

Abstract

The radiative forcing estimation of the polluted mineral dust is limited due to lack of morphological analysis, mixing state with the carbonaceous components and the hematite content in the pure dust. The accumulation mode mineral dust has been found to mix with anthropogenically produced black carbon, organic carbon and brown carbon during long range transport. The above features of the polluted dust are not well accounted in the optical models and lead the uncertainty in the numerical estimation of their radiative impact. The Semi-external mixing being a prominent mixing of dust and carbonaceous components has not been studied in details so far compared to core-shell, internal and external mixing studies. In present study, we consider the pure mineral dust composed of non-metallic components (such as Quartz, Feldspar, Mica and Calcite) and metallic component like hematite (Fe_2O_3). The hematite percentage in the pure mineral dust governs its absorbance. Based on this hematite variation, the hematite fraction in pure mineral dust has been constrained between 0–8%. The morphological and mineralogical characterization of the polluted dust led to consider the three sphere, two sphere and two spheroid model shapes for polluted dust particle system. The pollution gives rise to various light absorbing aerosol components like black carbon, brown carbon and organic carbon (comprising of HUmic-Like Substances, HULIS) in the atmosphere. The entire above discussed model shapes have been considered for the mineral dust getting polluted with (1) organic carbon (especially HULIS component) (2) Brown carbon and (3) black carbon by making a semi-external mixture with pure mineral dust. The optical properties (like Single Scattering Albedo, SSA; Asymmetry parameter, g and Extinction efficiency, Q_{ext}) of above model shapes for the polluted dust have been computed using Discrete Dipole Approximation, DDA code. For above model shapes, the SSA was found to vary depending on hematite content (0–8%) and model shape composition. For the two sphere BC-mineral dust cluster, hematite was found to be dominating absorber compared to that of black carbon as the $R_{\text{BC}}/R_{\text{dust}}$ decreases. (i.e. with increase of dust sphere size compared to black carbon

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sphere in the composite 2-sphere cluster). SSA was found to be very sensitivity for the hematite content when both of the spheres (i.e. mineral dust and BC) are nearly of same size. The two spheroid system composed of organic carbon and dust with 0% hematite (OCD¹-0) showed the maximum deviation of SSA (i.e. ~5%) compared to the two sphere system of same composition and hematite content (OCD-0). Increase in hematite from 0 to 8% caused maximum SSA deviation of ~20% for two sphere organic carbon-dust system (OCD) while the same has been observed to be ~18% for two spheroid organic carbon-dust system (OCD¹). SSA was found to be more sensitive to hematite content than that of particle shape. Compared to SSA, Asymmetry parameter, g was found to be more sensitive towards particle shape. For three-sphere model shapes with 0% hematite composed of black carbon-dust-dust (BCDD-0), brown carbon-dust-dust (BrCDD-0) and organic carbon-dust-dust (OCDD-0), the deviation of SSA and g relative to conjugate black carbon (BC), brown carbon (BrC) and organic carbon (OC) spheres are ~68% and ~31%, ~83% and ~31% and ~70% and ~33%, respectively. Thus modeled polluted dust optics will provide a better basis for radiative forcing estimation and many sensitivity studies.

1 Introduction

The main sources for the mineral dust are the deserts which inject mineral dust in the atmosphere (Goudie et al., 2003; Warren et al., 2007; Schepanski, et al., 2009; Houghton et al., 2001; Xuana et al., 2004; Laurent et al., 2008). Saharan desert itself is solely responsible for half of this amount at global scale. It not only influences the aerosol loading to Africa, the Atlantic Ocean, South America, and the East coast of USA but also to that of Europe. However, the radiative effect of mineral dust is not well quantified (Sokolik and Toon, 1999; Durant et al., 2009; Ginoux et al., 2001; Zender et al., 2003; Kalashnikova et al., 2004; Darmenova et al., 2009; McConnell et al., 2010) and the global radiative forcing due to dust is expected to be negative (Diaz et al., 2001; Myhre et al., 2003; Mahowald and Kiehl, 2003). The mineral dust is transported to long

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distances (Chiapello et al., 1997; Afeti and Resch, 2000; Moulin et al., 1997; Chin et al., 2007; Flaounas et al., 2009) where it gets polluted by mixing with carbonaceous components (forming a polluted dust system) (Jaffe et al., 2003; Maria et al., 2004; Lelieveld et al., 2002). The sources of carbonaceous part are very diverse, which include biomass burning, industrial and vehicular emissions. Thus formed mixture influences the aerosol size distribution, particle shape, hygroscopicity, cloud condensation nuclei and refractive index. The above characteristics of the complex particles govern the optical properties of such particle system whereas the magnitude and sign of the aerosol direct effect is determined by the optical properties of individual aerosols (Lesins et al., 2002).

The radiative effect of dust systems is evaluated by feeding their physical and chemical properties in the radiative models (Tegen et al., 1996; Liao and Seinfeld, 1998; Balkanski et al., 2007; Darmenova et al., 2009; McConnell et al., 2010; Satheesh and Ramanathan, 2000). The sources for the uncertainties in the radiative forcing estimation include mineral aerosol shape (Volten et al., 2005; Zhao et al., 2003; Wang et al., 2003; Kalashnikova and Sokolik, 2002; Mishra et al., 2008), proportion of hematite content in mineral dust (Mishra and Tripathi, 2008) and their mixing states with carbonaceous components (Chandra et al., 2004; Jacobson, 2001). The radiative forcing imposed by the aerosols in the atmosphere can be computed by summing over the entire aerosol population. The assumptions are made on the proportions how the different aerosol components are mixed together. The optical properties of polluted aerosols are numerically estimated with the assumptions that either the particles are mixed externally (where various aerosol species exists independently) (e.g., World Meteorological Organization, 1986; Deepak and Gerbers, 1983; Tegen et al., 1997; Lohmann et al., 1999; Stier et al., 2005; Lesins et al., 2002) or internally mixed (one or several small aerosol particles are imbedded in a larger host particle) (Shinozuka et al., 2009; Ackerman and Toon, 1981; Ch'ylek et al., 1995; Jacobson, 2000; Stier et al., 2006). Core-shell structure of the particle has also been considered in many studies (Moteki et al., 2007; Shiraiwa et al., 2007; Schnaiter et al., 2005; Bond et al., 2006). The real mixed

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state can be expected to lie somewhere in between the external and internal mixing extremes. The semi-external mixing (two or more aerosol particles are in physical contact and form an aggregate) is such a kind of mixing observed with the individual particle morphological analysis with Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), High Resolution Transmission Electron Microscopy (HRTEM) and X-ray analysis (Zongbo et al., 2002; Li et al., 2003; Clark et al., 2004; Alexander et al., 2008; Takahama et al., 2010). However, the semi-external mixing is rarely modeled (Mishchenko et al., 2004). As the climatic impact can be estimated based on the radiative forcing estimation so better accounting of the existing mixing state in the optical model will lead to better forcing estimation and hence the better prediction of climatic impacts using General Circulation Model, GCM. In general, the assumption for mixing in GCM is the external mixing (Tegen et al., 1997; Lohmann et al., 1999; Koch et al., 2006; Adams et al., 2001; Shinozuka et al., 2009; Penner et al., 2002; Pierce et al., 2007) which leads to erroneous predictions. Some GCM models also account for internal mixing and core-shell mixing (Menon et al., 2009). Among the carbonaceous components, black carbon (BC), brown carbon (BrC), and the organic carbon (OC) are thought to be major components which mix with the accumulation mode (0.1–1 μm radius) mineral dust during transport over the polluting zone. Among the carbonaceous components, BC is highest absorbing while OC is least and BrC lies in between.

The black carbon particles warrant special attention because of their complex role in climate (Menon et al., 2002; Koch and Genio, 2010; Hill and Dobbie, 2008; Garrett and Zhao, 2006; Ackerman et al., 2000; Penner et al., 1992), long-range transport (Ramamanthan et al., 2001; Rosen et al., 1981; Subramanian et al., 2010), and large surface area that may facilitate heterogeneous reactions (Chughtai et al., 2002). Black carbon (BC) aerosols are different to that of other atmospheric aerosols in terms of their absorbance, heating the air, and contributing to global warming (Hansen et al., 2000; Jacobson, 2001; Menon et al., 2002). Generally the BC aerosols are generated in the atmosphere due to incomplete combustion of fuel oil, coal, and biomass (Ramanathan and Carmichael, 2008; Zhang et al., 2009; Andreae and Merlet, 2001;

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Griffin and Goldberg, 1979). The BC particles are generally very hydrophobic but it is converted as hydrophilic because of physical and/or chemical changes occurring during atmospheric aging process (Weingartner et al., 1997; Bauer et al., 2010). The BC aerosols have been well studied in terms of their lifetime, concentration, sources and optical properties on a fixed wavelength (Ogren and Charlson, 1983; Hildemann et al., 1994; Pratsinis, 1994; Hansen et al., 1993; Köhler et al., 2001; Streets et al., 2001). The optical properties of mineral dust externally and internally mixed with BC have been modeled by many scientists (Fuller et al., 1999; Bond and Bergstrom, 2006; Lesins et al., 2002; Jacobson, 2000). Bond et al. (2006) estimated the optical properties of set of combinations of core-shell structure covering the whole range of possible scenarios. In their study, they considered the core to be made of light absorbing carbon while the shell to be of weakly absorbing material. However, detailed research on the optical properties of the mixture of BC with mineral dust (semi-externally mixed) is rarely available. The modeling of optical properties of such externally mixed polluted dust system is very important as has been discussed earlier.

The particles were found to form a semi-external mixtures while the formation of internal mixtures can not be ignored. Liu and Mishchenko (2007) demonstrated the effects of aggregation, fractal morphology, and refractive index on the optical properties of soot aerosols. When the different aerosols form an aggregate (semi-external mixtures) they lie in the near field zone of each other and their scattering and radiative properties can differ from those of composition-equivalent external mixtures. The differences may be too strong that it may influence the results of remote sensing studies of tropospheric aerosols and their radiative forcing estimations (Sato et al., 2003 and references therein) so to reduce the differences, the mineral dust shapes, variation of hematite content (Fe_2O_3) and the mixing state with all possible carbonaceous components should be accounted together with mineral dust nonsphericity.

To date, there is no rigorous and comprehensive polluted dust optical model available, which deals with the semi-external mixing of mineral dust with all kind of carbonaceous components together with particle nonsphericity and hematite variation (0–8%).

atmosphere support the semi-external mixing of the mineral dust with the carbonaceous components (Zongbo et al., 2002; Li et al., 2003; Clark et al., 2004; Alexander et al., 2008; Takahama et al., 2010). Based on the above analysis, the model shapes for the polluted dust systems have been considered and shown in the Fig. 1. Besides the shape, particle size, Aspect Ratio and possibility of existence of considered mixing combinations have been discussed in detail under morphology section (Sect. 3). The consideration of the trace amount of fly ash in some mixing combinations has also been justified in Sect. 3. The optical constants for the components of the polluted dust system have been considered in Sect. 4. The volume equivalent radius, VER of the semi-externally mixed composite dust has been varied from 0.1 to 1.0 μm based on the discussion in Sect. 3. The optical properties of the polluted dust particles described in Fig. 1 have been modeled using DDA (Draine and Flatau, 2004).

3 Morphology

The mineral dust particles are not spherical but are characterized by an uneven shape distribution as a function of size. Particle morphology is characterized by the aspect ratio, AR (Reid et al., 2003), a measure of the sphericity of a particle, which is equal to one for a spherical particle and greater than 1 for elongated particles such as ellipsoids.

The mineral dust particles collected in Niger (Nothern Saharan desert) on board the Facility for Airborne Atmospheric Measurements (FAAM) BAe-146 research aircraft, were studied to understand their morphology using a combination of SEM, TEM equipped with an energy dispersive X-ray detection system. The particles have been analyzed using the HISTOLAB program. The AR of about 31 000 sampled particles was studied and found to be practically independent of size for particles of diameter 0.1–10 μm . The upper limit of the AR was found to be 5, whereas the median was 1.7 (Chou et al., 2008). Kandler et al. (2007) reported the AR to be 1.64 for the Saharan Mineral Dust at Izana, Tenerife (Spain), however, Reid et al. (2003) showed a higher value of AR of 1.9 for African mineral dust collected over the Caribbean after being

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existence of 3-OC cluster (OCOCOC) also OC being the weak absorber; the modeled optical properties for the cluster will not make the major change. Because of these reasons the OCOCOC clusters have not been considered for simulation.

The studies (Alexander et al., 2008; Andreae and Gelencser, 2006; Yang et al., 2009; Chakrabarty et al., 2010; Moosmüller et al., 2009) highlighted the existence of brown carbon in the atmosphere in the spherical form and classified the brown carbon as a special class of OC which is highly absorbing at short wavelengths. Likewise OC, the brown carbon is expected to attach with the mineral dust to form various BrC-mineral dust systems like 1 BrC and 2 dust (BrCDD), 2 BrC and 1 dust (BrCBrCD). The 3-BrC (BrCBrCBrC) system has been modeled to compare with the conjugate BCBCBC system. Because the concept of brown carbon is recently evolved, as our best knowledge no particle image of brown carbon is available to support the 3-BrC model shape. The model shape with 1 BrC + 1 dust + 1 Fly-ash (BrCDF) has not been modeled because of unavailability of the particle images to support the model shape.

Based on electroscopic studies (Zongbo et al., 2002; Liet al., 2003; Clark et al., 2004; Alexander et al., 2008; Moffet et al., 2010) for the mineral dust polluted with BC and fly-ash, various mixing scenarios have been accounted by considering different combinations (spheres with different composition) as 1 BC and 2 dust (BCDD), 2 BC and 1 dust (BCBCD), 1 BC + 1 dust + 1 Fly-ash (BCDF), 3-BC (BCBCBC). The possibility of 3-dust cluster (DDD) is not supported through the chemistry involved in the cluster formation so this system has been ignored. Hereafter, the above combinations will be addressed in abbreviation as given in respective brackets. As the fly-ash has been reported in trace amount in the SEM images so the same has been accounted for the modeling purpose.

Thus using above morphological information and the mixing state, the model shapes have been decided (Fig. 1) for the numerical estimation of the polluted dust optics using DDA model.

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4 Polluted dust composition

The polluted dust has been considered to be a semi-external mixture of the pure mineral dust with the carbonaceous components mentioned in Table 2. The optical parameters for the pure mineral dust (composed of Quartz, Feldspar, Mica, Calcite and hematite) at 0.550 μm wavelength have been taken from the study by Mishra and Tripathi (2008) where they varied the hematite percentage to check its effect on the optical properties as hematite is the major absorbing component in the pure mineral dust. The hematite fraction in the global dust has been inferred to be 3.75–11.97% (Koven and Fung, 2006) so to check the sensitivity of the hematite content to the dust optical properties, the optical parameters for 0–8% hematite variation have been considered from Table 1. The optical parameters from Table 1 have been used for modeling the optics of mineral dust mixed with the carbonaceous components while the optical parameters for the carbonaceous components have been taken from Table 2.

A considerable part (~ 10 – $\rightarrow 50\%$) of the atmospheric OC is found to be water-soluble organic carbon (WSOC) (Agarwal et al., 2010; and reference therein; Facchini et al., 1999; Kanakidou et al., 2000). Further, a major fraction ($> 50\%$) of WSOC is composed of high molecular weight multifunctional compounds (Seinfeld, 2006) that contain aromatic, phenolic, and acidic functional groups (Graber and Rudich, 2006; Varga et al., 2001) and called as HUmic-Like Substances (HULIS) because they resemble with Humic Substances (HS) from terrestrial and aquatic sources.

Dinar et al. (2008) measured the refractive indices of the HULIS substance extracted from air pollution particles which were sampled from 18 May 2006 till June 2006 during daytime in an urban location (Weizmann Institute, Rehovot, Israel). The refractive index, ($m = 1.595 + i0.049$) has been measured at 532 nm using a dual-wavelength Cavity Ring Down Aerosol Spectrometer (CRD-AS). The molecular weight (M_N) and aromaticity of the HULIS extracted from the pollution particles have been found to be 460 and 16% respectively. This HULIS compound has been considered as the representative of aged organic carbon coming from pollution. In present study, for

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modeling the semi-externally mixed OC-mineral dust clusters, the above mentioned HULIS (i.e. the major fraction of organic carbon) has been considered and represented as OC hereafter.

5 Model details

The Optical properties such as SSA, asymmetry parameter, g and extinction efficiencies, Q_{ext} of the polluted dust systems (for the model shapes described in Fig. 1) with the volume equivalent radius, $\text{VER} < 1 \mu\text{m}$ have been computed using Discrete Dipole Approximation (DDA) (Draine and Flatau, 2004). The DDA model has been used for computing the light scattering of above mentioned polluted dust (semi-externally mixed) accounting for particle nonsphericity. The VER of the non-spherical particle (r_{eff}), refractive index (index of each component in the case of multi components), wavelength (μm), particle shape and the particle shape parameters are the inputs to the DDA code. The shape parameters govern the aspect ratio and extent of the target in X , Y and Z directions whose optical properties are to be modeled. The output of the code is extinction, scattering and absorption efficiencies (Q_{ext} , Q_{sca} and Q_{abs}), g , and the scattering matrix elements.

The DDA model contains routines to generate dipole arrays which represent targets of various geometries like spheres, ellipsoids, rectangular solids, cylinders, hexagonal prisms, tetrahedral, two touching ellipsoids, and three touching ellipsoids. The target has been selected based on our model shapes (Fig. 1). The target dimension is specified in the units of interdipole spacing, d and the same is fed in the model as the target shape parameters (SHPAR1, SHPAR2, SHPAR3, ...).

The target geometry is described in a coordinate system attached to the target which is referred as “Target Frame” (TF). X , Y and Z are the coordinates in the Target Frame. For simulating the optical properties of two-sphere and two-spheroid polluted dust systems, the target geometry considered in the DDA model is TWOSPH. This geometry is applicable to two touching homogeneous, isotropic spheroids, with distinct

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compositions. Setting all the dimension same in the geometry helps to generate two sphere particle systems. The parameter, NCOMP shows the number of components in the composite polluted particle. For the above mentioned two sphere and two spheroid dust systems, NCOMP = 2. The optical constant for each spheroid is fed in the model.

5 The BC-mineral dust composite two-sphere system has been modeled for a range of R_{BC}/R_{dust} values (~ 0.9 – 1.5) by varying their shape parameters.

For simulating the optical properties of three-sphere polluted dust systems, the target geometry considered in the DDA model is THRELL. This geometry is applicable to three touching homogeneous, isotropic ellipsoids of equal size and orientation, but
10 distinct compositions. The setting of the entire dimension same for each spheroid in the geometry, helps to generate three sphere particle systems. The parameter, NCOMP shows the number of components in the composite polluted particle. For the three sphere particle system, NCOMP = 3. The optical constant for each spheroid is fed in the model.

15 6 Results and discussion

6.1 The optical properties of two-particle system

6.1.1 Two-sphere BC-mineral dust system (varying BC and dust particle size in the cluster)

The optical properties of the two-sphere BC-mineral dust system have been modeled for varying sizes of BC and mineral dust. The morphology of such system (Fig. 2) has
20 already been discussed in Sect. 3.

Figure 3 shows the SSA of the two-sphere BC-mineral dust system for varying effective radius and with hematite variation from 0–8%. The SSA of two sphere BC-dust system has been modeled for decreasing R_{BC}/R_{dust} (from 1.5 to 0.9) with increasing effective radius (from 0.1 to 0.8 μm) of the composite particle (Fig. 2). The ratio,
25 $R_{BC}/R_{dust} = 1$ is for 0.64 μm effective radius of the system.

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the effective cluster size less than $0.22\ \mu\text{m}$. Beyond this size, the efficiency increases with increasing hematite till $0.32\ \mu\text{m}$ size. For the size window 0.32 to $0.6\ \mu\text{m}$, the effect of hematite content on Q_{ext} is nearly independent except around $0.5\ \mu\text{m}$ where it decreases for hematite $>6\%$.

For 0.6 to $0.72\ \mu\text{m}$, where the mineral dust sphere and the black carbon sphere in the 2-sphere cluster, are of nearly of same size, the Q_{ext} decreases with increasing hematite while the vice-versa holds good for the size window 0.75 to $0.8\ \mu\text{m}$ where the mineral dust sphere is bigger than that of black carbon sphere for the 2-sphere cluster.

6.1.2 Two-sphere (OC-dust and BrC-dust) and two spheroid (OC-dust and BC-dust) particle systems (for same size of the individual particles in the cluster)

Figure 6 shows the SSA of the two-sphere and two spheroid clusters comprising of organic carbon, brown carbon, black carbon and mineral dust with hematite content 0 and 8%. The computations have also been done for the hematite percentage 2, 4 and 6 but have not been shown in Fig. 6. The two sphere OC-dust and BrC-dust systems for 0 and 8% hematite have been represented as OCD-0, BrCD-0 and OCD-8, BrCD-8, respectively. The two spheroid OC-dust and BC-dust systems for 0 and 8% hematite have been represented as OCD¹-0, BCD¹-0 and OCD¹-8, BCD¹-8, respectively. The 2-spheroid BrC-dust system could not be considered because of unavailability of particle images which could support the spheroidal morphology of brown carbon. Here, the particles comprising a cluster have been considered of same size. For the comparison purpose, the optical properties of independent homogeneous spheres (of organic carbon (OC), brown carbon (BrC), black carbon (BC) and pure dust sphere with 4% hematite content, D-4) have also been considered. The comparison will be helpful in determining the effect of particle nonsphericity and semi-external mixing of heterogeneous components on the optical properties. Generally, the satellite retrieval algorithms account for the external mixtures of BC and dust homogeneous spheres.

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The SSA was found to vary depending on hematite content (0 to 8%) for any class of the cluster while different cluster classes showed the variation due to their semi-externally mixed combinations as discussed earlier. For all hematite percentage, the OCD clusters show the higher scattering signature compared to that of BrCD clusters until the effective radius of the cluster reaches to $0.55\ \mu\text{m}$ and also beyond this threshold size OCD clusters show higher scattering to that of BrCD-8 cluster. The sensitivity of SSA to hematite was found to be insignificant for OCD and OCD¹ systems compared to that of BrCD and BCD¹ systems for the effective size $<0.3\ \mu\text{m}$. The SSA of the pure dust sphere with 4% hematite content (D-4) showed the highest scattering signature compared to that of any particle cluster. SSA of all the particle clusters have been found in the range which is constrained by that of pure dust (D-4), pure BC and pure BrC spheres. The BC and BrC spheres show nearly same scattering signature for the effective radius $>0.4\ \mu\text{m}$. Among all the clusters, the SSA of the semi-externally mixed BrCD-8 cluster significantly differs to that of independent mineral dust, D-4 (~30% SSA variation) for effective particle radius $>0.5\ \mu\text{m}$ while the same is true for BCD¹-0 system for $<0.3\ \mu\text{m}$. The BrCD-0 system shows the SSA pattern which could be generated by averaging the SSA of the D-4 and BrC spheres for $>0.3\ \mu\text{m}$ effective radius. The OCD¹-0 spheroid system showed the maximum deviation of SSA (i.e. ~5%) to the OCD-0 sphere system and nearly the same case has been observed for OCD¹-8 system relative to that of OCD-8. Increase in hematite from 0 to 8% caused maximum SSA deviation of ~20% for OCD system while the same has been observed to be ~18% for OCD¹ system. This shows that the SSA is more sensitive to hematite content compared to that of particle shape. BC and BrC spheres show nearly size independent SSA which does not hold good for OC spheres.

Figure 7 shows the asymmetry parameter, g for the OCD, BrCD, OCD¹ and BCD¹ systems for the hematite content 0 and 8%. The g of OC, BrC, BC and D-4 spheres have also been considered for the reasons discussed earlier. The BrC independent sphere shows the highest asymmetry parameter and tends to the g value ~0.9 with increasing size. The g of independent OC and D-4 spheres are more sensitive to the

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particle size compared to that of OCD, BrCD, OCD^I and BCD^I clusters. In general, the g of the all the clusters and independent spheres have been found to be in the range 0.6 to 0.9. For all the considered hematite range, the g of OC-dust clusters (except OCD^I-8) have been found to be higher compared to that of independent OC and D-4 spheres for the effective size $<0.5 \mu\text{m}$. This shows that in the above size regime the g value of semi-externally mixed OC-dust clusters will be underestimated if we consider an external mixture of OC and dust in the aerosol optical model. At $0.8 \mu\text{m}$ effective size, the g values of BrCD-0, BrCD-8, OCD^I-0, OCD-0, OCD-8, and BCD^I-0 systems are overestimated if we consider an external mixture of OC, BrC, BC and D-4 spheres. At $1.0 \mu\text{m}$ effective size, the g values of OCD-8, BCD^I-8, OCD^I-8, BrCD-0, BCD^I-0 and OCD^I-0 systems are well constrained by the g values of independent OC, BrC, BC and D-4 spheres. The OCD^I-8 spheroid system showed the maximum deviation in g (i.e. $\sim 11\%$) to the OCD-8 sphere system while OCD^I-0 system showed the maximum variation (i.e. 8%) relative to that of OCD-0. This indicates that compared to SSA, g is more sensitive towards the particle shape. Increase in hematite from 0 to 8% caused maximum g deviation of $\sim 14\%$ for OCD system while the same has been observed to be $\sim 18\%$ for OCD^I system.

Figure 8 shows the extinction efficiency, Q_{ext} for the systems discussed in Figs. 6 and 7. No regular pattern of Q_{ext} with effective size could be observed for the considered 2-sphere and 2-spheroid clusters. For the effective size window $0.4\text{--}0.6 \mu\text{m}$, the extinction efficiency of the semi-externally mixed OCD, BrCD, OCD^I and BCD^I clusters have been estimated to be greater than that of OC, BrC, BC and D-4 independent spheres.

6.2 The optical properties of three-particle system

The optical properties of the 3-sphere particle system comprising of black carbon (BC), organic carbon (OC), brown carbon (BrC), fly-ash (F) and dust (D) with hematite content 0–8% have been modeled. The three spherical particles forming a cluster have been considered of same size in this study.

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Figure 9 shows the SSA of the 3-sphere clusters comprising of organic carbon, brown carbon, black carbon, fly-ash and dust with hematite content 0 and 8%. The organic carbon, dust and fly-ash clusters have been represented as OCDF-0 and OCDF-8 followed by hematite content. One OC sphere and two dust spheres in a cluster have been represented as OCDD-0 and OCDD-8 while two OC spheres and one dust sphere in a cluster have been represented as OCOCD-0 and OCOCD-8. One brown carbon and two dust spheres are denoted as BrCDD-0 and BrCDD-8, two BrC spheres and one dust sphere in a cluster have been represented as BrCBrCD-0 and BrCBrCD-8. The cluster comprising of 3-sphere BrC is denoted as BrCBrCBrC. The clusters comprising of black carbon, dust and fly-ash have been represented as BCDF-0 and BCDF-8 followed by their hematite content. One BC sphere and two dust spheres in a cluster have been represented as BCDD-0 and BCDD-8 while two BC spheres and one dust sphere in a cluster have been represented as BCBCD-0 and BCBCD-8. The cluster comprising of 3-BC spheres, is denoted as BCBCBC.

To assess the effect of particle nonsphericity and three spheres semi-externally mixed heterogeneous components on the optical properties of above mentioned systems, a comparison has been made with that of OC, BrC, BC and pure dust spheres with 4% hematite content (D-4).

The SSA of above discussed clusters have been found to vary depending on hematite content for every class of the cluster while different cluster classes showed the variation due to their semi-externally mixed combinations as discussed earlier. The OCDF-0 cluster with 0% hematite content shows same scattering signature as that of OCDD-0 cluster with 0% hematite content. The same scattering signature of the above clusters is due to the negligible difference between the refractive indices of fly-ash and dust with 0% hematite content. For 8% hematite content, the OCDF cluster shows the higher scattering compared to that of OCDD cluster. The replacement of one dust sphere from OCDD cluster with one organic carbon sphere reduces the scattering nature of newly formed OCOCD cluster for the considered hematite contents. The SSA of the OCOCD system has also been found to be reduced with increasing hematite

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content. The sensitivity of SSA to hematite for all the OC-dust clusters has been found to be significant with increasing size. Except for the effective radius 0.43–0.72 μm , the D-4 sphere shows the highest scattering signature compared to that of any particle clusters discussed in Fig. 9. At particle radius $\sim 0.57 \mu\text{m}$, the OCDF clusters (with all hematite percentage) show high SSA compared to that of that of OC and D-4 species which form the cluster.

Among the BrC-dust clusters, the BrCDD clusters show the higher scattering compared to that of BrCBrCD clusters with respective hematite content. The replacement of one dust sphere from BrCDD cluster with one brown carbon sphere reduces the scattering nature of newly formed BrCBrCD cluster for the considered hematite contents. The sensitivity of SSA to hematite was found to decrease with reducing number of dust spheres in a particle cluster. The SSA of the BrCDD and BrCBrCD systems show reduced SSA with increasing hematite from 0 to 8%. The SSA of the 3-sphere brown carbon cluster (BrCBrCBrC) has been found to vary significantly compared to that of independent BrC sphere for the effective cluster radius $< 0.6 \mu\text{m}$ while beyond this size the difference is negligible. For all the BrC-dust systems, the hematite sensitivity to SSA has been found to be significant for effective cluster size $> 0.43 \mu\text{m}$. The deviation of BC sphere to that of 3-sphere BC cluster (BCBCBC) has been found to be lesser compared to that of BrC sphere to BrCBrCBrC cluster for the effective cluster size $< 0.6 \mu\text{m}$. The reason for the above may be due to the fact that the effect of particle nonsphericity on the optical properties dominates in case of lesser effective absorbance of the cluster.

Among the BC-dust systems, the BCDF-0 cluster shows same scattering signature as that of BCDD-0. The same scattering signature of the above clusters is due to the negligible difference between the refractive indices of fly-ash and dust with 0% hematite content. The BCDF-8 cluster shows the higher scattering compared to that of BCDD-8 cluster. The replacement of one dust sphere from BCDD cluster with one black carbon sphere reduces the scattering nature of newly formed BCBCD cluster for the considered hematite contents. The SSA of the BCBCD system has also been

found to be reduced with increasing hematite content from 0 to 8.

Till date there are limited experimental observations for the measurement of the asymmetry parameter, g . The uncertainty involved in the measurement is significant. So the modeling tools play significant role in the numerical estimation of g by accounting real particle morphology, heterogeneous composition and the mixing states in the optical model.

Among all the accumulation mode dust-carbonaceous species clusters discussed in Fig. 9, the SSA for the BCDD-0 (or BCDF-0) shows the highest deviation $\sim 68\%$ (relative to BC sphere), for BrCDD-0 cluster $\sim 83\%$ (relative to BrC sphere) and $\sim 70\%$ for OCDD-0 (or OCDF-0) cluster (relative to OC sphere). The maximum deviations have been shown for the respective classes in the accumulation mode. These differences may be due to cluster nonsphericity and the cluster heterogeneity.

Figure 10 shows the asymmetry parameter, g for all the particle systems discussed in Fig. 9. The g of OC, BrC, BC and D-4 spheres have been modeled for the reasons discussed earlier in Fig. 9. The BrCBrCBrC cluster together with BrC sphere show the highest g for the accumulation mode polluted dust systems. The BCBCBC cluster together with BC sphere show slightly lesser g values. First of all, we consider the g of OC-dust systems. For the effective radius $< 0.6 \mu\text{m}$, all the semi-externally mixed OCDF clusters show the energy scattered in the forward direction (i.e. g) to be greater than that of OC and D-4 independent spheres while vice versa holds good for size range $0.73\text{--}0.9 \mu\text{m}$. Thus neglecting the semi-external mixing and consideration of independent component spheres will lead to underestimation of g values for the effective radius $< 0.6 \mu\text{m}$ and the vice versa for radius range $0.73\text{--}0.9 \mu\text{m}$. The asymmetry parameters of independent D-4 sphere is most sensitive to the particle size compared to that of OCDF, OCDD and OCOCD semi-externally mixed clusters.

Among the BrC-dust systems, for the effective radius range $0.72\text{--}0.9 \mu\text{m}$, the semi-externally mixed BrCDD clusters with the cluster constituents like dust and brown carbon show the energy scattered in the forward direction is smaller than that of energy scattered by BrC and D-4 independent spheres. Thus neglecting the semi-external

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mixing of BrCDD clusters and consideration of independent component spheres will lead to overestimation of g values for the effective radius range 0.72–0.9 μm .

In case of BC-dust systems, for the effective radius range 0.75–0.9 μm , the semi-externally mixed BCDF and BCDD clusters with the cluster constituents like dust, black carbon and fly-ash show the lesser g values than that of BC and D-4 independent spheres. This indicates that, for the effective radius range 0.75–0.9 μm , the g values are overestimated if we replace the semi-external mixing state (presented here in form of 3-sphere clusters) by external mixing of BC and D-4 spheres.

Among all the clusters discussed in Fig. 10, the g for the BCDD-0 (or BCDF-0) shows the highest deviation $\sim 31\%$ (relative to BC sphere), for BrCDD-0 cluster $\sim 31\%$ (relative to BrC sphere) and $\sim 33\%$ for OCDD-0 (or OCDF-0) cluster (relative to OC sphere).

Figure 11 shows the extinction efficiency, Q_{ext} for the carbonaceous species and dust mixture model clusters for which SSA and g have been shown in Figs. 9 and 10 respectively. As has been observed in Sect. 6.1.2 for the two particle systems, no regular pattern of Q_{ext} with effective size could be observed for the considered three sphere particle clusters. The Q_{ext} of the semi-externally mixed OC-dust, BrC-dust and BC-dust clusters have been estimated to be greater than that of OC and D-4 (for size window 0.34–0.7 μm), BrC and D-4 (for the size window 0.38–0.7 μm) and BC and D-4 independent spheres (for the size window 0.4–0.76 μm) respectively.

7 Conclusions

We modeled the optical properties of several cases of semi-external mixing of carbonaceous components and mineral dust. The main findings can be summarized as follows.

1. The SSA was found to vary depending on hematite content (0–8%) for any class of the cluster while different cluster classes showed the variation due to their

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semi-externally mixed heterogeneous combinations. The SSA of the pure dust sphere with 4% hematite content (D-4) showed the highest scattering signature compared to all the clusters considered in the study. The sensitivity of SSA to hematite was found to decrease with reducing number of dust spheres in a particle cluster.

2. The extinction efficiency of the semi-externally mixed clusters was estimated to be greater than that of component species spheres for a size window of 0.3–0.8 μm ,
3. In view of effective absorbance of two sphere BC-mineral dust clusters, hematite starts playing role compared to that of black carbon as the $R_{\text{BC}}/R_{\text{dust}}$ decreases (i.e. with increase of dust sphere size compared to black carbon sphere in the composite 2-sphere cluster). The sensitivity of SSA to hematite content was found to be significant when both of the spheres (i.e. mineral dust and BC) are nearly of same size.
4. The SSA of the semi-externally mixed BrCD-8 cluster differs most significantly to that of independent mineral dust, D-4 ($\sim 30\%$ SSA variation) for effective particle radius $> 0.5 \mu\text{m}$ while the same is true for BCD¹-0 system for $< 0.3 \mu\text{m}$. The OCD¹-0 spheroid system showed the maximum deviation of SSA (i.e. $\sim 5\%$) compared to the OCD-0 and nearly same deviation has been observed for OCD¹-8 system relative to that of OCD-8. Increase in hematite from 0 to 8% caused maximum SSA deviation of $\sim 20\%$ for OCD system while the same has been observed to be $\sim 18\%$ for OCD¹ system. This shows that the SSA is more sensitive to hematite content compared to that of particle shape. In general, the g of the all the clusters and independent spheres have been found to be in the range 0.6 to 0.9. The OCD¹-8 spheroid system showed the maximum deviation in g (i.e. $\sim 11\%$) compared to the OCD-8 sphere system while OCD¹-0 system showed the maximum variation (i.e. 8%) relative to that of OCD-0. This indicates that compared to SSA, g is more sensitive to the particle shape. Increase in hematite from 0 to 8%

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caused maximum g deviation of $\sim 14\%$ for OCD system while the same has been observed to be $\sim 18\%$ for OCD' system.

- 5 5. The replacement of one dust sphere from OCDD (BrCDD) cluster with one organic carbon (brown carbon) sphere reduces the scattering nature of newly formed OCOCD (BrCBrCD) cluster for the considered hematite contents. Among the accumulation mode dust-carbonaceous species, the SSA for the BCDD-0 (or BCDF-0) shows the highest deviation $\sim 68\%$ (relative to BC sphere), for BrCDD-0 cluster $\sim 83\%$ (relative to BrC sphere) and $\sim 70\%$ for OCDD-0 (or OCDF-0) cluster (relative to OC sphere).
- 10 6. The g value of clusters, like BCDD-0 (or BCDF-0) shows the highest deviation $\sim 31\%$ (relative to BC sphere), for BrCDD-0 cluster $\sim 31\%$ (relative to BrC sphere) and $\sim 3\%$ for OCDD-0 (or OCDF-0) cluster (relative to OC sphere). The differences in SSA and g may be due to cluster nonsphericity and the cluster heterogeneity.
- 15 7. The modeled optical properties of the accumulation mode mineral dust mixed with the carbonaceous species for hematite (0–8%) will be valuable input in the GCM to further reduce the uncertainty in the direct and indirect radiative forcing. Till date no GCM account for the semi-external mixing for the radiative forcing estimation. The database of the optical properties for the mono-disperse particles can be used for the quantified radiative forcing estimation corresponding to different class of clusters accounting particle nonsphericity and heterogeneity. As the semi-external mixing has not been accounted so far in GCM so the modeled optical properties can directly be used for the radiative forcing estimation.

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Table 1. Optical constants (at $\lambda = 0.55 \mu\text{m}$) of mineral dust component for varying hematite percentage obtained from Mishra and Tripathi (2008).

Composite particle component	Hematite (%)	N	K
Mineral dust	0	1.510	0.0001
	2	1.540	0.0039
	4	1.570	0.0080
	6	1.600	0.0125
	8	1.630	0.0174

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Table 2. Optical constants of carbonaceous components and fly-ash.

Composite particle component	Reference	N	K	$\lambda(\mu\text{m})$
Organic carbon, OC	Dinar et al. (2008)	1.595	0.0490	0.532
Brown carbon, BrC	Alexander et al. (2008)	1.670	0.2700	0.550
Black carbon, BC	Hess et al. (1998)	1.750	0.4400	0.550
Fly-ash	Liu and Swithenbank (1996)	1.500	0.0000	0.550

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Mineral dust mixed with the carbonaceous component	Two-sphere system	Two-spheroid system	Three-sphere system
Organic Carbon, OC 			  
Brown Carbon, BrC 			  
Black Carbon, BC 			   

 Mineral dust (with Hematite variation 0-8%)  Fly-ash

Fig. 1. The pure mineral dust (shown with yellow sphere) mixed with the carbonaceous components like Organic carbon, OC, Brown carbon, BrC, and Black carbon, BC shown with pink, dark tan, and black spheres respectively (First column, top to bottom). Semi-externally mixed polluted mineral dust two-sphere and two spheroid model shapes comprising of mineral dust with OC, BrC and BC respectively (Second and third column, top to bottom). BrC mineral dust two spheroid model shape has not been modeled as BrC occurs in spherical form. The three sphere model shape of mineral dust with OC, BrC and BC respectively (Fourth column, top to bottom). Some of the three sphere systems were modeled with fly-ash (shown with grey sphere) based on the experimental observations.

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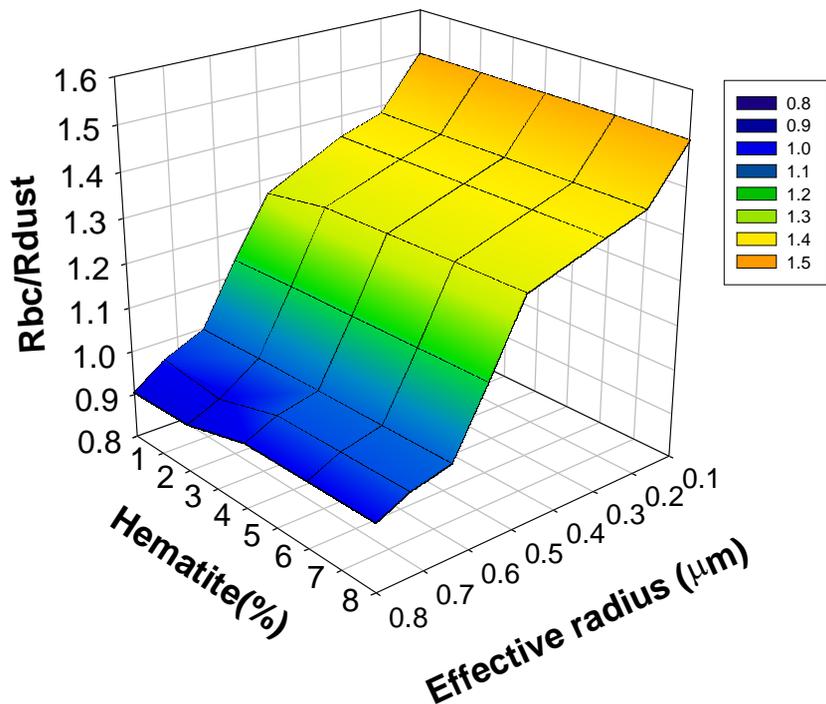


Fig. 2. The BC-mineral dust two sphere system with the ratio of radius of BC sphere (R_{BC}) to that of mineral dust sphere (R_{dust}) i.e. R_{BC}/R_{dust} for the effective volume equivalent radius of the two-sphere cluster with the varying hematite percentage in the mineral dust. Sizes of the individual particles in the cluster have been governed with the shape parameters in the DDA model. Increasing hematite causes increase in the absorption of the composite particle. Hematite variation is shown here to check the code sensitivity for the BC-mineral dust two sphere systems.

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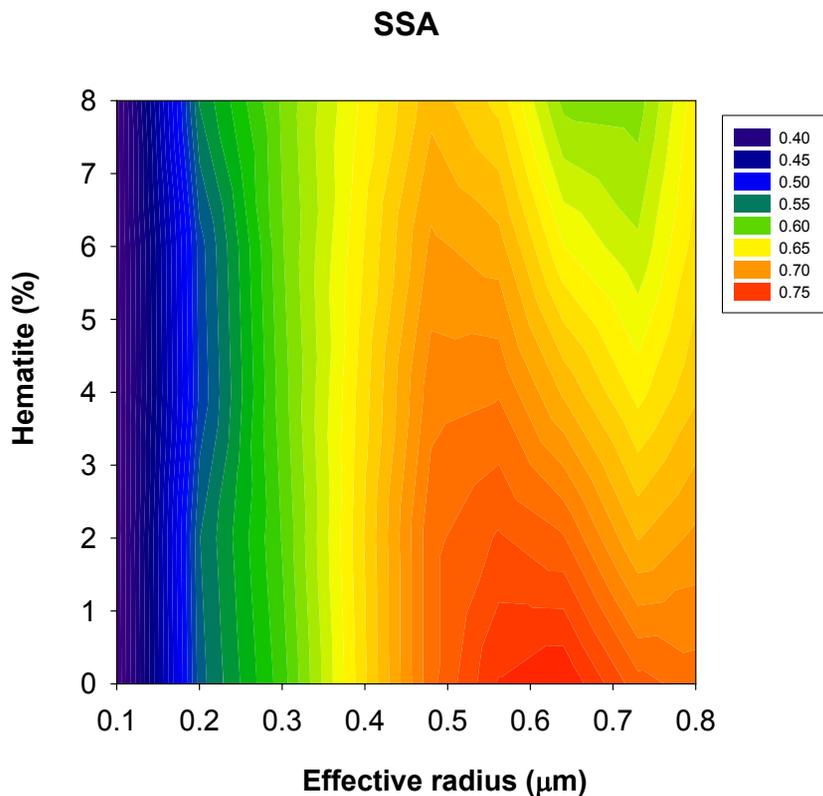


Fig. 3. The SSA of the BC-mineral dust two sphere system for the effective volume equivalent radius of the two-sphere cluster with the varying hematite percentage in the mineral dust.

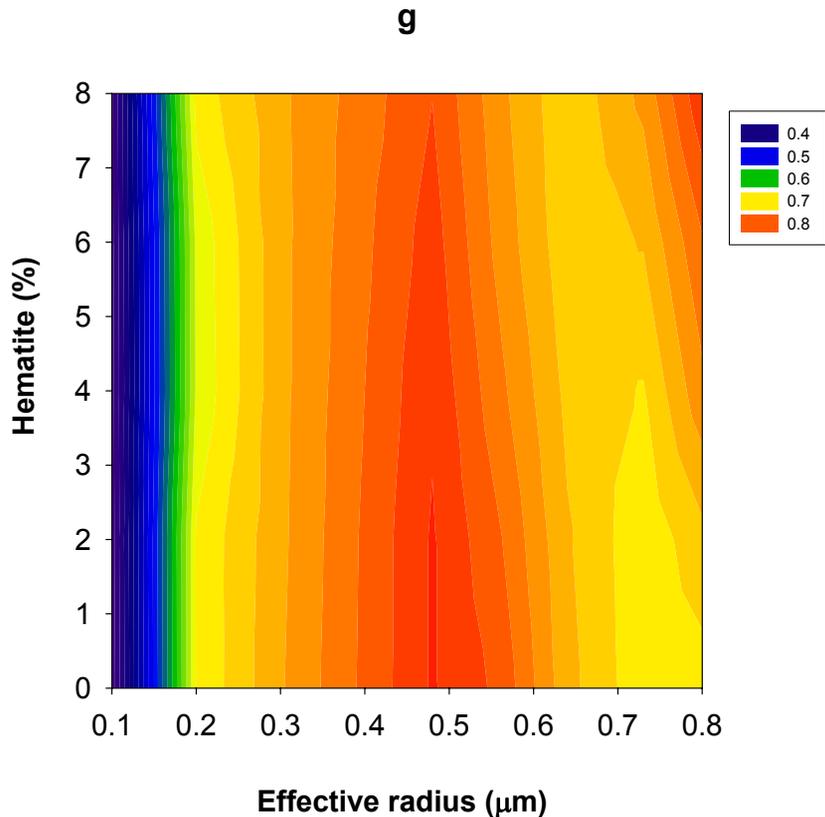


Fig. 4. The asymmetry parameter, g of the BC-mineral dust two sphere system for the effective volume equivalent radius of the two-sphere cluster with the varying hematite percentage in the mineral dust.

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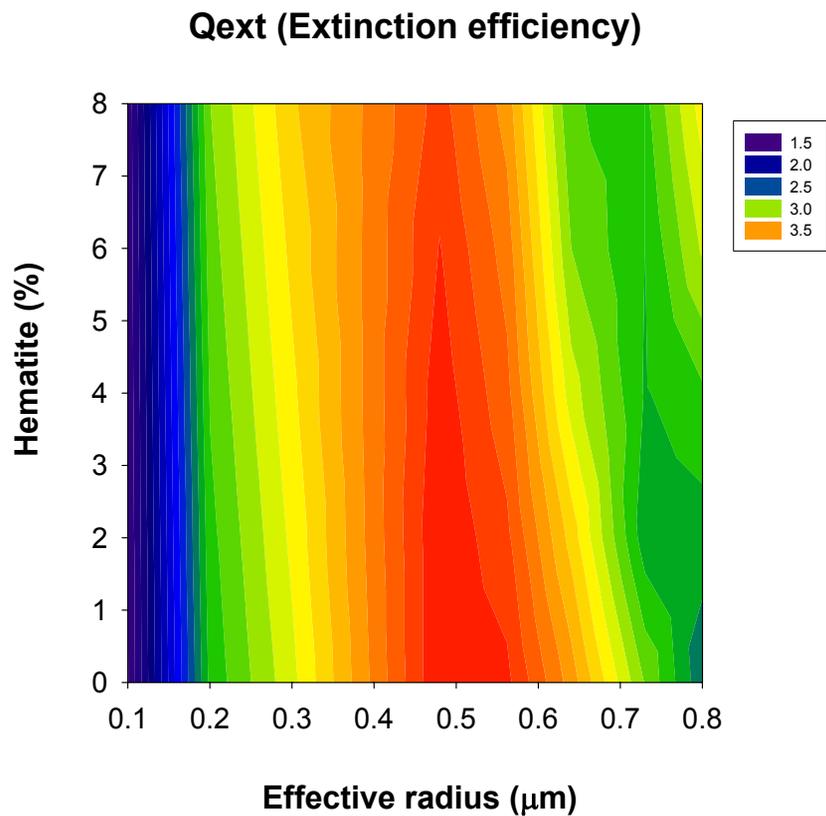


Fig. 5. The extinction efficiency, Q_{ext} of the BC-mineral dust two sphere system for the effective volume equivalent radius of the two-sphere cluster with the varying hematite percentage in the mineral dust.

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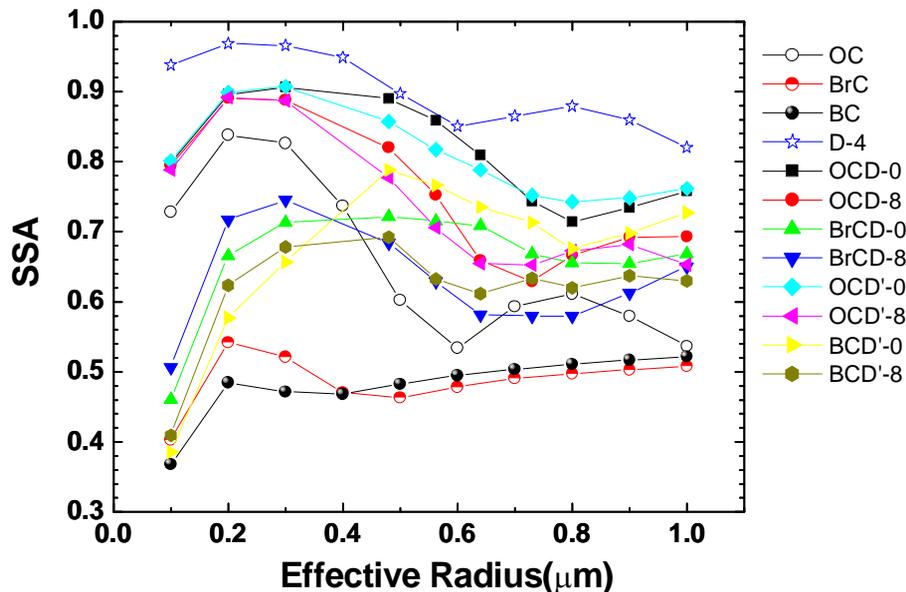


Fig. 6. The SSA of the 2-sphere OC-dust, BrC-dust and 2-spheroid OC-dust, BC-dust systems for the effective volume equivalent radius. The 2-sphere OC-dust and BrC-dust systems are denoted as OCD and BrCD respectively while the two spheroid systems OC-dust and BC-dust are denoted as OCD' and BCD' respectively. Each above mentioned nomenclature is followed by the hematite percentage. The above mentioned systems are compared to that of homogeneous spheres of organic carbon (OC), brown carbon (BrC), black carbon (BC) and dust with 4% hematite (D-4).

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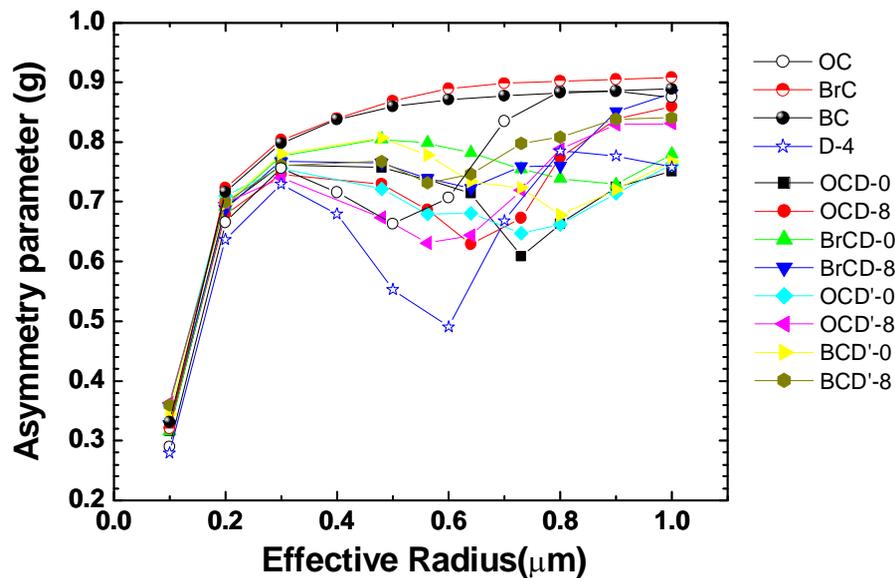


Fig. 7. The Asymmetry parameter, g for all the particle systems discussed in Fig. 6.

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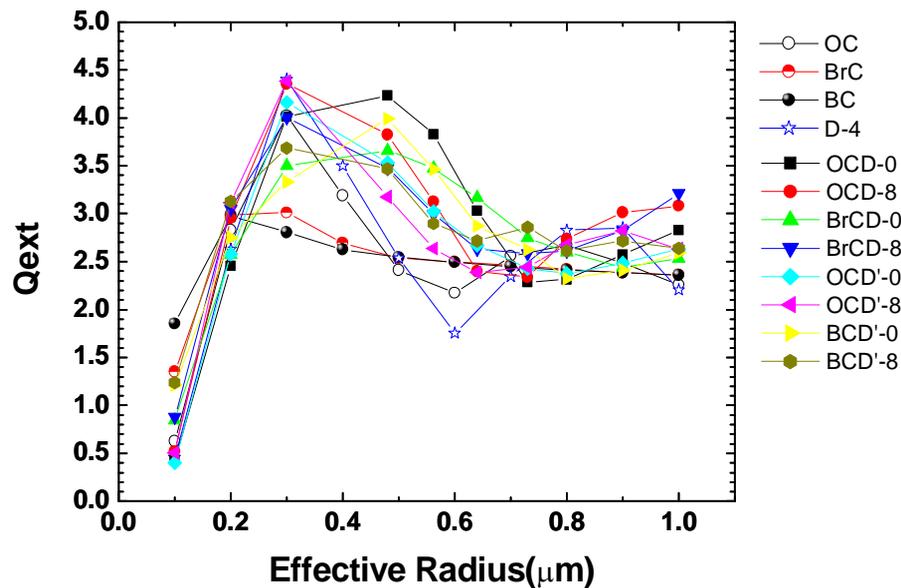


Fig. 8. The Extinction efficiency, Q_{ext} for all the particle systems discussed in Fig. 6.

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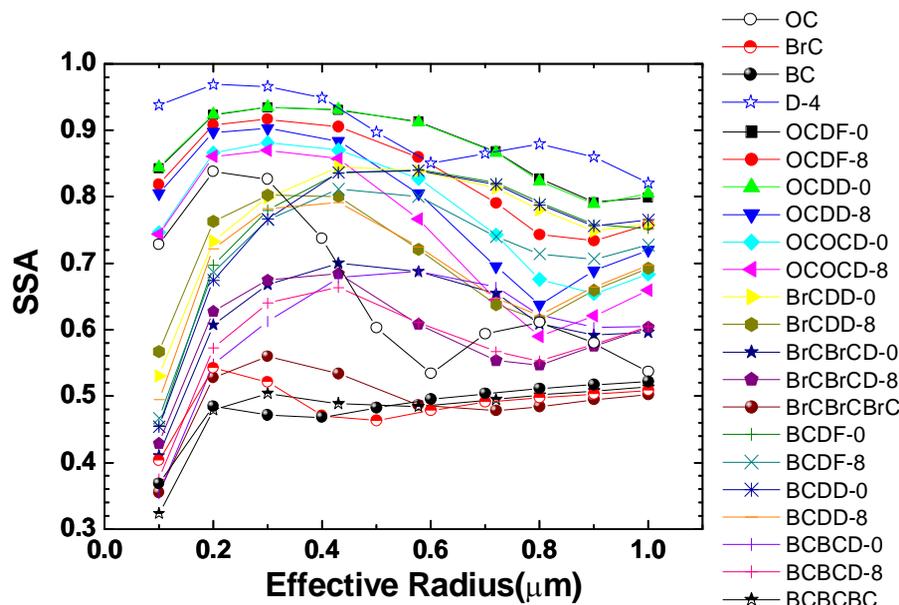


Fig. 9. The SSA of the 3-sphere OC-dust-flyash, OC-dust, BrC-dust, 3-sphere BrC, BC-dust-flyash, BC-dust and 3-sphere BC systems for the effective volume equivalent radius. The OC-dust-flyash and OC-dust systems are denoted as OCDF, OCDD and OCOCD respectively. The BrC-dust and BrC 3-sphere systems are denoted as BrCBrCD, BrCDD and BrCBrCBrC respectively. The BC-dust-flyash, BC-dust and BC 3-sphere systems are denoted as BCDF, BCDD, BCBCD and BCBCBC respectively. Each above mentioned nomenclature is followed by the hematite percentage. The 3-sphere systems are compared to that of homogeneous spheres of organic carbon (OC), brown carbon (BrC), black carbon (BC) and dust with 4% hematite (D-4).

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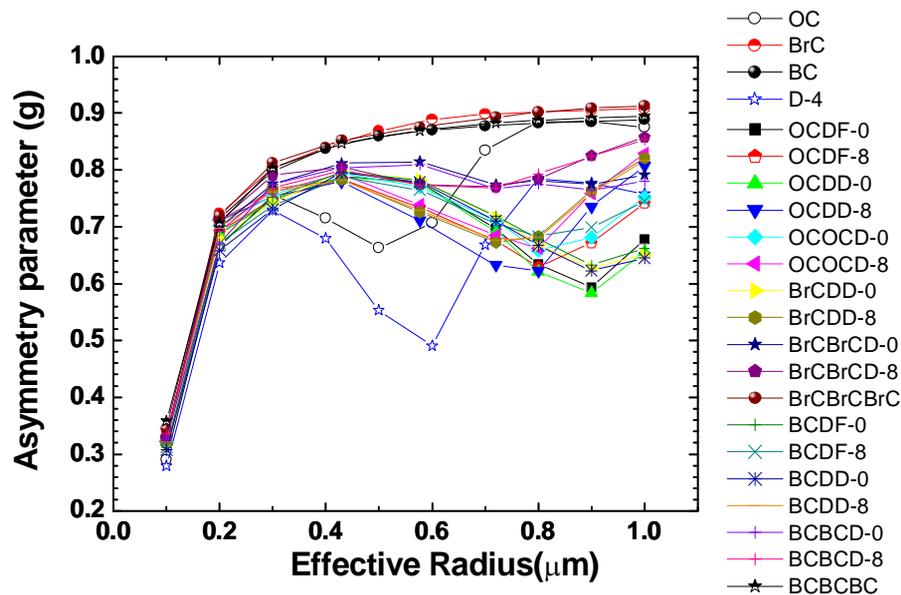


Fig. 10. The Asymmetry parameter, g for all the particle systems discussed in Fig. 9.

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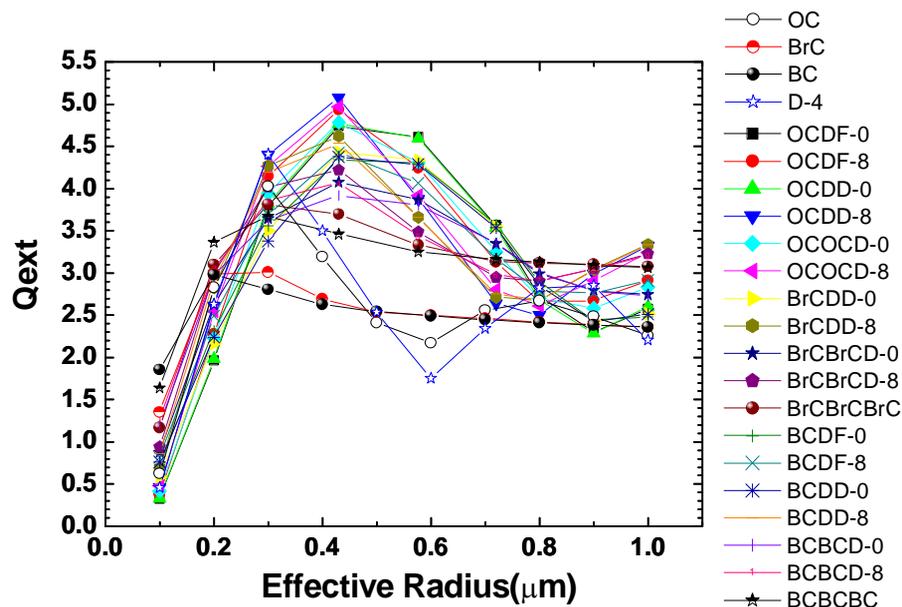


Fig. 11. The Extinction efficiency, Q_{ext} for all the particle systems discussed in Fig. 9.

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