A Comprehensive Database of the Optical Properties of Irregular Aerosol Particles for Radiative Transfer Simulations

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ABSTRACT: A database [Texas A&M University dust 2020 (TAMUdust2020)] of the optical properties of irregular aerosol particles is developed for applications to radiative transfer simulations involving aerosols, particularly dust and volcanic ash particles. The particle shape model assumes an ensemble of irregular hexahedral geometries to mimic complex aerosol particle shapes in nature. State-of-the-art light scattering computational capabilities are employed to compute the single-scattering properties of these particles for wide ranges of values of the size parameter, the index of refraction, and the degree of sphericity. The database therefore is useful for various radiative transfer applications over a broad spectral region from ultraviolet to infrared. Overall, agreement between simulations and laboratory/in situ measurements is achieved for the scattering phase matrix and backscattering of various dust aerosol and volcanic ash particles. Radiative transfer simulations of active and passive spaceborne sensor signals for dust plumes with various aerosol optical depths and the effective particle sizes clearly demonstrate the applicability of the database for aerosol studies. In particular, the present database includes, for the first time, robust backscattering of nonspherical particles spanning the entire range of aerosol particle sizes, which shall be useful to appropriately interpret lidar signals related to the physical properties of aerosol plumes. Furthermore, thermal infrared simulations based on in situ measured refractive indices of dust aerosol particles manifest the effects of the regional variations of aerosol optical properties. This database includes a user-friendly interface to obtain user-customized aerosol single-scattering properties with respect to spectrally dependent complex refractive index, size, and the degree of sphericity.

KEYWORDS: Radiative transfer; Databases; Aerosol optical properties

1. Introduction

Atmospheric aerosol impacts Earth's energy budget through their radiative effects and interactions with clouds and the ecosystem. Among various aerosol species, mineral dust is the most dominant aerosol type over deserts and arid/semiarid areas. Total global dust aerosol emissions range from 1.0 to $2.7 \times 10^3$ Tg yr$^{-1}$ (Zender et al. 2003; Kok et al. 2017). Airborne dust particles not only directly affect the surface–atmosphere energy budget by scattering and absorbing radiation but also indirectly affect Earth's atmospheric system. For example, they act as ice nuclei and affect ice cloud properties through the ice crystal formation process (Archuleta et al. 2005). Recent modeling studies suggest that atmospheric dust aerosol particles could affect tropical cyclones and rainfall (Reed et al. 2019; Thompson et al. 2019). Deposition of dust aerosol particles affects oceanic carbon uptake (Nickelsen and Oschlies 2015). Dust aerosol particles in the atmosphere are harmful to human health (Prospero 1999). Plumes of dust aerosols can be transported over a long distance (Duce et al. 1980). In consequence, dust aerosols are globally distributed, and thereby the impacts of dust aerosols on various weather and climate processes have been of great interest.

In addition, volcanic ash associated with a large eruption can have global climatic impacts lasting several months to several years (Hansen et al. 1992; Robock 2000). Although the average annual emission of volcanic ash is relatively low, the total amount of volcanic ash emission by a single volcanic event can be substantial. For example, the Eyjafjallajökull volcano emitted 11.4 Tg of volcanic ash in only 41 days in 2010 (Stohl et al. 2011), and the Pinatubo eruption in June 1991 emitted about ~1000 Tg of volcanic ash (Bluth et al. 1992). These aerosols are important not only for aerosol radiative cooling effect but also for iron fertilization for oceanic phytoplankton growth (Achterberg et al. 2013).

Continuous monitoring of dust aerosol and volcanic ash plumes is essential to assessing their climatic impacts. Satellite observations in conjunction with remote sensing techniques have been playing a vital role in the understanding of the global distribution of aerosol properties. For example, spaceborne Advanced Very High Resolution Radiometer instruments have been monitoring the global distribution of aerosol optical depth (AOD) since 1981 (Hsu et al. 2017). The Polarization and Directionality of the Earth's Reflectance (POLDER) aboard the Polarization and Anisotropy of Reflectances for Atmospheric Sciences Coupled with Observation from a Lidar (PARASOL) satellite (Fougnie et al. 2007) measures multidirectional total and polarized reflectances in visible and near-infrared bands and provides fine- and coarse-mode AODs, coarse-mode aerosol sphericity, and the Ångström exponent.

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(Formenti et al. 2018). The Cloud–Aerosol Lidar with Orthogonal Polarization (CALIOP) aboard the Cloud–Aerosol Lidar and Infrared Pathfinder Observations (CALIPSO) satellite reveals the three-dimensional distributions of aerosol properties (Winker et al. 2013). Moreover, CALIPSO deploys the Imaging Infrared Radiometer (IIR) to measure thermal infrared radiations in three split-window bands that are useful for dust plume detection (Chen et al. 2010).

However, the direct and indirect radiative effects of these aerosol particles in the surface–atmosphere coupled system remain highly uncertain. This is partly because aerosol optical properties that are fundamentally determined by particle sizes, chemical compositions, and complex morphologies of aerosol particles are not adequately modeled (Mishchenko et al. 2003; Huang et al. 2015). Laboratory measurements using a scanning electron microscopes (SEM) reveal that the shapes of dust aerosol and volcanic ash particles are irregular with a substantial variation among samples (e.g., Gao and Anderson 2001; Reid et al. 2003). Moreover, aerosol compositions are largely dependent on the aerosol origin location and the particle sizes (Di Biagio et al. 2017; Kandler et al. 2007), which result in complex variations of scattering properties of these aerosol particles. Nevertheless, spatially invariant spherical and spheroidal aerosol optical property models (Hess et al. 1998; Meng et al. 2010) have been extensively used for radiative transfer simulations and remote sensing applications. These simplifications hinder reliable aerosol property retrievals and aerosol radiative forcing estimates. To obtain robust observational constraints on the climatic impacts of aerosol particles, there is a pressing need to derive the optical properties of aerosol particles based on realistic particle shapes and inherent optical characteristics (i.e., the real and imaginary parts of the index of refraction).

This study develops a database [Texas A&M University dust 2020 (TAMUdust2020)] of the optical properties of irregular aerosol particles for various remote sensing applications involving radiative transfer simulations such as the shortwave polarimetric reflectance, thermal infrared brightness temperatures, or polarimetric lidar signals. Section 2 describes the present technical methodology, the definition of the aerosol particle shape model, the database structure, and the uncertainties of the database. Section 3 presents sensitivity studies of the single-scattering properties to the inherent optical and morphological characteristics of aerosol particles. Section 4 extensively compares simulated and in situ measured optical properties of dust aerosol and volcanic ash particles. Section 5 describes validation and applications concerning remote sensing and radiative transfer calculations. Section 6 summarizes this study.

2. Method

a. Particle shape model

The two major morphological parameters of particles important to the single-scattering properties are the aspect ratio (Merikallio et al. 2011) and the irregularity of the particle (Muinonen et al. 1996; Ishimoto et al. 2010). As natural mineral dust aerosols and volcanic ash particles show a variety of complex multifaceted irregular shapes (Reid et al. 2003; Riley et al. 2003), a single particle shape model may be inappropriate. To mimic the diversity of realistic particle morphological characteristics, we employ an ensemble of irregular hexahedral particles (Bi et al. 2010) as a particle shape model. First, we define a regular hexahedral particle with a specified aspect ratio (defined as a ratio of the largest particle dimension to the smallest particle dimension). Then, each face of a hexahedral particle is randomly tilted based on a two-dimensional Gaussian distribution (Yang and Liou 1998) with a parameter $\sigma = 1.0$ for the degree of surface tilt, in the form

$$P(Z_x, Z_y) = \frac{1}{\pi \sigma_T^2} e^{-[(Z_x^2 + Z_y^2) / \sigma_T^2]},$$  \hspace{1cm} (1)

where $Z_x = \partial Z / \partial x$ and $Z_y = \partial Z / \partial y$ are the slopes of a face along two axes orthogonal to the normal direction $Z$ relative to the untitled face. We define 20 irregular hexahedral particles, each of which is specified by the randomly tilted six faces of a regular hexahedron with an aspect ratio between 1.14 and 4.02, because most airborne mineral dust aerosol and volcanic ash particles have aspect ratios ranging from 1.3 to 2.5 (Kandler et al. 2011; Riley et al. 2003). The random face-tilting process allows particle shapes to be columnar or planar but highly irregular hexahedra, with no axes having an axis ratio of unity, which is consistent with mineral dust particles (Huang et al. 2020; and references cited therein).

Unlike a spheroid model, it is not straightforward to interpret the aspect ratio of an irregular (nonsymmetric) hexahedral particle as being equivalent to any particular complex dust aerosol particle in nature. Therefore, we use the degree of sphericity to quantify the particle aspherical characteristics, defined in the form (Wadell 1935)

$$\Psi = \frac{\pi^{1/3}(6V)^{2/3}}{A_{eq}},$$  \hspace{1cm} (2)

where $V$ and $A_{eq}$ are volume and the total surface area of a particle (subscript $eq$ stands for a particle ID). For a convex particle, the total particle surface area can be analytically obtained from the projected area ($A$) of the particle under the random orientation condition in the form of $A_{eq} = 4A$ (Vouk 1948).

To model the variability of particle shapes in different aerosol species, we consider various ensembles with different weightings (number mixing ratios) of the 20 irregular hexahedral particles. Figure 1 illustrates the particle geometry of individual irregular hexahedral particles and the number mixing ratio of individual particles for 5 ensemble models with specified ensemble-weighted sphericity ($\Psi$). In general, as the aspect ratio of an original regular hexahedron becomes larger, the degree of sphericity of an irregular hexahedral particle becomes lower. The number mixing ratio ($f_{mix}$) of the 20 particles ($N_{ens} = 20$) for an ensemble model is defined as

$$f_{mix} = e^{(\Psi_{j} - \Psi_{m})/\Delta \Psi} \sum_{j=1}^{N_{ens}} e^{(\Psi_{j} - \Psi_{m})/\Delta \Psi},$$  \hspace{1cm} (3)
where $\Psi$ is the mean degree of sphericity, and $\Delta\Psi$ is the difference between the maximum ($\Psi_{20}$) and minimum ($\Psi_{1}$) degree of sphericity, denoted as

$$\Psi = \frac{1}{N_{\text{ens}}} \sum_{j=1}^{N_{\text{ens}}} \Psi_j$$  \hspace{1cm} (4)

$$\Delta\Psi = \Psi_{20} - \Psi_{1},$$  \hspace{1cm} (5)

and $c$ is a coefficient iteratively obtained from a specified ensemble-weighted degree of sphericity

$$c = \frac{\pi^{1/3} \left( \frac{N_{\text{ens}}}{8^{1/3}} \sum_{j=1}^{N_{\text{ens}}} f_{\text{mix},j} V_{e,j} \right)^{2/3}}{\sum_{j=1}^{N_{\text{ens}}} f_{\text{mix},j} A_{e,j}}.$$  \hspace{1cm} (6)

As seen in Fig. 1, an ensemble model with a larger ensemble-weighted degree of sphericity has a higher mixing ratios of individual particles with larger degree of sphericity.

b. Light scattering computation

We use a suite of light scattering computational methods to calculate the single-scattering properties of irregular hexahedral particles. For very small size parameters ($kD \ll 1$, where $D$ is the diameter of a circumscribed sphere of a particle, $k = 2\pi/\lambda$, and $\lambda$ is wavelength), it is appropriate to use the Rayleigh scattering approximation (Bohren and Huffman 1983) with a $V/A$-equivalent sphere (Grenfell and Warren 1999).

For the small-to-moderate size parameter domain, the invariant-imbedding T-matrix method (IITM; Johnson 1988; Bi and Yang 2014 and references cited therein) provides numerically exact solutions of the single-scattering properties of nonspherical particles with a reasonable computational burden. Note that IITM is quite different from the conventional implementation of the T-matrix method in terms of the extended boundary condition method (EBCM; Waterman 1965, 1971; Mishchenko et al. 2002). In IITM, the computation of the T-matrix is based on an electromagnetic equation in a volume-integral form, and the scattering particle is considered as consisting of a number of concentric thin layers centered at the innermost small core. The T-matrix associated with the scattering particle is then computed in an iterative form with the initial value of the T-matrix associated with the aforementioned innermost core. Unlike IITM, EBCM is implemented by imposing electromagnetic boundary conditions in surface-integral forms. Because EBCM is based on a surface-integral technique, it is more challenging to implement EBCM for complex particle morphologies. As a result, the existing EBCM computational programs are mainly applicable to spheroid, circular cylinder, and Chebyshev particles (Mugnai and Wiscombe 1986; Mishchenko et al. 2002).

For the large size parameter domain, Yang and Liou (1996, 1997) developed the physical geometric optics method (PGOM). In PGOM, the principles of geometric optics are utilized to compute the near-field that is subsequently transformed to the corresponding far-field through the use of a rigorous electromagnetic relation. In practice, PGOM can be implemented in terms of a volume-integral technique (Yang and Liou 1997). Bi et al. (2011) and Sun et al. (2017) further improved the computational efficiency of the volume-integral based PGOM using computer graphics techniques. PGOM can also be implemented in terms of a surface-integral technique (Yang and Liou 1996). The equivalence of these two techniques is demonstrated by Yang et al. (2019).

Figure 2 compares the single-scattering properties of a single irregular hexahedron (particle ID 20) computed with IITM and PGOM. The real part ($m_r$) and imaginary part ($m_i$) of the refractive index are 1.5 and 0.005, respectively.
These computational techniques achieve convergence of the single-scattering property computations, including simulations for the 180° backscattering angle, for a dust aerosol particle in an intermediate size parameter range, similar to the ice crystal case demonstrated by Yang et al. (2019). Therefore, it is possible to provide reliable single-scattering properties of nonspherical atmospheric particles throughout practically the entire size parameter range based on a combination of IITM and PGOM.

A simplified version of the PGOM, which is known as the improved geometric optics method (IGOM; Yang and Liou 1996), is computationally much more efficient than PGOM. Unlike PGOM, which implements a rigorous near-field to far-field transformation, IGOM only considers the ray-spreading effect; as a result, the phase matrix computed by the conventional optics method (Cai and Liou 1982; Takano and Liou 1989) can be transformed to obtain a more accurate counterpart [see the technical details reported in Yang and Liou (1996)]. Overall, the simulations based on IGOM and PGOM are consistent, except that PGOM can explicitly account for interference of scattered fields associated with different beams, which can significantly reduce computational uncertainties around the backscattering angle (e.g., 170°–180°). For this reason, the optical properties computed with both IGOM and PGOM are applicable to remote sensing applications based on observations obtained by passive sensors. However, lidar backscatter simulations and solar reflectance simulations over quasi-backscatter sun-glint regions should use the optical properties computed with PGOM.

This study uses the Rayleigh scattering approximation for small particles with $kD < 0.03$ and IITM for small-to-moderate $kD$ particles to compute the single-scattering properties of individual irregular dust particles. Because the computational time of PGOM is proportional to the number of considered scattering angles, for large $kD$ particles, to reduce the computational burden for simulations of phase matrix elements, we use IGOM for forward to near-backward scattering angles, and PGOM for quasi-backscattering angles at which IGOM computations deviate from robustly accurate PGOM computations (see appendix A).

In addition, since the geometric optics methods do not take into account the edge effect, which is not negligible for particles with $kD < 50$ (Bi et al. 2010b; Lin et al. 2017), a semiempirical edge effect correction (Yang et al. 2013) is used to correct the extinction and absorption efficiencies computed with IGOM and PGOM. Furthermore, to obtain a smooth transition of the single-scattering property computations in the intermediate $kD$ range, the $kD$ threshold specifying the transition between IITM and the geometric optics methods is optimized independently for each particle. As a result, the $kD$ threshold ranges from 20 to 100, depending on the particle shape and absorptivity. This provides a reasonably smooth transition of the single-scattering properties of a particle through a range of particle sizes.

c. The single-scattering properties of an ensemble irregular hexahedral model

The single-scattering properties of the ensemble of irregular hexahedral particles are computed from the properties of the individual particles and their particle mixing ratios for each $kD$, $m_\ast$, $m_\varepsilon$, and $\Psi$. The individual particles are assumed to be randomly oriented. The phase matrix ($P$), extinction efficiency ($Q_{\text{ext}}$), single-scattering albedo (SSA; $\omega$), and asymmetry factor ($g$) of an ensemble model with a specified $kD$, $m_\ast$, $m_\varepsilon$, and $\Psi$ are given by

$$Q_{\text{ext}} = \frac{\sum_{j=1}^{N_{\text{ext}}} f_{\text{mix},j} A_j Q_{\text{ext},j}}{\sum_{j=1}^{N_{\text{ext}}} f_{\text{mix},j} A_j},$$

(7)
Fig. 3. An example of the phase matrix elements of the ensemble irregular hexahedral dust model. Bold black lines indicate the phase matrix elements of the ensemble-weighted average. Colors indicate the phase matrix elements of individual irregular hexahedral particles computed with IITM (solid lines) and IGOM–PGOM (dashed lines). The size parameter is (left) 20, (center) 70, and (right) 150. The complex refractive index and the degree of sphericity are 1.5 + i0.0001 and 0.75, respectively.

\[
\omega = \frac{\sum_{j=1}^{N_{\text{ext}}} f_{\text{mix}_j} A_j Q_{\text{sca},j}}{\sum_{j=1}^{N_{\text{ext}}} f_{\text{mix}_j} A_j Q_{\text{ext},j}}, \quad (8)
\]

\[
g = \frac{\sum_{j=1}^{N_{\text{ext}}} f_{\text{mix}_j} A_j Q_{\text{sca},j}}{\sum_{j=1}^{N_{\text{ext}}} f_{\text{mix}_j} A_j Q_{\text{sca},j}}. \quad (9)
\]
\[
\mathbf{P}(\theta) = \frac{\sum_{j=1}^{N} f_{\text{mix},j} A_{\text{Q}_{\text{sc}}j} \mathbf{P}_j(\theta)}{\sum_{j=1}^{N} f_{\text{mix},j} A_{\text{Q}_{\text{sc}}j}},
\]
where \(Q_{\text{sc}}\) is the scattering efficiency; and the phase matrix of randomly oriented particles accompanied with mirrored counterparts defined by Mishchenko and Yurkin (2017) is expressed as
\[
\mathbf{P}(\theta) = \begin{bmatrix}
P_{11}(\theta) & P_{12}(\theta) & 0 & 0 \\
P_{21}(\theta) & P_{22}(\theta) & 0 & 0 \\
0 & 0 & P_{33}(\theta) & -P_{43}(\theta) \\
0 & 0 & P_{43}(\theta) & P_{44}(\theta)
\end{bmatrix}.
\]

Figure 3 shows the phase matrix elements of individual irregular hexahedral particles and those of an ensemble model with \(kD\) of 20, 70, and 150. The ensemble model assumes \(m_r = 1.5, m_i = 0.0001,\) and \(\Psi = 0.75.\) For \(kD = 20,\) the phase matrix elements of all individual dust particles are computed based on IITM (as shown in solid lines). With this \(kD\) value, the phase matrix elements vary substantially with particle shapes and sizes. The phase matrix elements of an ensemble of irregular hexahedral particles show smoothed features compared to the individual counterparts. For \(kD = 70,\) the single-scattering properties are computed with IGOM–PGOM for particle IDs 1–11 and IITM for particle IDs 12–20. For \(kD = 150,\) all single-scattering property computations are performed with IGOM–PGOM. As \(kD\) increases, the phase matrix elements of an ensemble model tend to be insensitive to \(kD,\) except for \(P_{11}\) in forward directions where diffraction plays a role.

d. The database design and user interface

Table 1 shows the two sets of ranges of complex refractive indices and size parameters of the ensemble irregular hexahedral particles. We utilize a kernel technique to format the single-scattering properties of ensemble models (Twomey 1977; Dubovik et al. 2006) for shortwave (SW) and longwave (LW) domains. The ranges of the complex refractive index of dust aerosol and volcanic ash particles in the database are determined based on previous studies (e.g., Stegmann and Yang 2017), and are \(m_r = 1.37–1.7\) and \(m_i = 0.0001–0.1\) for the SW kernel, and \(m_r = 0.4–3.2\) and \(m_i = 0.001–4.0\) for the LW kernel. The \(\Psi\) ranges from 0.695 to 0.785. The size parameter ranges from the Rayleigh scattering domain to 11800 in the SW kernel, and up to 1470 in the LW kernel. Therefore, the TAMUdust2020 database covers practically the entire size range of typical dust particles. For example, the SW kernel allows a maximum diameter of 1000 \(\mu m\) at a wavelength of 532 nm. With a combination of SW and LW kernels, the TAMUdust2020 database contains the optical properties for particle maximum diameters from 0.01 to 375 \(\mu m\) at wavelengths from 0.2 through 200 \(\mu m.\)

The structure and user interface of the TAMUdust2020 database is illustrated in Fig. 4. A practically useful feature of the TAMUdust2020 database is flexibility for user’s needs. A user can specify wavelength bins, maximum diameter bins, spectrally dependent refractive indices, and a size-invariant sphericity to create a user-defined single-scattering property subdatabase. Therefore, it is straightforward to provide the single-scattering properties of aerosol particles customized for regional dependence of the aerosol optical, microphysical, and morphological characteristics based on in situ measured refractive indices and typical shapes of local aerosol particles.

e. Uncertainty in the database

The database has small uncertainties in the single-scattering properties of irregular aerosol particles associated with two error sources:

- A combination of multiple techniques in the computations of the single-scattering properties of some particles.
- Multidimensional linear interpolation for the kernels to obtain the single-scattering properties of a user-specified particle.

On the first error source, each computational method has inherent uncertainties caused by particular assumptions and
approximations. For example, the accuracy of the single-scattering computation with IITM depends on several assumed parameters such as the expansion order number of the T-matrix (Zhai et al. 2019). Although both PGOM and IGOM rely on the principle of geometric optics, there are several differences in their numerical implementations and assumptions such as the treatment of the interference among rays/beams (appendix A), the number of scattering azimuth angles, and the number of particle orientations. However, we consider these error sources to have a negligible impact on the accuracy of the single-scattering property calculations for a practical use, as demonstrated in Fig. 2. The uncertainty of the single-scattering properties due to a combination of multiple computational techniques is expected to be a few orders of magnitude smaller than the uncertainties from particle shape variations (Fig. 3).

On the second error source, despite computing a very large number of grids for each variable in both SW and LW kernels, the multidimensional linear interpolation could lead to the largest uncertainty in the database as the single-scattering properties have nonlinear dependence on all optical, microphysical, and morphological parameters. Therefore, in appendix B, we thoroughly evaluate the maximum absolute error due to multidimensional linear interpolation in the SW and LW kernels.

Figure 5 shows the statistics of the maximum absolute error of the extinction efficiency ($\varepsilon_{\text{max, } Q_{\text{ext}}}$), SSA ($\varepsilon_{\text{max, } \text{SSA}}$), and asymmetry factor ($\varepsilon_{\text{max, } g}$) in all grids of the two kernels. In general, maximum absolute errors in the SW kernel are smaller than those in the LW kernel as the SW kernel has finer grid resolutions. Also, these absolute errors show positive correlations with the size parameters for $kD < \sim 5$ since these single-scattering property values decrease toward zero with smaller size parameters. Most importantly, these absolute errors are several magnitudes smaller than the values of the single-scattering properties even with the worst cases in the LW kernel. We also estimate the maximum absolute errors of the phase matrix elements (not shown) for each kernel and find that potential interpolation-induced errors are $<0.01\%$ for $P_{11}$ and $<0.001\%$ for other normalized elements (relative to $P_{11}$) in the SW kernel, and $<1\%$ for $P_{11}$ and $<0.01\%$ for other normalized elements in the LW kernel. The maximum absolute errors of these single-scattering properties and phase matrix elements (corresponding to 95th percentiles) are found in the LW kernel for cases with $mr > 1$.

Note that typical errors in the single-scattering properties should be much smaller than these estimated maximum absolute errors since the errors induced in interpolation often tend to
errors in the single-scattering properties in the database may be an order of magnitude smaller than the estimated maximum absolute errors. Thus, the uncertainty in the database is negligible for most practical applications such as radiative transfer simulations and remote sensing.

f. Bulk scattering properties

In the following analysis, we compute the bulk scattering properties of various aerosol particles based on a specified particle size distribution (PSD). We use lognormal distributions for PSDs of dust and volcanic ash particles, defined as

\[
n(D) = \frac{dN}{dD} = \frac{N_0}{D \ln \sigma \sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{\ln D - \ln D_m}{\ln \sigma}\right)^2},
\]

(13)

where \(D_m\) is the median diameter, \(N_0\) is the particle number concentration, and \(\sigma\) is the geometric standard deviation.

The bulk extinction efficiency \(\langle Q_{\text{ext}}\rangle\), bulk SSA \(\langle \omega \rangle\), bulk asymmetry factor \(\langle g \rangle\), and bulk phase matrix \(\langle \mathbf{P} \rangle\) with a given PSD, \(\lambda\), \(m_r\), and \(\Psi\) are computed from the single-scattering property counterparts in Eqs. (7)–(10) as follows:

\[
\langle Q_{\text{ext}} \rangle = \frac{\int_{D_{\text{min}}}^{D_{\text{max}}} n(D) A(D, \Psi) Q_{\text{ext}}(D, m_r, m_i, \Psi) dD}{\int_{D_{\text{min}}}^{D_{\text{max}}} n(D) A(D, \Psi) dD},
\]

(14)

\[
\langle \omega \rangle = \frac{\int_{D_{\text{min}}}^{D_{\text{max}}} n(D) A(D, \Psi) Q_{\text{sca}}(D, m_r, m_i, \Psi) dD}{\int_{D_{\text{min}}}^{D_{\text{max}}} n(D) A(D, \Psi) dD},
\]

(15)

\[
\langle g \rangle = \frac{\int_{D_{\text{min}}}^{D_{\text{max}}} n(D) A(D, \Psi) Q_{\text{sca}}(D, m_r, m_i, \Psi) g(D, m_r, m_i, \Psi) dD}{\int_{D_{\text{min}}}^{D_{\text{max}}} n(D) A(D, \Psi) Q_{\text{sca}}(D, m_r, m_i, \Psi) dD}, \quad \text{and}
\]

(16)

\[
\langle \mathbf{P}(\theta) \rangle = \frac{\int_{D_{\text{min}}}^{D_{\text{max}}} n(D) A(D, \Psi) Q_{\text{sca}}(D, m_r, m_i, \Psi) \mathbf{P}(\theta, D, m_r, m_i, \Psi) dD}{\int_{D_{\text{min}}}^{D_{\text{max}}} n(D) A(D, \Psi) Q_{\text{sca}}(D, m_r, m_i, \Psi) dD}.
\]

(17)

3. Sensitivity results

Although previous studies have extensively investigated the sensitivity of the single-scattering properties to refractive index, size, and aspect ratio for spherical or spheroidal particles, it is of interest to quantitatively investigate these sensitivities using an ensemble of irregular hexahedral particles.

In particular, with the new particle shape model, we investigate the sensitivity of the single-scattering properties to the complex refractive index and the degree of sphericity.

Figure 6 shows the sensitivities of the single-scattering properties to \(m_r\) based on an irregular hexahedral ensemble model with \(m_i = 0.003\) and \(\Psi = 0.75\). The \(Q_{\text{ext}}\), SSA, and \(g\) show smooth transitions in an intermediate \(kD\) range where
two computational methods are used for the computations (Fig. 6c). A peak of $Q_{\text{ext}}$ occurs due to coherent interference between diffracted and reflection–refracted scattering waves. Since $m_r$ affects reflection–refracted scattering waves, $k_D$ at the peak in $Q_{\text{ext}}$ varies with $m_r$. The $g$ varies with $m_r$ in a $k_D$ range of 10–50 where coherent interference is prominent.

Figures 6d–i show the phase matrix elements for $k_D = 30$. A smaller $m_r$ shows weaker backscattering in $P_{11}$ because fewer incident electromagnetic waves are deflected to backward directions. Also, other phase matrix elements vary with $m_r$, especially in side-to-backward scattering angles in $P_{22}/P_{11}$ and forward-to-side scattering angles in $P_{23}/P_{11}$, $P_{33}/P_{11}$, and $P_{44}/P_{11}$. This confirms the usefulness of spaceborne/airborne polarimetric measurements for remote sensing of aerosol optical properties, which has been utilized for many aerosol remote sensing techniques (e.g., Herman et al. 2005; Stamnes et al. 2018).

Figure 7 shows the sensitivity of the single-scattering properties to $m_i$ based on an irregular hexahedral ensemble model with $m_r = 1.5$, and $m_r = 0.003$. The striking feature is that $Q_{\text{ext}}$ is less oscillatory versus $k_D$ for smaller $\Psi$ (Fig. 7d), which is similar to the characteristics for large aspect ratio particles (Huang et al. 2015). For compact particles that have large $\Psi$, a bundle of rays going through the particle interferes with diffracted fields, resulting in constructive and destructive interference of electromagnetic waves and causing oscillations in $Q_{\text{ext}}$ at intermediate size parameters. Also, $g$ varies moderately with $\Psi$ (Fig. 8c). For larger $\Psi$ particles with $kD = 30$, the phase function has stronger backward scattering intensity (Fig. 8d), and other phase matrix elements have apparent variations with $\Psi$ (Figs. 8e–i). In particular, the $P_{11}$ value in the 180° backscattering direction substantially

![Fig. 6. The single-scattering properties of the ensemble irregular hexahedral model with various real parts of the refractive index ($m_r$), including (a) the extinction efficiency ($Q_{\text{ext}}$), (b) single-scattering albedo (SSA), (c) asymmetry factor, (d) $P_{11}$, (e) $-P_{23}/P_{11}$, (f) $P_{22}/P_{11}$, (g) $P_{33}/P_{11}$, (h) $P_{43}/P_{11}$, and (i) $P_{44}/P_{11}$. The size parameter, degree of sphericity ($\Psi$), and imaginary part of the refractive index ($m_i$) are 30, 0.75, and 0.003, respectively.](https://example.com/figure6)
varies, implying the importance of an appropriate particle shape assumption to robustly simulate lidar signals (Bi et al. 2018).

4. Comparison between simulations and measurements
a. Laboratory-measured data and simulation setup

Laboratory measurements of aerosol particles are essential to a better understanding of the scattering properties of actual aerosol particles and play an important role in the validation of the light scattering computational techniques. The Instituto de Astrofísica de Andalucía (IAA) Cosmic Dust Laboratory (CoDuLAB) in Granada has conducted measurements of the scattering properties of various dust aerosols and volcanic ash particles. These measured data as well as previously obtained data in Amsterdam are archived in the Amsterdam–Granada light scattering database (Muñoz et al. 2012). This database includes the phase matrix elements in certain scattering angle ranges (e.g., $3^\circ$–$177^\circ$). PSDs derived from a Fritsch laser particle sizer (Konert and Vandenberghe 1997) or Mastersizer2000 measurements based on the Fraunhofer diffraction theory and/or Lorenz–Mie theory, refractive index experiments summarized from literature, and SEM images of various aerosol particles.

To assess the applicability of the TAMUdust2020 database to various aerosol particles, we compare the scattering properties of various bulk dust and volcanic ash samples obtained from the TAMUdust2020 database with those based on laboratory measurements. The bulk scattering properties are computed based on Eqs. (13)–(17) in which a projected-area-equivalent sphere diameter $D_A = 2 \sqrt{A/\pi}$ and analytically derived effective variance $\sigma_{eff} = \exp(\sigma^2) - 1$ are used instead of $D$ and $\sigma$. This is because PSDs of aerosol particles are estimated with the Fraunhofer diffraction theory for spheres. Table 2 shows optical and microphysical properties of 11 aerosol particle samples used for the following comparisons. Hereinafter, we omit angle brackets to describe the bulk optical properties for simplicity.

For the aforementioned reasons, we use optimized optical and microphysical properties, rather than the reference counterparts, in Table 2 to compute the bulk phase matrix elements of aerosol samples. The optimal effective particle radius and the effective variance of the aerosol...
samples are obtained through least squares fitting [Eq. (26) in Dubovik et al. 2006] of the phase matrix elements from the reference refractive indices with their uncertainties 50%–100% without considering the a priori and smoothness terms. We consider the entire range of $\Psi$ to obtain the optimal $\Psi$ of a given aerosol sample.

b. Comparison of the phase matrix elements

First, we compare simulations of the six independent phase matrix elements of several major aerosol particles at two wavelengths (441.6 and 632.8 nm) with those from laboratory measurements of natural samples. Typical dust aerosol particles consist of various silicate minerals that could be up to 80%–90% of the total mass (Kandler et al. 2007; Wagner et al. 2012). Di Biagio et al. (2017) demonstrate that the two major mineralogical components among silicate minerals are clay species (e.g., illite, kaolinite, and chlorite) and quartz followed by calcite and feldspar, with substantial regional variations of the mass fractions of individual mineralogical compositions. The mass fractions of total clay species in dust aerosol particles can reach up to 46%–92%, and quartz is the second most dominant component, in terms of the mass fraction ranging from 3% to 42%. Volcanic ash particles mostly consist of basaltic glass (Arnalds 2015), but the composition of volcanic ash particles is highly variable with location. Some volcanic ash particles include compositions originating from the upper mantle such as olivine (Fischer et al. 2005). We consider feldspar, quartz, red clay, olivine, and Pinatubo volcanic ash particles to examine the applicability of the TAMUdust2020 database to various major dust aerosol and volcanic ash particles through the comparison of the bulk scattering properties between measurements and simulations.

Figure 9 compares the full phase matrices of the five aerosol samples at wavelengths of 441.6 and 632.8 nm obtained from the TAMUdust2020 database with the laboratory-measured counterparts (Muñoz et al. 2000, 2001; Volten et al. 2001). As the phase matrix elements near forward and backward scattering angles are not available in the laboratory measurements, $P_{11}$ values are normalized at a scattering angle of 30\(^\circ\). The refractive index, effective radius, and effective variance of the five aerosol samples are listed in Table 2. The comparisons of $P_{11}$, $-P_{12}/P_{11}$, $P_{33}/P_{11}$, and $P_{44}/P_{11}$ of feldspar and small olivine samples at both the wavelengths show reasonable consistency between simulations and laboratory measurements, and the simulated $P_{44}/P_{11}$ is fairly consistent with measured counterparts. The simulations of $P_{22}/P_{11}$ are biased high compared to the laboratory-measured counterparts. However, measured $P_{22}/P_{11}$ values near forward scattering directions are somewhat lower than unity that is theoretically expected due to a major contribution from diffraction, implying potential systematic low biases in the laboratory-measured $P_{22}/P_{11}$ values, which was also indicated by Lin et al. (2018).

The simulated scattering properties of quartz and Pinatubo volcanic ash are robustly consistent with laboratory-measured counterparts for all phase matrix elements including their spectral dependence. As seen in the images of these particles in

Fig. 8. As in Fig. 6, but with various degree of sphericity ($\Psi$). The size parameter and the complex refractive index are 30 and $1.5 + 0.003i$, respectively.
Table 2. Optical and microphysical properties of aerosol samples used in this study. The unit of \( r_{\text{eff}} \) is \( \mu m \). The \( \lambda \) indicates the wavelength of the measured optical properties, where \( \lambda_1 \) is 647 nm for the Eyjafjallajökull sample and 632.8 nm for Sahara, Spurr Ashton, and St. Helens samples; and \( \lambda_1 \) and \( \lambda_2 \) for other samples are 441.6 and 632.8 nm, respectively. Note that the reference values of the refractive indices do not consider their spectral dependences.

<table>
<thead>
<tr>
<th>Samples</th>
<th>( m_r )</th>
<th>( m_i )</th>
<th>( r_{\text{eff}} )</th>
<th>( v_{\text{eff}} )</th>
<th>( \Psi )</th>
<th>( m_r )</th>
<th>( m_i )</th>
<th>( r_{\text{eff}} )</th>
<th>( v_{\text{eff}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Eyjafjallajökull</td>
<td>1.43</td>
<td>0.0001</td>
<td>6.43</td>
<td>5.9</td>
<td>0.695</td>
<td>1.43–1.59</td>
<td>0–0.004</td>
<td>4.0</td>
<td>5.9</td>
</tr>
<tr>
<td>Feldspar</td>
<td>1.5</td>
<td>0.001</td>
<td>1.0</td>
<td>1.0</td>
<td>0.695</td>
<td>1.5–1.6</td>
<td>0.0001–0.001</td>
<td>1.0</td>
<td>1.0</td>
</tr>
<tr>
<td>Green clay</td>
<td>1.5</td>
<td>0.001</td>
<td>1.55</td>
<td>1.4</td>
<td>0.695</td>
<td>1.5–1.7</td>
<td>0.0001–0.001</td>
<td>1.55</td>
<td>1.4</td>
</tr>
<tr>
<td>Loess</td>
<td>1.5</td>
<td>0.0005</td>
<td>3.9</td>
<td>2.6</td>
<td>0.695</td>
<td>1.5–1.7</td>
<td>0.0001–0.001</td>
<td>3.9</td>
<td>2.6</td>
</tr>
<tr>
<td>Olivine (S)</td>
<td>1.62</td>
<td>0.0001</td>
<td>1.3</td>
<td>1.8</td>
<td>0.695</td>
<td>1.62</td>
<td>0.0001</td>
<td>1.3</td>
<td>1.8</td>
</tr>
<tr>
<td>Pinatubo</td>
<td>1.53</td>
<td>0.0001</td>
<td>3.0</td>
<td>12.3</td>
<td>0.695</td>
<td>1.5–1.6</td>
<td>0.0001–0.001</td>
<td>3.0</td>
<td>12.3</td>
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<tr>
<td>Quartz</td>
<td>1.54</td>
<td>0.0001</td>
<td>2.3</td>
<td>2.3</td>
<td>0.695</td>
<td>1.54</td>
<td>0</td>
<td>2.3</td>
<td>2.3</td>
</tr>
<tr>
<td>Red clay</td>
<td>1.5</td>
<td>0.001</td>
<td>1.56</td>
<td>1.6</td>
<td>0.695</td>
<td>1.5–1.7</td>
<td>0.0001–0.001</td>
<td>1.5</td>
<td>1.6</td>
</tr>
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<td>Sahara</td>
<td>1.5</td>
<td>0.0001</td>
<td>8.7</td>
<td>2.0</td>
<td>0.71</td>
<td>1.5–1.7</td>
<td>0.0001–0.001</td>
<td>8.2</td>
<td>4.0</td>
</tr>
<tr>
<td>Spurr Ashton</td>
<td>1.48</td>
<td>0.0018</td>
<td>3.8</td>
<td>1.6</td>
<td>0.71</td>
<td>1.48–1.56</td>
<td>0.0018–0.02</td>
<td>2.7</td>
<td>4.9</td>
</tr>
<tr>
<td>St. Helens</td>
<td>1.48</td>
<td>0.0018</td>
<td>5.8</td>
<td>4.7</td>
<td>0.715</td>
<td>1.48–1.56</td>
<td>0.0018</td>
<td>4.1</td>
<td>9.5</td>
</tr>
</tbody>
</table>

Fig. 9, quartz and Pinatubo volcanic ash particles are highly irregular and faceted, which is very consistent with the assumed particle shape model in the database. This may partly explain the robust consistency of all phase matrix elements between simulations and laboratory measurements.

The simulations of \( P_{11} \) and \(-P_{12}/P_{11}\) of red clay samples show reasonable consistency with laboratory measurements. However, biases in the simulations of other phase matrix elements compared to the measured counterparts are similar to those based on a superspheroid model (Lin et al. 2018). The red clay samples show a layered structure according to the SEM measurements (Volten et al. 2001; Reid et al. 2003), which is inconsistent with the assumption of the homogeneous particle used in this study. This insufficient implementation of the particle medium inhomogeneity may result in systematic biases in the phase matrix element simulations (Lindqvist et al. 2014). In consequence, the uncertainty associated with particle morphological characteristics may lead to uncertainty in the optically measured/estimated refractive index of such particles, as well as bias estimates obtained through downstream applications (e.g., flux calculations and dust aerosol property retrievals). Although some idealized particle shapes can reproduce the phase matrix elements of red clays (Jin et al. 2016) with a “tuned” refractive index, further investigations are needed to properly model the single-scattering properties of red clays and particles with significant heterogeneity in the particle medium. In the current stage, the TAMUdust2020 database offers reliable \( P_{11} \) and \(-P_{12}/P_{11}\) values for red clay based on a suggested refractive index.

We further extend the comparisons of the scattering properties into more samples of mineral dust aerosol and volcanic ash particles with a focus on the first three phase matrix elements. Figure 10 shows \( P_{11}, -P_{12}/P_{11}, \) and \( P_{22}/P_{11} \) of dust aerosol samples including green clays, loess, and Saharan dust particles at wavelength 441.6 nm (if available) and 632.8 nm. Simulations of the phase matrix of green clay show robust consistency with the measured counterpart in \( P_{11} \) including spectral dependence. However, \(-P_{12}/P_{11}\) is moderately biased, and \( P_{22}/P_{11} \) is systematically biased similarly as with red clays, presumably due to insufficient characterizations of particle medium inhomogeneity. The first three phase matrix elements of loess dust particle samples are fairly consistent with measured counterparts, including \( P_{22}/P_{11} \) near the backscattering direction. The simulated Saharan dust samples show reasonable, fair, and poor consistencies in \( P_{11}, -P_{12}/P_{11}, \) and \( P_{22}/P_{11} \) with the measured counterparts, respectively. Overall, an irregular hexahedral ensemble model is fairly applicable to the simulations of \( P_{11} \) and \(-P_{12}/P_{11}\) for a majority of clay dust aerosol particles.

Figure 11 shows \( P_{11}, -P_{12}/P_{11}, \) and \( P_{22}/P_{11} \) of volcanic ash samples including Eyjafjallajökull, Spurr Ashton, and St. Helens samples at a wavelength of 632.8 or 647 nm. Overall, the ensemble irregular hexahedral model demonstrates almost perfect consistency between the simulations and measurements for \( P_{11} \) and \(-P_{12}/P_{11}\), and fair consistency in \( P_{22}/P_{11} \) simulations with the measured counterparts, but \( P_{22}/P_{11} \) is slightly overestimated.

As demonstrated with Figs. 10 and 11, an ensemble irregular hexahedral model can provide robust spectrally consistent \( P_{11} \) and fairly consistent \(-P_{12}/P_{11}\) simulations for dust aerosol and volcanic ash particles. Since past and current multispectral polarimetric passive sensor measurements typically rely on the first two phase matrix elements of the aerosol particles in a plume, the irregular hexahedral ensemble model can be
systematically used for dust aerosol property retrievals based on any combination of passive sensor measurements.

c. Comparison of the backscattering properties

A striking superiority of the TAMUdust2020 database is robust backscattering properties. It is of interest to demonstrate the performance of the backscattering simulations based on an ensemble irregular hexahedral model. We compute the lidar ratio ($S$) and depolarization ratio ($\delta$) of the 11 aerosol samples at each available wavelength (Figs. 9–11) using the same refractive indices and PSDs of these aerosol samples listed in Table 2. The bulk lidar and depolarization ratios are defined as (with omitting angle brackets):

$$S = \frac{4\pi}{\omega P_{11}(\pi)}$$  \hspace{1cm} (18)

$$\delta = \frac{P_{11}(\pi) - P_{22}(\pi)}{P_{11}(\pi) + P_{22}(\pi)}$$  \hspace{1cm} (19)

Figure 12 shows a scatterplot of $S$ and $\delta$ of these aerosol samples. In general, $S$ and $\delta$ of these aerosol samples range from 30 to 60 and from 0.25 to 0.45 at wavelengths of 441.6, 632.8, and 647 nm. Schuster et al. (2012) summarized $S$ estimated from the Aerosol Robotic Network (AERONET) in various locations over the African continent through the Middle East. For “pure” dust cases, $S$ ranges from 38.7 to 57.8 sr at 532 nm with substantial regional dependence. Several studies using ground-based lidars demonstrate that $S$ and $\delta$ range from 38 to 80 sr and 0.15 to 0.34 for Saharan dust events (Ansmann et al. 2003; Esselborn et al. 2009; Veselovskii et al. 2016), and from 39 to 45 sr and 0.2 to 0.35 in Asian dust events (Murayama et al. 2004; Hofer et al. 2017; Hu et al. 2020). In addition, these aerosol samples have weak spectral dependence of $S$ and $\delta$, with variations under 10% within a spectral domain. Although the selected 11 aerosol samples may not fully cover the range of airborne dust and volcanic ash particles, the variability of the bulk backscattering properties of these particles is mostly within those observed ranges.
To further validate the applicability of the ensemble irregular hexahedral model for backscattering properties, we compare the simulated $S$ and $\delta$ with the counterparts estimated from ground-based multiwavelength Raman lidar observations for two cases: Sahel dust loading, mainly from Mali, on 2–3 April 2015 (Veselovskii et al. 2020) and a Taklamakan dust plume on 9 April 2019 (Hu et al. 2020). The source regions of these dust events are confirmed with

FIG. 10. Comparison of (top) $P_{11}$, (middle) $-P_{12}/P_{11}$, and (bottom) $P_{22}/P_{11}$ of various dust aerosol particles between in situ measurements and simulations based on the TAMUdust2020 database, including (left) green clay, (center) loess, and (right) Saharan dust. The samples used in this study are listed in Table 2.

FIG. 11. As in Fig. 10, but for various volcanic ash particles, including (left) Eyjafjallajökull, (center) Spurr Ashton, and (right) St. Helens. The samples used in this study are listed in Table 2.
backward trajectory analysis. The bulk backscattering properties of dust aerosol particles in these two events are computed based on measured/assumed PSDs and refractive indices of dust aerosol particles sampled in a different period in these source regions (Di Biagio et al. 2019). The directly sampled aerosol properties should be representative of those in the 2015 and 2019 events as mineralogical characteristics do not change in a short time period.

In the bulk backscattering simulations, we assume the effective radius of dust plume particles to be 0.75 \( \mu \)m, estimated from AERONET for the Mali dust plume case, and to be 0.75 \( \mu \)m with \( \pm 100\% \) uncertainty for the Taklamakan dust plume case because collocated particle size measurements are not available. A degree of sphericity of 0.71 \( \pm 0.01 \) is used for the lidar simulations. Figure 13 shows \( S \) and \( \delta \) at available wavelengths for the two dust events based on observations and simulations. For the Mali dust plume case, \( S \) and \( \delta \) from simulations are fairly consistent with measured counterparts at 532 nm. In addition, the spectral dependence of \( S \) is successfully reproduced in the simulations. For the Taklamakan dust plume case, \( \delta \) at three wavelengths and \( S \) at 532 nm are reasonably consistent between measurements and simulations. However, the simulated \( S \) at 355 nm are systematically smaller than the measured counterparts, which lead to a weak spectral dependence that is not very consistent with measurements. This could be partly due to more strong absorption of the particles than those assumed in the simulations and the effects of small-scale surface morphological characteristics of atmospheric aerosol particles, which are not considered in the ensemble model.

\section{5. Applications}
\subsection{a. Bulk dust particle mixture models}

Figure 14 shows refractive indices of eight dust aerosol particle samples (Tunisia, Morocco, Libya, Algeria, Mauritania, Australia, Namib-1, and Namib-2) obtained from laboratory measurements (Di Biagio et al. 2017, 2019). In the SW range, \( m_t \) varies by an order of magnitude among samples, and shows moderate spectral variation. The spectral variations of both the \( m_t \) and \( m_i \) associated with various chemical compositions are much larger in the LW range than the SW range. For example, major peaks of \( m_i \) at 7.0, 8.7, 9.6, and 12.5–12.9 \( \mu \)m indicate the presence of calcite, feldspar, clay minerals, and quartz, respectively.

In this study, we develop three dust aerosol optical property models for northern Africa–Sahara (NAF-S), Australia (AUS), and southern Africa (SAF), according to Di Biagio et al. (2019). First, we obtain single-scattering property tables of the aforementioned eight dust aerosol samples from the TAMUdust2020 database using their corresponding dust refractive indices. Then we compute the bulk optical properties of each dust aerosol model considering a mixture of multiple dust aerosol samples. The NAF-S model is an equally weighted mixture of Tunisia, Morocco, Libya, Algeria, and Mauritania samples. The SAF model is based on an equally weighted mixture of Namib-1 and Namib-2 samples. The AUS model simply relies on the Australia sample. The \( \Psi \) in these models and the geometric standard deviations of PSDs are assumed to be 0.7 (unitless) and 2.8 \( \mu \)m, respectively. The effective radius \( (r_{\text{eff}}) \) of a nonspherical particle is defined as

\begin{equation}
    r_{\text{eff}} = \frac{3}{4} \frac{\int_{D_{\text{max}}}^{D_{\text{min}}} n(D)V(D, \Psi) dD}{\int_{D_{\text{min}}}^{D_{\text{max}}} n(D)A(D, \Psi) dD}.
\end{equation}
Figure 15 shows the spectral dependence of $S$, $Q_{\text{ext}}$, SSA, and $g$ of the bulk dust aerosol models, each with three $r_{\text{eff}}$. In the SW domain, $Q_{\text{ext}}$ primarily varies with $r_{\text{eff}}$ and has moderate spectral dependence for small particles since $Q_{\text{ext}}$ increases monotonically with increasing size parameters in this size parameter range (Figs. 6a, 7a, and 8a). The SSA, $g$, and $S$ have moderate spectral dependence and vary with both $r_{\text{eff}}$ and the dust refractive index. In particular, wavelengths $<0.6 \, \mu m$ have relatively larger $m_i$ values (Fig. 14), resulting in a smaller SSA, larger $g$, and larger $S$. In the LW domain, the three optical properties are sensitive to both the particle size and the index of refraction. Large dust aerosol particles ($r_{\text{eff}} > 1.0 \, \mu m$) have noticeable impacts on thermal infrared radiative transfer due to moderately large $Q_{\text{ext}}$ values. In addition, the variation of the refractive index in the LW range causes substantial variations of optical properties, especially in SSA.

b. Satellite sensor simulations with various AODs and effective radii of dust plumes

To demonstrate the usefulness of the TAMUdust2020 database for forward radiative transfer simulations involved in aerosol remote sensing applications, we perform simulations of spaceborne sensor observations from POLDER, CALIOP, and IIR for various dust plume cases. Figure 16 demonstrates the sensitivities of these spaceborne sensor signals to AOD and/or $r_{\text{eff}}$ in dust plumes using the NAF-S dust model, assuming a homogeneous single-layer dust plume located over ocean. The U.S. standard atmospheric gases profile and sea surface temperature of 300 K are used. The top height and geometric thickness of the dust plume layer are 4 and 2 km, respectively. AOD is defined at a reference wavelength of 0.55 $\mu m$.

For the POLDER simulations, the total and normalized modified polarized reflectances ($L_{\text{nmp}}$) in the 0.865 $\mu m$ channel are computed with an adding-doubling vector radiative transfer model (Huang et al. 2015); $L_{\text{nmp}}$ is defined as

$$L_{\text{nmp}} = \frac{\pi \eta (\mu_0 + \mu) \sqrt{Q^2 + U^2}}{\mu_0 E_0},$$

where $Q$ and $U$ are the second and third Stokes parameters, $\mu_0$ and $\mu$ are the cosines of solar and viewing zenith angles, $E_0$ is solar irradiance at the top of atmosphere, and $\eta = \pm 1$ (see C.-Labonnote et al. 2001). AOD is assumed to be 1. In the simulation, the solar zenith angle of 20°, relative azimuth angle of 0°, and various viewing zenith angles are considered. For $r_{\text{eff}} > 2 \, \mu m$, the total reflectance increases as $r_{\text{eff}}$ increases due to an increase of the ratio of $Q_{\text{ext}}$ between the POLDER band and the reference wavelength. For larger $r_{\text{eff}}$ cases, the total reflectance decreases with increasing $r_{\text{eff}}$ due to a pronounced reduction of SSA (Figs. 15 and 16a). On the other hand, the angular distribution of $L_{\text{nmp}}$ exhibits a monotonic increase with increasing $r_{\text{eff}}$ (Fig. 16b). For CALIOP simulations, the layer integrated attenuated backscatter (IAB), volume depolarization ratio at 532 nm
channel, and the color ratio using 532 and 1064 nm channels are computed with a Monte Carlo radiative transfer model (Hu et al. 2001). The IAB proportionally increases with increasing AOD, and also IAB is a function of dust aerosol \( r_{\text{eff}} \) due to the variation of \( Q_{\text{ext}} \) with \( r_{\text{eff}} \) (Fig. 16c). The volume depolarization ratio is sensitive to both AOD and \( r_{\text{eff}} \) (Fig. 16d). As AOD increases, the contribution of the Rayleigh scattering by molecules to the polarized backscattering signals decreases, resulting in a large volume depolarization ratio. As seen in Fig. 3, \( P_{22}/P_{11} \) at the backscattering direction decreases with increasing size parameter, contributing to the sensitivity of the volume depolarization ratio to \( r_{\text{eff}} \). Furthermore, Fig. 16e shows that the color ratio is sensitive to \( r_{\text{eff}} \) as well, due to a monotonic variation of the ratio of \( Q_{\text{ext}} \) between 1064 and 532 nm.

For the IIR simulations, the brightness temperature (BT) in the 12.05 \( \mu \text{m} \) channel and two brightness temperature differences (BTD; 8.65 \( \mu \text{m} \) or 10.6 \( \mu \text{m} \) minus 12.05 \( \mu \text{m} \)) are computed with a two-stream radiative transfer model (Iwabuchi et al. 2016; Saito et al. 2017). The BT in the 12.05 \( \mu \text{m} \) channel is sensitive to AOD and a large \( r_{\text{eff}} \) due to the absorption in and radiative emission from the dust plume layer. The magnitude of the radiative emission also depends on the bulk SSA of the dust aerosol model in the split-window bands. Small dust particles with \( r_{\text{eff}} < 1 \mu \text{m} \) are not radiatively important in the thermal infrared wavelengths since they have a small \( Q_{\text{ext}} \) value (Fig. 15) and therefore a small optical thickness at these wavelengths. For dust plumes with intermediate \( r_{\text{eff}} \), BTDs between the 10.6 and 12.05 \( \mu \text{m} \) channels decrease with increasing AOD (Fig. 16h), while BTDs between the 8.65 and 12.05 \( \mu \text{m} \) channels increase with increasing AOD (Fig. 16g) in accordance with the spectral dependence of the bulk \( Q_{\text{ext}} \) value that has a local maximum at \( \sim 10 \mu \text{m} \) (Fig. 15). This implies that the spectral dependence of a dust aerosol optical property model at thermal infrared wavelengths must be considered to quantitatively interpret the radiative signals.

c. Satellite sensor simulations with various bulk dust particle mixture models

To investigate the impacts of dust aerosol models on the satellite sensor simulations, we compare the same three satellite sensor simulations (POLDER, CALIOP, and IIR) and dust aerosol optical property models (NAF-S, SAF, and AUS).

Fig. 15. The bulk optical properties of three dust aerosol mixture models as shown in solid (NAF-S), dotted (AUS), and dashed lines (SAF) for (left) SW and (right) LW radiative domains. Line colors indicate effective radii.

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Figure 17 shows satellite sensor simulations for the same dust aerosol models and \( r_{\text{eff}} \) cases as in Fig. 15. The total reflectances computed based on the AUS dust aerosol model are systematically smaller than the other two dust aerosol models, in particular for large \( r_{\text{eff}} \) cases at smaller scattering angles (Fig. 17a) due to stronger absorption at 0.865 \( \mu \text{m} \) (Fig. 15). This implies the importance of a relevant choice of dust aerosol optical property model for aerosol retrievals with multiangle reflectance observations. The normalized modified polarized reflectance is less sensitive to particle absorptivity (Fig. 17b) but is known to be sensitive to particle shape (Huang et al. 2015).

For CALIOP simulations, the \( m_3 \) of a dust aerosol model has noticeable impacts on IAB due to high sensitivity of \( S \) to \( m_3 \), as seen in Fig. 6d. Figure 17c exhibits highest IAB for the SAF dust aerosol model, followed by NAF-S or AUS dust aerosol models, depending on \( r_{\text{eff}} \). The sensitivity of the volume depolarization ratio (Fig. 17d) to dust aerosol models is not negligible, but less important in particular for larger particles. The color ratio is also influenced by spectral dependence of the dust refractive index (Fig. 17e). Therefore, it is important to use an appropriate dust refractive index model to retrieve aerosol optical properties using lidar measurements.

The IIR simulations exhibit the largest impact of dust aerosol models on sensor signals among the three spaceborne sensors. Figure 17f shows the variation of BT in the 12.05 \( \mu \text{m} \) channel associated with differing absorptivities among dust aerosol models, as seen in Fig. 15. In particular, BTD is largely influenced by the spectral dependence of the dust refractive index at thermal infrared wavelengths (Figs. 17g,h). Also, Bi et al. (2020) show that simulations of thermal infrared spectra exhibit apparent differences among dust aerosol particle models. This is crucial for dust plume detection based on a fixed BTD threshold technique, because the minimum detectable AOD of a dust plume may be different among dust aerosol types, leading to regional biases of dust plume occurrence.

6. Summary and remarks

A comprehensive database of the single-scattering properties of irregular dust aerosol particles (TAMUdust2020) was developed using a combination of the IITM, IGOM, and PGOM. The database includes the single-scattering properties of an ensemble of 20 irregular hexahedral particles in the size parameter range from the Rayleigh scattering domain up to 11 800, in the refractive index range covering typical dust aerosol and volcanic ash particles, and in the degree of sphericity range from 0.695 to 0.785. The ensemble of 20 irregular hexahedral particles mimics the diversity of complex dust particle shapes in nature. The single-scattering properties of this ensemble, including full phase matrix elements, show sensitivities to variations in refractive index, size, and the degree of sphericity.

The bulk dust aerosol phase matrix elements of reported dust aerosol and volcanic ash samples simulated with the TAMUdust2020 database are reasonably consistent with those from laboratory measurements, implying that the assumed particle shape is robust. Also, the lidar and depolarization ratios of the bulk dust aerosol models computed based on the TAMUdust2020 database in almost all cases fall...
within the range of available lidar observations. Furthermore, ground-based lidar signals of dust plumes and simulated counterparts in Mali and Taklamakan dust cases exhibit robust consistency, indicating that the backscattering properties of particles in the TAMUdust2020 database are reliable.

In addition, we performed simulations of signals from spaceborne instruments such as POLDER, CALIOP, and IIR for various dust plume cases using the bulk dust aerosol optical properties computed from the TAMUdust2020 database, demonstrating sufficient sensitivity to not only AOD but also the effective radius. The TAMUdust2020 database in conjunction with a lidar simulator enables physical interpretation of CALIOP signals in terms of optical and microphysical properties for dust plumes consisting of coarse-mode nonspherical particles (Figs. 16c–e). As the bulk dust aerosol optical properties are physically consistent among different spaceborne sensor simulations, the TAMUdust2020 database is also useful for multi-satellite-based aerosol optical property retrievals, such as combined active–passive aerosol remote sensing. Previous dust modeling generally is based on a single global aerosol model, which may lead to systematic regional biases of aerosol detection and/or retrievals based on lidar or thermal infrared observations due to geophysical variations in dust aerosol chemical compositions and other characteristics. The TAMUdust2020 database allows user specification of dust plume properties (e.g., wavelength-dependent complex refractive index and user-specified aerosol particle diameters) to provide customized computation of single-scattering properties.

The TAMUdust2020 database is a useful resource for radiative transfer simulations and remote sensing involving atmospheric aerosols such as dust and volcanic ash plumes. This database is designed to improve aerosol optical property retrievals using ground-based, airborne, and spaceborne multisensor measurements. The A-Train satellite constellation has provided passive sensor measurements spanning from visible to thermal infrared channels, multiangle polarimetric observations, and lidar observations of dust aerosol plumes on a global scale for more than a decade. In addition, the future Earth Clouds, Aerosols and Radiation Explorer (EarthCARE) mission (Illingworth et al. 2015) will provide both spaceborne passive and active sensor measurements. The TAMUdust2020 database can exploit such current and future spaceborne active–passive sensor observations to significantly advance our knowledge of the detailed global climate effects of atmospheric aerosol particles.

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Data availability statement. The TAMUdust2020 database is publicly available at https://doi.org/10.5281/zenodo.4711247. Geometrical information of 20 irregular hexahedral particles is available as supplemental data.

APPENDIX A

A Combination of PGOM and IGOM

Both IGOM and PGOM are based on the principle of geometric optics and mapping techniques from the near-field to far-field with an electromagnetic surface integral equation. A notable difference between these two methods is whether or not the interference among diffracted fields and various ray-tracing fields in the scattering field is considered. IGOM partially considers the phase interference of outgoing waves through the incorporation of the ray-spreading effect; but PGOM utilizes rigorous electromagnetic integrals to account for the interference of scattered waves. This evaluation of electromagnetic integrals substantially increases the computational burden in PGOM. The effect of the abovementioned interferences mostly cancels out for randomly oriented particles except for the coherent backscattering enhancement (CBE).

The amplitude factor of CBE is derived by Zhou (2018), indicating the characteristic angular width of a CBE-induced backscattering peak. A critical off-backscattering angle \( \theta_c = \pi / 4D \).

The phase function with off-backscattering angles < \( \theta_c \) is amplified by CBE (Zhou 2018). To seamlessly combine IGOM and PGOM calculations, we define two thresholds of off-backscattering angles (\( \theta_{t1} = 80^\circ \) and \( \theta_{t2} = 40^\circ \)) that are sufficiently larger than the critical off-backscattering angle. IGOM calculations are used at off-backscattering angles > \( \theta_{t1} \); PGOM calculations are used at off-backscattering angles < \( \theta_{t2} \). Phase matrix elements at angles between \( \theta_{t1} \) and \( \theta_{t2} \) are obtained from IGOM and PGOM through an angle-dependent weighting average.

Figure A1 shows an example of \( P_{11} \) computed with IGOM and PGOM, and two thresholds of off-backscattering angles for the case of size parameter 150 and refractive index 1.5 + 0.005. The phase function at off-backscattering angles \( \theta_{t1} \) and \( \theta_{t2} \) are sufficiently larger than the angular width of the CBE-induced peak. Note that there are several differences in the assumptions and numerical implementations between IGOM and PGOM (section 2e), which induce a difference in the phase matrix elements at off-backscattering angles as seen in Fig. A1.

APPENDIX B

Estimation of the Error due to Multidimensional Linear Interpolation

The linear interpolation is performed with respect to the three parameters (herein referred to as \( x, y, \) and \( z \)) as described in section 2d and Eq. (12). Therefore, the error \( e_k \) due to the linear interpolation at \( (x, y, z) \) can be described through the Taylor series as

\[
e_k(x, y, z) \leq \frac{(x_{i+1} - x)(x - x_i)}{2} \frac{\partial^2 K}{\partial x^2} + \frac{(y_{j+1} - y)(y - y_j)}{2} \frac{\partial^2 K}{\partial y^2} + \frac{(z_{k+1} - z)(z - z_k)}{2} \frac{\partial^2 K}{\partial z^2}, \tag{B1}\]

where \( x \in [x_i, x_{i+1}], y \in [y_j, y_{j+1}], \) and \( z \in [z_k, z_{k+1}] \). The maximum absolute error \( e_{K,max} \) within a grid cube due to the multidimensional linear interpolation is estimated as

\[
|e_{K,max}(x, y, z)| \leq \frac{\Delta x^2}{8} \max_{x_i < x < x_{i+1}} |\partial^2 K/\partial x^2| + \frac{\Delta y^2}{8} \max_{y_j < y < y_{j+1}} |\partial^2 K/\partial y^2| + \frac{\Delta z^2}{8} \max_{z_k < z < z_{k+1}} |\partial^2 K/\partial z^2|, \tag{B2}\]

where \( \Delta x = x_{i+1} - x_i, \Delta y = y_{j+1} - y_j, \) and \( \Delta z = z_{k+1} - z_k \). To estimate \( \partial^2 K/\partial x^2, \partial^2 K/\partial y^2, \) and \( \partial^2 K/\partial z^2 \), we use the natural cubic spline polynomial fit in the neighboring grids of interest.

We apply this procedure to all grids in the kernels to obtain \( |e_{K,max}| \). Then, we use the kernel technique (section 2d) for both \( K \) and \( K + |e_{K,max}| \) and the error propagation formulae to estimate the maximum absolute error in the single-scattering properties due to multidimensional linear interpolation. Note that the maximum absolute error may be
slightly underestimated when third- or higher-order derivatives omitted in Eq. (B2) are not negligible.

REFERENCES


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