This Letter focuses on modeling the optical properties of red clay. Specifically, the measured phase matrix element $P_{22}$ of red clay shows a scattering-angle dependence that is quite different from those of other dust types, and has not been modeled successfully. The spheroidal particles cannot yield accurate agreement on all independent phase matrix elements of red clay simultaneously (failed for $P_{22}$). The $P_{22}$ element can be modeled well with the spheroidal model only by sacrificing the agreement on the other elements. However, the Koch-fractal model clearly outperforms the spheroidal model in reproducing all six phase matrix elements concurrently with a reasonable refractive index.

As one of the major atmospheric aerosols, mineral dust can directly absorb and scatter solar radiation and emit thermal radiation, and indirectly influence the radiative transfer by serving as cloud condensation nuclei or ice nuclei and, thereby, changing cloud properties [1]. The optical properties of mineral dust are fundamental for aerosol radiative effect and aerosol retrievals [2]. Considering the significant variation on dust microphysical properties and the lack of observational data, numerical modeling presents the only practical way to obtain the full-spectral optical properties of mineral dust.

Among the various optical properties, the 4-by-4 phase matrix $P$ describes the angular polarizing properties of a scattering event. It relates the components of the Stokes vectors of the incident and scattered radiation. There are 16 elements $P_{ij}$ in the matrix $P$. In scenarios for an ensemble of aerosols at random orientations (with equal numbers of particles and their mirror particles), the phase matrix has a simple block-diagonal form with six non-zero independent elements:

$$
P = \begin{bmatrix}
P_{11} & P_{12} & 0 & 0 \\
P_{12} & P_{22} & 0 & 0 \\
0 & 0 & P_{33} & P_{34} \\
0 & 0 & -P_{34} & P_{44}
\end{bmatrix}. \tag{1}
$$

For the physical interpretation, the $P_{12}$ is related to the degree of linear polarization of the scattered radiation under unpolarized incident light; and the $P_{22}$ can be related to the depolarization of linearly polarized incident light and is an indicator of particle nonsphericity [2]. Polarization properties become more and more important for aerosol observations [3,4], and have been used to infer the properties of atmospheric aerosols [5,6].

Because of the particle complexity, e.g., irregular geometry, surface roughness, and components [7], it is a great challenge to obtain dust optical properties. Significant efforts are devoted to both observations and simulations on the optical properties of a variety of dust particles. The Amsterdam–Granada light scattering database (AGLSD) provides laboratory-observed phase matrices of various mineral dust samples [8,9], and their results are widely regarded as standards to evaluate numerical models. Meanwhile, a large number of numerical models are developed and compared for computing the optical properties of dust particles [10]. The simplest dust model assumed is sphere, which generally performs poorly [11]. For non-spherical particles, spheroids are widely adapted to reproduce the light scattering properties of dust [11–14], and show great success. Subsequently, various models such as ellipsoids [15], polyhedra [13,16], Gaussian spheres [17], Koch-fractal polyhedra [18], agglomerated debris particles [19], and spatial Poisson–Voronoi tessellation [20], have been introduced for computing dust optical properties, and most of them have been found to perform well in reproducing laboratory-measured dust phase matrix elements.

Most of the aforementioned shape models for dust have been evaluated by comparisons against the measured phase matrix of feldspar, one of the mineral samples from the AGLSD. As the optical properties of dust samples vary, it is possible that a model validated with feldspar measurements does not work for other types of dust, making the applicability of the model...
uncertain. By comparing the laboratory measurements from the AGLSD for different aerosol types, we find that there are obvious differences, e.g., in their matrix element \( P_{22}/P_{11} \). Figure 1 compares three non-zero phase matrix elements of five different dust species. The upper left panel of Fig. 1 is a scanning electron microscope image of red clay [8,9], the focus of this Letter; the phase matrix elements, e.g., \( P_{11} \), \( P_{22}/P_{11} \), and \( P_{44}/P_{11} \), are given in the other three panels. Geometrically, all mineral dust is composed of mixtures of varying types of complex particles. For the five dust samples, the phase functions are qualitatively very similar, but show some differences in the backward hemisphere. The \( P_{22}/P_{11} \) of feldspar differs most obviously on the scattering-angle dependence from the other four samples (labeled by the markers), especially in the forward direction. Similar differences are also noticed for \( P_{44}/P_{11} \). Indeed, after comparing all the dust samples (over 20 kinds) included in the AGLSD, we find that the four samples shown with markers in Fig. 1, i.e., red clay, Sahara, olivine \( L \), and Sahara sand (Libya) [8,9], show similar variations on \( P_{22}/P_{11} \) and \( P_{44}/P_{11} \), which are quite different from those of other samples (such as the feldspar shown here as an example). Considering the differences shown in Fig. 1, quite different numerical models may be needed to consider the optical properties of some dust samples (e.g., red clay) from the AGLSD.

This Letter considers the phase matrix of the red clay, and carefully investigates the \( P_{22} \) and \( P_{44} \) elements. Both spheroidal and irregular particles are used to reproduce the phase matrix elements from laboratory measurements available at the AGLSD.

Spheroid is one of the simplest and most popular geometries used to model the optical properties of dust particles, and performs well on reproducing scattering properties of realistic aerosols [11–14]. Dubovik et al. [21] applied the model of polydisperse spheroids to dust retrievals. Huang et al. [6] tested the polarization properties of the spheroidal model based on the POLDER (POLarization and Directionality of the Earth’s Reflectances) observation. Overall, spheroids have significant advantages over spheres to represent the optical properties of dust particles, but some limitations have also been revealed. For example, it is difficult for spheroids to reproduce \( P_{22} \), i.e., the depolarization properties [10,11], and the spectral dependence of linear polarization predicted by spheroids is inconsistent [19].

The performance of spheroids to model light scattering by mineral dust has been systematically investigated by Merikallio et al. [11]. They found that some of the simulated phase matrix elements do not agree well with the measurements. However, the ranges of spheroidal refractive index and aspect ratio were quite limited. To better address the performance of spheroids, we significantly expand the ranges of the aspect ratio and the refractive index considered. We use aspect ratios (the ratio of the rotational to horizontal axes) of 1/2, 1/3, 1/4, and 1/5 for oblate spheroids and 2, 3, 4, and 5 for prolate spheroids. Considering the significant uncertainties on the refractive indices of mineral dust particles, the real part is set to 1.5, 1.75, and 2.0, and the values of 0.0001, 0.001, and 0.01 are used for the imaginary part. The invariant imbedding T-matrix method is used to calculate the optical properties of spheroids at the wavelength of 632.8 nm (as in the AGLSD) [22].

Figure 2 compares the six phase matrix elements of laboratory measurements for red clay particles and the numerical results based on spheroid assumption. The shaded areas show the scope of variation in the phase matrix elements for all spheroids of different aspect ratios and refractive indices. The range of variation indicates the possible values attainable with an ensemble of spheroids. Overall, the simulated results of oblate spheroids cover wider ranges of variations than those of prolate ones, and, thus, may give a more accurate agreement on the phase

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**Fig. 1.** Scanning electron microscope image of red clay samples and three phase matrix elements \( (P_{11}, P_{22}/P_{11}, \text{and } P_{44}/P_{11}) \) of some mineral dust samples measured by the AGLSD [8,9].

**Fig. 2.** Comparison of the phase matrix elements of red clay from laboratory measurements and simulations with spheroids at the wavelength of 632.8 nm. The yellow and blue shaded areas indicate the coverage by different prolate and oblate spheroids, respectively. The results of two particular cases are also shown with solid lines. Case 1 uses oblate spheroids with an aspect ratio of 1/3 and a refractive index of 2 + 0.01i, and Case 2 uses ones with an aspect ratio of 1/4 and a refractive index of 2 + 0.0001i.
matrix elements of red clay, especially for the $P_{22}/P_{11}$ and $P_{44}/P_{11}$. However, even with the extended ranges of the aspect ratio and the refractive index, the spheroidal model cannot fit all six non-zero phase matrix elements well simultaneously. Figure 2 also directly compares two special cases of spheroidal models (the solid lines) with the measurements, and each case indicates the results based on spheroids with a fixed aspect ratio and refractive index. For Case 1, the aspect ratio and refractive index used are $1/3$ and $2 + 0.01i$, respectively, and the results agree well with all matrix elements, except $P_{22}/P_{11}$. Case 2 shows the best fit that a spheroidal model can achieve for $P_{22}/P_{11}$, and the aspect ratio of $1/4$ and refractive index of $2 + 0.0001i$ are used. The agreement on $P_{22}/P_{11}$ is significantly improved, whereas the other five elements cannot match the measurements. It appears that spheroids are unable to provide satisfactory fits to all six phase matrix elements of red clay simultaneously, even with shape distribution considered, and the best fits are obtained with an unrealistically high real part of the refractive index. Although showing great success previously on modeling dust optical properties, the spheroidal model is lacking for the red clay, and a better model should be developed.

Considering the complex geometries of dust particles, a large amount of irregular particles has been introduced to model their optical properties. This Letter considers one of those really complex irregular particles, namely the Koch-fractal particles [23]. The Koch-fractal particle is a kind of concave polyhedrons based on tetrahedron elements, and it has great success in modeling the optical properties of natural particles, e.g., ice crystals [23] and mineral dust [18]. However, the potential of the Koch-fractal particle on modeling the optical properties of mineral dust has not been completely investigated yet.

The geometry of the Koch-fractal particles is specified by three parameters [18], i.e., the generation, the aspect ratio, and the irregularity factor. The Koch-fractal particle of the zeroth generation is a regular tetrahedron, and the particle becomes concave as tetrahedrons of the higher-order generation are added to the triangular surfaces of the lower-order generation. With the increase of generation number, the particles become more complex with small structures introduced. The second parameter, the aspect ratio of the Koch-fractal particles, is defined similarly to that of other geometries, the ratio of the particle height to the width. The third parameter, the irregularity factor, is used to specify the random movement of the vertices to generate irregular particles. The irregularity factor is defined by a real number between 0 and 0.5, and larger numbers correspond to more irregular particles. The details of the three parameters are given and illustrated in Macke et al. [23] and Liu et al. [18].

Figure 3 shows some examples of the Koch-fractal particle geometries. The left panel is the regular third-generation particle, and the middle and right panels show corresponding irregular ones with aspect ratios of 1 and 0.3, respectively. The irregularity factors of the two irregular particles are both 0.15. The rightmost particle is constructed by simply compressing the middle one in the vertical direction. It should be noticed that fairly small structures similar to surface roughness [7,24,25] are introduced by the third-generation tetrahedron.

We first calculate the optical properties of Koch-fractal particles with different shape factors (i.e., generation, aspect ratio, and irregularity factor) and refractive indices. A state-of-the-art combination of the pseudo-spectral time domain method and the improved geometric-optics method is used to calculate the optical properties following Liu et al. [18]. Second, the irregular Koch-fractal particle shape and the corresponding refractive index that give the best overall agreement with the observed phase matrix are chosen as the “optimal” model. The shape in the right panel of Fig. 3 (third generation, aspect ratio of 0.3, and irregularity factor of 0.15) with a refractive index of $1.8 + 0.0005i$ is the optimal model we found. Figure 4 shows the performance of the Koch-fractal particle on modeling the optical properties of sampled red clay aerosols, i.e., the phase matrix elements of the laboratory measurements and numerical results based on the Koch-fractal particles. The optical properties of the Koch-fractal particle are computed and integrated over the size distribution of the red clay sample. As seen from the figure, all six phase matrix elements of the Koch-fractal particles fit the measurements closely. The numerical model underestimates the phase function in the forward direction with scattering angles smaller than 30°, but it shows excellent agreement in the backward direction. For the other five non-zero phase matrix elements, the modeled results and measurements all show the same variations over scattering angles, whereas little differences are noticeable. We pay more attention.

![Fig. 3. Koch-fractal particles of the third generation. The left one is a regular one. The middle and right ones are irregular (irregularity factors of 0.15), and their aspect ratios are 1 and 0.3, respectively.](image)

![Fig. 4. Comparison between the phase matrix elements of numerical results based on Koch-fractal particles and laboratory measurements for red clay at the wavelength of 632.8 nm.](image)
to the $P_{22}/P_{11}$ element of red clay, because no numerical models have been found to successfully simulate it before. The modeled and measured $P_{22}/P_{11}$ values given in Fig. 4 agree closely with slight differences at scattering angles between 20° and 50°.

Compared with the spheroidal model given in Fig. 2, the results based on the Koch-fractal particles are much closer to the measurements, and the refractive index of $1.8 + 0.0005i$ is also closer to the suggested values of red clay [8,9]. The real part used may be still larger than that of real particles, and the erroneous refractive index may be compensating for a shape error or size error, or possibly both [15], which can hardly be determined from the available data. Furthermore, for both the spheroids and the Koch-fractal models, oblate particles with small aspect ratios are needed to match the measurements, which may be the reason for the $P_{22}$ behavior of red clay [26]. This indicates that models based on platy particles should be considered for radiative effects and remote sensing of mineral dust.

This Letter focuses on modeling the optical properties of a red clay aerosol sample, particularly its phase matrix. The $P_{22}/P_{11}$ and $P_{44}/P_{11}$ of red clay show distinct variations from those of other well-studied mineral dusts. The phase matrices simulated based on relatively simple spheroids and complex Koch-fractal particles are compared with the laboratory measurements. A wide range of geometric parameters and refractive indices are considered in the simulations. Both prolate and oblate spheroids show limited applicability on modeling the optical properties of red clay, although the oblate ones are better than the prolate ones. Oblate spheroids could perform well on five non-zero phase matrix elements, whereas they fail to reproduce the $P_{22}$ element. To improve the agreement on $P_{22}$, the agreement with the other elements becomes quite poor. Furthermore, an unrealistically high real part of the refractive index of 2 is needed. It is obvious that the numerical results based on the Koch-fractal particles give better agreement than the spheroidal model on all six phase matrix elements concurrently, and with a more realistic refractive index.

It should be noticed that the Koch-fractal model is just one example we used to reproduce the phase matrix of one type of dust, and other irregular models or spheroids with certain surface roughness that may yield similar success can be tested in future studies. Furthermore, the spectral performance of the Koch-fractal model and its performance for other dust particles are also interesting topics for further investigation.

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