



## Abstract

To determine a plausible range of mass extinction efficiencies (MEE) of terrestrial atmospheric dust from the near to thermal IR, sensitivity analyses are performed over an extended range of dust microphysical and chemistry perturbations. The IR values are subsequently compared to those in the near-IR, to evaluate spectral relationships in their optical properties. Synthesized size distributions consistent with measurements, model particle size, while composition is defined by the refractive indices of minerals routinely observed in dust, including the widely used OPAC/Hess parameterization. Single-scattering properties of representative dust particle shapes are calculated using the T-matrix, discrete dipole approximation and Lorenz-Mie light-scattering codes. For the parameterizations examined, MEE ranges from 0–1.2 m<sup>2</sup> g<sup>-1</sup>, with large contributions from non-spheres composed of quartz and gypsum. At near-IR wavelengths, MEE for non-spheres generally exceeds those for spheres, while in the IR, shape-induced changes in MEE strongly depend on volume median diameter (VMD) and wavelength, particularly for MEE evaluated at the mineral resonant frequencies. MEE distributions appear to follow particle geometry and are evidence for shape dependency in the optical properties. It is also shown that non-spheres best reproduce the positions of prominent absorption peaks found in silicates. Generally, angular particles exhibit wider and more symmetric MEE distribution patterns from 8–10 μm than those with smooth surfaces, likely due to their edge-effects. Lastly, MEE ratios allow for inferring dust optical properties across the visible-IR spectrum. We conclude MEE are significant for the parameter space investigated, and are a key component for applications including sea surface temperature retrievals and the study of direct aerosol radiative effects.

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## 1 Introduction

In Earth's atmosphere, dust particles both scatter and absorb solar and terrestrial radiation, with the radiative interactions critically depending on the bulk optical and microphysical properties of the constituent minerals. Previous works have clearly demonstrated the inherent difficulties in modeling dust due to the large uncertainties in their physicochemical properties (e.g., Sokolik et al., 1999; Reid et al., 2003b). More measurements of dust properties having greater spatial, temporal, and spectral coverage are absolutely essential, as these ultimately define the aerosol inputs used by radiative transfer and global climate models. The model inputs are represented by a set of wavelength dependent single-scattering parameters which are functions of the particle's mineral composition, geometric size, and morphology. These include the single-scattering albedo ( $\omega$  – the percentage of light extinction due to scattering), asymmetry parameter ( $g$  – a parameterization that describes the particle phase function), and extinction coefficient ( $\beta_{\text{ext}}$  – the amount of scattering and absorption per unit path length).

Another parameter commonly employed in aerosol studies is the mass extinction efficiency (MEE) ( $\alpha_{\text{ext}}$  – Hand et al., 2007 and references therein) which defines the total light extinction per unit mass of aerosol. Also referred to as the specific extinction cross-section (Gerasopoulos et al. 2009), MEE is the sum of the mass scattering and mass absorption efficiencies (MSE and MAE, respectively). MEE is particularly useful for converting observed aerosol mass into an equivalent optical depth ( $\tau$ ) for computing direct aerosol radiative effects (DARE – units of  $\text{W m}^{-2}$ ) essential to climate research (e.g., Myhre et al., 2001; Hansell et al., 2010). In previous works, this parameter has been determined experimentally using both field and laboratory measurements (e.g., Li et al., 1996; Maring et al., 2000; Clarke et al., 2004) and through model calculations (e.g., Hand et al., 2002; Quinn et al., 2004; Malm et al., 2005) at the visible wavelengths.

Prior research has demonstrated that MEE varies widely depending on the method employed and the conditions under which it is measured or calculated. Hand et

### A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**A sensitivity study of dust mass extinction efficiencies**

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



al. (2007) for example, conducted an extensive survey of ground-based estimates of visible ( $\lambda \sim 0.55 \mu\text{m}$ ) MEE for various aerosol types and size modes using published literature since 1990. Hand et al. (2007) showed that MSE (a major component of MEE at visible wavelengths) for fine and coarse-mode dust, varied from  $1.2 \pm 0.3$  to  $0.9 \pm 0.8 \text{ m}^2 \text{ g}^{-1}$  for theoretical and measurement methods, respectively.

Besides the reported variability in MEE at the visible wavelengths, there is limited or virtually no information on MEE in the near to thermal IR, which was the impetus for this study. In the IR, MEE is important for dust remote sensing applications, including Advanced Very High Resolution Radiometer (AVHRR) retrievals of sea surface temperatures (SST – for example, Arbelo et al., 2005), a topic later addressed in Sect. 5, and the retrieval of surface parameters and water vapor using Atmospheric Infrared Sounder (AIRS) spectra (e.g., DeSouza-Machado et al., 2006). In the IR, MEE can also be used to better characterize the longwave (LW) radiative energetics of the atmosphere. This work represents to the best of our knowledge, the first time that MEE for dust aerosol has been quantified over such a broad range of parameters.

The underlying goal of this study is to use combined analytical and numerical light-scattering models to build a well-defined, spectrally resolved dataset of plausible dust MEE values, as a function of particle chemistry, asphericity, and size, at key remote sensing wavelengths that span the near-IR ( $0.87\text{--}3.75 \mu\text{m}$ ) and thermal IR window ( $8\text{--}12 \mu\text{m}$ ) regions. This study analyzes dust properties that are considered extreme (e.g., single mineral compositions with strong absorption and large particle sizes and aspect ratios) to identify limiting cases of MEE and those properties that are more routinely observed in nature, to construct a full spectral envelope of MEE. Moreover, dust MEE values are compared with those computed at  $\lambda = 0.87 \mu\text{m}$  to help bridge the optical properties between the visible and IR wavelengths. Supplemental datasets of MEE/MAE for several key minerals (e.g., quartz) are available online. For access to the full MEE/MAE mineral datasets, please contact the authors.

Invariably, there are uncertainties in the model studies of light scattering, due in part to limitations in the numerical schemes employed and assumptions made for charac-

terizing particle properties (Nousiainen, 2009). Although an exact dust model is still far too complex to simulate (i.e., one that fully accounts for surface roughness/porosity, mineral partitioning, orientation, etc.), simplifications are made to best represent airborne dust particles employing common microphysical and chemistry parameterizations. Because global dust properties are so large and varied, we limit our analyses to the following:

1. Only the MEE and its component MSE and MAE of pure dust minerals are examined, excluding the possibilities of coatings by other aerosols (e.g., soot sulfates, etc.), as in the case of aged or transported dust.
2. The MEE is computed for common remote sensing wavelengths between  $\lambda=0.870\text{--}12\ \mu\text{m}$ .
3. The MEE of single mineral dust particles are mainly evaluated to address extreme cases in particle composition, that is, the full envelope of possible values is determined.
4. The Hess/OPAC dust parameterization and a two-component silicate-hematite dust mixture are also evaluated to address mineral compositions more frequently employed in contemporary dust research.
5. Size distributions have volume median diameters (VMD) in the range of  $1.6\text{--}20\ \mu\text{m}$ , with a baseline geometric standard deviation ( $\sigma_g$ ) of 2, which is later tested with a range in  $\sigma_g$  from  $1.7\text{--}2.3$ .
6. Irregular dust shapes are represented by common polyhedral geometries.
7. Although single dust shape distributions are mainly used, two possible shape scenarios are investigated: background dust and dust storm.

Potential benefits of this study include (1) promoting further insight into LW contributions of dust DARE, (2) allowing for improved retrievals of surface parameters (e.g.,

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



SST) and water vapor and (3) providing a reference by which field derived MEE data (e.g., from bulk mass and light scattering measurements) can be compared to, thus allowing for some improved measure of data interpretation.

The paper is arranged as follows: the dust chemistry and microphysical parameterizations pertinent to this study are presented in Sect. 2; an overview of the theory and numerical scheme used to compute the dust MEE is given in Sect. 3; the computational results and their implications are examined in Sect. 4, and finally a discussion and summary are given in Sects. 5 and 6, respectively.

## 2 Physicochemical properties of mineral dust

### 2.1 Mineralogy

Interactions of LW radiation with airborne minerals primarily occur due to the fundamental vibrational modes of the component dust molecules, where the number, intensity, and shape of the modes are dependent on the atomic masses, interatomic force fields, and molecular geometry (Salisbury et al., 1991). The optical constants of many common dust minerals that describe these interactions are well documented (e.g., Roush et al., 2007; Glotch et al., 2007).

For this study, the following major mineral classes were selected to characterize dust particle composition: silicates, clays, carbonates, sulfates, and iron oxides. Although other mineral classes abound in nature, (e.g., phosphates, oxides, sulfides, halides, etc.), literature surveys of dust chemistry from both laboratory and field measurements (e.g., Reid et al., 2003a; Matsuki et al., 2005; Kandler et al., 2006; Formenti et al., 2008; Chou et al., 2008) suggest these are the dominate classes. The refractive index datasets for the minerals employed in this study including their spectral ranges and reference sources are listed in Table 1. The minerals include: (1) quartz, muscovite, chlorite, and the clays kaolinite, montmorillonite, and illite, all from the silicate group, (2) calcite (i.e., calcium carbonate or limestone) and dolomite (calcium-

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



magnesium-carbonate) from the carbonate group, (3) the sulfate gypsum (hydrated-calcium-sulfate), and lastly, (4) the iron-oxide, hematite. The global significance of these minerals have been corroborated by numerous studies of dust samples from the Saharan desert during the PRIDE, SHADE, SAMUM, and AMMA field campaigns (Reid et al., 2003a; Formenti et al., 2003, 2008; Kandler et al., 2006; Chou et al., 2008, respectively), and Northern China (Arimoto et al., 2006; Jeong et al., 2008), two of the world's largest dust sources.

Prominent spectral features of these dust minerals are depicted in Fig. 1, where the imaginary component of the refractive index ( $k$ ), which is related to the absorption coefficient ( $\alpha'$ ) via the dispersion relation:

$$k = \frac{\alpha' \lambda}{4\pi} \quad (1)$$

is plotted as a function of wavelength ( $\lambda$ ) from 0.20–12.5  $\mu\text{m}$  (note, the wavelengths used in surface remote sensing applications are enclosed in light gray boxes for reference).

At  $\lambda \leq 8 \mu\text{m}$  for example, Fig. 1a shows several strong absorption bands including gypsum (red curve) likely attributed to combination tones of the sulfate ion and perhaps water ( $\lambda \approx 2.8, 4.6,$  and  $6 \mu\text{m}$ ), and those due to the carbonate ion in calcite and also dolomite at  $\lambda = 7 \mu\text{m}$  (green and blue curves, respectively – both scaled down  $5\times$ ). Weak absorbers are shown in Fig. 1b over the same spectral range.

Arguably, the most commonly observed spectral features can be found in the silicates, the largest mineral group, across the thermal IR window region. Here the phyllosilicate (e.g., clays and micas) and tectosilicate (e.g., quartz and feldspars) minerals compose much of the observed fraction of airborne dust. For example, the fundamental asymmetric stretching vibrations of the Si-O bonds ( $\nu_2$ ) give rise to the classic absorption feature of quartz centered at  $9.2 \mu\text{m}$  (dashed blue curve – Fig. 1c). Gypsum (red curve) centered at  $8.7 \mu\text{m}$  is also dominant but varies with spectral position, strength, and shape. Lastly, Fig. 1d illustrates the complex spectral features associated with common clay minerals and the mica, muscovite. Although wavenumbers ( $\nu$  – in  $\text{cm}^{-1}$ )

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



are typically employed in IR studies, we continue to use units of wavelength ( $\lambda$  – in  $\mu\text{m}$ ).

To elucidate the effects of strong absorption (i.e., particles with extreme refractive indices) on MEE, we mainly focus on single mineral dust particles. This is important, since large absorption features of individual minerals tend to average out in heterogeneous mixtures. Dust MAE is later examined in this study to help explain these strongly absorbing regions. The significance of evaluating the light scattering properties of individual minerals was also recently pointed out by Nousiainen et al. (2009).

In addition to single mineral dust particles, and to represent more complex dust mineralogies, we further examine the impact on MEE using the frequently used Hess/OPAC dust parameterization (Hess et al., 1998) for transported dust, and a two component internal dust mixture composed of the silicates (quartz, kaolinite, montmorillonite, and illite) and hematite (90% and 10%, respectively). The latter is determined by applying the Maxwell-Garnett (MG) Rule. Note that although the MG rule cannot predict the influence of cationic substitutions within crystals, which can lead to changes in the positions of spectral features, this will not impact the results of this study. Although recent estimates by Lafon et al. (2006), Formenti et al. (2008) and Lazaro et al. (2008), for example, report that the iron oxide content in mineral dust should not exceed 5%, we employ a 10% mixture as an extreme case of particle composition to help identify the bounds of the MEE spectral envelope. The hematite mixtures may be representative of Saharan dust, where hematite is commonly found (Linke et al., 2006).

The significance of birefringence (i.e., a particle's variable dielectric properties along each of its crystallographic directions) on the scattering of calcite flakes has been recently reported by Nousiainen et al. (2009). To account for a mineral's birefringent properties, we follow the work of Long et al. (1993) and compute an average of the refractive indices over each crystallographic direction, assuming randomly oriented particles. This procedure was performed for quartz, calcite, muscovite, hematite, and dolomite.

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





## 2.2 Particle size

Dust particle size is usually characterized as being log-normally distributed (D'Almeida et al., 1991; Seinfeld, 1998) either in terms of its particle number concentration [dN/dlog10(*r*)], surface area [dA/dlog10(*r*)] or volume [dV/dlog10(*r*)]. In many cases, particle sizes are distributed over several size modes, depending on such factors as geographic location, the age of the dust plume and the interactions of dust with other aerosols. The partitioning of size modes may be due to contributions from either fine or coarse mode dust particles, i.e., those with effective radii ( $r_{\text{eff}}$ ) less than or greater than 0.4  $\mu\text{m}$ , respectively (Reid et al., 2003b, 2008). To assess the impact of extreme size parameters on MEE, coarse-mode normalized volume size distributions based on the lognormal expression:

$$n_N(D_p) = N \left( (2\pi)^{1/2} \ln(\sigma_g) D_p \right)^{-1} \exp \left[ -\ln(D_p/r_g)^2 / 2 \ln(\sigma_g)^2 \right] \quad (2)$$

are constructed, where *N* is the particle number concentration and  $r_g$ , and  $\sigma_g$  are the effective radius and geometric standard deviation, respectively. The size distributions are consistent with measurements from past field campaigns; For example, PRIDE and UAE<sup>2</sup> (Reid et al., 2003b, 2008, respectively), SAMUM (Kahn et al., 2008; Schladitz et al., 2009), and AMMA, where Haywood et al. (2008) reported on results from AMMA/SOP0-DABEX and Zipser et al. (2009), focused on the NASA extension of AMMA (NAMMA) at the Cape Verde Islands.

Following the work of Reid et al. (2003b, 2008), we use the volume median diameter (VMD) as our size metric for dust. The computed VMD for this study include: 1.6, 3.0, 6.0, 9.0, 12.0, 18, and 20  $\mu\text{m}$ , although observations all place the VMD of coarse-mode dust in the 1.5–9  $\mu\text{m}$  range with a majority of reported values between 3–6  $\mu\text{m}$  (Reid et al., 2003b, 2008). All volume size distributions are then converted to mass spectra by multiplying the volume with the appropriate mass density ( $\rho$ ) of each mineral. Consistent with prior observations of dust particle size (Reid et al., 2003b), a baseline  $\sigma_g$  of 2.0 is employed for all calculations which we later adjust to test its effect on MEE.

### A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

## 2.3 Particle morphology

Dust particles are rarely spherical as evidenced from numerous prior works (e.g., Okada et al., 2001; Kalashnikova et al., 2002, 2004; Reid et al., 2003a; Kandler et al., 2008; Otto et al., 2008). Moreover, natural dust particles are found to be angular and jagged, likely due to the tendency of flaking of clay minerals and the dust particle's propensity to form aggregates, i.e., clusters of internally mixed minerals.

This study investigates the effect that particle asphericity has on dust MEE by employing a diverse but representative collection of dust particle morphologies, ranging from various axisymmetric geometries to those that are highly irregular. These dust shapes are based on observed microphysical parameters from field studies (e.g., Okada et al., 2001; Reid et al., 2003a; Kandler et al., 2008; Chou et al., 2008), shape information from previously published literature, and various mineralogical datasets that are publicly available via the world-wide web (e.g., <http://webmineral.com> and <http://mindat.org>).

Depending on a mineral's internal structure, particle shapes may take on various forms (Griffen, 1992). For example, calcite can display a variety of crystal habits including acute rhombohedra (Farmer, 1974), or prisms (<http://mindat.org>), while clays tend to form flat plates (Reid et al., 2003a). Although realistic dust particle morphologies and their distributions are far more complex, we baseline our study by analyzing monodispersed shape distributions (SD) of common geometrical shapes related to the minerals' crystal habits. Later in Sect. 4, the sensitivity of MEE to polydispersed SD is investigated.

In total, nine basic shapes are investigated: spheres, oblate and prolate spheroids, hexagonal columns and plates, hexahedrons (cubes and rectangles), tetrahedrons and irregular grains. The hexagonal and hexahedral structures make up the primary shapes used in this study (i.e., those that closest resemble reality and are nearest to what is known/documented), with the remaining shapes being secondary, since these too are possible and are commonly used in contemporary research. The rational and physical

### A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



bases for the shapes, along with particle densities are presented in Table 2.

### 3 Theory and numerical scheme

Hand et al. (2007) describe the theory for calculating MEE of aerosol particles. For convenience, a summary of the theoretical approach in the context of a uniform, homogenous dust mixture is provided, followed by methodology.

#### 3.1 Theoretical approach

The bulk single-scattering properties at wavelength  $\lambda$  for a homogenous ensemble of randomly oriented dust particles having identical shape parameters can be computed if the distribution of particle sizes is known. For a given number distribution  $n_N(D_p)$  in the size range  $D_{p1}$  to  $D_{p2}$ , and mineral composition specified by the complex refractive index ( $m$ ), the extinction coefficient ( $\beta_e$  – in units of  $\text{cm}^{-1}$ ) for dust can be written as:

$$\beta_e = \int_{D_{p1}}^{D_{p2}} \frac{\pi}{4} D_p^2 Q_e(m, D_p) n_N(D_p) dD_p \quad (3)$$

where  $Q_e$ , the optical extinction efficiency, is equal to the ratio of the extinction cross section ( $\sigma_e$ ) to the projected area of a volume-equivalent sphere:

$$Q_e(m, D_p) = \frac{4\sigma_e}{\pi D_p^2} \quad (4)$$

Note the wavelength dependency is implied in Eqs. (3) and (4). If the mineral density ( $\rho$ ) is known, Eq. (3) can be rewritten in terms of a mass distribution  $n_M(D_p)$ , and when normalized by the total mass concentration ( $M$ ), the dust MEE ( $\alpha_{\text{ext}}$  – units of  $\text{m}^2/\text{g}$ ) at

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



wavelength  $\lambda$  is defined as:

$$\alpha_{\text{ext}} = \int_{D_{p1}}^{D_{p2}} 1.5 \frac{Q_e(m, D_p)}{\rho D_p} n_M(D_p) dD_p \quad (5)$$

where the single particle MEE ( $\alpha_{\text{sp}}$  – units of  $\text{cm}^2/\text{g}$ ) is given by:

$$\alpha_{\text{sp}} = 1.5 \frac{Q_e(m, D_p)}{\rho D_p} \quad (6)$$

- 5 Similarly, the above equations can be employed to calculate both the single particle MSE and MAE.

## 3.2 Methodology

### 3.2.1 Light-scattering codes

To investigate the effects of particle asphericity on dust MEE, three light scattering codes are employed: Lorenz-Mie, T-matrix, and Discrete Dipole Approximation (DDA). The first two methods, used to simulate rotationally symmetric and smooth particles (e.g., spheres, spheroids, and cylinders), are fully described in Mishchenko (1994, 1998). Similar to the Finite Difference Time Domain (FDTD) method (e.g., Yang and Liou, 1995), DDA (e.g., Draine and Flatau, 2004) is a numerical technique for solving the electromagnetic scattering problem used to compute the single-scattering properties of irregularly shaped, inhomogeneous particles.

This study uses DDSCAT program version 6.1 (Draine and Flatau, 2004) for computing the optical extinction efficiencies ( $Q_e$ ) of irregularly-shaped dust particles. In brief, the DDA method discretizes an arbitrarily shaped particle into an array of point dipoles (i.e., polarizable points) on a cubic lattice, which interact with a monochromatic plane wave characterized by wavelength  $\lambda$  and incident polarization vector  $\mathbf{e}_0$ . The

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



computed single-particle extinction efficiency ( $Q_e$ ) averaged over random orientations of the particle is given by:

$$\langle Q_e \rangle = \frac{1}{8\pi^2} \int_0^{2\pi} d\delta \int_{-1}^1 d\cos\Theta \int_0^{2\pi} d\phi Q(\delta, \Theta, \phi) \quad (7)$$

where angles  $\delta$ ,  $\Theta$ , and  $\phi$  specify the particle's orientation in the lab frame. Considering the point symmetry of our particle shapes and the demanding computational requirements of DDA over all prescribed dust parameterizations and wavelengths,  $\langle Q_e \rangle$  was computed by averaging over a total of  $N=12$  orientation angles. Sensitivity of the model results to an increase in particle orientations (e.g.,  $N=1050$ ) for an asymmetric kaolinite-hematite grain mixture, for example, reveals absolute differences in  $\langle Q_e \rangle$  [ $\langle Q_e \rangle_{N=12} - \langle Q_e \rangle_{N=1050}$ ]  $\leq 0.08 \text{ m}^2 \text{ g}^{-1}$  (Fig. 2), with the maximum difference corresponding to the mineral's peak absorption bands. Since all particles in this study with the exception of irregular grains are rotationally symmetric, we expect any errors with using a reduced set of orientation angles to be at most  $\sim 0.08 \text{ m}^2 \text{ g}^{-1}$  across the thermal IR.

Following Draine (2000), accurate DDA calculations of the optical cross-sections (within several percent) are achieved if (1) an adequate number of dipoles ( $N$ ) are specified ( $N > 10\,000$ ), (2) the inter-dipole separation ( $d$ ) is smaller than the wavelength of incident radiation ( $\lambda$ ):  $|m|k' \cdot d < 1$ , where  $m$  is the particle's complex refractive index, and  $k'$  is the free-space wave number ( $2\pi/\lambda$ ), and (3) the refractive index is not too large:  $|m - 1| < 2$ .

The above criteria are illustrated in Fig. 3 assuming  $N=10\,001$  dipoles, where Fig. 3a and b show the maximum inter-dipole separation and extreme refractive index ( $m$ ) test, respectively, for select minerals across the window region. Although gypsum slightly exceeds the  $m$  test threshold at  $9 \mu\text{m}$  (Fig. 3b), the error should not significantly impact the MEE results. All DDA computations are performed using  $N > 10\,001$  dipoles.

**A sensitivity study of dust mass extinction efficiencies**

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper

### 3.2.2 Numerical approach

For this study we compute MEE and MAE at discrete wavelengths from the near to thermal IR. Model simulations are evaluated at the wavelengths  $\lambda=0.87, 1.04, 1.6, 2.12, 3.75, 8, 8.6, 9, 10, 11,$  and  $12\ \mu\text{m}$  (Lorenz-Mie and T-matrix) and  $\lambda=8, 8.5, 9, 9.5, 10, 10.5, 11, 11.5, 12,$  and  $12.5\ \mu\text{m}$  (DDA). These wavelengths were chosen since they are commonly used in ground and satellite-based remote sensing such as those from AERONET (Holben et al., 1998) and the MODIS and MISR programs (e.g., Levy et al., 2007; Kahn et al., 2007). Because dust tends to be spectrally flat in the visible,  $\lambda=0.87\ \mu\text{m}$  can represent wavelengths down through the green and its use avoids the extreme computational cost at the shorter wavelengths. This point is later addressed in Sects. 4 and 5. Furthermore, wavelengths at  $\lambda=12\ \mu\text{m}$  reach the most commonly used satellite IR bands.

The refractive indices of all mineral datasets (Table 1) are pre-processed to include only the selected wavelengths. Exceptions made are for those minerals where there was little or no information available on the refractive indices at the near-IR wavelengths, including muscovite, dolomite, calcite, and chlorite. These minerals were therefore only evaluated from  $\lambda=3.75\text{--}12\ \mu\text{m}$ . For illite and kaolinite, we combined the near-IR and IR datasets from T. Roush into one spectral dataset. As previously noted (Sect. 2.1), effective refractive indices were computed for birefringent minerals, and the MG Rule was applied to create two component internal mixtures of silicates and hematite.

The Lorenz-Mie and T-matrix light scattering codes were employed for particle sizes in the range of  $0.05\text{--}12\ \mu\text{m}$ , for spheres and spheroids, respectively. Aspect ratios for spheroids were varied as follows: oblate (1.4, 1.8, 2.3, and 2.8), and prolate (0.3, 0.5, and 0.8). Limitations in the size parameter for DDA ( $\chi < 15$ ) imposed additional constraints for accurately computing  $\langle \alpha_{\text{sp}} \rangle$  for coarse-mode particles at the visible wavelengths. For this reason, we only use DDA to compute the size integrated MEE spectra for each discrete shape in the thermal IR. However, to help understand the dis-

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



create shape effect on MEE at visible wavelengths ( $\lambda=0.86\ \mu\text{m}$ ), we use the computed  $\langle\alpha_{\text{sp}}\rangle$  from Kalashnikova et al. (2004) for several angular shapes composed of a 10% hematite-quartz mixture.

The bulk MEE ( $\langle\alpha_{\text{ext}}\rangle$ ) for a monodispersed SD was numerically computed for each set of dust parameters at each wavelength ( $\lambda$ ) using the expression:

$$\langle\alpha_{\text{ext}}\rangle = \sum_{i=\lambda_1}^{\lambda_2} \left\{ \sum_{j=D_{p1}}^{D_{p2}} (\alpha_{\text{sp}})_j (n_M(D_p))_j \cdot \Delta D_p \right\} \quad (8)$$

where  $j$  is a summation over particle size ( $D_p$ ) and  $\alpha_{\text{sp}}$  is the single-particle MEE. The bulk MAE was computed in a similar manner. For polydisperse SD, such as those used to assess the two possible dust scenarios described in Sect. 2, we weight  $\langle\alpha_{\text{ext}}\rangle$  according to how much each mineral habit contributes to the total MEE.

The total parameter space for dust MEE is given by 13 possible mineral compositions (9 pure minerals+3 silicate-hematite mixtures+Hess/OPAC dust parameterization), 14 particle morphologies (6 angular+7 spheroidal+1 spherical), 7 particle sizes, and 11 channels covering the near-IR and IR regions of the spectrum, including the 10 sub-divided window channels. This yields a total of 13,468 possible dust MEE values employed in this study.

## 4 Model results

First the significance of dust absorption (MAE) on the total MEE over the thermal IR is examined. Next, plausible ranges and trends of MEE are presented, followed by its sensitivity to the dust parameterizations. Lastly, spectral MEE are compared to identify relationships in the optical properties.

### A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## 4.1 Dust absorption

MAE plays a major role in the extinction properties of mineral dust throughout the thermal IR, yet is nearly zero across the near-IR ( $MSE \gg MAE$ ), except when hematite is added to the mixture (not shown). Examples of prominent absorption features for common minerals in the IR are clearly illustrated in Fig. 4. Here we show normalized MAE ( $\times 100\%$ ) for several representative silicates (quartz, kaolinite, illite), sulfate (gypsum), mica (muscovite), and for reference, the OPAC/Hess dust parameterization. The color bar represents the percentage of particle extinction due to absorption and the horizontal and vertical axes are the particle size (VMD) and wavelength ( $\lambda$ ), respectively. Noteworthy are the regions of enhanced MAE (color-coded red and yellow), which are later referred to as “hot-spots” or areas that are characterized by the minerals’ strong absorption features (reststrahlen bands), and their dependency on VMD. Pockets of weaker absorption (color-coded blue) are those regions marked by corresponding increases in scattering or MSE. The partitioning of the dust particle’s MAE and MSE is also shown as a function of VMD. The MAE distribution for quartz (Fig. 4a), for example, distinguishes three distinct regimes of particle absorption, which includes the resonant peaks near 8.3, 9.2 and 12  $\mu\text{m}$ , separated by a scattering region from  $\lambda=10\text{--}11 \mu\text{m}$ , where particle absorption is nearly zero (refer to Fig. 1c).

Apparent at the IR wavelengths is the reduction in fractional MAE as VMD increases, which means that scattering generally contributes more to the MEE of larger size particles; this being analogous to the simple Fresnel reflectance of a solid surface (Salisbury, 1991). Compared to quartz, the clays kaolinite (Fig. 4b) and illite (Fig. 4c), and the mica muscovite (Fig. 4d) exhibit broader spectral ranges of particle absorption throughout much of the window region. Interestingly, the MAE distribution for Hess/OPAC (Fig 4e) is similar to that of the quartz and clays, which comes as no surprise since Hess/OPAC is essentially a heterogeneous dust mixture consisting of the silicate minerals (note that the refractive indices of OPAC/Hess are predominantly derived from D’Almeida (1991), which in turn reference Shettle and Fenn (1979) and Volz (1973).

### A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





We also plot the sulfate gypsum (Fig. 4f) which exhibits strong absorption around 8  $\mu\text{m}$  and then transitions over to a region dominated mostly by scattering.

## 4.2 Ranges in Dust MEE

Following Eq. (8), dust MEE values were computed and subsequently grouped according to wavelength to determine a maximum plausible range of MEE for the channels investigated. Note the discussion that follows reflects the entire parameter space over which this study was conducted, and illustrates the impact of extreme dust chemistry and microphysics on MEE. Numerical tables of MAE/MEE for the common dust minerals kaolinite, gypsum and quartz, are publicly available on-line. The full datasets can be provided upon request to the author.

Figure 5a illustrates the variability in MEE over all spectral channels (near-IR-IR), where the values at each wavelength represent the maximum MEE over the entire range of the seven particle sizes for each composition and shape combination (13 minerals and shapes, respectively). Figure 5b is an enlarged view of the same plot but in the thermal IR showing the minerals which correspond to the maximum MEE at each wavelength. For convenience, the curves are color-coded according to particle morphology: white for spheroids (oblate/prolate), red for spheres, and yellow for angular particles. In addition, the Hess/OPAC parameterization for three particle morphologies (blue curves) are shown for comparison. Note the MEE values between channels are interpolated and therefore do not have any physical meaning.

In Fig. 5a, two dominant peaks are clearly evident: one at  $\lambda=0.87\ \mu\text{m}$  and the other at  $\lambda=9.0\ \mu\text{m}$ , with maximum MEE values for all shape/composition combinations clustered near  $\alpha_{\text{ext}}=1.18$  and  $1.28\ \text{m}^2\ \text{g}^{-1}$ , respectively (note the  $0.87\ \mu\text{m}$  peak does not include angular particles). A third smaller peak is also apparent at  $\lambda=10\ \mu\text{m}$ . The MEE differ by about an order of magnitude with minimum values falling below  $\alpha_{\text{ext}}=0.1\ \text{m}^2\ \text{g}^{-1}$ . Both maxima are attributed to non-spherical particles (oblate spheroids) with the first being mostly composed of gypsum, with contributions from the clays illite, kaolinite, and montmorillonite, and also the clay-hematite mixture, since

### A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



hematite is a strong absorber at the visible/near-IR wavelengths. Although quartz does contribute to the first peak, its presence mainly dominates the second maximum due to the strong absorption band centered at  $9.2\ \mu\text{m}$  (Fig. 5a).

Interestingly in Fig. 5b, the resonance peak for a quartz sphere (dashed red curve) appears to be blue-shifted by almost  $0.5\ \mu\text{m}$  with respect to a quartz non-sphere (e.g., an oblate spheroid – dashed white curve), but is also observed for the quartz angular particles (yellow curves) as well (this is clearly shown later in Fig. 7). Note the black arrow denotes the spectral shift between the quartz particles. A large spectral shift was also detected for the clay minerals, where montmorillonite for example, which has a strong absorption peak around  $\lambda=9.6\ \mu\text{m}$  (Manghnani et al., 1964) showed spheres and angular particles to differ by nearly  $1\ \mu\text{m}$ . Although the observed spectral shifts are likely to be overestimated due to the coarse resolution in the computed MEE spectra, the results clearly demonstrate, similar to that reported by Hudson et al. (2008), that Mie simulations can not accurately reproduce the peak positions of silicate minerals, i.e. quartz ( $9.2\ \mu\text{m}$ ) and clays ( $\sim 10\ \mu\text{m}$ ). On the other hand, non-spherical shapes are able to better reproduce the minerals' true spectral features and should be used when modeling dust aerosol.

If we restrict dust particle size to what is commonly measured in the field, i.e. VMD between  $3\text{--}6\ \mu\text{m}$  (Reid et al., 2003b), and use an aspect ratio of 1.8 for oblate spheroids, consistent with observations (Reid et al., 2003a; Chou et al., 2008), then a more representative range of MEE spectra are given as shown in Fig. 6, where panels (a–c) are for a VMD of  $3.5$ ,  $4.5$ , and  $5.5\ \mu\text{m}$ , respectively. As before, the curves are color-coded according to particle morphology and the blue curves depict the Hess/OPAC parameterization. The same two dominant peaks at  $\lambda=0.87\ \mu\text{m}$  and  $\lambda=9.0\ \mu\text{m}$  (including the third smaller peak at  $\lambda=10\ \mu\text{m}$ ) have maximum MEE values approaching nearly  $\alpha_{\text{ext}}=0.9$  and  $0.8\ \text{m}^2\ \text{g}^{-1}$ , respectively, with minimum values about an order of magnitude smaller. The bifurcation in the spheroidal MEE spectra (white curves), are due to the extreme differences in aspect ratios, where the upper/lower groups represent aspect ratios of 1.8 and 0.5 (prolate), respectively. The MEE spectra

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



for prolate spheroids are smaller due to the larger projected areas.

At the shorter wavelengths ( $\sim\lambda=0.87\ \mu\text{m}$ ), an increase in VMD generally causes the dust MEE to decrease with a change rate that depends on both particle morphology and chemistry. For example, the MEE for all spherical mineral particles vary as  $\text{VMD}^{-a}$ , where  $a\approx 1$  (Reid et al., 2003b), however in the case of spheroids, “ $a$ ” is found to vary more slowly (i.e.,  $a<1$ ) which becomes smaller ( $a\ll 1$ ) as the asphericity of the particles increase (e.g., aspect ratio goes from 1.4–2.8 or 0.5–0.3). In the thermal IR on the other hand, all dust particles, independent of morphology, seem to exhibit a relatively slower decrease in MEE ( $a\ll 1$ ) when the VMD becomes larger, and in some cases the MEE is found to even increase with VMD (e.g., kaolinite). In the IR, “ $a$ ” is also sensitive to wavelength since the absorptive properties of each mineral exhibit a strong spectral dependence.

### 4.3 Sensitivity of MEE to dust microphysics and chemistry

#### 4.3.1 Preliminary assessments and trends

Examples depicting changes in MEE in response to perturbations in the dust physicochemical properties are illustrated in Fig. 6. Presented are MEE surface plots corresponding to each combination of dust parameters, where the rows and columns represent particle mineralogy and morphology, respectively. Here the VMD is defined for a coarse-mode size distribution with the baseline geometric standard deviation ( $\sigma_g$ ) of 2.0 which we later adjust to assess the corresponding changes in MEE (see Sect. 4c). The color bar depicts the MEE intensity where an upper value cutoff of  $\alpha_{\text{ext}}=0.6\ \text{m}^2\text{g}^{-1}$  was chosen to help resolve the fine structure detail in the MEE distributions.

The panels share common regions which exhibit higher MEE intensity values ranging from  $\sim 0.3$  to  $>0.6\ \text{m}^2\text{g}^{-1}$  which, like the MAE distributions in Fig. 4, are referred to as “hot-spots”. These sharply contrast against the background MEE which are typically  $\leq 0.2\ \text{m}^2\text{g}^{-1}$ . The dark central bands from about 6–8  $\mu\text{m}$  are likely numerical artifacts due to interpolation of points between 3.75 and 8.0  $\mu\text{m}$  (gray boxed regions). A quick

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



inspection of the panels immediately reveals several interesting features.

1. The shapes and positions of hot-spots vary depending on mineral type, and the particle's respective size and shape. Particularly notable are the differences between minerals, where the hot-spots are related to absorption band number, position, shape and depth. For example, the quartz prism in Fig. 7h exhibits two hot-spots in the IR: one due to the dominant fundamental asymmetric Si–O–Si stretching vibration near 9.2  $\mu\text{m}$  and another that is less apparent due to the weaker symmetric Si–O–Si stretching vibration around 12  $\mu\text{m}$  denoted by the black arrow (Farmer, 1974). Evidently, the latter region is not so easily discerned in the smooth particles (e.g., compare Fig. 7e through g). Kaolinite plates (Fig. 7i), on the other hand, have one hot-spot centered near 10  $\mu\text{m}$ . The central positions of the hot-spots are nearly consistent with the peak vibrational frequencies of each mineral as noted by Karr et al. (1975): quartz (9.2  $\mu\text{m}$ ) and kaolinite (9.6–9.7  $\mu\text{m}$ ).
2. At the shortest wavelength ( $\lambda=0.870 \mu\text{m}$ ), the MEE increases as VMD decreases, since particle size is on the order of the incident wavelength.
3. The hot-spots appear to follow the particle's geometry, particularly spheroids, and are evidence for shape dependency in the optical properties. For example at the shorter wavelengths, the hot-spots associated with spherical particles (Fig. 7a, e, and i) appear to be more rounded and distributed symmetrically over the size and wavelength domains (VMD $\sim$ 1–6  $\mu\text{m}$  and  $\lambda=0.870$ –3.75  $\mu\text{m}$ , respectively), whereas those for oblate spheroids (spheres stretched along the equatorial axis – see Fig. 7c, g, and k) are more elongated with respect to particle size and are more narrowly confined in wavelength. Notable differences in the hot-spots of angular particles are also apparent both in intensity and position (e.g., gypsum – Fig. 7d and quartz – Fig. 7h), and are consistent with the spectral features reported in previously published literature (e.g., Karr et al., 1975; Farmer et al., 1974; Salisbury et al., 1991). For example, the spectral shifts in MEE between spheres and angular quartz particles (Fig. 5b) can be seen by comparing

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**A sensitivity study of dust mass extinction efficiencies**

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Fig. 7f and h. Generally, sharp-edged particles tend to produce wider and more symmetric MEE distribution patterns from about 8–10  $\mu\text{m}$  compared with spheres and spheroids, perhaps due to the edge effects in the optical properties of the particles.

5 Trends noted in the MEE spectra are as follows. At the shorter wavelengths ( $\lambda=0.87 \mu\text{m}$ ), MEE generally tends to increase when going from spheres to spheroids (e.g., Figs. 5 and 6). To evaluate the shortwave effects on MEE due to discrete shapes, we use the computed  $\langle Q_e \rangle$  from Kalashnikova et al. (2004) for a quartz-hematite mixture (10%) consisting of 1  $\mu\text{m}$  sized particles. The single particle MEE for several geometries including plates, irregular grains, tetrahedrons, and rectangles were calculated  
10 and were found to lie in the range of 0.81–1.19  $\text{m}^2 \text{g}^{-1}$ , about 2–3 times greater than that of spheres (e.g., red curves in Figs. 5 and 6). Increases in MEE at short wavelengths are primarily due to enhancements in scattering (MSE), when MAE tends to zero.

15 Across the thermal IR, changes in MEE due to shape are a strong function of VMD and wavelength, particularly if MEE is evaluated at the mineral resonant frequencies where the absorption coefficients are high. At these frequencies, MAE and consequently MEE generally tend to increase when going from spheres to spheroids, particularly for larger particles; however, outside of these strongly absorbing regions, both  
20 MAE and MEE tend to decrease. Similar changes in MEE and MAE are also apparent when going from spheres to the discrete angular shapes. The behavior of MEE in the IR is strongly linked to changes in MAE which ultimately depend on both wavelength and VMD.

25 Next the effects of each parameter on the MEE spectra are examined in the order of particle chemistry, size, and morphology.

### 4.3.2 Particle chemistry

To illustrate the impact of chemistry on dust MEE, we analyze dust grains with a VMD of about  $3\ \mu\text{m}$ , which roughly correspond to the median size of the MEE distributions (Fig. 8a). Note that granular particles have been routinely observed (Reid et al., 2003a) and are commonly employed in optical dust models (Kalashnikova et al., 2005). The MEE spectra for six pure minerals (quartz, gypsum, illite, kaolinite, montmorillonite, and muscovite), one clay-hematite mixture (kaolinite-hematite), and one bulk dust parameterization (Hess/OPAC) are given. Immediately apparent are two dominant peaks in the spectra, one narrowly positioned at  $9\ \mu\text{m}$  and another centered around  $10\ \mu\text{m}$ . Incidentally, a smaller third peak is also visible around  $\lambda=12\ \mu\text{m}$  due to quartz. The second peak is more broadly distributed over wavelength than the first since there are a greater number of absorption bands, particularly for the clay minerals in the range of  $\sim 9\text{--}11\ \mu\text{m}$ . Note that both quartz and gypsum exhibit the strongest peaks over the thermal IR. This also includes the quartz-hematite mixture (not shown).

Adding hematite to clays, shown by the green curve (squares) for a kaolinite-hematite mixture, decreases MEE in the strongly absorbing region of  $\lambda=9\text{--}11\ \mu\text{m}$ . At  $\lambda=10\ \mu\text{m}$ , for example, MEE is reduced by almost 14% when a 10% hematite inclusion is added to kaolinite (Sect. 2a), although this is likely overestimated since the iron-oxide content in mineral dust typically does not exceed 5% (Lafon et al., 2006). Further analysis of this effect is illustrated in Fig. 9, where the change in MEE (Fig. 9a), MAE (Fig. 9b), and MSE (Fig. 9c) are shown after hematite has been added (i.e.  $\Delta\text{MXE}=\text{MXE}_{\text{hematite}}-\text{MXE}_{\text{no hematite}}$ , where X=E, A, and S, respectively). Here, positive values denote regions of enhanced absorption and scattering due to the presence of hematite. Because kaolinite is a much stronger absorber than hematite in the thermal IR (compare Fig. 1c and d), the addition of hematite increases kaolinite's absorption efficiency (Fig. 9b) for all particle sizes at wavelengths between  $\lambda=8\text{--}9\ \mu\text{m}$ . This effect is sensitive to particle size where  $\Delta\text{MXE}$  falls off with an increase in VMD. This is also observed, albeit a weaker affect, in the MSE (Fig. 9c). Likewise, where absorption is

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



weaker in kaolinite ( $\lambda \sim 8\text{--}9\ \mu\text{m}$ ), the added hematite increases the kaolinite's absorption efficiency (Fig. 9b). These same patterns are similar for the other clays.

Lastly, it is evident that the MEE spectrum corresponding to the Hess/OPAC parameterization (Fig. 8a, dashed black curve) is a heterogeneous mixture of silicates and clays. Displaying a central peak around  $10\ \mu\text{m}$ , the spectrum resembles those for the clays, particularly illite; however from about  $11\text{--}12\ \mu\text{m}$  the spectrum looks more similar to quartz. For many dust applications in the thermal IR, the Hess/OPAC parameterization represents a reasonable approach for modeling dust; particularly in regions where clays dominate. Where potential problems may arise however, is when the main dust component is either quartz or gypsum which can lead to errors in MEE of up to 100% for wavelengths between  $8\text{--}9\ \mu\text{m}$ . This corresponds to the widely used  $8.6\ \mu\text{m}$  channel used in many remote sensing applications. It is important to point out that these bulk dust models may miss the larger absorption features that are otherwise present in a homogeneous dust mixture. Potential errors may include the retrieval of key dust and surface parameters, and the quantitative assessment of DARE used in climate research.

### 4.3.3 Particle size

To illustrate the impact of particle size on dust MEE, we again choose to analyze granular quartz particles. Figure 8b shows the resulting MEE spectra as a function of particle VMD which varies from  $1.6\text{--}20.0\ \mu\text{m}$  using the reference geometric standard deviation ( $\sigma_g$ ) of 2.0. Note the largest changes in MEE occur at the peak absorbing wavelength ( $\sim \lambda = 9\ \mu\text{m}$ ) for particle sizes with a VMD in the range of  $\sim 1.6\text{--}6\ \mu\text{m}$ . At the remote sensing channels ( $\lambda = 8.6, 11, \text{ and } 12\ \mu\text{m}$ ), MEE sensitivity to particle size is greatest at  $\lambda = 8.6\ \mu\text{m}$ , where absolute differences in MEE can exceed  $0.15\ \text{m}^2\ \text{g}^{-1}$ . For wavelengths between  $\lambda = 8.1\text{--}9.9\ \mu\text{m}$  and greater than  $\lambda = 12\ \mu\text{m}$ , MEE clearly increases as VMD decreases, consistent with the shortwave calculations of Reid et al. (2003b). (It is important to note that Reid et al., 2003b employed spheres and the refractive indices of Shettle and Fenn, 1979). Curiously, the correlation between MEE and VMD was

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



not observed between  $\lambda=10\text{--}12\ \mu\text{m}$ , which may be directly related to the behavior of quartz particles at these wavelengths (refer to discussion on MEE vs. VMD – Sect. 4b).

To assess the sensitivity of MEE to changes in the  $\sigma_g$ , we performed a series of tests in which  $\sigma_g$  was adjusted to  $\pm 0.3$  of the reference value (2.0). The absolute differences in MEE were largest at the wavelengths where peak absorption occurs. For granular quartz and Hess/OPAC dust models, MEE were  $\sim \pm 0.04\ \text{m}^2\ \text{g}^{-1}$  and  $\pm 0.02\ \text{m}^2\ \text{g}^{-1}$  within their reference values at 9 and 10  $\mu\text{m}$ , respectively. Hence the Hess/OPAC model is less sensitive by a factor of about 2 to changes in  $\sigma_g$ , which could be related to the heterogeneity of its dust composition.

#### 4.3.4 Particle shape

To illustrate the impact of particle shape on dust MEE, we again choose quartz particles with a size distribution characterized by the median VMD of  $\sim 3\ \mu\text{m}$ . In Fig. 8c, the results for spheres, spheroids, rectangles, grains, prisms, and the two shape distributions (SD1 and SD2) are given.

Apparent are the large differences in MEE between smooth and angular particles, particularly between 8–10  $\mu\text{m}$ . The spheres and oblate spheroids (OS), for example, exhibit large spectral peaks at 8.5 and 9  $\mu\text{m}$ , respectively, which are not seen in the angular particles, likely due to the edge effects. As previously noted in Sect. 4.1, the Mie solutions for the quartz resonance peak at 9.2  $\mu\text{m}$  is blue-shifted nearly 0.5  $\mu\text{m}$ , and is incorrectly positioned near 8.5  $\mu\text{m}$ . The spheroids and angular particles on the other hand are much closer to the true resonance frequency of quartz.

For angular particles, the sensitivity appears to be largest in the wavelength range of 9–10.5  $\mu\text{m}$ , with rectangles/hexagonal prisms yielding maximum MEE. At 9  $\mu\text{m}$ , for example, absolute differences between angular shapes approach  $\sim 0.2\ \text{m}^2\ \text{g}^{-1}$ . At the remote sensing wavelengths, MEE sensitivity to shape is not as strong, but appears to be largest at  $\lambda=12\ \mu\text{m}$ .

Lastly, we evaluate MEE spectra for two dust scenarios: SD1 (background dust) and SD2 (dust storm) similar to those described in Kalashnikova et al. (2002) and are

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





defined as:

1. SD1 – 20% spheres+50% angular+30% oblate spheroids (background dust)
2. SD2 – 5% spheres+75% angular+20% oblate spheroids (dust storm)

Although dust storms may contain giant-sized particles that exceed our maximum VMD of 20  $\mu\text{m}$ , the size range employed in this study along with the SD2 model, allow for a reasonable characterization of a dust storm's impact on MEE.

For a polydispersed SD, weighting factors are applied to the total MEE corresponding to each mineral habit. For example, in background dust, spheres are mixed with spheroids and angular particles and are weighted by the factors 0.20, 0.30, and 0.50, respectively. Since the SD is a weighted mixture of the mineral habits, the resulting MEE spectra (SD1/SD2) appear to be much smoother (Fig. 8c – red/blue curves). Note that by adding more angular particles to the distribution, the magnitude of MEE spectra increases in the 9–12.5  $\mu\text{m}$  range, whereas between 8–9  $\mu\text{m}$ , the effects of the smooth particles dominate.

#### 4.4 Comparisons of MEE between the near and thermal IR

To identify spectral relationships in the optical properties of dust between the near and thermal IR, ratios of MEE are analyzed over all possible particle compositions and sizes using spheres and spheroids. Since the Hess/OPAC and kaolinite-hematite optical models are frequently applied in dust research (e.g., Balkanski et al., 2007; Hansell et al., 2008; Huang et al., 2009), we specifically focus on these compositions to help illustrate these relationships.

To this end, MEE at the near-IR wavelength of  $\lambda=0.870 \mu\text{m}$  are compared to those at the IR wavelengths (i.e.  $\lambda=3.75, 8.0, 8.6, 9, 10, 11,$  and  $12 \mu\text{m}$ ). Because dust tends to be spectrally flat in the visible,  $\lambda=0.870 \mu\text{m}$  can represent wavelengths down through  $\sim\lambda=0.55 \mu\text{m}$ , and can thus be used as a proxy for estimating optical properties across the visible-IR spectrum. For example, MEE derived from bulk mass and light scattering

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



measurements at the visible wavelengths can be converted to an equivalent in the IR for use in radiation transfer and climate modeling studies. To put these comparisons into context for remote sensing purposes, emphasis is given to the center-wavelengths corresponding to AVHRR channels 3, 4, and 5 ( $\lambda=3.75$ , 10.8, and 12.0  $\mu\text{m}$ , respectively).

Computed MEE ratios ( $\alpha_{\text{IR}}/\alpha_{\text{NIR}}$ ) between the near-IR ( $\lambda=0.870 \mu\text{m}$ ) and IR channels (3.75–12  $\mu\text{m}$ ) are listed in Table 3 for spheres and oblate spheroids (OS – aspect ratio=1.8) using the two prescribed dust compositions, with VMDs of 1.5, 3, and 6  $\mu\text{m}$ . To better illustrate the dependence of particle size on MEE ratios, the data from Table 3 are shown plotted in Fig. 10 for spheres and oblate spheroids (aspect ratio=1.8), using the Hess/OPAC dust parameterization. The gray/black curves denote the spherical/spheroidal particle geometries, respectively, while the markers indicate the AVHRR IR channels. The mean IR/visible optical depth ratio reported by DeSouza-Machado et al. (2006) (0.425) falls within the range presented here ( $\alpha_{\text{IR}}/\alpha_{\text{NIR}}=0.3\text{--}0.7$ ) assuming a Hess/OPAC spherical dust model for the commonly observed VMDs of 3–6  $\mu\text{m}$  at  $\lambda=10.8 \mu\text{m}$  (channel 4).

Apparent is the rapid increase and convergence ( $\alpha_{\text{IR}}/\alpha_{\text{NIR}}=1$ ; broken black line) of the MEE ratios for both shapes as particle VMD increases, an effect attributed to the changing particle size parameter (i.e.,  $\alpha_{\text{NIR}}>\alpha_{\text{IR}}$  for small particles, and  $\alpha_{\text{IR}}>\alpha_{\text{NIR}}$  for large particles). Depending on particle size, the ratios display a shape dependency, where spheroids tend to have a greater impact (i.e., larger MEE) at visible wavelengths ( $\alpha_{\text{NIR}}/\alpha_{\text{IR}}\geq 1$ ) than do spheres for particle sizes with VMDs  $\leq \sim 9 \mu\text{m}$ . For larger particles (VMDs  $> \sim 9 \mu\text{m}$ ) however,  $\alpha_{\text{NIR}}/\alpha_{\text{IR}}\leq 1$  and the MEE ratios are nearly insensitive to shape. The magnitude of the ratio effectively tracks the relative significance of dust extinctive properties between the visible and IR wavelengths.

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## 5 Discussion

The efficacy of this study can be demonstrated with a simple example. Suppose the research objective is to estimate optical properties in the IR to approximate dust impacts on AVHRR SST retrievals. For simplicity, we assume dust particles are spherical and that MEE at  $0.55\ \mu\text{m}$  can be derived from bulk mass and light scattering/absorption measurements. Following Reid et al. (2003b), an average MEE of  $\sim 0.65\ \text{m}^2\ \text{g}^{-1}$  at  $0.55\ \mu\text{m}$  is implied for Saharan dust after adding the contributions from scattering ( $0.5\pm 0.1\ \text{m}^2\ \text{g}^{-1}$ , Maring et al., 2000) and absorption ( $0.08\ \text{m}^2\ \text{g}^{-1}$ , personal communication with D. Savoie). Applying the Hess/OPAC dust model for a particle VMD of  $3.0\ \mu\text{m}$  (Table 3), the corresponding MEE at 3.75, 10.8, and  $12.0\ \mu\text{m}$  are estimated to be  $\sim 0.4$ , 0.21, and  $0.20\ \text{m}^2\ \text{g}^{-1}$ , respectively. The MEE in turn translates into a dust IR aerosol optical depth (AOT) of around  $\tau=0.4$ , 0.21, and 0.20 at the three wavelengths, respectively, assuming a column dust load of  $1\ \text{g}\ \text{m}^{-2}$ . Incidentally, the visible AOT ( $0.55\ \mu\text{m}$ ) is  $\tau=0.65$ . Using the estimated channel AOTs and accounting for the atmospheric state, surface properties and dust distribution in a radiative transfer model, the dust effect can be calculated by the difference in brightness temperature (BT) between channels 4 and 5 (i.e., BT4-BT5) of the AVHRR with and without dust. If, for example, the dust top/bottom is 2.0/0.5 km, respectively ( $\Delta z=1.5\ \text{km}$ ), in an atmosphere characterized by a mid-latitude summer profile, the SST will be negatively biased by  $\sim 1\ ^\circ\text{C}$ .

Similar studies can also be performed to estimate perturbations in the longwave energetics of the column atmosphere, which can help not only lead to an improved assessment of DARE, but also the ensuing surface-air exchange processes and general circulation of the atmosphere.

## 6 Summary

Sensitivity analyses were performed over an extended range of dust microphysical and chemistry perturbations, to determine a plausible range of MEE for terrestrial atmo-

### A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



spheric dust, at wavelengths commonly used in remote sensing spanning the near to thermal IR. The following major conclusions were noted:

- In the frequently observed dust size range (VMD=3–6  $\mu\text{m}$ ), two dominant peaks were identified: one at  $\lambda=0.870 \mu\text{m}$  and the other at  $\lambda=9 \mu\text{m}$ , with maximum MEE values reaching nearly  $\alpha_{\text{ext}}=0.90$  and  $0.80, \text{m}^2 \text{g}^{-1}$ , respectively. Both maxima were attributed to non-spherical particles with the near-IR peak composed primarily of gypsum, clay minerals and the clay-hematite mixture. The second peak was mostly attributed to quartz due to the strong Si–O stretch resonance at  $9.2 \mu\text{m}$ .
- Mie spherical solutions for quartz spheres in the thermal IR are blue-shifted by  $\sim 0.5 \mu\text{m}$  compared with spheroids and angular particles. As shown in previous studies, spherical particles are not able to accurately reproduce the resonance peaks commonly found in silicate minerals.
- The shapes in MEE distributions appear to follow particle geometry, particularly for oblate spheroids. This provides more evidence for shape dependency in the optical properties of mineral dust.
- Generally, angular particles have wider and more symmetric MEE distributions from 8–10  $\mu\text{m}$  than those with smooth surfaces, likely due to their edge-effects.
- At shorter wavelengths ( $\lambda=0.87 \mu\text{m}$ ), MEE tends to increase when going from spherical to non-spherical particles. Single particle MEE for several angular geometries was found to be about 2–3 times greater than that of spheres. Increases in MEE at short wavelengths are primarily due to enhancements in scattering (MSE), when MAE tends to zero.
- In the thermal IR, changes in MEE due to particle shape strongly depend on VMD and wavelength, particularly if MEE is evaluated at the mineral resonant frequencies where MEE and MAE generally tend to increase when going from spheres to non-spheres; however, outside of these strongly absorbing regions, MEE and MAE tend to decrease.

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



This study not only bounds the MEE of dust aerosols for the parameter space examined, but it also provides a mechanism for linking the spectral optical properties of dust between the visible and IR wavelengths. Potential applications for the derived data include remote sensing of atmospheric and surface parameters, computing LW DARE, and providing a reference for comparing field derived MEE data.

## Appendix A

### List of key acronyms and symbols

ACE-Asia	Aerosol Characterization Experiment – Asia
AMMA	African Monsoon Multidisciplinary Analyses
DABEX	Dust and Biomass Burning Experiment (during AMMA)
DARE	Direct aerosol radiative effect
MAE	Mass absorption efficiency
MEE	Mass extinction efficiency
MSE	Mass scattering efficiency
NAMMA	NASA African Monsoon Multidisciplinary Analyses
OPAC	Optical properties of aerosols and clouds
PRIDE	Puerto Rican Dust Experiment
SAMUM	Saharan Mineral Dust Experiment
SHADE	Saharan Dust Experiment
SOP0	Special Observing Period 0 (during AMMA)
UAE <sup>2</sup>	United Arab Emirates Unified Aerosol Experiment
VMD	Volume median diameter
$\alpha_{sp}$	Single particle mass extinction efficiency
$\alpha_{ext}$	Mass extinction efficiency

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



$k$	Imaginary component of refractive index
$\alpha'$	Absorption coefficient
$\nu_2$	Fundamental asymmetric stretching vibration
$\nu$	Wavenumber ( $\text{cm}^{-1}$ )
$\beta_e$	Extinction coefficient
$Q_e$	Optical extinction efficiency
$\sigma_e$	Extinction cross section
$m$	Complex refractive index
$k'$	Free-space wave number

**Supplementary material related to this article is available online at:**  
<http://www.atmos-chem-phys-discuss.net/10/17213/2010/acpd-10-17213-2010-supplement.pdf>.

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## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**A sensitivity study of dust mass extinction efficiencies**

R. A. Hansell et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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**A sensitivity study of dust mass extinction efficiencies**

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



- Malm, W. C., Day, D. E., Carrico, C., Kreidenweis, S. M., Collett Jr., J. L., McMeeking, G., Lee, T., Carillo, J., and Schichtel, B.: Intercomparison and closure calculations using measurements of aerosol species and optical properties during the Yosemite aerosol characterization study, *J. Geophys. Res.*, 110, D14302, doi:10.1029/2004JD005494, 2005.
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**A sensitivity study of dust mass extinction efficiencies**

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Wendisch, M., and Trautmann, T.: Solar radiative effects of a Saharan dust plume observed during SAMUM assuming spheroidal model particles, *Tellus B*, 61, 270–296, 2008.

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## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Table 2.** Densities and shapes of common dust minerals.

Mineral	$\rho^a$ g cm <sup>-3</sup>	Crystal habit <sup>a</sup> (documented)	Primary shapes in study
<b>Silicates</b>			
Quartz	2.65	Hexagonal prism; pyramid on end (Huggins et al. 1922)	Hexagonal column (HC) <sup>e</sup>
Kaolinite	2.60	Pseudo-hexagonal crystal plate; could be fibrous or spherical <sup>b</sup>	Hexagonal plate (HP) <sup>e</sup>
Illite	2.75	Pseudo-hexagonal crystal plate <sup>b</sup>	Hexagonal plate <sup>e</sup>
Montmorillonite	2.35	Pseudo-hexagonal crystal plate <sup>b</sup>	Hexagonal plate <sup>e</sup>
Muscovite	2.82	Crude hexagonal cross-section; platy <sup>b</sup>	Hexagonal plate <sup>e</sup>
Chlorite	2.95	Barrel/tabular with hexagonal outline – compact/platy <sup>c</sup>	Hexagonal column <sup>e</sup>
<b>Carbonates</b>			
Calcite	2.71	Rhombohedral (e.g., skewed rectangle). Tabular form, prism; long spiny crystal <sup>d</sup>	Rectangle/Cube
Dolomite	2.87	Rhombohedral <sup>d</sup>	Rectangle/Cube
<b>Sulfates</b>			
Gypsum	2.30	Tabular (rectangular); Bladed rosettes <sup>d</sup>	Rectangle/Cube
<b>Iron-oxides</b>			
Hematite	5.3	Rhombohedral <sup>d</sup>	Rectangle/Cube
<b>Secondary shapes in study</b>			
<b>Tetrahedron</b> – although Si–O bonds are tetrahedral, additional cationic groups usually precludes this configuration. Nevertheless the possibility is considered.			
<b>Grain</b> – In planetary/astrophysical studies, dust is commonly modeled as irregular-sized dust grains (e.g., Kalashnikova et al., 2005; Draine and Weingartner et al., 1996); For this the Draine and Weingartner (1996) model (DW96), an array of 13 identical cubes, is employed.			
<b>Spheroid</b> – analysis of dust samples (Reid et al., 2003a; Okada et al., 2001) reveals particles to usually be oblate spheroids (aspect ratio ~1.4–1.9). Here oblate and also prolate spheroids are examined.			
<b>Sphere</b> – although usually not observed in nature, the possibility is considered; also serves as a reference for past works.			

<sup>a</sup> Data obtained from online mineral databases, past literature, and field studies

<sup>b</sup> Glotch et al. (2007); Reid et al. (2003a); Kalashnikova et al. (2004)

<sup>c</sup> Kerr (1959)

<sup>d</sup> Farmer (1974); <http://mindat.org>

<sup>e</sup> (HC) Aspect ratio  $L/2a_{\text{eff}}=2$  ( $L$ =column length;  $a_{\text{eff}}$ =effective radius); (HP)  $L/2a_{\text{eff}}=0.5$

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**A sensitivity study of dust mass extinction efficiencies**

R. A. Hansell et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Table 3.** MEE ratios ( $\alpha_{IR}/\alpha_{NIR}$ ) – from the near to thermal IR.

Shape	Wavelength/Channel $\lambda$ ( $\mu\text{m}$ )	Kaolinite-hematite mixture VMD ( $\mu\text{m}$ )			OPAC/Hess Parameterization <sup>a</sup> VMD ( $\mu\text{m}$ )		
		1.5	3	6	1.5	3	6
Sphere	3.75/(3)	0.48	0.83	1.11	0.17	0.59	1.00
	8.0	0.03	0.05	0.11	0.05	0.12	0.25
	8.6	0.09	0.14	0.22	0.09	0.22	0.37
	9.0	0.40	0.50	0.63	0.15	0.45	0.83
	10.0	0.38	0.77	1.11	0.15	0.53	1.00
	10.8/(4)	0.31	0.63	1.00	0.08	0.33	0.71
	11.0	0.29	0.63	1.00	0.07	0.28	0.63
	12.0/(5)	0.05	0.19	0.43	0.10	0.31	0.63
	3.75/(3)	0.03	0.14	0.33	0.04	0.14	0.32
	8.0	0.01	0.01	0.02	0.03	0.04	0.07
Spheroid (OS 1.8)	8.6	0.02	0.04	0.06	0.07	0.09	0.14
	9.0	0.13	0.17	0.25	0.08	0.14	0.27
	10.0	0.05	0.14	0.31	0.06	0.14	0.31
	10.8/(4)	0.05	0.11	0.26	0.04	0.08	0.17
	11.0	0.05	0.11	0.24	0.03	0.06	0.14
	12.0/(5)	0.01	0.02	0.05	0.06	0.09	0.18

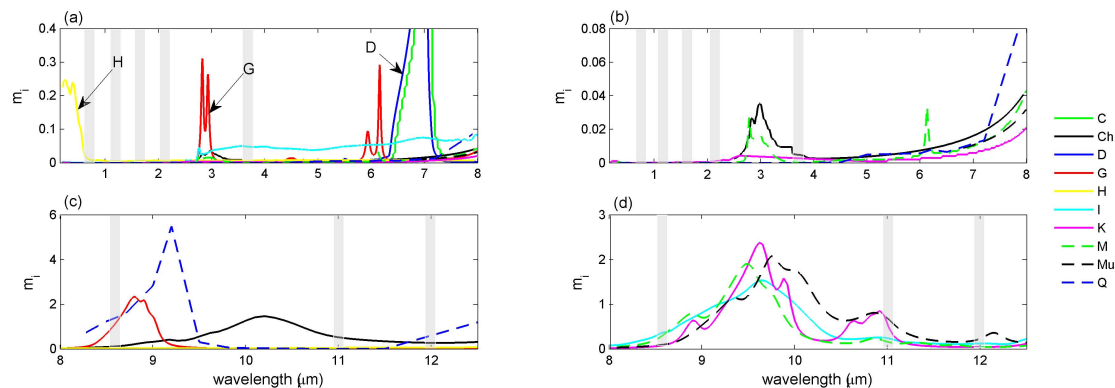
Note: AVHRR channels 3, 4 and 5 are denoted as (3), (4), and (5), respectively

<sup>a</sup> See Fig. 10 for plot of OPAC/Hess MEE ratios using spheres and spheroids.



## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.



**Fig. 1.** Imaginary component of refractive index for common dust minerals, with vertical axes re-scaled to resolve spectral features. Gray shaded regions represent common remote sensing channels. Shown are the minerals calcite (C – solid green), chlorite (Ch – solid black), dolomite (D – solid blue), gypsum (G – solid red), hematite (H – solid yellow), illite (I – solid aqua), kaolinite (K – solid magenta), montmorillonite (M – dashed green), muscovite (Mu – dashed black) and quartz (Q – dashed blue). **(a)** strong absorbers for  $\lambda \leq 8 \mu\text{m}$  with D and C scaled down 5 $\times$ , **(b)** weak absorbers for  $\lambda \leq 8 \mu\text{m}$ , **(c)** Q, G, Ch, and H in the window region, and **(d)** same as (c) except for the clays and Mu.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

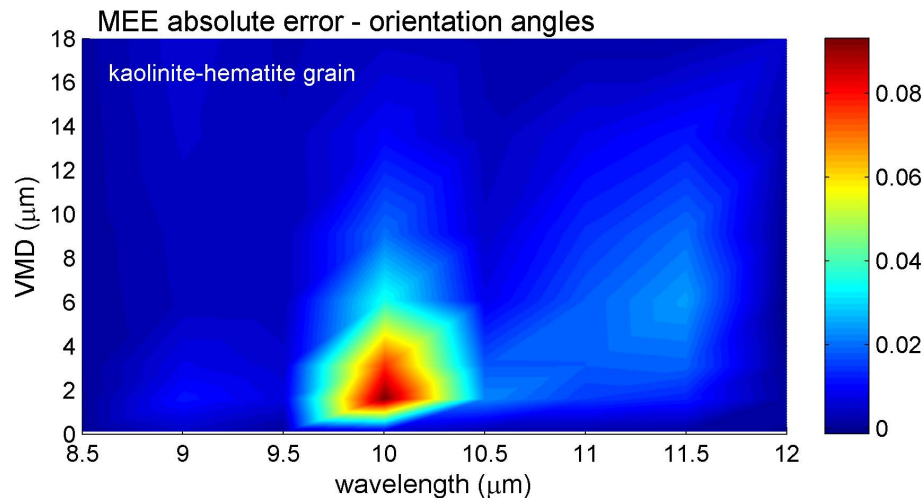
Back

Close

Full Screen / Esc

Printer-friendly Version

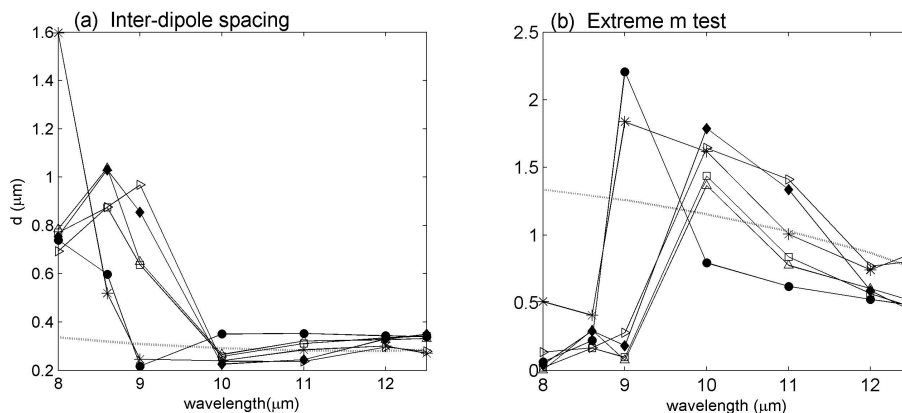
Interactive Discussion



**Fig. 2.** Absolute error in MEE between using a reduced number of orientation angles ( $N=12$ ) versus an extended set ( $N=1050$ ) for an asymmetric kaolinite-hematite grain mixture (units are in  $\text{m}^2 \text{g}^{-1}$ ). See text for details.

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.



**Fig. 3.** DDA applicability criteria versus wavelength ( $\lambda$ ) and mineral composition for **(a)** maximum inter-dipole spacing “ $d$ ” ( $\mu\text{m}$ ) and **(b)** large refractive index “ $m$ ”. Shown are the minerals gypsum (black circle), hematite (dashed line), illite (open square), kaolinite (solid diamond), montmorillonite (upward triangle), muscovite (tilted triangle), and quartz (asterisk). See text for details.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

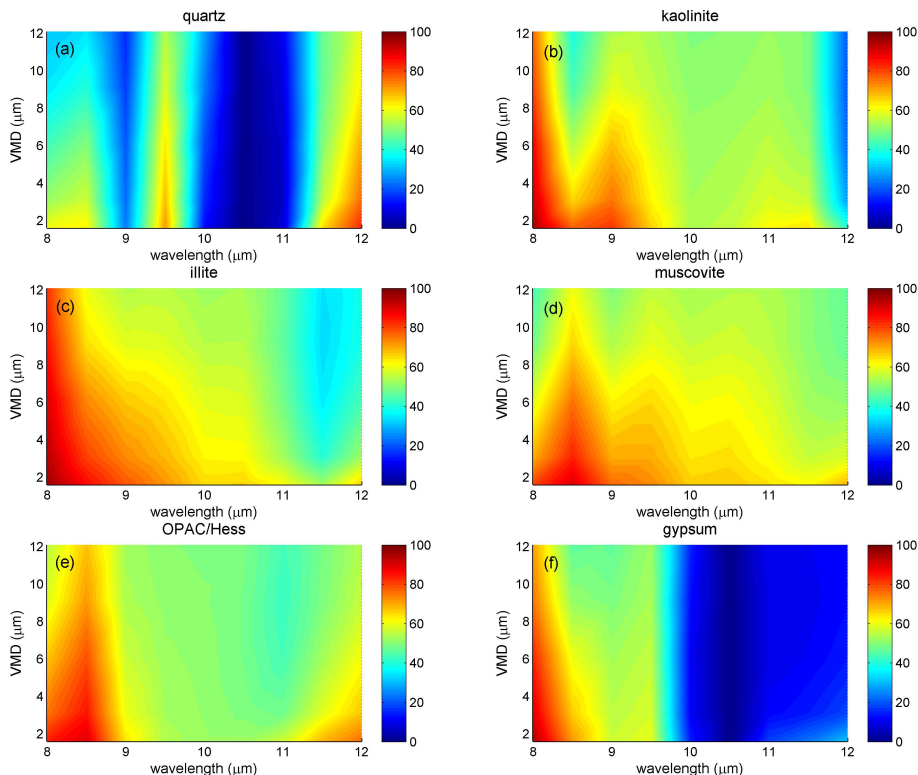
Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





**Fig. 4.** MAE as a percentage of MEE for common dust minerals across the thermal IR including **(a)** quartz (rectangle), **(b)** kaolinite (plate), **(c)** illite (plate), **(d)** muscovite (grain), **(e)** Hess/OPAC (plate) and **(f)** gypsum (rectangle). The vertical and horizontal axes are the particle VMD and wavelength, respectively. The color bar denotes the percentage of particle extinction due to absorption. Note the unique positions and shapes in the absorption features for each mineral composition. See text for details.

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

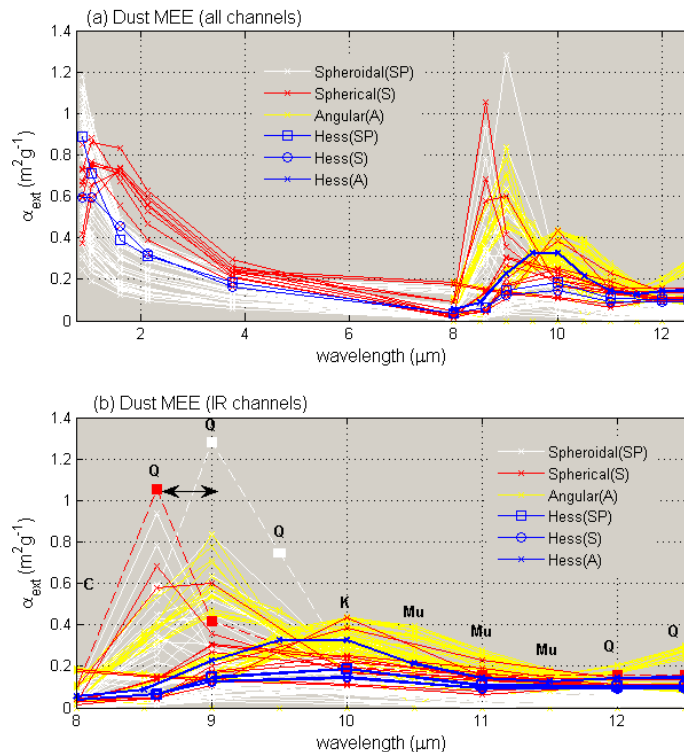
Printer-friendly Version

Interactive Discussion



## A sensitivity study of dust mass extinction efficiencies

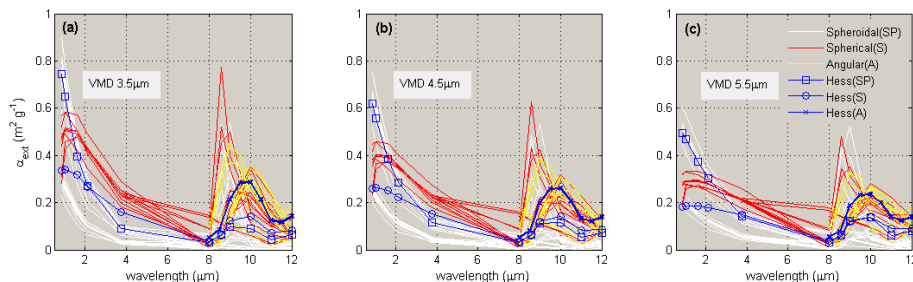
R. A. Hansell et al.



**Fig. 5.** (a) Maximum dust MEE over all dust parameterizations and wavelengths (channels) where white, red and yellow curves denote spheroids, spheres and angular particles, respectively. The Hess/OPAC model is represented by the blue curves, for spheroids (square), spheres (circle), and angular particles (cross). (b) Same as (a) but in the thermal IR. The black arrow denotes the spectral shift between spherical and non-spherical quartz particles (dotted red and white curves, respectively). The letters C (calcite), Q (quartz), K (kaolinite), and Mu (muscovite) represent the minerals that have the maximum MEE at each wavelength. See text for details.

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

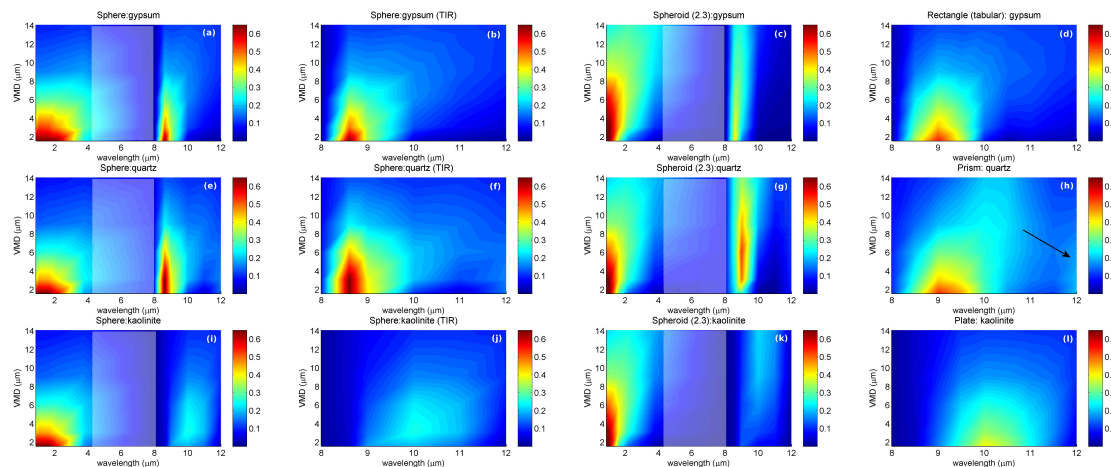


**Fig. 6.** Dust MEE over all particle shapes, compositions and wavelengths (channels) with VMD corresponding to the frequently observed size range of **(a)** 3.5  $\mu\text{m}$ , **(b)** 4.5  $\mu\text{m}$ , and **(c)** 5.5  $\mu\text{m}$ . Following the same color scheme in Fig. 5, shapes consist of oblate and prolate spheroids with aspect ratios of 1.8 and 0.5, respectively, spheres, and angular particles (calculated only for IR wavelengths) over all 13 mineral compositions. The Hess/OPAC model is represented by the blue curves.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

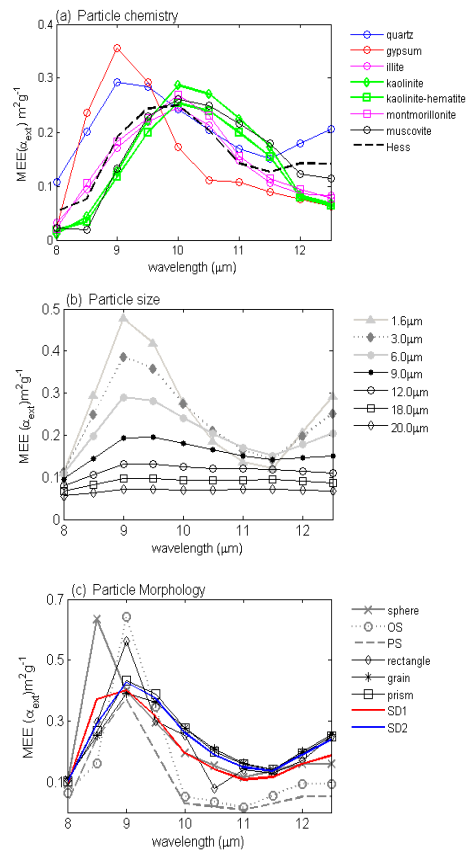


**Fig. 7.** Surface plots depicting MEE distributions (units are in  $\text{m}^2 \text{g}^{-1}$ ) for several key minerals as functions of particle chemistry (rows), particle morphology (columns – note the 2nd and 4th columns are only for IR wavelengths), and particle size (VMD) and wavelength  $\lambda$  (vertical and horizontal axes, respectively). Columns 2 and 4 compare MEE for spheres and angular particles in the thermal IR (TIR). Note the changes in MEE distributions (hot-spots) when the dust parameters are perturbed.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

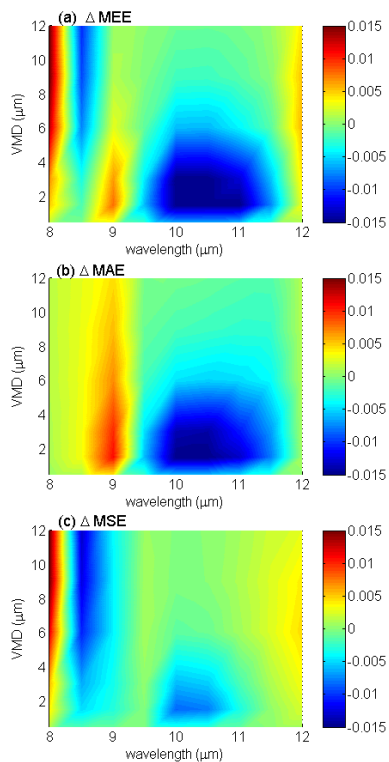


**Fig. 8.** Sensitivity of MEE to dust microphysics and chemistry. **(a)** Particle chemistry for the minerals quartz (blue-circle), gypsum (red-circle), illite (magenta-circle), kaolinite (green-diamond), kaolinite-hematite (green-square), montmorillonite (magenta-square), muscovite (black-circle), and Hess/OPAC (dashed black line). **(b)** Particle size (VMD – from top to bottom) for 1.6 (solid triangle), 3.0 (solid diamond), 6.0 (solid gray circle), 9.0 (solid black circle), 12.0 (open circle), 18.0 (open square), and 20 μm (open diamond). **(c)** Particle morphology for sphere (cross), oblate (circle) and prolate (dotted curve) spheroid, rectangle (diamond), grain (asterisk), and prism (square). SD1 (red curve) and SD2 (blue curve) are the particle shape distribution scenarios. See text for details.



## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.

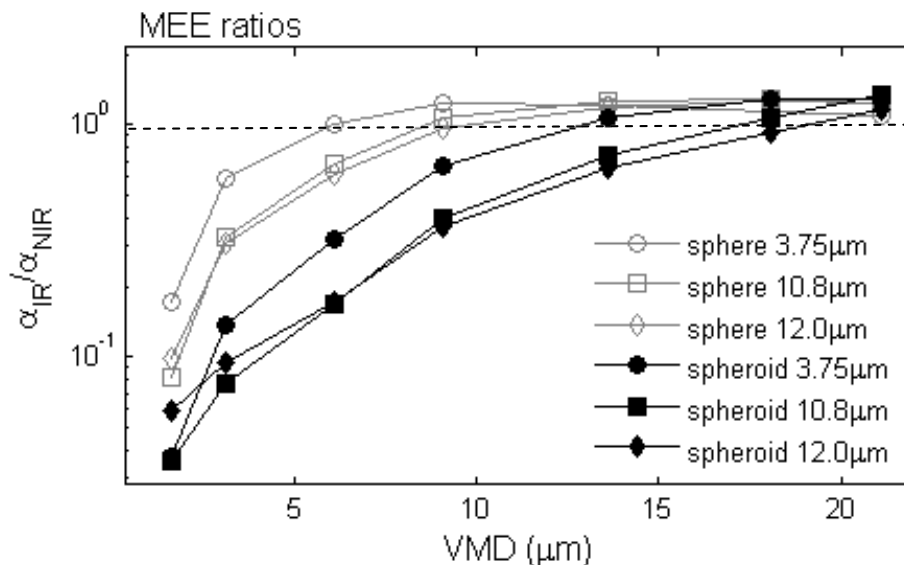


**Fig. 9.** Sensitivity of MEE to a clay-hematite mixture. Difference plots are shown for (a)  $\Delta MEE$ , (b)  $\Delta MAE$ , and (c)  $\Delta MSE$ . All units are in  $\text{m}^2 \text{g}^{-1}$ . See text for details.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

## A sensitivity study of dust mass extinction efficiencies

R. A. Hansell et al.



**Fig. 10.** Log scale of MEE ratios ( $\alpha_{IR}/\alpha_{NIR}$ ) versus VMD for the Hess/OPAC parameterization using **(a)** spheres (gray curves), and **(b)** oblate spheroids with aspect ratio 1.8 (black curves) at wavelengths corresponding to the centers of AVHRR channels 3 (3.75  $\mu\text{m}$  – circles), 4 (10.8  $\mu\text{m}$  – squares), and 5 (12.0  $\mu\text{m}$  – diamonds), respectively. Refer to Table 3.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

