over the three shapes) and spherical dust for accumulation mode.
monthly difference in SSA and $g$ and report the values at 0.5 μm wavelength (Fig. 5). Qualitatively, we get similar changes (i.e. increase in $C_{ext}$ and SSA and decrease in $g$) for non-spherical shape, but the magnitudes are smaller. Dust optical properties in OPAC 4.0 are calculated using T-matrix for accumulation mode and geometric optics for coarse mode. The results imply that the non-spherical dust particles scatter more shortwave radiation than by spherical counterparts, especially for moderately absorbing dust (Mishra et al., 2015). Also, the scattering is enhanced in backward direction as evident from a decrease in $g$.

Since we have used T-matrix and OPAC 4.0 to study non-spherical dust, we also compare optical properties at mid-visible wavelength of accumulation spheroid dust from both the codes (Fig. 6) to examine the robustness of the two methods. We observe that spherical dust is more absorbing (lower SSA) and slightly more forward scattering in OPAC 4.0 relative to T-matrix. Extinction cross-section ($C_{ext}$) of spherical dust is higher by 7% in OPAC 4.0 than in T-matrix, mainly because of a larger absorption cross-section as the scattering cross-section ($C_{ext} \times$ SSA) are pretty close (within 3%) in both the codes.

These changes in spectral optical properties are clearly reflected in the changes in estimated dust DRF (Fig. 7). In this figure dust DRF from T-matrix (non-spherical accumulation mode dust) and OPAC (spherical accumulation mode dust) has been compared. Mean seasonal dust DRF at TOA assuming spherical shape are $-0.13 \pm 0.10, 0.02 \pm 0.05, -0.23 \pm 0.10$ and $-0.08 \pm 0.03$ W m$^{-2}$ respectively for the pre-monsoon, monsoon, post-monsoon and winter seasons. When the dust is considered non-spherical in shape, mean ($\pm 1\sigma$) seasonal dust DRF at TOA switches to cooling with respective values for the four seasons as $-1.72 \pm 0.41, -0.63 \pm 0.32, -2.27 \pm 0.74$ and $-0.88 \pm 0.26$ W m$^{-2}$ (using T-matrix). Mean surface albedo at Delhi NCR considered in this study are 0.20, 0.33, 0.17 and 0.20 for the pre-monsoon, monsoon post-monsoon and winter seasons respectively. We note that surface albedo and all other non-aerosol parameters (e.g. ozone and water vapor) remains same in our estimation of DRF for non-spherical dust. Therefore, the changes in dust DRF can only be attributed to changes in optical properties due to change in particle shape. Larger scattering (due to increase in SSA and $C_{ext}$) and backscattering (due to decrease in $g$) combine to result in more shortwave radiation reflected back to TOA, leading to an enhanced cooling at TOA. Mean ($\pm 1\sigma$) seasonal surface dust DRF of $-0.68 \pm 0.14, -0.34 \pm 0.08, -0.82 \pm 0.23$ and $-0.33 \pm 0.09$ W m$^{-2}$ respectively for the pre-monsoon, monsoon, post-monsoon and winter seasons for spherical shape changes to $-2.98 \pm 0.61, -1.45 \pm 0.39, -3.65 \pm 1.05$ and $-1.47 \pm 0.42$ W m$^{-2}$. Larger surface dimming in case of non-spherical shapes can be attributed to larger attenuation of solar radiation (as evidenced from enhancement in $C_{ext}$) in the atmosphere. Net result is negligible change in atmospheric heating (difference between TOA and surface DRF), because switch to TOA cooling is balanced by increase in surface dimming. Larger dust TOA (more cooling) and surface DRF for both spherical and non-spherical dust in the post-monsoon season relative to the winter season can be attributed to larger dust fraction (Fig. 1), because surface albedo are same. Though highest surface albedo is observed in the monsoon season, magnitudes of the DRF are not high. Largest DRF in the pre-monsoon season can be attributed to largest dust fraction (Fig. 1). Our dust DRF (mean annual value of $1.33 \pm 0.7$ W m$^{-2}$) is larger than the values reported in IPCC Fifth Assessment Report ($-0.14 \pm 0.11$ W m$^{-2}$). This may be attributed to smaller global average dust AOD relative to dust AOD over Delhi NCR, because larger dust DRF was reported where dust AOD is much higher (e.g. Liao and Seinfeld, 1998). Also, dust RI used here and in other studies may differ resulting to the difference observed in DRF.

To understand how much these changes would translate into composite aerosol DRF, we replace the optical properties of spherical accumulation mode dust by the non-spherical counterparts in an external mixing mode. Left panel of (Fig. 8) shows the mean monthly difference in aerosol DRF at TOA and surface between composite aerosol with non-spherical and spherical dust. As explained earlier for only dust DRF, the enhanced TOA cooling is apparent in even composite aerosol DRF when dust particles are considered non-spherical. The cooling varies in the range 0.45 to 1.8 W m$^{-2}$. Similarly, the surface dimming (larger negative aerosol surface DRF for consideration of non-sphericity) enhances in the range 0.2 to 2.4 W m$^{-2}$. Comparison of changes in dust DRF with the changes in composite aerosol DRF due to particle non-sphericity reveals that the magnitude of TOA cooling enhancement is suppressed when non-spherical dust is externally mixed with other spherical particles by 5–50%. On the contrary, surface DRF does not show much change even when non-spherical dust is externally mixed with other spherical species.

These results have important climatic implications. Outgoing shortwave flux at TOA increases by 0.57 to 3.3% (right panel of Fig. 8), when non-spherical dust is included in external mixing. This implies that in case non-spherical particles are present in the atmosphere and are not considered in the look-up table of the retrieval algorithm, angular scattering may change relative to reality impacting aerosol
4. Uncertainty analysis

The estimated aerosol optical properties and the resulting radiative forcing have uncertainties that demand detailed attention to fully comprehend the significance of the results presented so far. Two most important factors which may contribute to uncertainty in the estimated aerosol optical properties and hence the forcing are RI of dust and its size distribution. For our work, we have considered RI to be 1.545 + 0.0055i, where 1.545 is the real and 0.0055 is the imaginary part of the RI. We have considered a possible range of RI (1.56 ± 0.03) + (6.5 ± 4.6)i for dust in the IGB (Agnihotri et al., 2015) and estimated the possible uncertainty range in the estimated aerosol radiative forcing. Our calculations show that SSA and g can increase by 3.8% and 0.2% respectively if the dust RI decreases to its lowest possible value and SSA and g can decrease by 6.8% and 5.0% respectively if dust RI increases to its highest possible value. These changes in optical properties translate to 0.2 W m⁻² increase in TOA forcing and 0.4 W m⁻² decrease in surface forcing for the lowest possible value of dust RI. Similarly for the upper range of dust RI, TOA forcing decreases by 0.3 W m⁻² whereas surface forcing increases by 0.6 W m⁻². To summarize, the error in dust RI may lead to an uncertainty in TOA and surface radiative forcing by ±0.2–0.3 and ±0.4–0.6 W m⁻² respectively. Similarly to account for the uncertainty due to size distribution, we consider two parameters - \( R_{\text{mod}} \) and \( \sigma \) (for a log-normal size distribution). In our study, we used size distribution parameters from OPAC database (Hess and Schult, 1998). Only information available from the Indo-Gangetic Basin on direct measurement of dust size distribution is from Kanpur (500 km southeast of Delhi) during winter of 2004–2005 (Dey and Tripathi, 2007), which showed a 15.7% and 19% deviation from OPAC database. The magnitude of deviation of size distribution parameters in other seasons and elsewhere including Delhi is not known. Our sensitivity study for ±20% variation would provide a best possible uncertainty estimate. Sensitivity study shows that a decrease of \( R_{\text{mod}} \) and \( \sigma \) by 20% translates to an increase of SSA by 1.9% and \( g \) by 3.7% at 0.5 \( \mu \)m wavelength. This results in a decrease of TOA forcing by 0.2 W m⁻² and surface forcing by 0.6 W m⁻². On increasing size distribution parameters by 20%, SSA and \( g \) decrease by 0.2% and 6.2% respectively; which translates to an increase of TOA and surface warming by 0.4 W m⁻² and 0.5 W m⁻² respectively. It implies that a decrease in size distribution parameters lead to more cooling at TOA and surface; while the increase would cause more warming. We note that additional uncertainty may arise due to any uncertainty in optical database of non-spherical shapes used in OPAC 4.0 and T-matrix, which is beyond the scope for this work. Overall, our results demonstrate the importance of consideration of non-spherical shapes of dust particles during computation of aerosol radiative forcing in the IGB, where dust is a dominant aerosol species.

5. Summary and conclusions

Several recent studies (Vinoj et al., 2014; Das et al., 2015) have pointed out a strong dynamic impact of dust DRF on the Indian summer monsoon circulation. Our results emphasize on improving the treatment of particle shape in estimation of aerosol DRF, especially when
climate change.


