



15



Abstract

The lidar backscattering properties of Asian dust particles, namely the lidar ratio (S) and backscattering depolarization ratio (δ), were studied using a discrete dipole approximation (DDA) model. The three-dimensional morphology of the dust particles was reconstructed in fine detail using the focused ion-beam (FIB) tomography technique. An index based on the symmetry of the scattering phase matrix was developed to assess the convergence of random orientation computation using DDA. Both the S and δ exhibit an asymptotic trend with dust particle size: the S initially decreases while the δ increases with size, before both approach their asymptotic values. The lidar properties were found to have statistically insignificant dependence on effective sphericity. The presence of strongly absorbing minerals, such as magnetite, can greatly reduce the dust's single-scattering albedo and δ . Utilizing the robust asymptotic trend behavior, two parameterization schemes were developed: one to estimate the δ of a single dust particle given its size, and the other to estimate the δ of dust particles with a lognormal particle size distribution given the effective radius. The parameterization scheme was compared with results based on the TAMUdust2020 database, showing hexahedrals to reasonably represent realistic geometries with similar physical properties.



34 **1. Introduction**

35 Dust aerosols are an important component of the Earth System, interacting with Earth's energy,
36 water, and carbon cycles. Directly, dust aerosols scatter and absorb both shortwave and longwave
37 radiation, influencing the planet's energy balance (Tegen et al., 1996; Miller and Tegen, 1998;
38 Myhre et al., 2013; Song et al., 2018, 2022). By scattering incoming solar radiation, dust aerosols
39 contribute to cooling the atmosphere and surface regionally, impacting temperatures and
40 affecting atmospheric circulation patterns (Evan et al., 2006; Lau and Kim, 2007; Zhang et al.,
41 2022). For example, Asian dust transportation to the Tibetan Plateau causes direct radiation
42 effects which result in enhancement to the monsoon season (Lau et al., 2006). In addition, the
43 absorption of longwave radiation, particularly thermal infrared radiation, by dust particles leads
44 to surface warming effect by 0.54 W m^{-2} on a global average (Song et al., 2022). Indirectly, dust
45 aerosols serve as nuclei for cloud condensation and ice nucleation, altering cloud microphysical
46 properties and precipitation patterns (Atkinson et al., 2013; Field et al., 2006; Kanji et al., 2017).
47 These indirect effects can further influence regional and global climate dynamics by modifying
48 cloud albedo and distribution (Johnson et al., 2004; Huang et al., 2006; Helmert et al., 2007;
49 Amiri-Farahani et al., 2019).

50

51 The transport of this dust also has far-reaching implications. The long-range transport of Asian
52 dust is frequently observed on the United States' west coast with considerable impacts on the air
53 quality and climate (Yu et al., 2012; Creamean et al., 2014; Wu et al., 2015). Moreover, the
54 deposition of dust aerosol during the long-range transport brings essential nutrients such as iron
55 and phosphorus from terrestrial sources to marine ecosystems, facilitating the working of
56 biogeochemical cycles across vast distances (Baker et al., 2003; Yu et al., 2015b; Westberry et



57 al., 2023). Asian dust deposition in the East China Sea stimulates phytoplankton growth and
58 primary productivity, influencing marine food webs and carbon cycling (Kong, S. S.-K. et al.,
59 2022).

60

61 Lidar is an important tool for remote sensing measurements of airborne dust particles. As
62 demonstrated in many previous studies (Omar et al., 2009; Burton et al., 2012), it allows us to
63 distinguish dust aerosols from clouds and other types of aerosols, track their long-range transport
64 and study their evolution as they interact with the environment such as clouds, atmospheric
65 gasses, and other aerosols. Among others, elastic backscattering lidars are one of most widely
66 used types of lidar. For example, Cloud-Aerosol Lidar and Infrared Pathfinder Satellite
67 Observations (CALIPSO), is a NASA-French satellite mission that implements a two-
68 wavelength elastic lidar Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) at 532
69 nm and 1064 nm wavelengths (Winker et al., 2009). Ground-based lidar networks such as the
70 NASA Micro-Pulse Lidar Network (MPLNET) use single wavelength measurements for
71 extinction, backscattering, and depolarization profiles (Welton et al., 2001).

72

73 Lidar ratio (S) and depolarization ratio (δ) are two most important parameters for lidar-based
74 remote sensing of aerosols and clouds. For a single dust particle, the S , referred to as the
75 extinction-to-backscatter coefficient, is defined as (Platt, 1979; Ansmann et al., 1992; Mattis et
76 al., 2002; Liu et al., 2002)

77

$$S = \sigma/\beta = \frac{4\pi}{\omega P_{11}(\theta_s = \pi)}, \quad (1)$$



78 where σ is the extinction coefficient and ω and P_{11} are the bulk optical properties of polydisperse
79 dust particles, the single-scattering albedo and phase function of the dust particle, respectively.
80 For the purposes of this paper, P_{11} is normalized to 1 when integrating across all scattering
81 directions. $\beta = P_{11}(\theta_s = \pi)C_{sca}$ is the backscattering coefficient at the exact backscattering
82 direction; defined for a normalized particle size distribution as

$$\beta = \int_{-\infty}^{\infty} P_{11}(r_v, \theta_s = \pi) C_{sca}(r_v) n(r_v) d \ln r_v, \quad (2)$$

83 Where r_v is the volume-equivalent sphere radius and $n(r_v) = dN/d \ln r_v$ defines a normalized
84 particle size distribution ($n(r_v)$). For Raman lidar and high spectral resolution lidar systems, the
85 lidar ratio can be derived directly from the observed extinction and backscatter without
86 assumptions about the composition (Müller et al., 2007). However, for elastic backscattering
87 lidars, the lidar ratio cannot be directly measured. As a result, assumptions need to be made about
88 the composition of the atmosphere. Therefore, it is fundamentally important for elastic lidars like
89 CALIOP and MPLNET to convert the direct attenuated backscatter observations to an extinction
90 profile (Young et al., 2018) and derived quantities such as dust aerosol optical depth (Yu et al.,
91 2015a; Song et al., 2021).

92

93 Depolarization ratio δ is the ratio of the perpendicular or cross-polarized component to the
94 parallel component of polarized backscattering signal. For backscattering lidar the
95 depolarization ratio is defined as

96

$$\delta = \frac{1 - \frac{P_{22}(\theta_s = \pi)}{P_{11}(\theta_s = \pi)}}{1 + \frac{P_{22}(\theta_s = \pi)}{P_{11}(\theta_s = \pi)}}, \quad (3)$$



97 where P_{ij} is the ij -th element of the particle's scattering phase matrix (Kong, S. et al., 2022). δ
98 is highly useful for aerosol and cloud classifications. First of all, if lidar backscattering is
99 dominated by single scattering, δ is close to zero for spherical or quasi-spherical particles like
100 smoke aerosols and water droplets. In contrast, δ is notably greater for nonspherical particles
101 like dust aerosols and ice crystals. Therefore, δ is often used for aerosol type (Kim et al., 2018)
102 and cloud phase classifications (Hu et al., 2009). Moreover, the considerable δ differences
103 between spherical fine particles and nonspherical coarse dust particles also enables the separation
104 of dust extinction from the total extinction profile retrieved by CALIOP (Yu et al., 2015; Song
105 et al., 2021).

106

107 Because of the fundamental importance of S and δ for lidar based dust remote sensing, previous
108 studies have made substantial effort to understand the connection between dust particle
109 properties, e.g., shape and size, and their lidar characteristics, in particular the S and δ (e.g.,
110 Dubovik et al., 2006; Gasteiger et al., 2011; Liu J. et al., 2015; Kahnert et al., 2020; Saito et al.,
111 2021; Saito and Yang, 2021; Kong, S. et al., 2022). The common methodology used in these
112 studies is to use light scattering models, such as the T-matrix (Mishchenko et al., 1996; Bi and
113 Yang, 2014b) and Discrete Dipole Approximation (DDA) model (Draine and Flatau, 1994, 2013;
114 Yurkin and Hoekstra, 2007, 2011), to compute the scattering properties including S and δ of dust
115 aerosols and then study the potential dependence on particle properties. Although these studies
116 have greatly improved our understanding and paved the foundation for the current aerosol
117 retrieval algorithms, they share a common limitation as they all used hypothetical dust particle
118 shape models, such as spheroid (Dubovik et al., 2006), irregular polyhedron (Saito et al., 2021),
119 Gaussian random sphere (Muinonen et al., 1996; Liu J. et al., 2015; Kahnert et al., 2020), tri-



120 axial spheroids (Meng et al., 2010; Huang et al., 2023), and super-spheroid (Kong, S. et al., 2022)
121 to simulate dust particle shapes that are weakly or *not* constrained by observations. The reason
122 for this is probably two-fold. Most microscopic observations of dust particles in the literature are
123 two-dimensional (2D) images based on scanning or transmission electron microscopes (SEM or
124 TEM), while three-dimensional (3-D) observations are extremely rare. In addition, the
125 implementation of complex shapes in scattering models is also a challenging task. For example,
126 until recently the widely used T-matrix code based on the extended boundary condition method
127 (Mishchenko et al., 1996) is primarily applicable only to rotationally symmetric particles such
128 as spheroid. It is worth noting that the T-matrix method implementation based on the invariant
129 imbedding T-matrix method is applicable to arbitrary shapes (Bi and Yang, 2014a). Aware of
130 the limitation of hypothetical dust particle shape, these studies often use dust scattering
131 properties from laboratory measurements as benchmark to select an optimal set of hypothetical
132 shapes that are able to generate similar scattering properties, e.g., lidar characteristics, as
133 measurements (Saito et al., 2021; Kong, S. et al., 2022). Nevertheless, the use of hypothetical
134 instead of realistic dust shape inevitably leads to some important questions. Is the match of the
135 dust scattering properties a result of a good shape model or a fortunate coincidence? If an optimal
136 shape model is selected based on one set of dust scattering observations (e.g., δ at 532 nm), can
137 this model automatically simulate other scattering properties (e.g., δ at other wavelengths)?
138 Obviously, one way to address the above questions is to use realistic shape models in the
139 computation of dust scattering properties. A few studies have made attempts along this direction.
140 For example, Lindqvist et al. (2014) developed a so-called stereogrammetric surface retrieval
141 method to construct 3-D dust shapes from 2D SEM dust images. Järvinen et al. (2016) compared
142 the lidar backscattering properties based on the constructed 3-D dust shapes with laboratory



143 measurements and found reasonable agreements. An important finding from this study is that δ
144 of realistic dust particles at 532 nm first increases with particle size but seems to approach an
145 asymptotic constant value of ~ 0.30 for coarse dust particles.

146

147 The main objective of this study is to better understand the lidar backscattering properties of dust
148 particles with realistic shapes. The dust shape models used here are based on the focused ion-
149 beam (FIB) tomography technique, aided by the energy dispersive X-ray spectroscopy (EDX)
150 and SEM imaging, developed by Conny et al. (2014) and Conny and Ortiz-Montalvo (2017),
151 which as far as we know is the most direct and faithful measurement of the shape and
152 morphology of single dust particles. In addition to shape measurement, the EDX is used to
153 measure the mineral composition of dust particles, which in turn enables the estimation of the
154 complex refractive index (CRI) of dust particles. Based on the measured dust particle shape and
155 estimated CRI, Conny et al. (2019, 2020) simulated and studied the scattering properties such as
156 single scattering albedo and phase functions of the dust samples using the DDASCAT model
157 (Draine and Flatau, 1994, 2013).

158

159 In this study, we focus on the lidar backscattering properties of realistic dust samples obtained
160 from FIB tomography measurements (Conny et al., 2019). For simplicity, we will refer to these
161 dust samples as "FIB dust samples." We are particularly interested in the following questions:
162 How do the S and δ of realistic dust samples vary with particle size, shape, mineral composition,
163 and lidar spectral channel? The remaining portion of the paper is organized as follows: First, in
164 Section 2, we introduce the dust samples used in this study, along with their origins and
165 properties. We also explain the Amsterdam Discrete Dipole Approximation (ADDA) model and



166 introduce a convergence index to determine the number of orientations necessary for calculating
167 the optical properties under the random orientation condition. In Section 3, we examine how the
168 lidar backscattering properties of the dust samples depend on dust properties, including size,
169 shape, and mineral composition. In Section 4, we present two dust δ parameterization schemes:
170 one to estimate the δ of a single dust particle based on its size, and the other to estimate the δ of
171 dust particles with a lognormal particle size distribution based on the effective radius. Finally, in
172 Section 5, we summarize the main findings and conclusions of this study.

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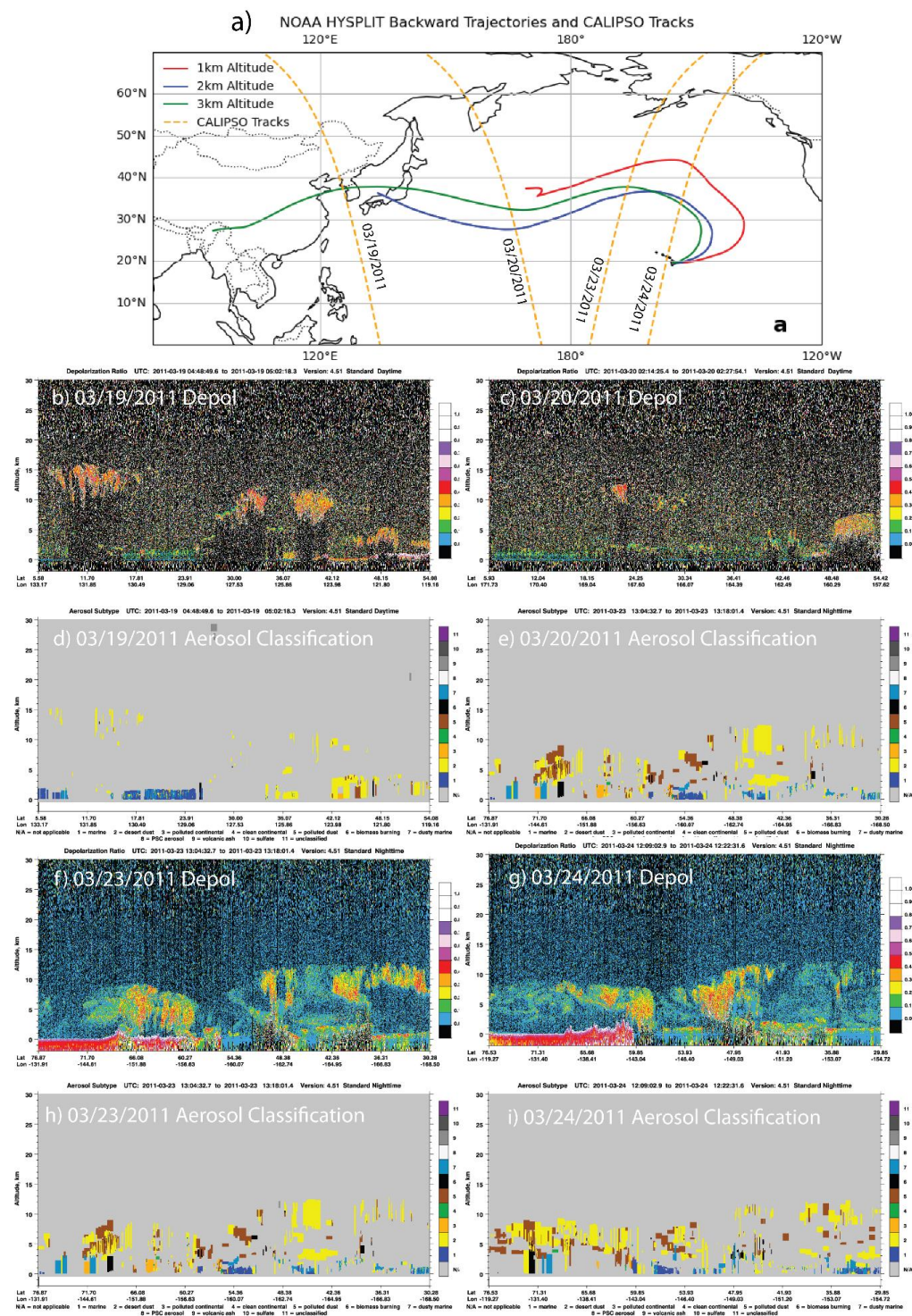
174 **2. Data and model**

175 **2.1. FIB Dust Samples**

176 The fourteen FIB dust particles were obtained from the Mauna Loa Observatory (19° 32' 10"N,
177 155° 34' 34"W) on the island of Hawaii between March 15 and April 26, 2011. Six of these
178 particles were collected during the daytime. Following Conny et al. (2019), these particles will
179 be referred to as the "D" sample (e.g., "3D" indicates that the sample was collected during the
180 daytime of day 3). The other eight particles were collected at night and are referred to as "N"
181 samples. The properties of these particles, including their shape, size, and composition, as well
182 as the measurement techniques, have been extensively documented in (Conny et al., 2019, 2020).
183 Conny et al., (2019) analyzed the back trajectories from the Mauna Loa Observatory during this
184 time interval, suggesting that their samples likely originated as Asian dust. Out of curiosity, we
185 collocated the CALIOP observations with the back trajectories from the Hybrid Single-Particle
186 Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015; Rolph et al., 2017) from
187 March 25, 2011, 0000 UTC to March 18, 2011, 0000 UTC, starting from the Mauna Loa
188 Observatory. The lidar depolarization ratio observations and aerosol classification (Figure 1d



189 and e) results show large amounts of dust along the later portion of the projected path between
190 March 23 and March 24, 2011. The back trajectories and CALIOP observations confirm that the
191 FIB dust samples are likely long-range transported Asian dust particles, consistent with Conny
192 et al. (2019).





194

195 **Figure 1.** (a) NOAA HYSPLIT Backward Trajectory paths from March 25th, 2011 0000 UTC
196 to March 18th, 2011 0000 UTC starting from Mauna Loa Observatory shown in solid lines.
197 Vertical dashed lines show CALIPSO tracks intersecting with the modeled dust paths.
198 Depolarization ratio and aerosol subtype classification for CALIPSO tracks intersecting with
199 modeled dust paths from NOAA HYSPLIT Backward Trajectory for March 19th, 20th, 23rd,
200 and 24th, 2011 (b-e, respectively). Through δ and aerosol subtype a dust plume was found to be
201 present.

202

203 2.2. Dust particle shape and refractive index

204 As emphasized above, the primary advantage of using FIB dust samples for this study is that the
205 shape and composition of these samples are directly measured. To determine the dust shape, the
206 FIB uses a gallium ion beam, milling through each particle in 15 nm to 20 nm increments. This
207 process results in a stack of 100 to 200 cross-sectional images with dimensions of 1024 by 884
208 pixels for each particle. These cross-sectional images are then combined to reconstruct highly
209 detailed 3-D dust shapes, composed of three-dimensional pixels or voxels as illustrated by an
210 example in Figure 2.

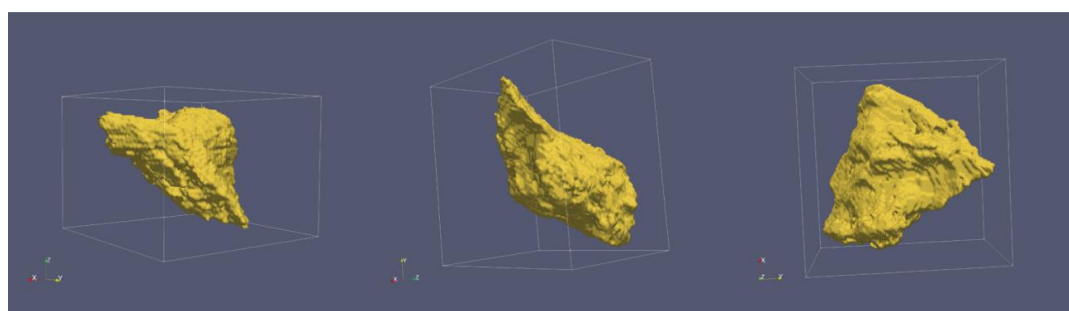
211

212 The collection of dust samples spans a range of sizes. In this study, we quantify this for irregular
213 geometries using the volume equivalent sphere radius. Using this metric, our library covers a
214 range from 0.46 μm to 0.93 μm in volume equivalent radius (r_v). The particle geometries are
215 also assigned two aspect ratios, where orientation is determined through principal component
216 analysis of the voxel coordinates. This analysis aligns the longest axis along the z-direction and



217 the greatest variation from this axis with the x - and y -directions, aligning with an intuitive
218 understanding of defining aspect ratios in three dimensions. The aspect ratios of these particles
219 vary from 0.629 and 0.398 (particle 2N Ca-S) to more symmetrical particles with aspect ratios
220 of 0.582 and 0.575 (particle 4N1 CaMg).

221



222

223 **Figure 2.** Orthographic projection of a sample dust particle from the FIB reconstructed
224 database, 3D Ca-Rich.

225

226 In addition to the FIB-based dust shape reconstruction, Conny et al. (2019) also performed the
227 element composition and mineral phase analysis for the FIB dust samples using the SEM and
228 energy-dispersive X-ray spectroscopy (EDX). They found that the dust samples can be loosely
229 classified into three categories based on the element compositions, the mainly Calcium
230 Magnesium based (Ca-Mg), the Calcium rich (Ca-rich) ones and lastly those primarily composed
231 of Calcium Sulfide (Ca-S). We shall follow their classification and naming convention in this
232 study (see Conny et al., 2019). To determine the refractive index of the dust samples, Conny et
233 al. (2019) first estimated the volume fractions of possible mineral phases in the particles based
234 on the composition analysis results. Then, the complex refractive index of each particle was
235 determined through the average Maxwell-Garnett dielectric function based on the estimated



236 volume fraction of each mineral phase. It should be noted that the iron-phase composition in the
237 particle was assumed to be either siderite, hematite, or magnetite which has different complex
238 refractive index. Moreover, two sets of complex refractive index were used for each iron-phase
239 mineral to account for the variability induced by optical anisotropy. The combination of mineral
240 differences and refractive index variability lead to several sets of final refractive index after the
241 Maxwell-Garnett average. Take the 3D Ca-Rich particle in Figure 2 for example. Table 1
242 provides the possible complex refractive index at 589 nm from Conny et al. (2019). Interested
243 readers are referred to their study for more information.

244

245 Table 1. The possible complex refractive index at 589 nm of the 3D Ca-Rich particle in Figure
246 2 from Conny et al. (2019).

Iron-phase mineral	Minimum	Minimum	Maximum	Maximum
	Refractive Index Real	Refractive Index Imaginary	Refractive Index Real	Refractive Index Imaginary
Magnetite	1.532	2.14E-02	1.660	2.36E-02
Hematite	1.544	2.32E-03	1.681	2.28E-03
Siderite	1.508	1.34E-05	1.648	1.34E-05

247

248 In this study, we are interested in the dust scattering properties at three commonly encountered
249 lidar wavelengths, namely, 355 nm, 532 nm, and 1064 nm. For simplicity, we assume the same
250 refractive index from Conny et al. (2019) for all three wavelengths, which is probably reasonable
251 only for the 532 nm. On the other hand, because we assume the refractive index to be invariant
252 with wavelength, the wavelength variation essentially corresponds to the variation of dust



253 particle size parameter, allowing us to focus on the impact of dust particle size on the lidar
254 scattering properties. The impacts of the spectral variation of refractive index will be investigated
255 in the future studies.

256

257

258 **2.3. ADDA model and convergence index of random orientation.**

259 In this study, we utilize the ADDA model to compute the single scattering properties, including
260 the extinction cross section σ_e , single scattering albedo ω , and scattering phase matrix $\mathbf{P}$, of each
261 FIB dust particle. The scattering properties of dust particles depend on not only their size, shape,
262 and refractive index, but also their orientations with respect to the incident light. In this study we
263 assume that dust particles are randomly oriented. The theoretical basis and numerical
264 implementation of the ADDA model have been well documented (Yurkin and Hoekstra, 2007,
265 2011). It has been used in numerous previous studies to compute the scattering properties of
266 aerosol and cloud particles (Yang et al, 2013; Gasteiger, 2011; Collier et al, 2016). The process
267 to generate the inputs from the FIB shape measurements for the discrete dipole approximation
268 (DDA) model has been described in detail in Conny et al. (2019). We use the same inputs and
269 configurations in this study. The only difference is that we use the ADDA model while Conny
270 et al. (2019) used a different DDA model, DDSCAT, by Draine and Flatau (1994). The reason
271 we cannot directly use the DDA simulation results from Conny et al. (2019) is twofold. Firstly,
272 their computations are conducted for an incident light at the 589 nm wavelength, whereas we are
273 interested in lidar wavelengths of 355 nm, 532 nm, and 1064 nm. Secondly, as will be explained
274 later, we will need a greater number of orientations to simulate random orientation for $\mathbf{P}$ and
275 lidar backscattering properties (Konoshonkin et al., 2020) than may be sufficient for the σ_e and



276 ω to converge. In the remainder of this section, we will introduce a practical method to determine
277 if a sufficient number of orientations have been used in the ADDA simulations to ensure
278 convergence in the results for random orientation computations.

279

280 For a particle with an irregular shape and arbitrary orientation, the scattering phase matrix $\mathbf{P}$
281 that relates the incident and scattering Stokes parameters is a 4x4 matrix with 16 elements

282

$$\begin{bmatrix} P_{11}(\theta_s) & P_{12}(\theta_s) & P_{13}(\theta_s) & P_{14}(\theta_s) \\ P_{21}(\theta_s) & P_{22}(\theta_s) & P_{23}(\theta_s) & P_{24}(\theta_s) \\ P_{31}(\theta_s) & P_{32}(\theta_s) & P_{33}(\theta_s) & P_{34}(\theta_s) \\ P_{41}(\theta_s) & P_{42}(\theta_s) & P_{43}(\theta_s) & P_{44}(\theta_s) \end{bmatrix}, \quad (4)$$

283 where θ_s is the scattering angle. If the particle is randomly oriented, for any orientation its
284 reciprocal orientation is equally likely. Because of the reciprocal symmetry, the scattering phase
285 matrix for randomly oriented particle with irregular shape reduces to (van de Hulst 1957;
286 Mishchenko et al., 2002; Mishchenko and Yurkin, 2017)

287

$$\begin{bmatrix} P_{11}(\theta_s) & P_{12}(\theta_s) & P_{13}(\theta_s) & P_{14}(\theta_s) \\ P_{12}(\theta_s) & P_{22}(\theta_s) & P_{23}(\theta_s) & P_{24}(\theta_s) \\ -P_{13}(\theta_s) & -P_{23}(\theta_s) & P_{33}(\theta_s) & P_{34}(\theta_s) \\ P_{14}(\theta_s) & P_{24}(\theta_s) & -P_{34}(\theta_s) & P_{44}(\theta_s) \end{bmatrix}. \quad (5)$$

288 The symmetry property of the $\mathbf{P}$ matrix for randomly oriented particles in Eq. (5) provides a
289 basis to assess the convergence of random orientation simulations in ADDA. For example,
290 utilizing the fact that $P_{41} = P_{14}$ for randomly oriented particles we can define a convergence
291 index (CI) random orientation as

292



$$CI = \int_0^\pi (P_{14}(\theta_s) - P_{41}(\theta_s))^2 d \cos(\theta_s). \quad (6)$$

293 As such, CI approaches zero when random orientation computation converges. It should be
294 noted that that CI can also be defined based on other symmetric elements of the phase matrix
295 such as $P_{21} = P_{12}$, $P_{31} = -P_{13}$. For practical applications, we usually assume that particles are
296 randomly oriented with an equal number of mirror particles. Under such a condition, or if the
297 particle in question has mirror symmetry itself, the phase matrix has only 6 independent
298 elements in the form (van de Hulst 1957; Mishchenko and Yurkin, 2017; Yang et al., 2023):
299

$$\begin{bmatrix} P_{11}(\theta_s) & P_{12}(\theta_s) & 0 & 0 \\ P_{12}(\theta_s) & P_{22}(\theta_s) & 0 & 0 \\ 0 & 0 & P_{33}(\theta_s) & P_{34}(\theta_s) \\ 0 & 0 & -P_{34}(\theta_s) & P_{44}(\theta_s) \end{bmatrix}, \quad (7)$$

300 and a CI based on P_{12} or P_{34} must be used.

301

302 In the context of ADDA, the orientation of a particle with respect to the incidence is defined
303 using three Euler angles α , β , and γ . To specify a certain orientation, the particle is rotated first
304 α on the z -axis, then β on the y -axis, and finally γ across the new z -axis through the zyz
305 convention (Yurkin and Hoekstra, 2020). Then, to produce the scattering properties for a
306 randomly oriented particle, ADDA averages across a large number of orientations. ADDA can
307 do this internally through specified number of evenly spaced intervals across α , β , and γ . For α
308 and β , ADDA calculates the scattering properties for the new orientation while for γ , or the self-
309 rotation angle, it equivalently rotates the scattering plane to improve computational time. It
310 calculates orientations in intervals of $2^n + 1$ for each of α , β , and γ resulting in $(2^n + 1)^3$ total



orientations. To assess if the random orientation convergence has been achieved, one can examine the behavior of CI as well as other scattering properties of interest, as a function of the number of orientations. An example using the 3D Ca-Rich dust particle is shown in Figure 3 for $n = 1, 2, \dots, 6$. As expected, all properties converge to asymptotic values as n increases from $n = 1$ (i.e., 8 orientations) to $n = 6$ (i.e., 262,144 orientations). On the other hand, it is important to note that the scalar properties such as extinction efficiency and asymmetry factor (Figure 3a), and S and δ (Figure 3b) have converged when $n = 4$, while the CI based on certain phase matrix elements (Figure 3c) only converged after $n = 5$. Based on this result, we employ $n = 5$ for the computations in this study. The results in Figure 3 clearly show that although one can assess the convergence of random orientation computation by observing the asymptotic behavior of scalar properties, the CI based on phase matrix elements is a more robust index supported by fundamental physics.

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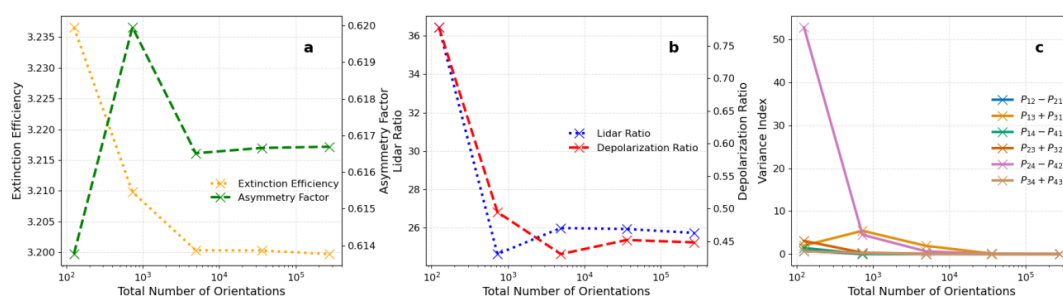


Figure 3. (a) Change in single-scattering albedo and asymmetry factor with increasing orientations for a representation of a randomly oriented particle. (b) The S and depolarization ratio's convergence as the number of orientations used increases. (c) Convergence Index for each of dust particle 3D Ca-Rich's Mueller index pairs at 532 nm. Note figures start at $n = 2$.

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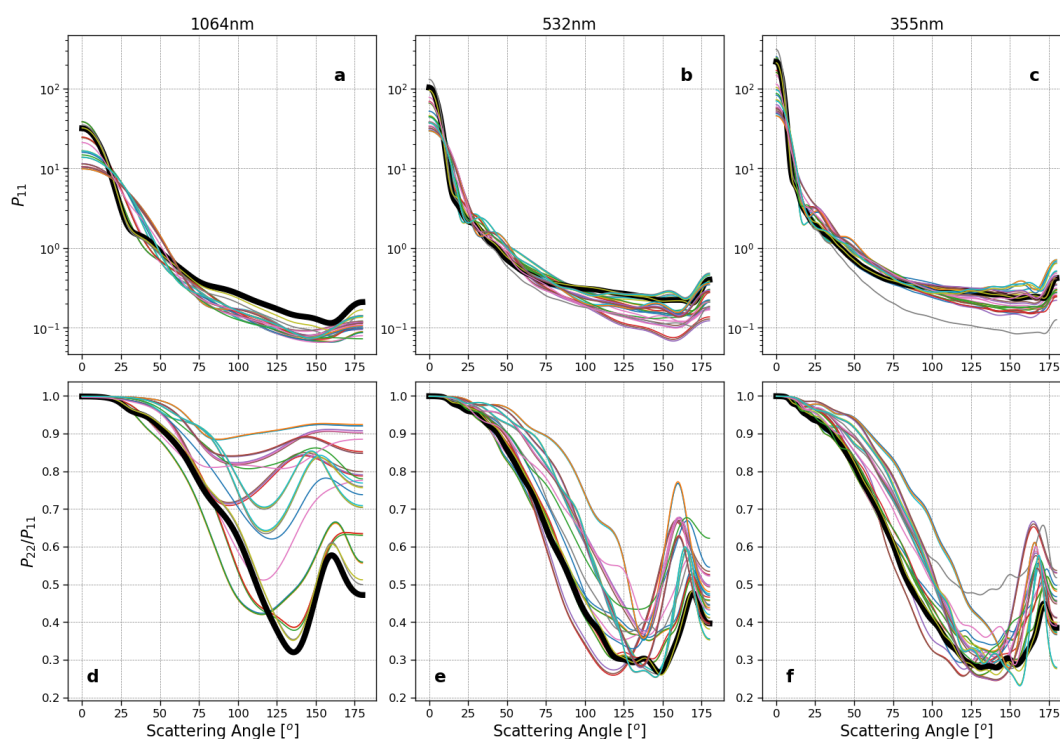
330 Thus, the error in computations of optical properties through ADDA is strongly tied to the
331 number of orientations used. We find in section 3 constraining refractive index through
332 mineralogy and size through proper characterization of particle size distribution are the largest
333 potential sources of error in these calculations, as ADDA's integration error has been set to less
334 than 10^{-5} and the geometries used are highly detailed, with individual dipole sizes on the order
335 of $\sim 20 \text{ nm}^3$. This makes the numerical error negligible compared to the error in chosen
336 parameters, convergence level, and sample size through the limited set of geometries. The *CI* is
337 a tool to minimize computational error while considering computational cost.

338

339 With the help of the newly developed *CI*, we computed the scattering properties of the FIB dust
340 samples for three commonly encountered lidar wavelengths 355 nm, 532 nm, and 1064 nm. For
341 each wavelength, more than 60 ADDA simulations are carried out corresponding to different
342 particles, as well as different refractive indices for each particle as explained above (see section
343 2.2). Figure 4 shows the phase matrix elements P_{11} and P_{22}/P_{11} for the FIB dust samples for the
344 three lidar wavelengths. Given the realistic morphology and extensive computational methods
345 of determining these optical properties, the FIB dust samples shown in Figure 4 can serve as a
346 benchmark for future studies on mineral dust scattering properties. As one can see in Figure 4 a-
347 c, the values of P_{11} in the forward scattering directions increases systematically from 1064 nm to
348 532 nm, and 355 nm, which can be explained by the increase of size parameter $x = \pi D/\lambda$ as
349 wavelength decreases. In Figure 4 d-f, the P_{22}/P_{11} shows considerable decreases from 1064 nm
350 to 532 nm, down $\sim 13\%$ on average. In contrast, the changes are relatively small from 532 nm to
351 355 nm. These features will help us understand the spectral dependence of S and δ shown and
352 discussed in the next section.



353



354

355 **Figure 4.** P_{11} and P_{22}/P_{11} for each particle geometry. Results for (a, d) 355 nm, (b, e) 532 nm,
356 (c, f) 1064 nm of each iron-containing mineral phase's minimum refractive index. Highlighted
357 in black is particle 3D Ca-Rich.

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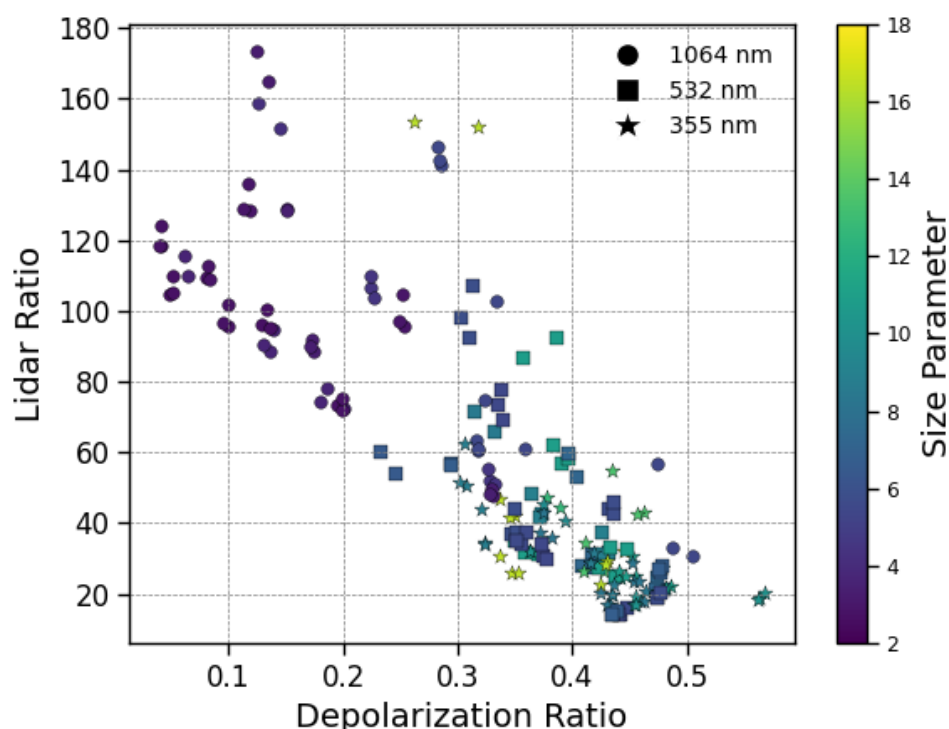
360 3. Sensitivities of lidar ratio and depolarization ratio to particle properties

361 3.1. Sensitivity to dust particle sizes

362 In lidar-based aerosol remote sensing, the $S - \delta$ diagram is often used to classify aerosols into
363 different types (Burton et al., 2012; Illingworth et al., 2015). The $S - \delta$ diagram for the FIB dust
364 samples is shown in Figure 5. Notably, S is negatively correlated with δ when the results for all



three wavelengths are combined (correlation coefficient of 0.83). Specifically, the δ at 1064 nm is smaller than the corresponding values at 532 nm and 355 nm, while the opposite is true for the S . The results for 532 nm and 355 nm largely overlap with each other. Recall that the same CRI is used for all three wavelengths, so these spectral differences are caused by the size parameter difference, i.e., the relative size of the particle with respect to the lidar wavelength. To further illustrate this point, we plotted the S and δ separately as a function of the dust particle size parameter, shown in Figure 6. Note that the size of the irregular particle can be defined in different ways; here, we adopt the volume-equivalent size.



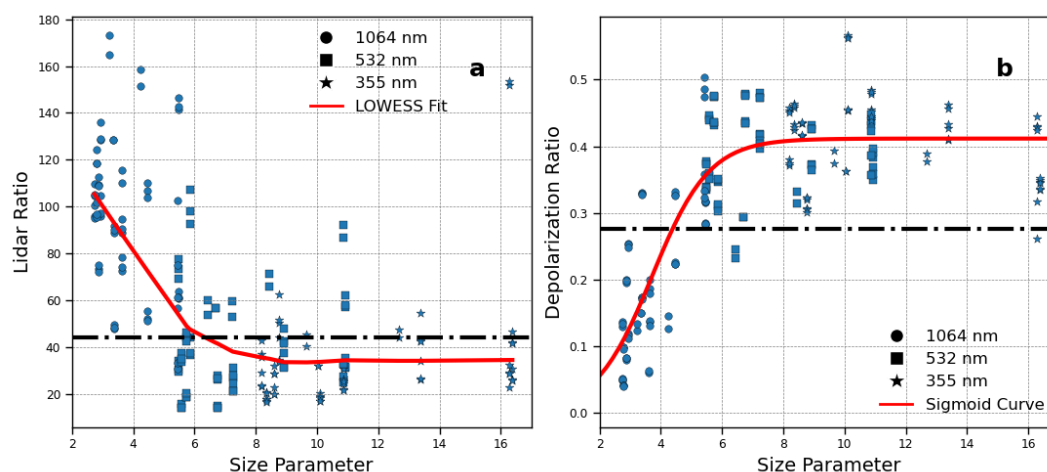


375 **Figure 5.** S - δ graph of FIB dust particles at each of 355 nm, 532 nm, and 1064 nm
376 wavelengths for the refractive index of each mineral type found present in the particle.

377

378 Figure 6 reveals an interesting asymptotic behavior of lidar properties with respect to size, where
379 S (Figure 6a) and δ (Figure 6b) first decreases and increases, respectively, with size parameters
380 and then seemingly approach their asymptotic values. We use a locally weighted scatterplot
381 smoothing regression (or LOWESS) to fit the trend in lidar optical properties with size
382 parameters. We find that both S and δ plateau around size parameter $x \approx 8$ and then approach to
383 their asymptotic values, $S = 35sr^{-1}$ and $\delta = 0.41$. The asymptotic behavior of lidar properties
384 have also been reported in several previous studies. For example, the S and δ based on the so-
385 called super-spheroid dust model in Kong, S. et al. (2022) showed a similar asymptotic behavior
386 for the size parameter range between 2 and 20 (see their Figure 3), and so is the laboratory
387 measured dust δ in Järvinen et al. (2016) (see their Figure 9).

388



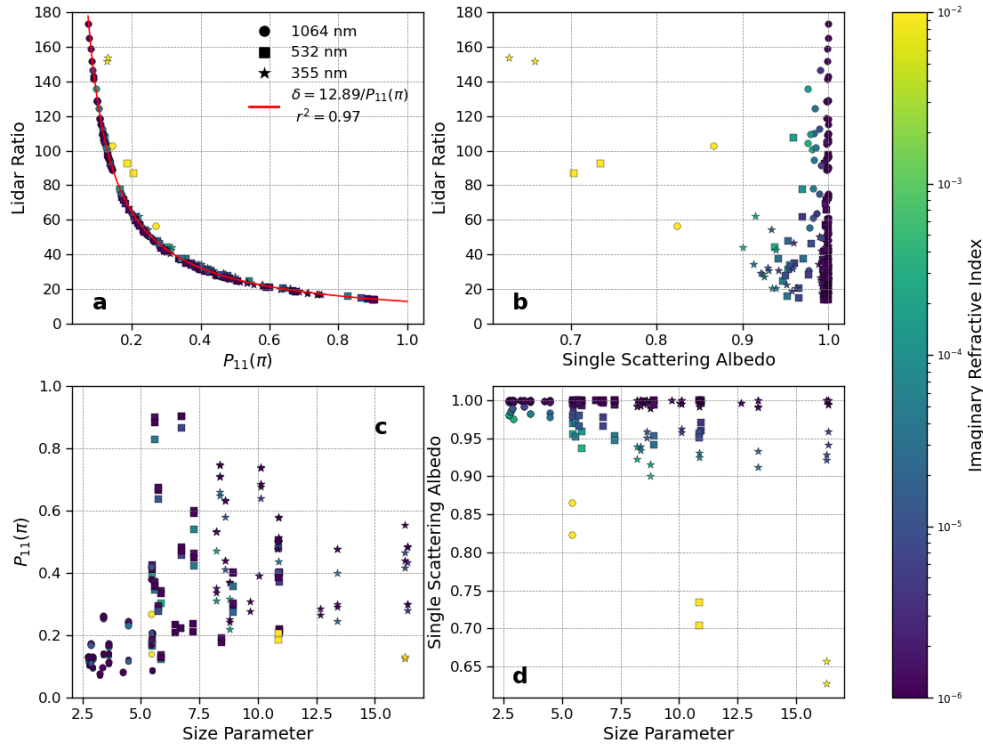
389



390 **Figure 6.** Relationship between dust particle size parameter and (a) S and (b) δ . The red line is
391 a LOWESS fit of the data for S and a Sigmoid function for δ . The black lines correspond to (a)
392 $S = 44 sr^{-1}$, the S used for CALIPSO's aerosol classification of dust (Kim et al., 2018) and (b)
393 $\delta = 0.277$, the median observed δ of the dust transport region using CALIOP (Liu Z. et al.,
394 2015).

395

396 Since S is a function of both $P_{11}(\pi)$ and the ω , we investigate their relative roles in determining
397 the size dependence of S . Figure 7a shows that the values of S lie closely around the $1/P_{11}(\pi)$
398 line, with the r-square value around 0.97 for a simple regression of $S = 12.9/P_{11}(\pi)$. In contrast,
399 single-scattering albedo ω plays a lesser role in S among the particles tested due to greater
400 similarities in values (Figure 7b). However, the outliers in Figure 7a correspond to points with
401 much lesser ω in Figure 7b, particularly the FIB sample 3D Ca-rich (see Figure 2) using the
402 magnetite refractive index, which has an imaginary refractive index of 0.021 to 0.024, an outlier
403 with a magnitude ten times greater than the other mineral types present (See Table 1). In Figure
404 7c and d, we plot the variation of $P_{11}(\pi)$ and ω respectively as a function of size parameter.
405 Although the variability of $P_{11}(\pi)$ is quite large, especially in the size parameter range between
406 5 and 10, it generally increases with size parameter. In contrast, the ω in Figure 7b shows a slight
407 decrease with size. These results indicate that $P_{11}(\pi)$ plays a more dominant role than the ω in
408 determining the size dependence of S in these dust samples.



409

410 **Figure 7.** S as a function of a) P_{11} and b) ω . c) P_{11} and d) ω as a function of dust size

411 parameter $x_v = 2\pi r_v/\lambda$. The color of each dot corresponds to the imaginary of the imaginary

412 index.

413

414 Following the same thought for the above S analysis, we analyze the role of $P_{11}(\pi)$ and $P_{22}(\pi)$

415 in determining the asymptotic behavior of δ in Figure 6b. It is seen in Figure 8a and b that both

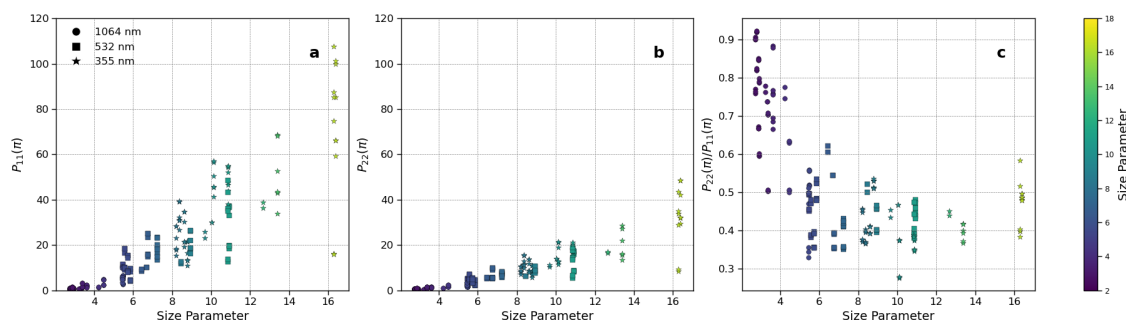
416 $P_{11}(\pi)$ and $P_{22}(\pi)$ increase with dust size. Interestingly, their ratio $P_{22}(\pi)/P_{11}(\pi)$ first

417 decreases with size and then seems to approach an asymptotic value of 0.4 when dust particles

418 are large. So, the result suggests that the asymptotic trend of δ with respect to dust size is a result

419 of the asymptotic behavior of $P_{22}(\pi)/P_{11}(\pi)$.

420



421

422 **Figure 8.** a) $P_{11}(\pi)$, b) $P_{22}(\pi)$ and c) $P_{22}(\pi)/P_{11}(\pi)$ as a function of the dust particle size
423 parameter.

424

425

426 3.2. Sensitivity to dust shape and sphericity

427 Several studies have shown that constraining particle morphology is important for quantifying
428 the δ of dust particles (Dubovik et al., 2006; Saito et al., 2021; Liu J. et al., 2015; Kahnert et al.,
429 2020; Kong, S. et al., 2022). As explained in the introduction, most of these studies are based on
430 simple hypothetical shape models such as ellipsoid and irregular hexahedrons. In this section,
431 we investigate the dependence of δ on dust sphericity based on the FIB dust samples. As
432 explained in session 2.2, in the baseline simulations each dust sample has different sizes and CRI
433 that corresponds to laboratory measured dust mineralogy. As a result, the differences in δ
434 between different sample particles in the baseline simulations are caused by not only shape but
435 also size and CRI differences. To eliminate the influence of size and CRI and focus on the effect
436 of sphericity, we carried out an additional set of ADDA computations for the 532 nm wavelength,
437 where we used the same CRI of $n = 1.5 + 0.005i$ and the same volume-equivalent radius of 0.5
438 μm for all the FIB particles but kept the original shape of each particle. The use of the common



439 size and CRI allows us to investigate the dependence of δ on the sphericity index defined as
440 follows (Wadell, 1935; Saito and Yang, 2022):

441

$$\Psi = \frac{\pi^{1/3}(6V)^{2/3}}{A_s}, \quad (8)$$

442 Where Ψ is the sphericity, V is the volume of the particle, and A_s is the surface area. By
443 definition, a sphere is $\Psi=1$, and a perfectly spherical particle has a δ of 0. However, due to the
444 irregularity of the FIB dust sample geometries, their Ψ , more specifically the surface area, is
445 heavily impacted by the level of granularity in the voxel size, similar to the well-known coastline
446 paradox (Steinhaus, 1954). Therefore, we employ the effective sphericity as the average
447 projected area of a particle is not susceptible to the same issues of increasing value with precision
448 (Vouk, 1948; Saito and Yang, 2022):

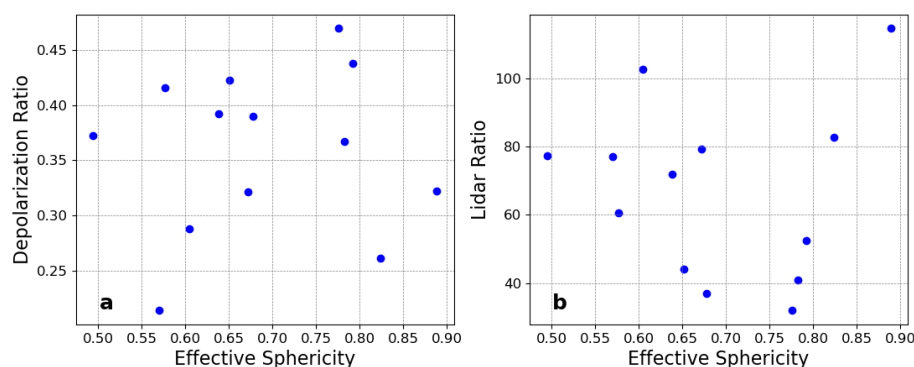
$$\Psi_{eff} = \frac{\pi^{1/3}(6V)^{2/3}}{4A_{proj}}, \quad (9)$$

449

450 Where Ψ_{eff} is the effective sphericity and A_{proj} is the average projected area across all
451 projection directions. This gives us a wide range of effective sphericity between 0.49-0.89. As
452 shown in Figure 9, we find no clear relationship between effective sphericity and δ or S (null
453 hypothesis rejected with $p > 0.05$ for both S and δ). This may be a result of a limited set of
454 geometries of the FIB dust samples. It could also be due to the limitation of the effective
455 sphericity index in Eq. (9) failing to capture the subtle dependence of δ on dust particle shape.
456 Note that other previous studies have also found weak dependence of δ in particle sphericity
457 (e.g., Kong, S. et al., 2022). Further studies are warranted to better understand the relationship



458 between the δ and morphology of dust particles. But overall, our results seem to suggest that the
459 impact of particle sphericity on δ and S is less important than particle size.



460

461 **Figure 9. (a)** Effective sphericity dependence of δ . **(b)** Lidar ratio variance with effective
462 sphericity. A common volume is used by constraining the volume equivalent sphere radius to
463 $0.5 \mu\text{m}$ for each particle as well as a refractive index of $n = 1.5 + .005i$. A wavelength of
464 532 nm was used.

465

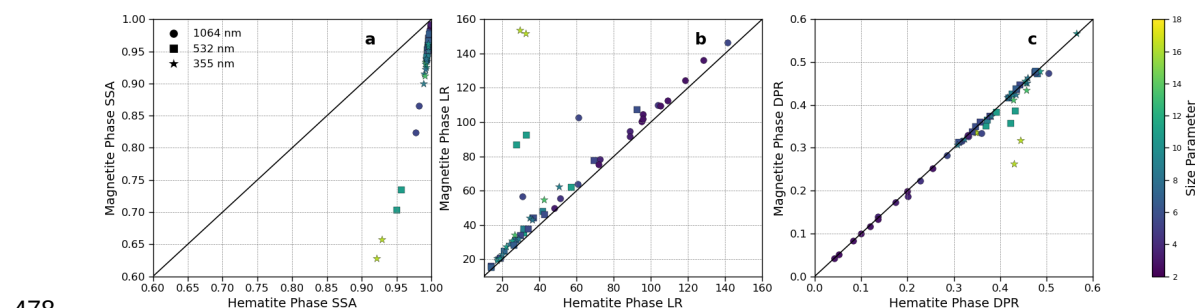
466

467 3.3. Sensitivity to dust mineralogy

468 Each particle from Conny et al.'s study (2019) was determined to have different amounts of iron
469 in its composition through their EDX spectroscopy tests. Using this data, they determined the
470 refractive index of each particle with the Maxwell-Garnett dielectric function described in
471 section 2.3. The tests resulted in the percentage of elements by mass and volume, but did not
472 reveal the mineral phase within the dust. To account for this, the study uses various possible iron
473 containing mineral phases for each particle to determine the refractive index, as these phases
474 have the greatest variability in possible refractive index for these particles. They also account for
475 birefringence through a minimum and maximum value for refractive index. Each particle was



476 given a hematite phase, while some had magnetite, ankerite, and/or siderite present. Interested
477 readers are directed to Conny et al. 2019 for further details.



478
479 **Figure 10.** Variation of a) ω , b) S , and c) δ for each particle with its magnetite phase and
480 corresponding hematite phase.

481 Each of these mineral phases has a different CRI, with magnetite being the most absorbing of
482 the iron-containing phases present (see Table 1). This results in considerable variations (up to
483 32%) in single scattering albedo (Figure 10a), particularly for 3D Ca-Rich particles, which have
484 the highest iron content by mass, ranging from 11.4 % to 7.90 %. In contrast, the next most iron-
485 dense particle (4N1 Ca-Mg) contains only 4.35 % to 1.56 %. Accompanying the reduction in
486 single scattering albedo, the S becomes systematically larger (Figure 10b), and the δ becomes
487 smaller (Figure 10c) when hematite is replaced by magnetite. These results underscore the
488 critical role of dust mineralogy in influencing the SSA of dust particles, as highlighted in
489 previous studies (Li et al., 2021; Song et al., 2022, 2024). However, the effects on lidar-derived
490 δ and S are comparatively smaller than the impacts on SSA and dust particle size. An important
491 caveat to keep in mind when interpreting these results is that the same dust CRI has been used
492 for all three wavelengths, as mentioned earlier. Dust absorption typically increases with
493 decreasing wavelength in the visible to ultraviolet spectral region, which is not accounted for in
494 our computations. Therefore, the impacts of mineralogy on lidar properties at the 355 nm



495 wavelength, where dust can have strong absorption, may be underestimated. We will leave this
496 for future studies because the spectral dependence of dust CRI is still highly uncertain due to the
497 lack of reliable observations.

498

499

500 **4. Parameterization schemes for dust δ**

501 The results in Section 3 indicate that particle size plays a dominant role in determining the dust
502 δ of FIB dust particles. As shown in Section 3.1, the dust δ exhibits an asymptotic trend with
503 increasing size (see Figure 6b), a pattern also noted in several previous studies (Kong, S. et al.,
504 2022; Järvinen et al., 2016; Kemppinen et al., 2015 a, b). The robustness of this asymptotic trend
505 inspired us to develop two parameterization schemes for δ as a function of dust size, which will
506 be introduced in this section. One scheme is designed for single particles, while the other is
507 intended for ensembles of particles with a particle size distribution. We hope that these
508 parameterization schemes can be used to efficiently estimate the δ of dust particles without
509 resorting to time-consuming scattering simulations.

510

511 The parameterization for single particles is straightforward. To model the asymptotic trend of
512 individual particle δ with dust particle size, we employed a sigmoid function as follows:

$$\delta(x) = \frac{\delta_{\infty}}{1+e^{-a(x+b)}} = \frac{0.41}{1+e^{-1.09(x-3.7)}}, \quad (10)$$

513

514 Where $x = \pi r_v/\lambda$ is the particle size parameter. The sigmoid function has three parameters: δ_{∞}
515 is the asymptotic value of δ when the size parameter is large. The other two parameters a and
516 b control the shape of the sigmoid function. After a nonlinear curve fitting, we find $\delta_{\infty} = 0.41$,



517 $a = 1.09$ and $b = -3.7$ ($R^2 = 0.72$). This simple parameterization can be used to estimate
518 the δ of a single dust particle given its size and the wavelength of interest.

519

520 Next, we will use Eq. (10) to construct a parameterization scheme for the volumetric
521 depolarization ratio, $\langle\delta\rangle$ of a dust plume following the widely used lognormal particle size
522 distribution ($n(r_v)$) giving us a value for δ for the ensemble of particles. To this end, we need to
523 first make an approximation. For a given dust particle size distribution $n(r_v) = dN/d\ln r_v$, the
524 rigorous definition of the volumetric δ is given by

$$\langle\delta\rangle = \frac{1 - \langle P_{22} \rangle / \langle P_{11} \rangle}{1 + \langle P_{22} \rangle / \langle P_{11} \rangle}, \quad (11)$$

525 where $\langle P_{11} \rangle$ and $\langle P_{22} \rangle$ are the bulk scattering phase matrix elements after the averaging over
526 $n(r_v)$. For example,

$$\langle P_{11} \rangle = \frac{\int_{-\infty}^{\infty} P_{11}(r_v) C_{sca}(r_v) n(r_v) d \ln r_v}{\int_{-\infty}^{\infty} C_{sca}(r_v) n(r_v) d \ln r_v}, \quad (12)$$

527 where C_{sca} is the scattering cross section of dust particle with the size of r_v . We found that it is
528 difficult to use Eq. (11) to estimate $\langle\delta\rangle$, because neither $\langle P_{11} \rangle$ nor $\langle P_{22} \rangle$ can be easily
529 parameterized with size parameter. To avoid this difficulty, we propose the following
530 approximate way to estimate the $\langle\delta\rangle$ as

$$\langle\delta\rangle \approx \frac{\int_{-\infty}^{\infty} \delta(r_v) C_{sca}(r_v) n(r_v) d \ln r_v}{\int_{-\infty}^{\infty} C_{sca}(r_v) n(r_v) d \ln r_v}, \quad (13)$$

531 where r_{vg} is the volume median radius, which allows us to use the simple parameterization in
532 Eq. (10). The accuracy of this approximation will be evaluated momentarily. Here, we convert



533 from size parameter to volume median radius through $x_{vg} = 2\pi r_{vg}/\lambda$ as δ will vary with
534 wavelength. Next, we need to specify the $C_{sca}(r_v)$ of single particles. Unfortunately, the size
535 parameter span of the FIB dust samples is too small to cover the whole dust $n(r_v)$. To solve this
536 problem, we use the TAUMdust2020 database to estimate $C_{sca}(r_v)$. The TAMUdust2020 is a
537 comprehensive database by Saito et al. (2021) that covers the scattering properties of 20 irregular
538 hexahedral shape models over the entire practical range of particle sizes, wavelengths, and CRI
539 of mineral dust particles. Based on the regional dust models recommended by Saito et al. (2021),
540 an ensemble-weighted degree of sphericity of 0.7 is elected to represent the dust particles. For
541 the dust CRI, we use the data from Song et al. (2022) to interpolate the TAMUdust2020 to obtain
542 the $C_{sca}(r_v)$. In Song et al. (2022), three sets of dust CRI corresponding to the low, mean, and
543 high concentration of hematite were used to compute the dust scattering properties and their
544 direct radiative effects. Here we adopt the CRI corresponding to the mean concentration of
545 hematite. Note that the CRI from Song et al. (2022) is spectrally dependent with increasing
546 absorption with decreasing wavelength (see their Figure 2), which means that the 355 nm has
547 the strongest absorption among the three lidar wavelengths considered here. Finally, for the dust
548 $n(r_v)$, we use the lognormal distribution

549

$$n(r_v) = \frac{dN}{d \ln(r_v)} = \frac{N_0}{\sqrt{2\pi}(\sigma_g)} \exp \left[-\frac{\ln^2(r_v/r_{vg})}{(\sigma_g)^2} \right], \quad (14)$$

550

551 Where N_0 is a constant and we use a fixed standard deviation of $\sigma_g = 0.529$, the same standard
552 deviation of the fine mode dust from AERONET's $n(r_v)$ for Cape Verde from Dubovik et al.
553 (2002) used later in Figure 12.



554

555 Using the combination of the $\delta(x)$ parameterization in Eq. (10), the $C_{sca}(r_v)$ from the
556 TAMUdust2020 database and the lognormal $n(r_v)$ in Eq. (14), we computed the volumetric dust
557 depolarization ratio $\langle\delta\rangle$ based on the proposed approximation in Eq. (13). The result for the 532
558 nm $\langle\delta\rangle$ as a function of the effective size parameter is shown in Figure 11a. It is not surprising
559 to see that the volumetric dust $\delta\langle\delta(x_{vg})\rangle$ resembles the $\delta(x)$ for the single particles in terms of
560 its size dependence. Further simplification is possible through a fitting of the newly bulk
561 averaged depolarization ratio. We find the depolarization of the FIB realistic particles are well
562 approximated by the following hyperbolic tangent equation:

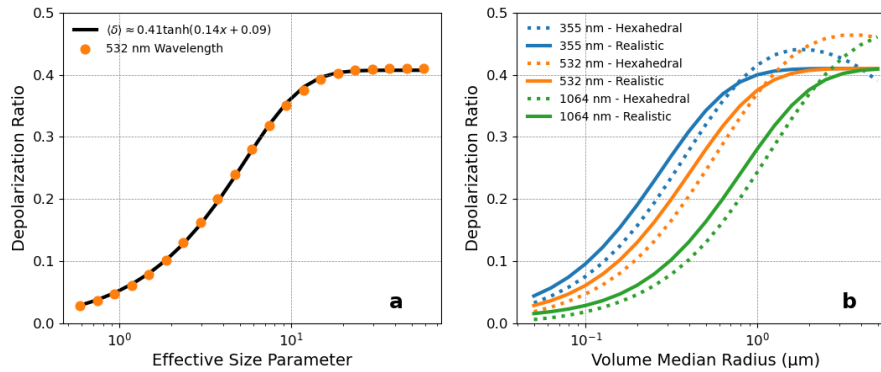
$$\langle\delta(x_{vg})\rangle \approx 0.41 \tanh(0.14x_{vg} + 0.09), \quad (15)$$

563 with an r-squared value of 0.79 as shown in Figure 11a. While this function is fitted for a
564 wavelength of 532 nm in particular, we found that the results for the 355 nm and 1064 nm
565 wavelengths are almost identical. This is probably because we used the same $\delta(x)$
566 parameterization for all three wavelengths, and only different C_{sca} due to the use of spectrally
567 dependent CRI in Song et al. (2022). It turns out that the C_{sca} plays a minimal role in the δ value
568 making Eq. (15) a reasonable approximation for all three lidar wavelengths given an effective
569 particle size parameter, x_{vg} . This is supported by the comparison results shown in Figure 11b.
570 The solid lines correspond to the volumetric $\langle\delta\rangle$ for the three wavelengths predicted based on
571 the parameterization Eq. (13). The dotted line corresponds to the $\langle\delta\rangle$ of irregular hexahedral
572 computed based on the TAMUdust2020 database using the Song et al. (2022) dust CRI. It is
573 important to note that the computation for irregular hexahedral is based on the rigorous definition
574 of δ in Eq. (11) without any approximation. Evidently, the two sets of $\langle\delta\rangle$ agree reasonably well
575 in terms of both spectral and size parameter dependencies. Interestingly, a decreasing trend was



576 observed for the 355 nm δ based on the irregular hexahedral when r_{vg} is larger than about 2 μm
 577 to 3 μm , which is not seen in either our parameterization or hexahedral results for other
 578 wavelengths. As mentioned above, in the computation for the irregular hexahedral we used the
 579 spectrally dependent CRI that has a higher absorption at 355 nm. Recall the result in Figure 10c
 580 that indicates δ to decrease with dust absorption. We believe that this decreasing with size trend
 581 of δ for large r_{vg} is a result of stronger absorption at 355 nm, reflected in a decrease in SSA for
 582 those particles.

583



584

585 **Figure 11:** (a) Parameterization of realistic δ for effective size parameter using a hyperbolic
 586 tangent function. (b) Depolarization Ratio predicted for a monomodal size distribution with
 587 varying volume-equivalent median diameter. The δ for realistic geometries was derived
 588 through parameterized approximations, while hexahedral shapes used P_{11} and P_{22} parameters.

589

590 The utility of the simple parameterization scheme in Eq. (15) is further demonstrated in terms
 591 of simulating the spectral dependence of δ as shown in the following case. Here, we use the
 592 climatological dust $n(r_v)$ retrieved by the AERONET at Cape Verde as reported in Dubovik et



593 al., (2002) (Figure 12a) to compute three sets of volumetric dust $\langle \delta \rangle$ for the three lidar

594 wavelengths using the following three methods:

- 595 1. In the first method (black solid lines in Figure 12b), dust scattering properties are based
596 on the irregular hexahedral model from the TAMUdust2020 database. The dust CRI is
597 spectrally dependent from the Song et al. (2022). The $\langle \delta \rangle$ is computed based on the
598 rigorous definition in Eq. (11) with $\langle P_{11} \rangle$ and $\langle P_{22} \rangle$ averaged over $n(r_v)$.
- 599 2. In the second method (blue dashed lines in Figure 12b), same as the first method except
600 that the $\langle \delta \rangle$ is computed based on the approximation method in Eq. (13).
- 601 3. In the third method (red dotted lines in Figure 12b), the $\langle \delta \rangle$ for each wavelength is
602 simply predicted using the parameterization in Eq. (15) by converting the x_{vg} to r_{vg} .

603 As such, the comparisons between the three methods enable us to assess the uncertainty
604 associated with each step of approximation. For example, the comparison between method 1 and
605 2 can help us understand the uncertainty associated with the $\langle \delta \rangle$ computation using the
606 approximation method in Eq. (13). The comparison of method 3 to the other two methods helps
607 us understand the overall accuracy of our simple parameterization.

608

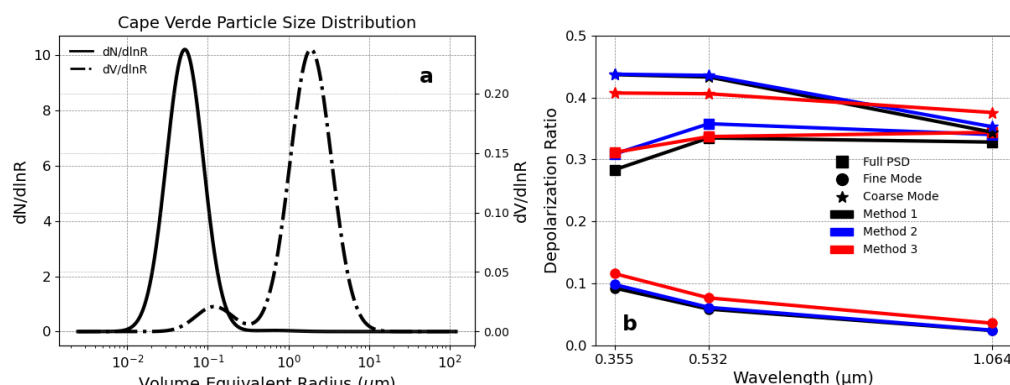
609 In order to use the full $n(r_v)$ with method 3, a weighting by backscatter coefficient is utilized
610 such that (Mamouri and Ansmann, 2014)

$$\langle \delta \rangle = \frac{\beta_f \delta_f (1 + \delta_c) + \beta_c \delta_c (1 + \delta_f)}{\beta_f (1 + \delta_c) + \beta_c (1 + \delta_f)}, \quad (16)$$

611

612 where β is calculated from the TAMUdust2020 database.

613



614

615 **Figure 12.** (a) Dust particle size distribution for Cape Verde using AERONET, adapted from
 616 Dubovik et al. (2002). Vertical line indicates fine/coarse mode cutoff. (b) Depolarization Ratio
 617 of fine and coarse mode for hexahedral dust and FIB reconstruction using approximation
 618 methods 1, 2, and 3 as described previously.

619 The comparisons shown in Figure 12 are promising. First of all, all three methods simulate a
 620 substantially smaller δ for the fine mode than the coarse mode. Secondly, the fine mode δ based
 621 on all three methods exhibits a decreasing trend with wavelength which is a result of the fast-
 622 increasing trend of δ with dust particle size parameter for fine mode dust particles (See Figure
 623 6). The differences in the fine mode δ between the three methods are mostly smaller than 0.05,
 624 with the method 3 result based on the simple parameterization scheme slightly larger than the
 625 other two methods. Finally, for the coarse mode dust δ , the results based on the simple
 626 parameterization (method 3) are close to spectrally neutral and smaller than the other two
 627 methods for 355 and 532 nm, while the use of TAMUdust2020 decreases δ at 1064 nm.
 628 Interestingly, the full-size distribution δ s based on the other two methods exhibit an inverse “v”
 629 shape, with the maximum at the 532nm and decreasing toward both 355 nm and 1064 nm. Such
 630 an inverse “v” shape spectral signature of dust δ has also been observed recently by (Haarig et



631 al., 2022) over Leipzig, Germany, in February and March 2021 during a transported Sahara dust
632 event (see their Figure 5). As aforementioned, our δ parameterization scheme does not take into
633 account the spectral dependence of dust CRI and the corresponding change of absorption. In
634 method 2 and 3, we use the CRI from Song et al. (2022) which has a stronger absorption at 355
635 nm, which leads to a decrease of δ from 532 nm to 355 nm. Therefore, our results indicate that
636 the inverse “v” shape spectral signature of dust δ is a result of competing effects of dust size and
637 absorption. The decrease of δ from 532 nm to 1064 nm is the result of dust size while the decrease
638 from 532 nm to 355 nm is a result of dust absorption. Despite this limitation, the overall accuracy
639 of our parameterization scheme is satisfying, partly due to the error cancellation between the
640 overestimation of the fine mode δ and underestimation of coarse mode δ . For example, after
641 summation of fine and coarse modes, the δ of the whole $n(r_v)$ for the 532nm wavelength is
642 $\langle \delta \rangle \approx 0.335$ based on method 1, while method 3 based on our simple parameterization is $\langle \delta \rangle \approx$
643 0.334.

644

645 Comparing the dust δ of the full $n(r_v)$ to that of fine mode δ and coarse mode δ also gives us
646 interesting results. Both fine and coarse modes individually decrease with wavelength despite
647 the inverse “v” shape spectral signature of the full $n(r_v)$. This characteristic is quite nicely
648 explained by an interpretation of Eq. (16). Across each wavelength, $\beta_f < \beta_c$ so $\langle \delta \rangle$ is greater
649 than a simple average of both fine and coarse modes. But β_c increases with wavelength.
650 Therefore, despite δ_f and δ_c decreasing spectrally, δ_c has a greater weighting in the equation. In
651 other words, more of the backscattered signal is due to larger particles as wavelength increases,
652 which are the particles exhibiting greater depolarization. The competing factors of β and δ
653 further reinforces the absorption and size impact on δ .



654

655 The utility of this parameterization likely comes from the inverse problem. Given the reliance
656 on TAMUdust2020 for β , reconstructing the δ from a $n(r_v)$ still requires use of theoretical
657 libraries for some amount of calculation. However, given a retrieved backscattering coefficient,
658 δ , and $n(r_v)$, using Eq. (15) and (16) creates a succinct method of retrieving β_f and β_c , separating
659 fine and coarse fraction of dust according to Mamouri and Ansmann (2014).

660

661 Specifically in coarse mode analysis, there are some limitations of our study. The sigmoid
662 parameterization leads to a very flat parameterization of δ for particles greater than $1\ \mu\text{m}$ in
663 volume equivalent radius seen in both Figure 11b and 12b which may be further refined with
664 larger particles, currently unavailable due to computational cost. It is also important to note our
665 study uses a wavelength-independent refractive index based on 589 nm, causing this work to
666 miss some spectral dependency that may cause the coarse mode differences in each wavelength
667 when using the globally averaged refractive index (see Figure 11b). The competing effects of
668 size and mineral composition of dust particles has been observed in studies of spectral
669 dependence of δ (Haarig et al., 2022), which we will investigate in future studies.

670

671

672 5. Conclusions and summary

673 In this study, we utilized the ADDA model to compute the scattering properties of FIB dust
674 samples and derived the S and δ at three widely used lidar wavelengths: 355 nm, 532 nm, and
675 1064 nm. The advantage of this study compared to previous work is the use of realistic dust
676 shapes reconstructed through the FIB tomography technique. The characterization of single



677 scattering properties of these realistic samples through rigorous computational techniques should
678 serve well as benchmark data for the dust scattering community. We investigated the dependence
679 of dust S and δ on dust particle size, shape, and mineral composition. The results lead to the
680 following conclusions:

- 681 • Both the S and δ exhibit an asymptotic trend with dust particle size: the S initially
682 decreases while the δ increases with size, before both approach their asymptotic values.
- 683 • The lidar properties were found to have only a weak dependence on effective sphericity.
- 684 • The presence of strongly absorbing minerals, such as magnetite, can greatly reduce the
685 dust's single scattering albedo and δ , especially at 355 nm.

686 In addition to these scientific findings, the convergence index introduced in Section 2.3 and the
687 δ parameterization schemes described in Section 4 may be useful for future research on light
688 scattering by nonspherical particles and lidar-based remote sensing. The CI can be used to assess
689 the convergence of random orientation computation using the DDA method. The δ
690 parameterization scheme in Eq. (15) can be used to estimate the δ of dust with a lognormal $n(r_p)$,
691 which can help us understand the variation of dust size based on δ observations and the
692 separation of fine and coarse model dust (Mamouri and Ansmann, 2014).

693

694 *Author Contributions.* ZZ proposed and conceptualized the work, providing both funding
695 acquisition and supervision of the project. All authors contributed to review & editing of the
696 manuscript. JD helped to design the convergence index. MS and PY provided the
697 TAMUdust2020 database for comparison to the FIB dust particles and MS provided insight into
698 particle sphericity. JZ and QS assisted in data curation and interpretation for FIB dust lidar
699 property results and global refractive index data. ASL and ZZ contributed to the original draft



700 preparation of the manuscript. ASL contributed to the methodology, data collection,
701 interpretation and analysis and data visualization under ZZ supervision and project
702 administration.

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