

Response to RC1

We would like to thank the reviewers for their comments and suggestions, which have helped us significantly improve the manuscript. In the revised version, we have made the following major changes:

- Streamlined introduction and reduction of discussion on the radiative effects of dust
- Equations and quantity definitions moved to a newly introduced “Theoretical Background” section
- Minor revisions to most plots for improved clarity
- Clarifications to results and discussion in section 5
- Additions to limitations in conclusion section

Below are point-to-point responses to reviewer comments.

The study uses 14 realistic Asian dust particles with sizes from $r = 0.46$ to $0.93 \mu\text{m}$ and describe their scattering properties by using the discrete dipole approximation (DDA). They calculate lidar ratios and depolarization ratios at 3 commonly used lidar wavelengths based on their realistic particles with the limited size range. They reveal an asymptotic behavior of the lidar ratio and depolarization ratio with increasing size parameters and develop a parameterization for the later one. The study is interesting and contributes to the challenging task of modelling the scattering properties of irregularly shaped mineral dust particles. The DDA technique allows to create any particle shape which has advantages above predefined particles shapes. However, it is difficult to extend it to large size parameters, where the asymptotic behavior might be helpful. The manuscript can still be improved and therefore, I recommend to consider my major revisions listed below.

Major comments:

1. Size

Your studied particles range roughly between $1 - 2 \mu\text{m}$ in diameter. Is this sufficient to realistically describe atmospheric mineral dust? The fine mode or sub-micrometer mode is missing but contributes to the optical properties observed with lidar. And on the other end, the large particles are missing as well. It is a major limitation of the study and hampers a good comparison to real world observations with lidar. Please discuss how representative your particle size range is for atmospheric observations.

Response: You are right that our dust samples are limited in terms of their sizes and CRI (see our reply to CRI below). The dust samples in our study are provided by the NIST group who did some comprehensive single-particle analysis of Asian dusts (see Conny et al. 2017, 2019). Their techniques, in particular the focus-ion beam (FIB) tomography, are expensive and labor intensive. So they were only able to analyze those dust samples presented in our study.

On the other hand, the advanced techniques provide rich information on the microphysical and optical properties of dust particles. In particular, the FIB tomography enables extremely detailed rendering of the 3D shape of the dust particle. As pointed out in the introduction, the use of realistic dust particle shape is one of the novelties of this study in comparison with many previous ones.

Going back to your questions, we believe that our samples are highly representative of the real world dust particles in the similar size range, although as you pointed out the size range is indeed limited. We have pointed out this limitation in the discussion, lines 661-664. Additional discussion to this limitation is added to lines 388-391 and 510-511, and lines 708-717. To alleviate this limitation, we add to the study the results based on the irregular hexahedral to check that the behaviors of lidar scattering properties, e.g., DPR vs size, learned from our dust samples also apply to other shapes and to a larger size range. As we showed in Section 5, the results based on two shape models agree reasonably well. It is also aligned with the lab measurements results from Järvinen et al., 2016.

In summary, although the size range of our dust samples are limited, we believe that the lessons learned are general and applicable to realistic dust.

Because you don't vary CRI nor shape, there is no additional information in using different wavelengths. If you would stick to one wavelength (e.g., 532 nm), you would just cover the size parameters from 5.5 to 11, this is much less than in Järvinen et al., 2016. And from this, you cannot draw the conclusions presented in Sect. 4. Now, you just add calculations at other wavelengths, in principle you could take any wavelength to cover the size parameter space from 0.1 to 20. And in fact, you're just covering the size parameter space from 2.7 to 16.5. So, the smallest size parameters, i.e., the fine mode, is not included. Please start your figures at 0 and not at 2 (Fig. 6-8). If you take for example Fig. 12a and mark the covered size range of your particles, you will see that just a small part of the size distribution is covered.

Response: We don't fully agree that "Because you don't vary CRI nor shape, there is no additional information in using different wavelengths." First of all, dust shape does not change with lidar wavelengths. Second, in the visible region, the variation of dust CRI is mainly due to the presence of iron oxide (See Sokolik et al. 1993). For dust particles with low iron oxide content, the CRI in the SW spectral region does not change much (Zhang et al., 2024).. In other words, for real-world dust with low iron oxide content, the only significant differences between the three lidar wavelengths are size parameters. Therefore, our analysis is still meaningful. Yes, we could add other wavelengths, but the 355 nm, 532 nm and 1064 nm are widely used for dust remote sensing, and it is why they are selected for our study. As shown in section 5 (Fig. 12), even with the simplest set for three wavelengths, our results can still simulate the spectral DPR behavior observed by Haarig et al. (2022). Finally, it is important to note that a large variety of CRI values have been used in our computations based on the assumed iron oxide compositions (see Section 3.2). This ensures our computations to capture some variability of dust CRI.

Nevertheless, we carried out the following sensitivity study to further understand the impacts of spectrally dependent CRI on the LR and DPR. To specify CRI variation with spectral

wavelengths, we used the percentage of hematite by volume given in Conny et al., 2019 to assign new refractive indices to each particle based on a database of dust refractive index available in Obiso et al., 2024. The LR and DPR results (red dots) based on the new spectrally-dependent CRI are shown in Figure R1 below, overlaid with our original results (blue). Although there are some differences, the two sets of computations follow the same general trends (i.e., LR decreasing and DPR increasing with size parameter). This suggests that our conclusion still holds when using the spectrally dependent CRI. This is not surprising because as mentioned above we have used a variety of CRI values (although they are not spectrally dependent) in our computations.

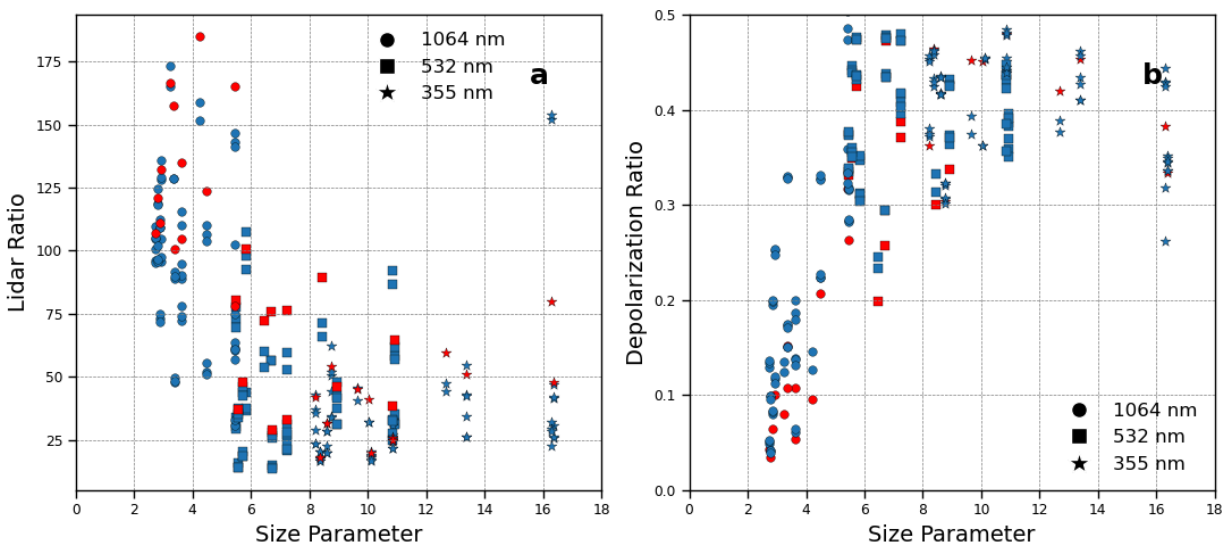


Figure R1. Lidar Ratio and Depolarization Ratio of dust particles as a function of size parameter. Red points represent the use of a wavelength-dependent CRI based on hematite content of the particle, while blue points indicate the optical properties using the original 589 nm wavelength refractive indices reported in Conny et al., 2019.

We have updated manuscript Figures 6-8 to start at size parameter 0.

2. CRI

If you cannot include the spectral dependence of the CRI, i.e., the increase towards the UV, I would omit the results at 355 nm. In case you want to keep the results at 355 nm, please find a way to mimic a realistic increase in the imaginary part of the CRI. Otherwise, your discussions might be misleading.

The complex refractive index (CRI) is an important quantity. However, you missed completely to set your results in the context of previous observations. The first study which comes into my mind is the one by Di Biagio et al., 2019.

Response: First of all, as explained in section 3.2, the values of CRI at the 532 nm are derived from laboratory measurements (e.g., energy-dispersive X-ray spectroscopy see more details in Conny et al. 2019). Extending the CRI from 532 nm to 355 nm and 1064 nm, in a meaningful way, would require knowledge of dust mineralogical compositions and the CRI of each individual component. This is not a trivial task and it is beyond the scope of this study.

We also want to note here that the CRI of the dust minerals are still poorly understood and subject to large uncertainty. Take for example the CRI of hematite—one of the most important absorbing minerals, the imaginary part of hematite often in the literature often differ by several orders of magnitude (see Go et al. 2021; Di Biagio et al. 2019). See also Figure R2 below from Go et al. 2021. Even if we had taken into the consideration of CRI spectral variation due to dust mineralogy, the results would have been qualitative and subject to large uncertainty.

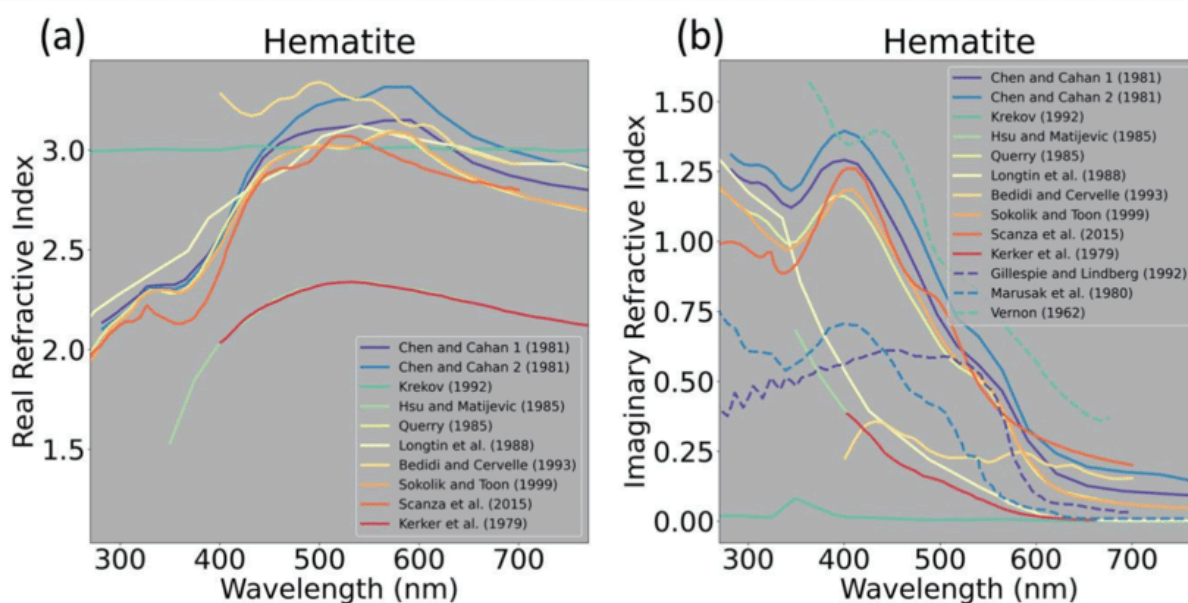


Figure R2. A figure from Go et al. 2021 (Their Figure 3) Plots of previously published hematite refractive indices at 300–700 nm: (a) real part and (b) imaginary part. Note the large uncertainty among previously published data.

Finally, it should be noted that although we used spectrally invariant CRI for our FIB dust particle computation, we compared the scattering properties of FIB particles to those of TAMU irregular hexahedra that used a spectrally dependent CRI. The CRI used for the TAMU hexahedra (shown in Figure R3 below) is adopted from Song et al., 2022 which combined data from several previous studies. Despite the constant CRI limitation, our work still captures a decrease in SSA and DPR for the particles similarly to the hexahedra using a spectrally dependent CRI.

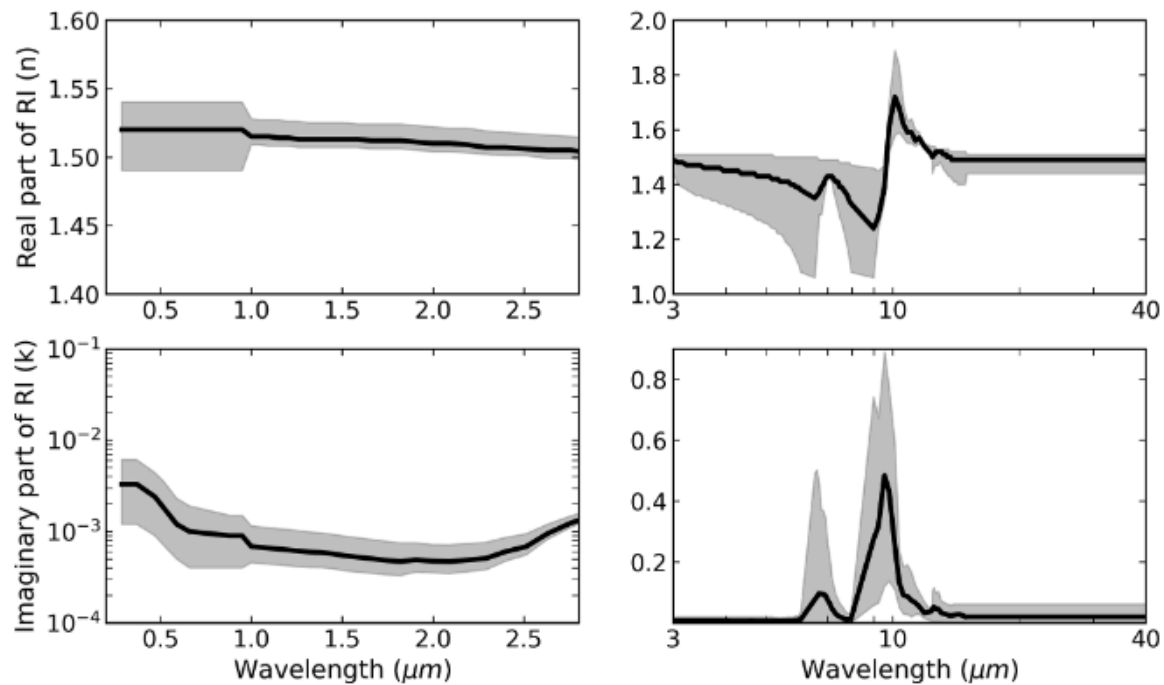


Figure R3. Spectral refractive index from Song et al. 2023 used to derive the scattering properties of irregular hexahedra from the TAMU2020 dust database.

The CRI of dust is highly uncertain and pertains to the particular mineralogy of the dust particle. We discuss the limitations of CRI, particularly for 355 nm wavelength, in lines 494-497, ... Additionally, the CRI of the hexahedra which we compare our study to used the globally averaged dust refractive index from Song et al., 2022, deriving the values from Di Biagio's study. Di Biagio is an author for both works. Despite the constant CRI limitation, our work still captures a decrease in SSA and DPR for the particles similarly to the hexahedra using a spectrally dependent CRI.

3. Asian Dust

The term "Asian dust" is widely used in literature, especially to separate it from Saharan dust. However, Asia is a huge continent and at some point, you should be more specific about the source region, which is probably in the Gobi Desert. Dust from Central or West Asian (Middle Eastern, Persian or Arabian) deserts might exhibit different optical properties.

And there are differences in the optical properties, especially in the lidar ratio, between Asian and Saharan dust, which was summarized by Floutsi et al., AMT 2023 based on observations of Hofer et al., ACP 2020. A lidar ratio of 35 sr might be not that bad for Asian dust, but not for (West) Saharan dust.

Response: To our knowledge, there is no evidence to suggest that morphology of dust particles is strongly tied to regional origin. Therefore, while these dust particles are suspected to be of Gobi origin, we believe the FIB dust samples to be useful for characterization of atmospheric dust more generally.

To try to better address the origin of the dust, we run a back trajectory of the dust particles from the location found in the CALIPSO track as a new starting point to attempt to determine the desert of origin, in Figure R4.

NOAA HYSPLIT MODEL
Backward trajectories ending at 0400 UTC 19 Mar 11
GHDA Meteorological Data

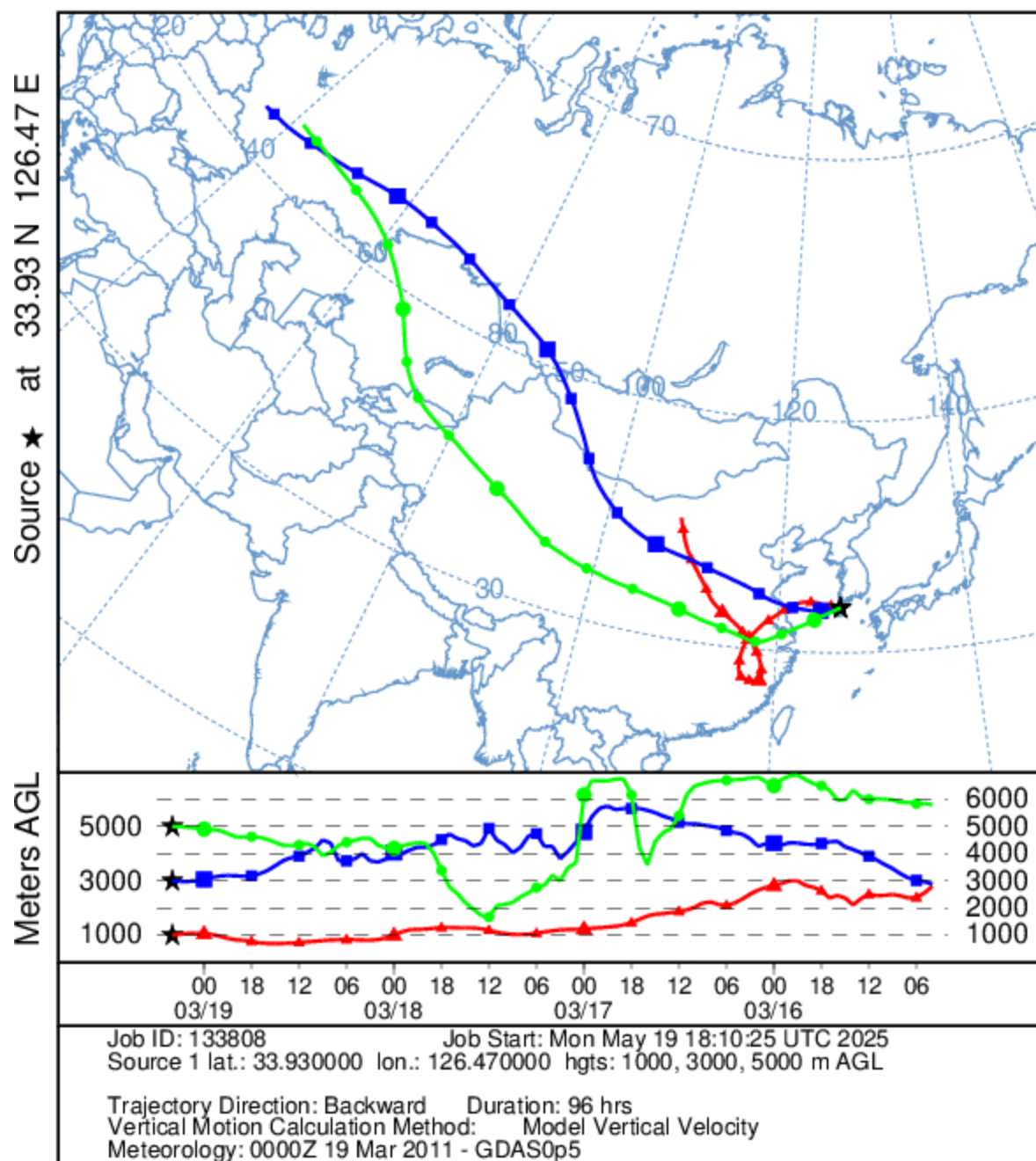


Figure R4. HYSPLIT backtracking trajectories from the intersection of CALIOP and original HYSPLIT trajectories seen in Figure 1 of the manuscript.

This seems not to fully address the question of desert origin. Notably, the 5000m line drops in height significantly just East of the Qaidam Basin, so this could be a reasonable guess (circled in red). We have added mention of the Gobi origin to the manuscript.

4. Asymptotic Behavior

The measurements of Järvinen et al., 2016, show an asymptotic behavior for the depolarization ratio as you mentioned correctly. But you are hiding that this plateau was found at around 0.30 and not 0.41. This is a significant difference. Does your model overestimate the depolarization ratio of mineral dust? And why? What could be the reason? Asian dust was included in the study of Järvinen et al., 2016. Kahnert et al., 2020, used the laboratory results of Järvinen et al., to test various modelling parameters. Please take these two studies seriously and discuss the differences to your results.

Response: We cited Järvinen et al. (2016) multiple times throughout our manuscript, clearly indicating our careful consideration and respect for their findings. Therefore, we disagree with the characterization that we are "hiding" results or not taking their study seriously. Given the inherent variability in Asian dust particle properties, it is entirely plausible that neither our study nor Järvinen et al. (2016) alone can fully represent all Asian dust conditions.

Could our study potentially overestimate the depolarization ratio (DPR) for certain mineral dust particles? Indeed, it is possible for specific cases. However, as clearly explained earlier, we have confidence in the accuracy and representativeness of our modeling results for the particular dust samples analyzed in this study.

More fundamentally, rather than fixating solely on the exact numerical value of the DPR asymptotic plateau, our intention was to emphasize the significance and practical utility of the observed asymptotic behavior itself. Our developed parameterization scheme for DPR in Section 5 can, in fact, be adapted effectively to match observations from Järvinen et al. (2016), underscoring its broader applicability and robustness.

Finally, we would like to highlight that differences among various studies naturally arise due to distinct methodologies, dust samples, and experimental setups employed. Expecting identical numerical outcomes across studies is unrealistic. Instead, it is more meaningful to acknowledge shared conclusions or common underlying behaviors, such as the observed asymptotic nature of the DPR, as confirmed by both our study and Järvinen et al. (2016).

Järvinen et al. (2016)'s lower plateau is stated in lines 143-145. For single particle simulations, Kong, S. et al., 2020 using the hypothetical Super-Spheroid Model shows great variability in DPR throughout the coarse mode. It seems reasonable for differences between single particle simulations such as these and bulk scattering properties observed by Jarvinen et al to arise. Compared to our bulk averaged results, their study also uses a standard deviation between 1.2

and 2.0 for lab measurements, significantly greater than our 0.529. Järvinen et al.'s work notes the sensitivity of results to PSD themselves throughout section 2.4. Clearly, a wider PSD would result in a lesser DPR when an asymptotic relationship exists as both studies observe.

We also suspected the measured scattering angle of 178 through lab measurements as opposed to the 180 degrees we use to be of importance. Setting our results to 178 degrees backscatter, we find the following:

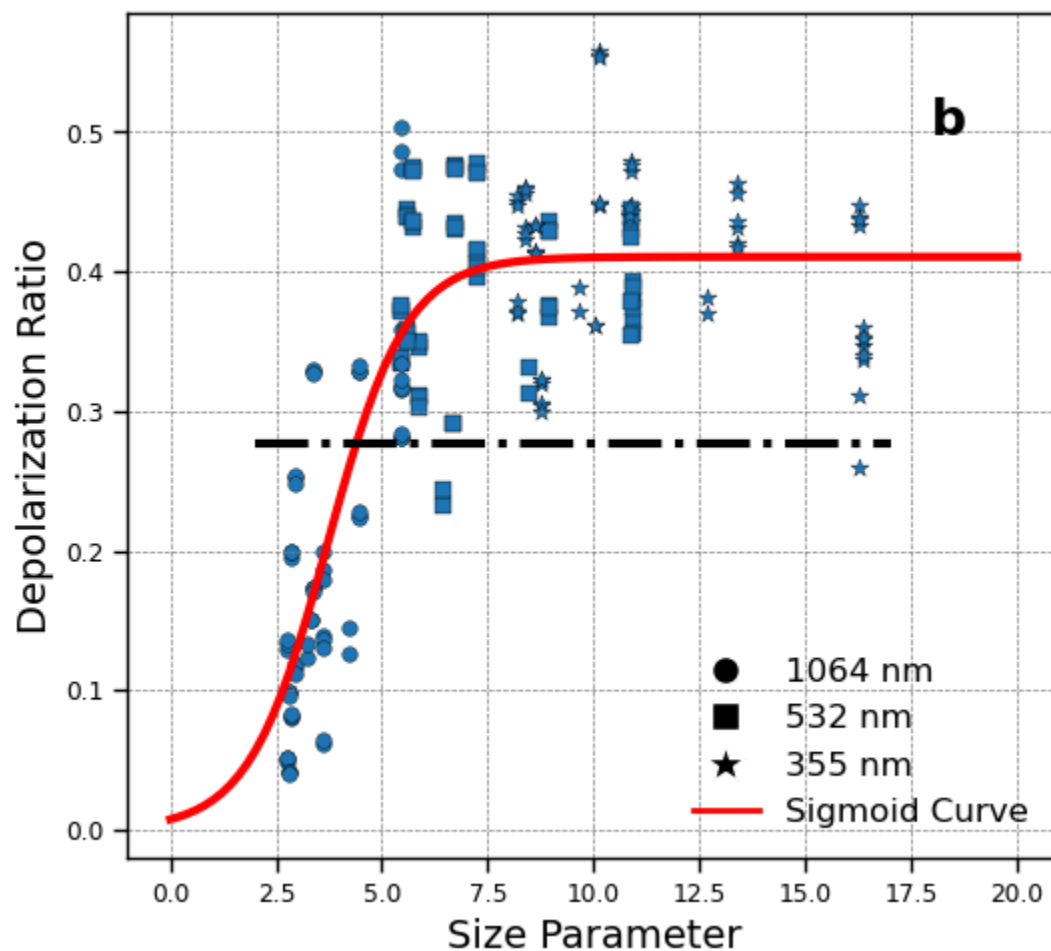


Figure R5. Lidar DPR as a function of dust size parameter derived from the scattering phase function P11, P12 and P22 at the 178 degree scattering angle.

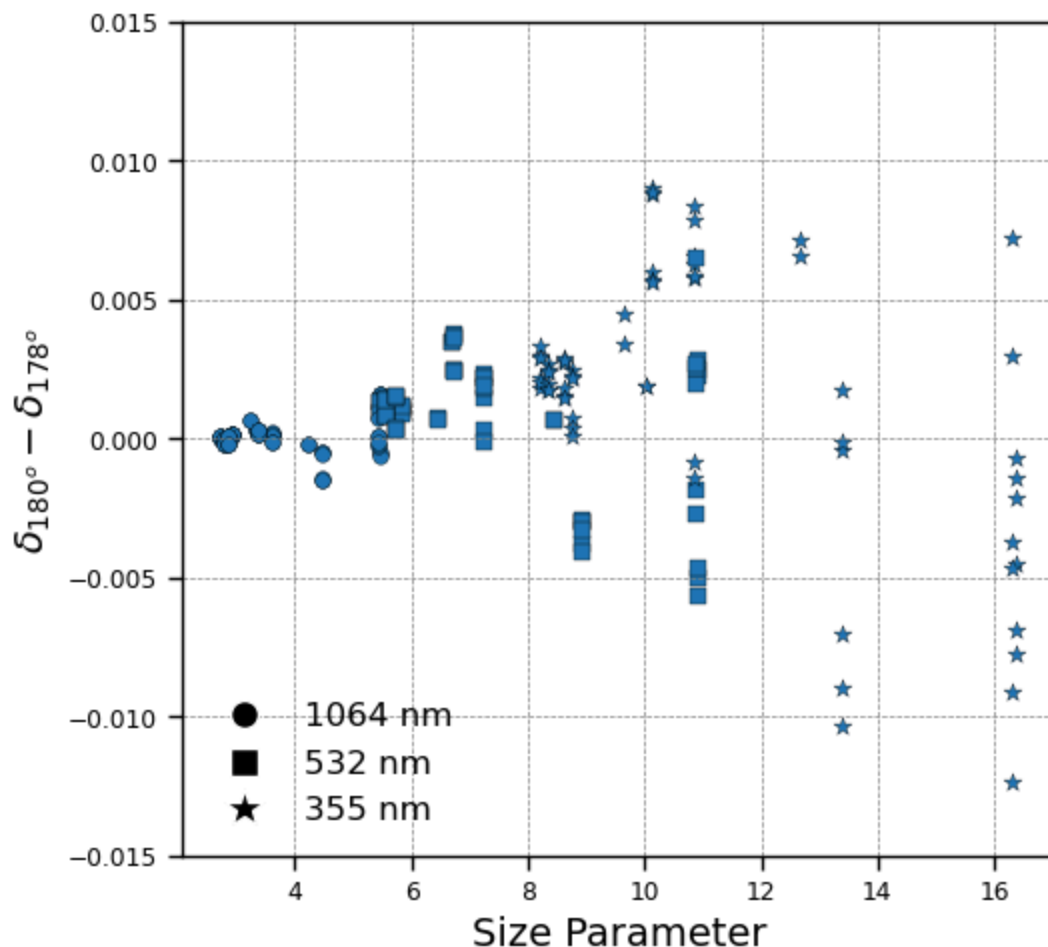


Figure R6. The difference between the DPR at 180 and 178 degree scattering angle.

Obviously, the results are very minimally affected by this change. Looking at the change between the two in Figure R6, We find in addition to the limited variance, no clear trend positive or negative for which would be greater. However, we note greater variance for larger particles. This likely comes from a decrease in P11 and P22, but an increase in P12, previously negligible in the DPR values at 180 degrees.

L511-518: The asymptotic value of the depolarization ratio (0.41) is quite high compared to approximately 0.3 in Järvinen et al., 2016. How do you explain the differences? If I as a user would like to apply a parameterization like your eq 10, I would apply it rather to the measured data from Järvinen than to the purely modelled data. It is too far from the observations and maybe linked to some limitations in the model. Even if you use realistic shapes, it is still a model.

Response: It is entirely up to the user to decide which parameterization—ours or that of Järvinen et al. (2016)—best suits their application. We do not claim that our model is superior.

As discussed earlier, neither our model nor that of Järvinen et al. can fully represent the wide diversity of Asian dust properties on its own.

It is also important to clarify that our parameterization in Eq. (10) describes the DPR of a single particle, whereas the values reported in Järvinen et al. (2016) represent bulk scattering properties averaged over particle size and shape distributions. Recognizing that bulk DPR is more relevant for practical applications, we extended our analysis to develop an approximation and parameterization for bulk scattering DPR, resulting in the DPR–effective radius relationship presented in Eq. (15). This parameterization enables users to construct bulk DPR for monomodal, bimodal, or even multimodal particle size distributions.

In this regard, our scheme provides greater flexibility and broader applicability than that of Järvinen et al. (2016), which—with an asymptotic value fixed at ~ 0.30 —would likely underpredict DPR values commonly observed in measurements, particularly those exceeding 0.30.

As with any modeling approach, our results are constrained by the assumptions and input data used. Nonetheless, the purpose of our parameterization is to offer a physically interpretable and practical framework for characterizing the asymptotic behavior of DPR, thereby supporting a more comprehensive understanding of dust scattering across a range of atmospheric conditions.

Furthermore, in Fig. 11: Why don't we see an asymptotic behavior for the irregular hexahedra? It seems to decrease for 355 nm after reaching a maximum. This finding questions your derived plateau.

Response: This is an interesting observation discussed in lines 575-582.

And to further add, you did the calculations up to a size parameter of 16.5 (Fig. 6). And by purely looking at Fig 6b, I would not be sure if the plateau continues to exist above $x = 12$. Who knows what will happen for larger size parameters?

Response: We acknowledge the reviewer's concern. Indeed, calculating scattering properties for large size parameters using ADDA, especially for particles with highly irregular geometries like the FIB-reconstructed dust used in this study, poses significant computational challenges and is a known limitation of the method. While it is true that extrapolating beyond the maximum size parameter considered in this study introduces uncertainty, we believe the evidence supports the validity of our parameterization within the range relevant for typical dust particle size distributions.

Specifically, the parameterization is not limited to a single particle case, but is further evaluated in Section 5 through comparisons with bulk scattering properties across various dust PSDs

(including dust with size parameters much larger than $x=12$), including those derived from TAMUdust2020. Additionally, similar asymptotic behavior has been reported in other studies (Järvinen et al. (2016) and Kong et al. 2021) using different particle models and methods and for a larger size parameter range, lending credibility to the observed plateau.

While no model can guarantee perfect behavior beyond its tested range, the combination of model-based evidence, comparison with prior studies, and the agreement with representative PSDs provides a strong justification for the robustness and practical utility of our parameterization.

I know that you are still far from lidar observations in the atmosphere. However, the spectral slope of the depolarization ratio was measured for Saharan dust (see literature, which comes close to the shape in Fig. 12) and for dust from the Taklamakan dessert by Hu et al., 2020.

5. Data availability

A statement about the data and code availability is missing although it should be included in the ACP style file. Please ensure the availability and traceability of the used data.

Minor comments

- The overall impression is that the manuscript would have benefited if the authors would have spent another month to carefully check the manuscript. There are several minor, but annoying issues which could have been eliminated, e.g., figure captions which mention different quantities than shown in the figure (e.g., Fig. 3), changing symbols for lidar ratio and depolarization ratio (Fig. 10) or color coding with the same quantity as shown on the x-axis (Fig. 8). Furthermore, a more careful literature study would have been great.

Response: We have made the suggested changes to the mentioned plots.

- The introduction is not really an introduction but already describes the theoretical background. I would move all equations to a separate section and keep a more clear and straight forward structure of the introduction.

Response: Equations have been moved to a new section.

- Furthermore, the first paragraph of the introduction discusses extensively the radiative forcing of mineral dust, but this is of minor importance for the presented study. Please

reshape the introduction and reduce it to the parts relevant for the present study. To my opinion, the first paragraph can be reduced to 2 sentences.

Response: Done

- All figures missing the unit of the lidar ratio and probably some other units as well.

Response: Added steradians.

- The unit of the lidar ratio is sr and not sr⁻¹ as used throughout your manuscript.

Response: Fixed.

- The size parameter is defined quite late (L411) and later on defined differently (L514). Please define it earlier and keep one convention (2 pi or just pi).

Response: This is corrected and now stated once, on line 252.

- You are discussing Asian dust, but through an US American perspective (e.g., lines 51-53) omitting a long tradition of Asian dust research in Japan, but also in China and Korea, which are countries much stronger affected by Asian dust. Please add the respective literature.

Response: Lines 51-53 discuss the long range transport and effect of dust on the western United States, which seems relevant to a discussion of the optical properties of dust morphologies based on those obtained from deposition in Hawaii. We have added more discussion of dust in Asia throughout section 1.

- This American perspective continues when solely name MPL Net and CALIPSO omitting European and Asian lidar networks which already use much more advanced lidar systems. The new EarthCARE satellite measures not only the elastic backscatter like CALIPSO but is equipped with an HSRL channel to measure directly the extinction coefficient and so the lidar ratio. The products are described by Donovan et al., AMT 2024.

Response: We discuss MPL Net and CALIPSO because the observations used in the study come from CALIOP and AERONET. This is why we focus primarily on the 532 nm wavelength. Regardless, we have added references to EarthCARE.

- L73-78 You're talking about the optical properties of a single particle and at the same time introduce the bulk properties. Please keep it well separated.

Response: Rephrased this.

- L138-141 Kemppinen et al., 2015a,b used realistic dust shapes as well for their DDA calculations. The 2 papers are cited later (L504), but should be already mentioned here.

Response: The discussion of L138-141 is in regard to using the observed morphology of dust as a direct model for dust shape. Kemppinen uses a voronoi tessellation-based geometry to add detail to mineral occlusions, but the overall shapes of the model do not come from direct imaging measurements. Nevertheless, their mention was added.

- Fig 1: Please be sure what you want to show. 4 CALIPSO cross sections are a lot and less would be sufficient as well. The captions are not readable at all and the plots are only understandable for people familiar with CALIPSO. Which color represents dust? The dashed lines in 1a are not vertical.

Response: As described in the plot, yellow (or 2) describes desert dust, with brown (or 5) corresponding with polluted dust. This is the standard aerosol classification for CALIOP data and not unique to this work. The curvature of the dashed lines is due to the flat projection of the Earth; these were renamed to “North-South running lines” to assist readers. Figure is remade and simplified.

- The date format is changing throughout the manuscript. MDY – Month Day Year – is not a well-defined date format, even if it is commonly used in the United States. Please choose to go from specific to general (DMY) or from general to specific (YMD).
- L365-367: Quantitatively, the same behavior for the spectral depolarization ratio and lidar ratio was measured by Haarig et al., 2022. However, the values are different.

Response: Specifically regarding an inverse relationship between lidar ratio and depolarization ratio, this does seem to be the case. However, Haarig’s study shows depolarization ratio to be greatest at 532 nm wavelength of the three, and the change from 355 nm to 532 nm in lidar ratio to be statistically insignificant. It is important here to note that the discussion on lines 365-367 is in regards to single particle values, which can vary greatly and do not often exhibit the same properties as large-scale observations of the atmosphere. It would be more meaningful to compare their results to ours in section 5, where we discuss bulk scattering properties of the realistic dust model, as cited in lines 629 and 671.

- Eq 4,5 & 7 are not a real equation, but only a matrix. Please write them as equations.

Response: Corrected.

- L393: Which dust transport region you are referring to? I would guess you are referring to Asian dust over the Pacific when you speak about dust transport region.

Response: This refers to the Atlantic dust transport region as discussed in the cited literature (Liu Z. et al., 2015). Specificity is added to the manuscript.

- L454-457: Please compare to Saito & Yang, GRL 2021 and Gasteiger et al., TellusB 2011.

Response: Neither of these studies directly compare sphericity to lidar properties to show a clear statistical trend. They do however agree with our understanding that morphology plays an important role in optical properties, as we had great variation between properties and particle shape. However, size was generally important in each study.

We can, however, use the TAMUdust2020 database to look at sphericity effects ourselves. Using the Global average refractive index from Di Biagio's work and the hexahedral database at 532 nm wavelength we can see another database's sensitivity of sphericity with lidar properties

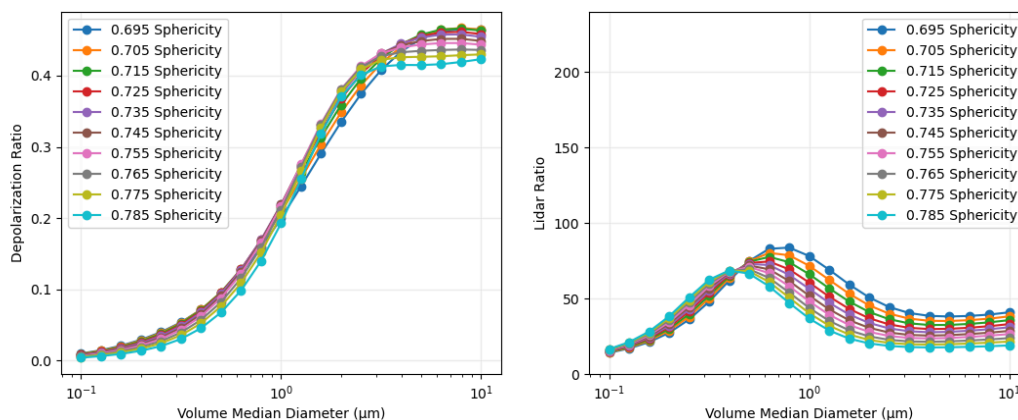


Figure R7. Depolarization ratio and Lidar ratio of TAMUdust2020 with varying volume median diameter and effective sphericity. This data uses a logarithmic monomodal particle size distribution with $\sigma = 0.47$ and the global mean refractive index for dust from Di Biagio et al., 2019.

As seen in Figure R7, there is a small dependency on sphericity, with a trend of decreasing depolarization ratio and lidar ratio with increased sphericity for larger particles in particular. However, there are some limitations to these results. Firstly, the range of effective sphericity in the library available is very limited, from 0.695 and 0.785. There are also multiple inversion points as size increases, with the opposite relationship occurring for smaller particles and lidar ratio or particles between ~1μm to 3μm diameter. Additionally, and what further limits the conclusion of this test, is that through the TAMUdust2020 database, sphericity is not strictly changing the individual particle being used, rather, it is a difference in weighting of each particle in the library's contribution to the output optical properties. A higher sphericity increases the relative importance of the more spherical particles in the database when producing an output, rather than having a single particle for each sphericity value. This means if a high sphericity particle has a particularly low depolarization and lidar ratio, increasing its weighting smoothly decreases the lidar and depolarization ratios, whereas with the FIB dust data, each particle individually has a separate sphericity across a much larger range, introducing greater noise to

our sphericity comparison.

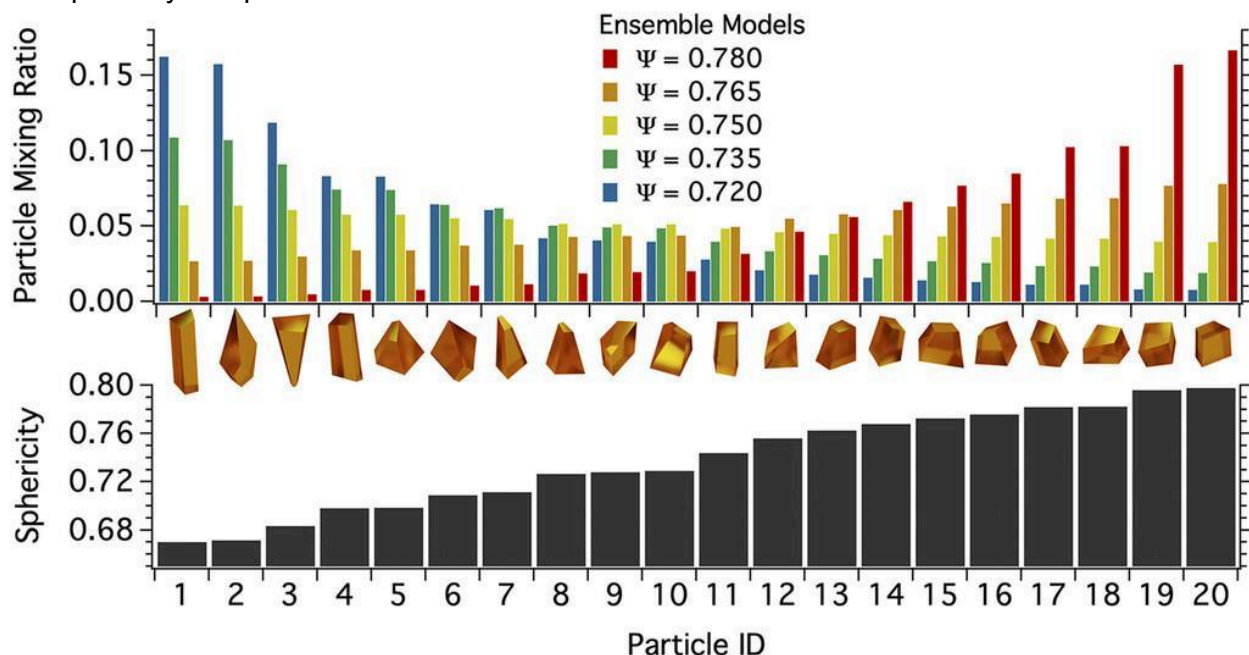


Figure R8. From Saito et al., 2021, mixing ratio of each particle for TAMUdust2020's optical properties. Each sphericity used in the ensemble model corresponds to weighted results of each particle ID rather than an individual geometry.

- L699 ASL does not appear in the list of coauthors, probably it refers to the first author.

Response: Correct.

- L700: “ASL contributed to the methodology, data collection, interpretation and analysis and data visualization” – But who has done the data collection and analysis? If ASL just contributed to it, someone else had to do it. But who?

Response: As described in Lines 694-702, JD helped to design the convergence index, JZ and QS assisted in data curation and interpretation for FIB dust lidar property results and global refractive index data, and ZZ supervised the project.

- It seems that the manuscript was made in word – it is recommended to use a latex environment instead. This will prevent that figure captions are given on the next page and not below the figure, and that formulas have different sizes. Furthermore, with latex the references are given in a consistent manner. In your manuscript some references are cited with the initials of the first author, e.g., L 58, but most not.

Response: The initials are due to multiple authors with the same name and year of publication. Attributing “a” or “b” would imply the authors to be the same person. MS Word follows the recommendations of EGU.

Technical corrections

- L65 gases
- L531: r_{vg} is not used in eq 13.
- L692: fine and coarse mode dust
- Burton et al., 2012 – the reference appears twice in your list.

Response: We have added the above technical corrections to the manuscript.

References (which are not already in the paper):

Di Biagio, C.; Formenti, P.; Balkanski, Y.; Caponi, L.; Cazaunau, M.; Pangui, E.; Journet, E.; Nowak, S.; Andreae, M. O.; Kandler, K.; Saeed, T.; Piketh, S.; Seibert, D.; Williams, E. & Doussin, J.-F.: Complex refractive indices and single-scattering albedo of global dust aerosols in the shortwave spectrum and relationship to size and iron content, *Atmospheric Chemistry and Physics*, **2019**, *19*, 15503-15531

Donovan, D. P.; van Zadelhoff, G.-J. & Wang, P.: The EarthCARE lidar cloud and aerosol profile processor (A-PRO): the A-AER, A-EBD, A-TC, and A-ICE products, *Atmospheric Measurement Techniques*, **2024**, *17*, 5301-5340

Floutsi, A. A.; Baars, H.; Engelmann, R.; Althausen, D.; Ansmann, A.; Bohlmann, S.; Heese, B.; Hofer, J.; Kanitz, T.; Haarig, M.; Ohneiser, K.; Radenz, M.; Seifert, P.; Skupin, A.; Yin, Z.; Abdullaev, S. F.; Komppula, M.; Filioglou, M.; Giannakaki, E.; Stachlewska, I. S.; Janicka, L.; Bortoli, D.; Marinou, E.; Amiridis, V.; Gialitaki, A.; Mamouri, R.-E.; Barja, B. & Wandinger, U.: DeLiAn -- a growing collection of depolarization ratio, lidar ratio and Ångström exponent for different aerosol types and mixtures from ground-based lidar observations, *Atmospheric Measurement Techniques*, 2023, *16*, 2353-2379.

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Hu, Q.; Wang, H.; Goloub, P.; Li, Z.; Veselovskii, I.; Podvin, T.; Li, K. & Korenskiy, M.: The characterization of Taklamakan dust properties using a multiwavelength Raman polarization lidar in Kashi, China, *Atmospheric Chemistry and Physics*, **2020**, *20*, 13817-13834

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Response to RC2

We're grateful to the reviewers for their comments and suggestions, which significantly improved the manuscript. In the revised version, we have made the following major changes:

- Streamlined introduction and reduction of discussion on the radiative effects of dust
- Equations and quantity definitions moved to a newly introduced "Theoretical Background" section
- Minor revisions to most plots for improved clarity
- Clarifications to results and discussion in section 5
- Additions to limitations in conclusion section

Below are point-to-point responses to reviewer comments.

Comments:

1. Please capitalize the 'p' in the title "properties" to maintain consistency with the other words in the title.

Response: Done.

2. Page 6 (48): The sentence, 'If lidar backscattering is dominated by single scattering, δ is close to zero for spherical or quasi-spherical particles like smoke aerosols and water droplets,' is only partially accurate. Theoretical calculations show that even a small deviation from perfect sphericity can produce a finite depolarization ratio. Additionally, the term 'smoke aerosols' can refer to both fresh and aged soot, which may form super-aggregates with irregular shapes. Therefore, the near-zero depolarization ratio observed for soot particles may also be due to their high imaginary refractive index, which reduces their depolarizing ability. I encourage the authors to revise this statement accordingly.

Response: We have removed "or quasi-spherical" from our statement, and changed smoke to sulfate to avoid confusion over shape. Results from Bi et al. (2018) suggest, especially for larger particles, small changes from an ellipsoid aspect ratio of 1 have high depolarization ratio.

Lei Bi, Wushao Lin, Dong Liu, and Kejun Zhang, "Assessing the depolarization capabilities of nonspherical particles in a super-ellipsoidal shape space," Opt. Express **26**, 1726-1742 (2018)

3. Page 16 (48): The term 'scattering phase matrix P' is somewhat unusual. Most textbooks refer to this simply as the 'scattering matrix,' which includes the phase function as its first element. Please review this usage.

Response: We have reworded it to 'scattering matrix' throughout the text.

4. Page 19 (48): The statement 'Fig. 4 can serve as a benchmark for future studies on

mineral dust scattering properties' is somewhat surprising. To me, this appears to be a basic phase function calculation without a clear or coherent explanation. Similar plots are widely available in the literature. Furthermore, the graphs represent scattering calculations at different size parameters, which are dimensionless quantities. From the figure, what I observe is that larger particles (with higher size parameters) exhibit stronger forward scattering, which is expected, as scattering in the forward direction is roughly proportional to the particle diameter to the fourth power (D^4)-a well-known behavior in the scientific community.

Response: This is in regards to a benchmark comparison for future theoretical shape models, given the shapes are tied to physical observations and the lack of single particle lab measurements of lidar properties. The statement was adjusted to improve clarity.

5. In line 405, page 23 (of 48): A space is needed after " $P_{11}(\pi)$ ".

Response: Thank you for the correction.

6. Line 514, page 29 (48): The expression for the size parameter is written incorrectly- specifically, the factor of 2 is missing.

Response: This was previously defined in the paper with the factor of 2, so we have removed the second incorrect definition.

7. In the caption of Fig. 7, please correct the sentence: 'The color of each dot corresponds to the imaginary of the imaginary index.'

Response: Fixed.

Response to RC3

We appreciate the reviewers' insightful feedback, which was instrumental in enhancing our manuscript. In the revised version, we have made the following major changes:

- Streamlined introduction and reduction of discussion on the radiative effects of dust
- Equations and quantity definitions moved to a newly introduced “Theoretical Background” section
- Minor revisions to most plots for improved clarity
- Clarifications to results and discussion in section 5
- Additions to limitations in conclusion section

Below are point-to-point responses to reviewer comments.

Major comments:

1. Unclear description of refractive indices used

There are 14 particles and 3 wavelengths. If I understood correctly, in the main part of the study two different refractive indices were used for each of these 42 combinations (of particle/wavelength). However, the description around line 238-248 is a bit vague in my view. Which refractive indices did you actually use? In line 242 and 245, what does 'possible' mean? Please improve the description.

Response: Each particle's refractive index at 532 nm was estimated based on the possibility of the iron-phase composition found from SEM and EDX. This results in up to three different mineral phases for each particle, each with two refractive indices provided for variability induced by optical anisotropy. This means there are up to six refractive indices used for each particle, or far greater than 42 combinations. The description in the manuscript is improved, please refer to Conny et al., 2019 for more in depth explanation.

2. Data availability

Since the number of cases is not too large, I recommend to make the input and output data available in tables. For example, a list of radii, aspect ratios, used refractive indices for the 14 particles (in the main text), as well as a list of the obtained depolarization ratios and lidar ratios (in an appendix).

Response: We have added a data availability section and put the reference data in a publicly accessible netcdf format. Given the number of simulations, a table of all results would be very long.

3. Size

A volume-equivalent radius range from 0.46 to 0.93 μm is covered by the 14 particles, which is quite a narrow range. Since the authors assumed a wavelength-independent refractive index, the size range is a bit extended if viewed in size-parameter space as done in this study. However, the size range and also the size sampling remains limited. You might scale the size of the existing particles to get a better sampling and range but I understand that it takes considerable effort to do the additional simulations. Nonetheless, I would recommend to reconsider this possibility. In any case, adding some more discussion of this limitation is recommended.

Response: This is the primary motivation of section 5, as the lack of coarse mode particles requires the parameterization we produce here to replicate the size range of dust in the atmosphere. Based on the fitting of our parameterization, replicability of hexahedral dust results in section 5, and the commonality of the asymptotic behavior across particle geometries and laboratory measurements in other studies, we believe this to be evidence of the size parameter range to sufficiently capture the variability of dust DPR. We have added additional discussion to this point.

4. Fine-mode non-dust particles

In the discussion of your results I didn't find mentioning of the fact that in most cases desert dust aerosol contains a fine mode of non-dust aerosol particles which affects in particular lidar measurements at short wavelength. For example, these kind of particles are considered as WASO particles in the mineral dust mixture of OPAC [1]. Another example is the SAMUM campaign, where they were observed and considered in the optical modeling study of Gasteiger et al. (2011) (already referenced in the manuscript). I think it is necessary to take into account the effect of fine mode non-dust in the discussion of your results.

Response: The scope of this work is limited to dust particles, in fact, mostly limited to individual dust particles. Optical properties of non-dust particles are left to studies on other aerosols in our work. Nevertheless, if one wished to include them in our parameterization, it would be quite simple to expand the adapted equation 16 to the original work of Mamouri and Ansmann, 2014, including the non-dust term.

5. Settings of ADDA

Nowhere is mentioned which version of ADDA and which settings of ADDA you are using. Did

you just use the default settings? Did you try other settings? Please provide more details to make the study as reproduceable as possible.

Response: We used all the default settings of ADDA following the user manual, except for the orientation averaging. Therefore, we didn't reiterate the default settings in the manuscript but focused on how to treat the averaging over orientations. Section 3.3 is related to addressing this. These are the "settings" that can be adjusted with ADDA. The other values that can be adjusted aside from orientation are refractive index, size, and shape, detailed in section 3.2 and 3.1. We add mention of the use of ADDA version 1.4.0 to line 258. We used the default scattering angles as it was sufficient for our interests; it contains the backscattering angle of 180 degrees

6. Definitions in introduction

Lidar-related parameters are defined in the introduction. I find this unusual and would recommend to move this part into section 2. (If Copernicus guidelines allow it to be in the Introduction, I am also OK with it)

Response: We have moved the equations to a new separate section.

Minor and technical suggestions / comments:

Line 51: 'this dust' should be replaced by 'dust aerosol'.

Response: Replaced.

Line 55: 'facilitate the working of' should be 'being part of'.

Response: This has been adjusted.

Line 74-79: There is an inconsistency: You start with 'For a single dust...' but the equation 1 is for bulk aerosol.

Line 81: There is an inconsistency: Beta is a bulk property, while C_sca is a single particle property.

Response: These two issues are addressed together through rewriting the lines to better separate single particle and bulk discussion.

Line 88: It unclear why 'it is fundamentally important'. Please improve the argumentation.

Response: 'It' in this sentence was supposed to refer to the lidar ratio rather than the process of converting between extinction and backscatter. The sentence was rewritten to better communicate.

Line 98 and 101+102: The fact that delta is used for aerosol and cloud classification is repeated here within a few lines.

Response: Removed the redundancy.

Line 176: 'The fourteen FIB dust particles' should be reformulated to something including 'particles measured by FIB' or 'sampled by FIB'.

Response: In line with the previous work of Conny et al., (2019) we have labeled the particles "asian dust particles sampled by FIB" when referring to it for the first time then "asian dust" afterwards.

Figure 1: Labels are hardly readable.

Figure 1: Plots c) and e) are not compatible (different lat/lon).

Figure 1 / I195-201: Please explain better the content of the figure.

Response: Figure 1 and text remade for readability.

Figure 2: The coordinate system axes are hardly visible.

Response: Fixed.

Line 231-232: Could be simplified to 'In this study we follow the ... convention of Conny et al. (2019)'.

Response: We have altered this for better clarity.

Line 237-238: Use plural 'which have ... indices'.

Response: Done.

Line 260: The Extinction cross section is usually labelled as C_{ext} .

Response: Fixed to align with the rest of the text.

Line 261-262: The scattering properties also depend on wavelength which is missing here.

Response: Added.

Line 291: 'for' is missing .

Response: Added.

Line 293: Insert 'the' before 'random'.

Response: Done.

Line 294: 'that' is repeated.

Response: Removed.

Line 303-304: Insert 'by' before the three angles.

Response: Inserted.

Line 310 / 315: For $n=6$ the equation results in $(2^6+1)^3=274625$ orientations while 262144 orientations are reported; it is not consistent.

Response: You are correct, as well as for $n=1$, and is fixed in the manuscript.

Line 318: What was exactly the convergence criterion? A threshold?

Response: The threshold we use is based on the elbow method, we found generally for the particles a clear flattening to the curve in figure 3c for the various particles between $n=3$ to $n=6$, depending on the geometry. The Convergence Index shows when there is diminishing returns on computational time, and thus is based on a user's allowable level of error in their computations.

Figure 3c: The y-axis has label 'Variance Index' which is not defined. Do you mean the CI?

Response: Yes, CI is correct. Fixed.

Figure 3: I would try to add the n values somewhere in the plots to make the approach clearer. E.g. as labels at the top? If this is not possible, improve the legend such that it becomes clear immediately that n=2 to n=6 is covered.

Response: Vertical Lines were added for clarity.

Line 325: Insert 'number of' before 'orientations'.

Response: Done

Line 326: Is (a) and (b) also for particle 3D?

Response: Yes, we have made a note in the manuscript.

Line 326: The label should mainly explain the content of the Figure and not the interpretation. Therefore I suggest to write 'S and linear depolarization ratio as function of the number of orientations'.

Response: We have fixed the caption.

Line 335: The meaning '~20 nm³' is unclear. Do you really mean a volume?

Response: Corrected to 10³ nm³. Each particle contains over 100,000 dipoles in their geometry file, so they are highly detailed. So, for example, taking particle 3D Ca-rich, it has a volume of 0.481 μm³, with 144051 dipoles, so each dipole has a volume of ~3,339 nm³ in this particular particle.

Line 341: 'For each wavelength, more than 60 ADDA simulations are carried out': I thought it would be 14 particles and 2 refractive indices at each wavelength. Please clarify which simulations were performed.

Response: See explanation of RI above. Each mineral phase of each geometry has 2 refractive indices.

Line 349/350: The scattering angle is missing here.

Response: Added ($\theta = \pi$).

Figure 4: Is there one line for each of the 14 particles? Are there more lines? Please explain better what is shown here.

Response: Each minimum refractive index, or half the total simulations run at each wavelength, as described in the caption. I have added this to the main text as well. This was done for readability.

Line 361: Size should be singular.

Response: Corrected.

Figure 6a: There is quite some spread at size parameter 16-17. Why is there an outlier? What is special in that case?

Response: For individual particle measurements, there is often considerable variability in optical properties (see Kong, S. et al., 2022). This is the outlier discussed at length in section 4.3, and lines 404-407. In fact, it is the same particle highlighted throughout the manuscript, 3D Ca-rich, bolded in figure 3 and shown in figure 2. We suspect the primary reason for such outlying optical properties has to do with the high iron oxide content compared to the other particles in the study.

Line 393: The wavelength of the depolarization (i.e. 532nm) could be added.

Response: This was added for clarity.

Line 432: 'session' -> 'section'

Response: Thank you, corrected.

Line 484: It is unclear to me what the range 11.4% to 7.9% represents. Uncertainty of the iron mass in one particle? Please explain better.

Response: Different mineral types have different densities, thus varying the mass percent composition. Depending on the mineral type assigned to the iron-based mineral found through EDM, the iron phase mass could differ.

Line 489-490: This sentence is unclear to me. Please improve.

Response: Added “of mineralogy” to what effect we are referring to.

Equation 11: Scattering angle missing.

Response: Added.

Line 531: r_{vg} is not used in the equation. What does 'median' refer to here?

Response: Moved to equation 14.

Line 540: 'elected' doesn't sound right to me. Maybe 'selected'?

Response: Changed to 'selected'

Line 551-553: The sentence should be made clearer.

Response: The sentence is there to both define the variables in equation 14 and set the standard deviation we use for the remainder of the study. I have separated into two sentences and reworded the phrasing to be more clear.

Line 571: Eq. (13) does not contain a parameterization. Please check the equation number.

Response: Fixed to equation 15.

Line 587: 'Diameter' is not consistent with the label in plot b.

Response: Changed to 'Radius'.

Line 588: 'through parameterized approximations': Please be more explicit, e.g. mention equation XX

Response: Altered to mention equation 15.

Line 616: There is no vertical line in Fig 12a.

Response: Removed.

Line 618: 'as described previously' should be rewritten to 'as described in the text'

Response: Changed

Line 619: I suggest not to start the paragraph like this. 'promising' might be better suited as a conclusion that you may get after a discussion of the results.

Response: We rephrase the paragraph as a whole.

Line 638: You write that the decrease of the depolarization ratio from 532nm to 355nm is a result of absorption. But how can it be an absorption effect if the decrease is observed only for the full PSD and not for fine mode and not for the coarse mode (like in Fig. 12b)? To me it looks more like a size effect.

Response: When wavelength decreases, the size parameter of the dust increases. Therefore, the DPR would be expected to increase, as larger dust particles exhibit greater DPR within the considered size parameter range. And, when looking at the fine and coarse mode individually, DPR does increase. However, the DPR for the full PSD decreases. This must be accounted for through the backscatter coefficient weighting in equation 16 β_{coarse} increases with wavelength, meaning C_{sca} must increase with wavelength as P_{11} decreases with wavelength for coarse mode particles in particular. Thus, the decrease in Full PSD DPR must come from greater fine mode relative weighting in equation 16, which must come from lesser scattering or greater absorption.

Line 656-657: What do you mean with 'theoretical libraries for some amount of calculation'?

Response: Changed "theoretical libraries" to "simplified theoretical geometries". The backscattering as a function of size comes from the TAMUdust2020 hexahedral database for combining multiple modes.

Line 619-670: In particular this part should be reworked to make it clearer.

Response: We have reworked the section, in particular adding clarity to which methods we are comparing in each part.

Line 673: 'FIB particles' should be reformulated, see comment on Line 176.

Response: In line with the previous work of Conny et al., (2019) we have labeled the particles "asian dust particles sampled by FIB" when referring to it for the first time then "asian dust" afterwards.

Line 685: Absorption also greatly increases S.

Response: Added.

Line 688: 'CI' should be replaced by 'convergence index'

Response: Changed.

Line 690: Add 'size distribution'.

Response: Done.

Line 692: This section lacks a summary of the limitations of the study / of the proposed parameterization.

Response: We have added some limitations to the conclusions, in particular for the parameterization coming from Section 5.