

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 ' $\sim 20 \text{ nm}^3$ ' is unclear. Do you really mean a volume?

Response: Corrected to 10^3 nm^3 . 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 $0.481 \mu\text{m}^3$, with 144051 dipoles, so each dipole has a volume of $\sim 3,339 \text{ nm}^3$ 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 is 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.