

In this response letter, comments from the reviewer are highlighted in black font, our responses are in blue font, and modifications made to the manuscript are indicated in red font.

Reviewer #1

This study investigates differences in the retrieval refractive complex refractive indices using different optical models. In their baseline case, optical properties were calculated assuming inhomogeneous super-spheroid model, and were then compared with homogeneous super-spheroid models and the homogeneous sphere models. Optical properties were calculated for wavelengths 355, 532, 633, 865, 1064 nm. Three particle size distributions (S, M, and L) or combinations of them were used in the simulations.

Overall, the work is interesting, and the evaluation of different aerosol optical models is relevant to the scientific community. However, certain sections of the text are not clear or are confusing. For instance, the authors refer to a numerical simulation as a “measurement” and, at times, as “experiments”. Additionally, the baseline simulation is referred to as a “dust sample”. These nomenclature makes some parts of the text unclear. Also, the authors evaluate the retrieval refractive indices in what they called “ideal” and “realistic” scenarios. However, the “ideal case” corresponds to a simulation where size and scattering truncation are not corrected, and it is unclear why they would evaluate and label such a case as “ideal”.

In the “realistic case”, which appears to be the appropriate scenario for evaluation, they found that using either the homogeneous super-spheroid model or the homogeneous sphere model was unable to match the optical properties for the large particle sizes. The reason for this is attributed to “differences in size distribution between the dust samples and the models.” However, it is not clear why there are differences in the size distribution between the baseline and the model simulations.

Thank you for providing us with detailed and valuable feedback. We greatly appreciate your insights and concerns regarding certain sections of the text.

1. Nomenclature Clarification:

We acknowledge the confusion might be caused by referring to numerical simulations as ‘measurements’ or ‘experiments’ and the baseline simulation as a ‘dust sample’. To accurately reflect the nature of our simulations, we have revised the terminology by using terms ‘numerical simulation’ and ‘baseline case’ to avoid potential ambiguity.

2. Clarification of ‘Ideal’ Case:

We have revised the explanation regarding the ‘ideal’ case in the text to emphasize that it refers to a scenario where the size distributions and scattering coefficients of the baseline case can be accurately obtained and used in the retrieval. This implies that the inversion models share an identical size distribution with the baseline case. Additionally, we have made corresponding revisions to Table 1 in the manuscript to avoid any confusion regarding the terminology used.

3. Explanation of Differences in Size Distribution:

Essentially, the inversion models imperfectly represent the non-sphericity and inhomogeneity of the baseline case. This imperfect representation leads to discrepancies in the size distribution when using the OPC for particle sizing. We have provided a more detailed

explanation of this difference in relevant sections of the main text. Additionally, we have included a more comprehensive analysis of the differences in size distribution between the baseline and model simulations in Sect. 3.4.

Specific Comments:

Title: The title mentions short-wave refractive indices only, but the manuscript evaluates five wavelengths 355, 532, 633, 865, and 1065nm, ranging from near ultraviolet (NUV) to near infrared (NIR).

We understood that this term “short-wave” may have different wavelength ranges in different contexts. For example, Di Biagio et al. (2019) use the term to refer to the 370–950 nm range, while Go et al. (2022) define it as 0.185–4.0 μm to distinguish the terrestrial radiation and the solar radiation. In our study, we specifically focus on the wavelength range of 355 to 1064 nm, as stated in the abstract.

To address this concern, we have added an explanation in the introduction section when the term "short-wave" is first introduced.

“It should be noted that the short-wave (355-1064 nm in this study) refractive indices of dust...”

Page 2, Line 36: It would be useful to include a brief description of what a homogeneous super-spheroid model actually is.

Thank you for your suggestion. We have included a brief introduction to the super-spheroid model in this section.

“To improve upon the spheroid model, the super-spheroid model, which extends the dimensions of both the sphere and spheroid models, has been proposed to provide a more comprehensive framework for describing the shape of dust particles. Initial studies have shown that homogeneous super-spheroid dust models align well with laboratory measurements...”

Page 4, Line 99: “Dust samples of different sizes and optical databases for homogeneous models were prepared. Inhomogeneous super-spheroid dust models were regarded as the “dust samples.””. It is not clear what the authors mean by “prepared”. Do you mean selected? From where? Which datasets were used to define the “dust samples”? It would be more clear if you define “dust samples” as the “baseline case”.

Thank you for your feedback. We have revised these sentences and used the term of “baseline case” instead of “dust samples”.

Page 4, Line 99-108: For clarity, I would suggest the authors to rewrite the steps of the numerical experiment in a more directly, avoiding stating that quantities were measured, and refrain from calling an optical model/simulation a “dust sample”.

Thank you for your suggestion. We have revised the description of the numerical simulations to avoid ambiguity.

Page 4, Line 113: “Two correction processes were considered for the measurements of “dust

samples” to mimic actual laboratory experiments.” This sentence is not clear. Were these processes used in the numerical simulation? If so, please make sure to clarify that in the sentence.

Thank you for your suggestion. We have made revisions to clarify the correction processes used in the numerical simulation.

“In the numerical simulations, we have incorporated two correction processes based on the actual laboratory experiments. The first correction is the size correction, which is employed to determine the geometric size of the particles from imaginary OPC measurements...The second correction is the scattering truncation correction, which is associated with the unavoidable technical limitations in measurements of scattering coefficients.”

Page 4, Line 123: For clarity, I suggest replacing: “experiments...” by “numerical simulations”, here and throughout the entire manuscript.

Thank you for your suggestion. We have made the corrections accordingly.

Page 4, Line 124: “Note that E1 represents an ideal situation in which no instrument defects need to be corrected for the measurements, ...” It is not clear what the authors are calling “instrument defects” here. The corrections the authors mentioned previously (scattering truncation and size) corrections should not be refer to as instrument defects. The corrections are needed because instruments are limited in their measurement capacity and not because of a defect (broken or failure).

Thank you for your feedback. You are correct. The term ‘instrument defects’ cannot accurately represent the corrections needed for measurements. We have revised these sentences and referred to the ‘instrument defects’ as unavoidable technical limitations.

Table 1, Caption: Does the instrument bias the authors are referring to correspond to the same two corrections mentioned previously?

Thank you for your feedback. We have revised Table 1 and its caption in the manuscript to address the confusion. The updated table is as follows:

Table 1: A brief description of the four numerical simulations. Target scattering coefficient denotes the scattering coefficient of the baseline case.

Numerical simulations		Target scattering coefficient used for inversion models	
		the same as the baseline case	the scattering coefficients of the baseline case with scattering truncation correction
Particle size of inversion models	the same size as the baseline case	E1	E2
	the size of the baseline case with size correction	E3	E4

Page 11, Line 275: It is not clear why the authors decided not to include uncertainties in the absorption coefficient measurements in their simulations. The authors mentioned “corrections were validated using measurements”. Why does the validation of corrections by measurement change the necessity of including them in the simulations?

Thank you for your comment. The corrections for absorption coefficients are conducted using empirical formulas in the laboratory measurements. It is difficult to verify these complex corrections through numerical simulations. These corrections were validated in (Di Biagio et al. (2017a). Therefore, we assume that the absorption coefficient can be obtained accurately within a certain uncertainty, which is set to 30% based on previous studies (Di Biagio et al., 2019).

We have revised the sentence to clarify the process of correcting dust particles and the assumption made regarding the absorption coefficient in the numerical simulations. The revised sentence is as follows:

“Given that the corrections for dust particles are conducted using empirical formulas and are challenging to verify through numerical simulations. In addition, the corrections have been validated using measurements from the Multi-Angle Absorption Photometer (MAAP) and Cavity Attenuated Phase Shift Extinction (CAPS) (Di Biagio et al., 2017a). As a result, we assume that the absorption coefficient can be accurately obtained within a certain uncertainty, and thus, no additional correction is applied in the numerical simulations.”

Page 12, Line 285: “Due to the sparse nature of the measurements, it was almost impossible for them to fall on the grid points of the look-up tables... These average refractive indices were referred to as the exact values.” This sentence requires clarification. What measurements are you referring to? Are the authors saying that the calculated scattering and absorption coefficients could not be obtained with the optical parameters represented in the authors’ look-up tables?

Apologies for any confusion. What we meant is that it is unlikely for the target value (the scattering coefficients and absorption coefficients of the baseline case) to perfectly align with the grid points of the look-up tables due to their sparse nature. Therefore, we took the average of the four refractive indices corresponding to the four smallest values of RMSD and used that average as the exact retrieved value. The revised sentence is as follows:

“However, due to the sparse nature of the look-up table, it is highly unlikely for the target value to fall directly on the grid points. Therefore, we average the four refractive indices corresponding to the four smallest values of RMSD.”

Page 17, Line 384: “Note the significant discrepancy emerged between the “dust samples” and the homogeneous super-spheroid models as the size increased, which was inconsistent with the findings in Figure 5.” Can the authors explain why the discrepancy is larger in this case (E3/E4) compared to E1/E2? Specifically, in the E4 case where the model has both scattering and size correction, why would the discrepancy be larger in this case?

Apologies for the confusion about the explanation of E1/E2. E1 refers to a scenario where the size distributions and scattering coefficients of the baseline case can be accurately obtained and used in the retrieval. This implies that the inversion models share an identical size distribution with the baseline case. However, in E3/E4, the size distributions of the inversion

models are derived from the size distribution of the baseline case with size correction. We have revised the explanation of E1/E2 accordingly.

Page 21, Line 428: “The E4 scenario represented measurements closer to those obtained in the laboratory, while E1 was considered an ideal scenario.” According to Table 1, E4 corresponds to the cases where both size and scattering truncation corrections were applied, while E1 corresponds to a case “without correction”. It is not clear why the authors would call the case without correction “ideal”.

Apologies for the confusion. The previous reply clarifies that E1 refers to a scenario where the size distributions and scattering coefficients of the baseline case can be accurately obtained and used in the retrieval.

Page 22, Line 441: Instead of using the term “measurement”, please clarify what exactly you are referring to. The same applies to lines 446, 447, and 450.

Thank you for your suggestion. We have replaced the term "measurement" with "baseline case" to indicate the optical properties of the baseline case directly, thereby avoiding ambiguity.

Figure 11: It is difficult to distinguish between each line in the plots. Using different colors and/or line styles may improve clarity.

Thank you for your suggestion. We have adjusted the line styles and line widths in Figure 11 and modified the line widths in Figure 10 to improve clarity.

Page 25, Line 518: “However, when measuring their refractive indices in the laboratory, it is ...”. Refractive indices of aerosol particles are not usually directly measured in the laboratory, but are derived from absorption or extinction measurements assuming an optical model.

Apologies for the oversight. We have updated the word ‘measuring’ to ‘determining’.

Page 25, Line 530: “Under an ideal scenario, where no instrumental defects needed to be corrected, the look-up tables for the homogeneous super-spheroid models were able to fit the measurements at any size.”. There are a few issues here: First, the size and scattering truncation corrections are not “defects”. A nephelometer measuring scattering in an angular range from 7 to 170 degrees is not a defect. Extrapolation (or correction) is the procedure used to obtain the total integrated scattering. Second, I would not refer to an “ideal scenario” as a case where $\beta_{\text{scat}}(0 \text{ to } 180)$ is approximated by $\beta_{\text{scat}}(7 \text{ to } 170)$. Was a solution found using the super-spheroid model fitted with an approximation $\beta_{\text{scat}}(7 \text{ to } 170)$? What does that say about the consistency of the model?

Thank you for your feedback. We have revised the term 'defects' to 'unavoidable technical limitations' to accurately reflect the nature of the challenges faced in the measurements. Regarding the clarification about case E1, we apologize for the confusion. In case E1, the target scattering coefficients for the inversion models in retrieval are indeed the same as those

of the baseline case (beta_scatter(0 to 180)). The case E2 refers to the numerical simulations with scattering truncation correction. The conversion from beta_scatter(7 to 170) to beta_scatter(0 to 180) leads to slight inconsistency between the homogeneous super-spheroid model and the baseline case in E2.

Page 26, Line 552: “The retrieved refractive indices were found to be size-dependent. As the size increased, the imaginary part decreased.”. Where is this shown in the authors analysis? How do you explain this result if you have assumed the same composition in each simulation? Was that observed for both the homogeneous and inhomogeneous models?

The discrepancies in the size distribution mainly contribute to this observation, as illustrated in Figures 7 and 9 in the manuscript. Notably, this phenomenon is specific to homogeneous models, which utilize a single refractive index for the entire model, whereas inhomogeneous models incorporate different refractive indices for different components. The refractive indices assumed for the inhomogeneous models are considered to be independent of size (see Sect. 2.2). A comprehensive explanation is provided in Sect. 3.2.

“However, it should be noted that the refractive indices obtained at small sizes may not be applicable to large sizes as they depend on the sizes in the retrieval process when assuming a homogeneous model (Figure 9). The size-dependent refractive indices are more evident in E3/E4 than in E1/E2 due to significant discrepancies in size distribution between the baseline case and the inversion models. Specifically, the imaginary parts decrease as the size increases, as shown in Figure 7 and Figure 9. The variation of imaginary parts with particle size is less evident when the inversion models share an identical size distribution with the baseline case. However, the variation of imaginary parts is more pronounced at 355 nm in E1 compared with other wavelengths, primarily due to the particle’s relatively larger size parameter at 355 nm, which leads to variations in optical properties with the size parameter.”

Page 26, Line 556: Please define “true values”. Are the authors referring to the complex refractive indices obtained with the inhomogeneous super-sphere model? Why are they not shown?

The term “true values” refers to the optical properties of the baseline case, which specifically pertains to the inhomogeneous super-spheroid model. Only the optical properties of the baseline case (extinction coefficients, SSA, asymmetry factor, phase matrix) can be compared. It is important to note that the baseline case does not utilize a single refractive index for the entire model, which is a characteristic exclusive to homogeneous models.