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Atmospheric Chemistry and Physics Discussions

Interactive comment on " Effects of particle shape, hematite content and semi-external mixing with carbonaceous components on the optical properties of accumulation mode mineral dust" *by* S. K. Mishra et al.

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Reviewer#2

1)In the abstract, comparing the three-sphere model shapes to a spherical mineral dust particle with the same hematite content and VER might be more appropriate? Since the paper is interested in the radiative forcing estimation, and the whole mineral dust accumulation mode, the differences in optical properties for a distribution of particle

sizes also might be more appropriate?

Response: The authors are thankful to the reviewer for the valuable suggestion. As the size averaged optical properties are more appropriate compared to that of monodisperse hence earlier discussion of monodisperse optical properties has been removed from the abstract in the revised manuscript. The size averaged optical properties for the polluted dust systems have been compared to that of size averaged dust sphere with 4% (D-4) and 8% (D-8) hematite (Section 6.3, Second para, Line 561-573). The key results have been incorporated in the abstract.

2)p31256(7–10): The aerosol direct effect is not solely determined "by the optical properties of individual aerosols" as could be inferred by this sentence; e.g. Haywood & Shine (1995, Geophys. Res. Lett., 22 p603) - surface reflectance can change the sign of the effect for absorbing particles.

Response: The correction has been made in the revised manuscript. (2nd para of Introduction section, line # 106-108)

3)The introduction has several statements, which are not supported by the references given. For example:

Query: p31256(2–4) Maria et al. (2004) does not describe pollution of mineral dust by carbonaceous components but the oxidation of pre-existing OC ("the organic carbon compounds [. . .] were most likely associated with the dust source and subsequently oxidized on the dust particles during transport."). This reference supports the overall premise that OC is present on mineral dust particles, but in the context of its use here, it is not valid.

Response: The reference Maria et al. (2004) has been removed and the appropriate references (Sullivan and Prather, 2007; Yang et al., 2009b) have been inserted in the revised manuscript which support that the OC from pollution associates with mineral dust. (1st para of Introduction section, line # 84)

Query: p31256(17–18) The first reference (Chandra et al., 2004) deals mainly with the coating of sulfate and sea salt with carbonaceous components and vice versa. Mineral dust is included as an additional external mixture, but not dwelt upon at any point.

Response: The reference (Chandra et al., 2004) has been removed and the appropriate reference (Bauer et al., 2010) has been inserted in the revised manuscript. The inserted reference supports the fact that the uncertainties in radiative forcing may be due to mixing state of carbonaceous components with mineral dust. (3rd para of Introduction section, line # 115-116)

Query: p31257(2–7) These are the principle references supporting the semi-external mixing and are used throughout the work to justify choices of particle shape chosen. I do not find them overly convincing. Li et al. (2003) says that "only minor amounts of mineral dust particles occur [. . .] Some are aggregated with sea-salt, and some are coated with ammonium sulfate". There is no mention of semi-external mixtures between dust and carbon. I could find no mention of mineral dust at all in Alexander et al. (2008). Zongbo et al. (2002) (should be Shi et al.) says that mineral grains had "trace amounts of soot aggregates adhering onto the surfaces" in the description for Fig. 2a. In the figure, the soot aggregates were so small as to be almost imperceptible; certainly not large enough for the rdust=rcarbon used later in calculations. This was also true of Clarke et al. (2004) Fig. 2. Takhahama et al. (2010) shows mineral dust aggregated to OC and is the most convincing reference.

Response: The references Li et al. (2003) and shi et al. (2002) have been removed from the revised manuscript.

The references supporting the black carbon-dust semi-external mixing are:

i)Alexander et al. (2008, Science supplement)

The TEM image (figure S1, page #5) is of the particles sampled during flight RF13 of Asian Aerosol Characterization Experiment (ACE-Asia) in spring 2001. The sample

was taken at latitude of 30 m above the Yellow Sea during heavy dust events. In the sample, the high level of heterogeneous mixing was found amongst the carbon spheres, soot carbon, large spherule soot, ammonium sulfate particles, mineral dust and silica spheres. The two-sphere and three-sphere systems can clearly be visualized by looking at Figure S1. The size of the individual spheres in the two-sphere system was found to be nearly same. Some systems were also traced with varying sizes of the individual spheres.

To study the sensitivity of the individual sphere size in the two-sphere system, the computations were performed with varying sizes of individual sphere in the system in the initial part of the paper. However, to avoid complexity and to reduce the computational burden, the size of the individual sphere/spheroid in the two-spheroid and the three-sphere systems, has been considered to be the same.

ii)Derimian et al. (2008, ACP)

The Figure 5d (page #3629) shows the soot clusters attached to mineral dust which was found on the fine filter from the dust event on 3 February 2001 at University of Negev, Israel. The whole soot cluster in the compact form can be considered as a composite sphere/spheroid. Based on the SEM images, this soot cluster can be considered to attach with the spherical/spheroid shape dust particle.

iii)Clarke et al. (2004, JGR)

SEM image (Figure 2, top right, page # 5) shows particles at 5500 m altitude (over the Yellow sea) during ACE-Asia. In the Figure, the aluminosilicates (square) was found to be semi-externally attached to more typical soot carbon aggregates (solid circle). The aluminosilicates are the major part of the mineral dust as Mica and Feldspar. The composite particle can be modeled as 2-spheroid particle system comprising black carbon and mineral dust spheroids. However, the size of the individual spheroid does not seem to be equal but to avoid the computational burden, size of the individual spheroids has been considered to be the same.

The Figure (Figure 2, middle left, page # 5) shows the cluster of aluminosilicates (square), soot carbon aggregates (solid circle) and large spherical graphitic carbon (carbon cenosphere shown with open circle). These particles were sampled at 700 m altitude. This image supports the three-sphere particle system.

iv)Shi et al. (2005, JGR, page # 4& 5)

Figure 2(c) shows the samples from the Asian Dust Storm (ADS100) collected after the 16 March 2002 dust storm episode in Beijing, China. SEM image shows the mixing of soot, minerals and coal fly ash with different spherical/spheroid shape attachments.

The size segregated chemical composition and the particle number distribution for the accumulation mode polluted dust (above discussed ADS 100 sample) has been shown in Figure 3(b). The size segregated chemical composition of accumulation mode polluted dust confirms the presence of mineral dust, soot and coal fly-ash.

v)Arimoto et al. (2006, Global and Planetary Change, page # 42)

Figure 9 shows the field emission SEM image of RF13 sample collected from 600 m leg over Yellow Sea. The Ca aluminosilicate (denoted as 'a' in Figure) is the major part of the mineral dust which is found to be semi-externally mixed with the black carbon spherule aggregates (soot, denoted as 'c' in Figure). The three-sphere system (BCBCD) can easily be visualized.

Information on size:

For 1 micro-meter effective radius (VER) of the two-sphere and three-sphere systems, the radius of each sphere = 0.7955 μm (in two-sphere system) radius of each sphere = 0.6959 μm (in three-sphere system)

For 0.1 micro-meter effective radius (VER) of the two-sphere and three-sphere systems, the radius of each sphere = 0.0793 μm (in two-sphere system) radius of each sphere = 0.0693 μm (in three-sphere system)

The considered size range of individual spheres in the polluted systems (used for the computation) has been found to be consistent with the SEM images.

Query: p31257(7–8) The authors state that "the semi-external mixing is rarely modeled (Mishchenko et al., 2004)". In this reference's abstract, Mishchenko states: "It is concluded that aggregation is likely to have a relatively weak effect on scattering and radiative properties of two-component tropospheric aerosols and can be replaced by the much simpler external mixture model in remote sensing studies and atmospheric radiation balance computations". As such there would almost certainly be a better references to use in a paragraph justifying calculations involving semi-external mixtures.

Response: The optical properties of semi-externally mixed polluted mineral dust are rarely modeled and limited to two-sphere particle system with strong size gradient between the spheres (Mishchenko et al., 2004). The optical properties of such particle systems have been found to be same as that of bigger sphere in the two-sphere system. To the best of our knowledge, there is no literature available on semi-external mixing of spheres with nearly same size.

4)I would assume that optical calculations are made at _550 nm since refractive indices are given at this wavelength in Table 2?

Response: The optical calculations have been made at wavelength 550 nm.

5)There is as lot of redundancy between sections 2 & 5 and between sections 3 & 4. These could be presented in a more coherent manner.

Response: The suggestions have been incorporated in the revised manuscript.

6)Section 5 has unnecessary details such as computer parameters e.g. NCOMP.

Response: The unnecessary details have been removed in the revised manuscript.

7)What does Fig. 2 show? Surely if the percentage of hematite in the mineral dust is changing, this should have no effect on the overall volume equivalent radius? Isn't the key along the left hand side the same as the z-axis (Rbc/Rdust)? Why does changing the effective radius change Rbc/Rdust?

Response: In the two-sphere BC-dust system, the optical properties have been studied for the ratio RBC/Rdust for a given effective radius and for varying hematite content (0, 2, 4, 6 and 8%). The RBC/Rdust is calculated for a given effective radius of BC-dust system with a given hematite content in dust sphere. So, to check the consistency of the model calculated RBC/Rdust for different hematite content, figure 2 has been demonstrated. It is clear from figure 2 that the RBC/Rdust is independent of hematite content for a given effective radius. The color code in figure 2 shows the variation of RBC/Rdust with varying effective radius. RBC/Rdust is independent of hematite content. Figure 2 has been deleted in the revised manuscript and the figure numbers have been changed accordingly.

8)Figures 3–5: How many calculation points for hematite % and effective radius go into these contour plots?

Response: For figure 3-5: calculation points for hematite % = 5 (0, 2, 4, 6, 8 %) calculation points for effective radius = 8 (from 0.1 to 0.8 μm) Total points = 40.

9)Figures 6–11: It might be interesting to see differences between new dust models and D-4 as well as absolute values.

Response: The authors are thankful for the valuable suggestion. The suggestion has been incorporated for the size averaged optical properties and has been discussed in section 6.3 in the revised manuscript.

New references added in the revised manuscript

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Derimian, Y., Karnieli, A., Kaufman, Y. J., Andreae, M. O., Andreae, T. W., Dubovik, O., Maenhaut, W., and Koren, I.: The role of iron and black carbon in aerosol light absorption, Atmos. Chem. Phys., 8, 3623–3637, 2008.

Sullivan, R. C., and Prather, K. A.: Investigations of the diurnal cycle and mixing state of oxalic acid in individual particles in Asian aerosol outflow, Environ. Sci. Technol., 41(23), 8062–8069, 2007.

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