Responses to reviewers

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Review comments in black and responses in blue

We thank the two reviewers and the editor for their constructive comments that helped to improve the manuscript. Below, we include point-by-point responses to the comments, and describe the corresponding changes that we have made in the revised manuscript.

Reviewer #1: Dr. Lei Bi

This paper investigated the single-scattering properties and lidar observables of tri-axial ellipsoids at short wavelengths using a shape distribution from observations. The novelty of this paper is that the aspect ratio distributions were constrained by observations. The influences of model shapes were illustrated by comparing to spheroidal and spherical models, and the impacts of smaller-scale surface textures were also discussed. The ellipsoidal dust models were found to be substantially superior to spherical models and better than spheroid models in many ways except at the backscattering angle which might be caused by the inaccuracy of computation methods and smaller-scale textures. The tri-axial ellipsoidal dust models were believed helpful in improving climate models and remote sensing retrievals. This paper was well written and organized.

We thank the reviewer for their positive comments. We carefully address these comments below.

I have a few comments:

1) The database used and the calculation details should be more specific. It could be better to state which method (the Lorenz-Mie theory, the T-matrix method, the DDA, the IGOM) was used at what size parameter range. In particular, what is the aspect ratio range of particles in available databases? It looks that the aspect ratio range from observations is much larger than those of spheroid and tri-axial ellipsoid models. Since the database was developed a long time ago, I am wondering whether the database was updated.

We used the original database published in Meng et al. (2010), which hasn't been updated since then to our knowledge. Specifically, in terms of the size parameter range in the database, the DDA is used at 0.025 – 40, the T-matrix method is used at 0.025 – 40, the Lorenz-Mie method is used at 0.025 – 1000, and the IGOM is used at 10 – 1000 (after Table 2 of Meng et al., 2010). To address this comment, in lines 177-182 of the revised manuscript, we added "Lorenz-Mie theory was used for spherical particles with size parameter x of 0.025 – 1000, the T-matrix method was used for particles with x of 0.025 – 40, the discrete dipole approximation was used for x of 0.025 – 40, and the improved geometric optics method was used for x of 10 – 1000 (see Table 2 of Meng et al., 2010). At overlapping size parameters, results from different methods were averaged. These four methods together cover the size parameter range from Rayleigh to the geometric optics regimes".

In terms of the aspect ratio in the database, the ranges of length-to-height ratio and width-to-height ratio are both between 1 to 3.3. The reviewer is right that the aspect ratio from observations is larger than the upper limit of the database (i.e., 3.3). For particles with an aspect ratio larger than 3.3, we assume its aspect ratio is 3.3. Future database development to include these highly aspherical shapes is highly recommended. To address this comment, in lines 213-216 of the revised manuscript, we added "Note that the upper limits of LWR and HWR are both 3.3 in the Meng et al. (2010) database, whereas observations find dust particles can be more aspherical (Huang et al., 2020). For a dust particle with a LWR (or HWR) larger than 3.3, we assume its LWR (or HWR) is 3.3. Future database development to include these highly aspherical shapes is highly recommended".

2) The spheroidal model developed by Dubovik was used for comparison. Is it possible to update this model by using new shape distribution compiled from the observation? In this case, it will be easier to understand impact of tri-axial ellipsoids.

We thank the reviewer for this helpful suggestion. We didn't update Dubovik et al. (2006)'s spheroidal model with the observed shape distribution, because the problem with Dubovik et al. (2006)'s spheroidal model is more the shape definition and less the specific shape distribution. In fact, Dubovik et al. (2006)'s shape distribution is similar to our shape distribution of the length-to-width ratio (i.e., Huang et al., 2020). Although Dubovik et al. (2006) is thus reasonable for the length-to-width ratio, they neglected that the dust height is much smaller than the dust width (see Figs. 2c and 2e of Huang et al., 2020) and therefore they underestimated dust asphericity. It's difficult for Dubovik et al. (2006)'s spheroidal model to address this problem because of the nature of the spheroidal dust shape (i.e., assuming that the dust height equals the dust width).

3) Line 49-52 "These problematic dust shape assumptions of aerosol models and retrieval algorithms generate biases in dust single-scattering properties… To facilitate accounting for more realistic dust shape in aerosol models…". I suggest the authors add a few sentences to review the progress in studying the impact of dust asphericity. Other different dust models are also developed in the community to overcome the shortcoming of spheroidal dust models. For example, superspheroid (or superellipsoid) models (Lin et al. JGR, 126, e2020JD033310.https://doi.org/10.1029/2020JD033310, 2018) showed their significant superiority in simulating optical properties of dust comparing to spherical and spheroidal models. The model was also further used to study lidar observations (Kong et al. JGR, 127, e2021JD035629. https://doi.org/10.1029/2021JD035629,2022).

Thank you for the suggestion. We have added these recent advances in line 41 of the revised manuscript.

4) Besides, Line 149-150 "we for the first time (to our knowledge) account for the observation constrains on dust shape in obtaining dust single-scattering properties". I would like to mention the following papers. Bi et al. (Applied Optics, 48(1), 114–126, 2009; https://doi.org/10.1364/AO.48.000114) utilized the ratios of the three axes of ellipsoids obtained by Ghobrial & Sharief (IEEE Transactions on Antennas and Propagation, 35(4), 418–425, 1987) in simulating the single-scattering properties of triaxial

ellipsoidal dust particles. Actually, the above study provided the technique readiness for the development of a tri-axial ellipsoidal database, which was used in this study.

Thank you for pointing this out. In the revised manuscript, we removed "for the first time (to our knowledge)" from line 149, and added the references in line 40.

5) Line 159-160 "LWR … commonly referred to as the aspect ratio … Fig. 1". However, in Figure 1c, the aspect ratio (commonly referred to) should be because the LWR=1 for oblate spheroid in this figure. A little bit confusion.

We removed "also commonly referred to as the aspect ratio" from line 160 of the revised manuscript.

6) Line 207 "…, we used Monte-Carlo sampling to randomly generate a large number…". It is difficult to understand this procedure. Why did the authors use a Monte-Carlo method instead of directly multiplying the two probabilities from Eqs. (9) and (10).

We thank the reviewer for suggesting a simpler way of integration. Although the results are the same between the two ways of integrations, we took the reviewer's suggestion, and revised the sentence to "These weighting factors were calculated from the two lognormal distributions of LWR and HWR (Eqs. 9 and 10)" in line 212 of the revised manuscript.

7) Line 328-329 "First, most aerosol models underestimate the extinction efficiency (Qext) and mass extinction efficiency (MEE) by 20% to 180% …" The authors summed the extinction cross sections of all the 121 ellipsoid models and then divided by the projected area of the volume-equivalent sphere in Eqs. (11) and (18). Therefore, the extinction efficiency could be larger than 2 such that the difference of Qext in Fig. 2a-b was mostly caused by the definition in Eq. (11). I suggest the authors recalculate these two parameters by the projected area of the ensemble of 121 particles. The difference is expected to be smaller. Otherwise, more related discussion is necessary to clarify the understanding of the extinction efficiency, although different definitions (if used consistently) have no impact in the final calculation (e.g., the extinction coefficient).

We thank the reviewer for this question. We define a particle's extinction efficiency as the extinction per unit projected area of a volume-equivalent sphere (after Kok et al., 2017). We do this because extinction efficiency in this way ties directly to the mass extinction efficiency (MEE), and MEE is key to climate models to determine dust direct radiative effect (nicely reviewed and emphasized in Kok et al., 2023, Nature Reviews Earth & Environment). In particular, the fact that our extinction efficiency is enhanced by ~40% on average over that of spherical particles means that the MEE is enhanced by that ~40%.

But we understand that this new definition deviates from the standard definition of the extinction per unit projected of the aspherical particle (as in Meng et al., 2010). To address this comment, we attached the figure below to compare the two definitions.

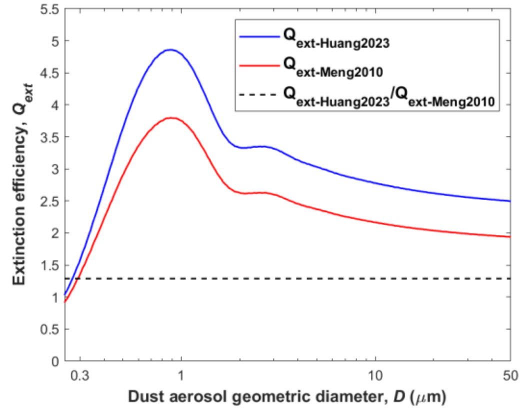

Figure R1. Comparison of the two different definitions of extinction efficiency. Meng et al. (2010) database calculated the extinction efficiency with regards to the projected surface area of the ellipsoidal dust particle (blue line), whereas this manuscript (Huang et al., 2023; red line) takes the extinction efficiency with regards to the projected surface area of the volume-equivalent sphere (after Kok et al., 2017; 2023). The black dashed line shows the ratio between the two definitions. The ratio is a constant (i.e., 1.29) resulting from the shape-integrated ratio between the projected surface area of an ellipsoidal dust particle and the projected surface area of the volume-equivalent spherical dust particle (i.e., SA_w integrated over the 121 shapes). Since dust shape is invariant with dust size (see Section 2.2 of main text), it makes sense that this ratio is a constant. Note that, although here we take the wavelength of $550\ nm$, real part of dust refractive index of 1.53, and imaginary part of $0.002\ i$, the constant ratio (1.29) is solid for other wavelengths and refractive indices.

8) The hexahedral dust optical model was compared in this study. The first paper that proposed the hexahedral dust model is Bi et al (Applied Optics, 49, 334-342. https://doi.org/10.1364/AO.49.000334, 2010).

We added the references in line 40 of the revised manuscript.

Reviewer #2, Dr. Qixing Zhang

This paper presents a study of single scattering properties of tri-axial ellipsoidal particles with observationally constrained dust shape distributions. The calculated tri-axial ellipsoidal dust topics are compared with the spherical dust optics, spheroidal dust optics and the AGLSD laboratory observations. The major novelty of this work is using the observationally constrained shape distribution, both LWR and HWR, for tri-axial ellipsoidal dust optics calculation, and the results seems promising.

I believe that a perfect dust optical model should meet the requirement of both radiative effects estimation in global aerosol model and remote sensing retrievals. Usually, the remote sensing retrievals need more detailed shape descriptors to reproduce the angle resolved observations. As mentioned in the paper, currently most remote sensing retrieval algorithms approximate dust aerosols as spheroidal particles with a shape distribution conflicts with observations. I think this is a good paper trying to solve this problem. The ability of the model to better reproduce the laboratory measured scattering matrices is convincing. Overall, the paper is well written and well organized.

We thank the reviewer for their positive comments. We carefully address these comments below.

Some comments:

1) The authors compared the calculation results with lidar field observation results. I am not sure whether the field observation meet the requirement of single scattering or not. Did the authors consider the multiple scattering effect when analyzing the results? And the aerosol mixing state? There are so many unknown factors in the field, it may be challenging to compare the model optics with field observation.

This is a good question. The eight lidar observations were collected near dust source regions, where newly emitted dust (also often called fresh dust) dominates the signal and for which mixing with other aerosols can thus be reasonably assumed to be small. In addition, dust multiple scattering is negligible relative to dust single-scattering (Kahnert and Scheirer, 2019, Optics Express).

Past studies have therefore used these field observations of lidar ratio and linear depolarization ratio to validate a range of simulations of single-scattering properties. For example, Saito and Yang (2021) compared their developed single-scattering properties of hexahedral dust particles against the same lidar field observations used in this manuscript. Tesche et al. (2019) also used the same lidar field observations to validate their single-scattering properties of spheroidal dust particles. We thus believe that using the eight lidar field observations to evaluate single-scattering databases is well-established.

To address this comment, in lines 269-270 of the revised manuscript, we added “We neglect the minor effects of dust multiple scattering and dust mixing with other aerosols on the observation results, as Tesche et al. (2019) and Saito and Yang (2021) did”.

2) As mentioned in the paper, “more laboratory observations of the scattering matrices of atmospheric dust aerosols with simultaneous measurements of these samples’ microphysical properties, namely their size distribution, refractive index, and shape distribution.” There are other laboratory measurement results of dust published (Liu, et al, JQSRT, 2019,229: 71-79, https://doi.org/10.1016/j.jqsrt.2019.03.010, and Liu, et al, AMT, 2020, 13, 4097-4109, https://doi.org/10.5194/amt-13-4097-2020), if possible, more comparisons between calculation results and laboratory measurement results are encouraged.

Thank you for pointing out these references. We have added citations to these recent papers in line 571 of the revised manuscript.

3) Line 205, “To obtain these weighting factors, we used Monte-Carlo sampling to randomly generate a large number of volume-equivalent …” Did the authors generate the particles for each volume-equivalent diameter D? The authors have already known the distribution of LWR and HWR, why need this Monte-Carlo sampling? Please clarify it.

We thank the reviewer for suggesting a simpler way of integration. Although the results are the same between the two ways of integrations, we took the reviewer's suggestion, and revised the sentence to "These weighting factors were calculated from the two lognormal distributions of LWR and HWR (Eqs. 9 and 10)" in line 212 of the revised manuscript.

4) Line 520, "they overestimate the lidar ratio (Fig. 7b) by underestimating the backscattering intensity by a factor of ~2", and Line 535, "Kemppinen et al. (2015) added surface roughness to smooth particles with sharp corners and found that surface roughening can reduce the backscattering intensity." From the results presented in Fig 3-5, it seems that more detailed shape model will further widen the gap between modeled and measured backscattering intensity.

We thank the reviewer for the good question. It remains unclear that adding surface roughness will increase or decrease the gap with regards to backscattering intensity. The reviewer is correct that Kemppinen et al. (2015)'s results indicate an increase in the gap. However, Kemppinen et al. (2015)'s results were based on relatively fine dust particles with a size parameter less than 10 (i.e., diameter around 1.8 micrometers at 550 nm wavelength). A range of papers find that dust aerosols are much coarser and can be as large as 50 micrometers (Kok et al., 2017; Ryder et al., 2019; Adebiyi et al., 2023). No study has been conducted to investigate the results of roughening coarse and super-coarse dust particles. As a result, it remains unclear that adding surface roughness for the ensemble of dust with various sizes will increase or decrease the gap. More work to observe and simulate surface roughness of dust with various sizes is needed.

To address the comment, in lines 544-550 of the revised manuscript, we added "Although Kemppinen et al. (2015)'s results indicate that adding surface roughness can widen the gap between modelled and measured backscattering intensity, Kemppinen et al. (2015)'s results were based on relatively fine dust particles with a size parameter less than 10 (i.e., diameter around 1.8 μm at 550 nm wavelength). A range of studies find that dust aerosols are much coarser and can be as large as 50 μm (Kok et al., 2017; Ryder et al., 2019; Adebiyi et al., 2023). No study has been conducted to investigate the results of roughening coarse and super-coarse dust particles. As a result, it remains unclear that adding surface roughness for the ensemble of dust with various sizes will increase or decrease the gap between modelled and measured backscattering intensity.".

5) The optics database (Meng et al. 2010) was used in this paper. I am not sure whether this database is accessible or not. I suggest to give some information about this database in the part of Code/Data availability.

In the revised manuscript, we added "The Meng et al. (2010) database is provided by Drs. Ping Yang and Bingqi Yi upon request" in line 652.

References:

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