Responses to suggestions of Referee #2

Thank you for reading the manuscript and providing very useful comments and suggestions to improve the paper. The replies to the referee comments are given below. The referee comments are in blue with our responses in black. The modifications in the revised manuscript can be found in the track change version of manuscript.

Based on the reviewers' comments, we have revised the structure of Sect. 2 and relocated some equations, sentences, and paragraphs in Sect. 2. We have added a new subsection (Sect. 2.3: Synthetic aerosol mixture), which mainly incorporates content previously included in Sects 2.1 and 2.2.

Sentences and equations that were only moved, without any changes to their wording, do not appear in the tracked changes version. This was done to ensure smoother readability.

In this manuscript, the authors propose a method for decomposing aerosol components using particle depolarization ratios measured from lidar at two different wavelengths. The methodology is presented in detail for cases involving mixtures of two and three aerosol components, respectively. Case studies are conducted for mineral dust from Arabian, Asian, and Saharan sources. The method is comprehensively described, and the case studies provide a thorough experimental validation. Given the increasing availability of multi-wavelength lidar measurements, this work represents a valuable contribution to the lidar and aerosol observation communities. However, I have one major comment regarding the methodological analysis, along with several technical and minor comments. Additionally, the manuscript would benefit from proofreading by a native English speaker to improve clarity and flow.

Major comment:

The proposed method, e.g. for the mixture of two components (Eqs. 5 - 9), relies on input parameters from Table 1. These parameters undoubtedly influence the results of the aerosol decomposition, and therefore, a more comprehensive sensitivity analysis that what it has for now is needed. In Section 2.3 and Table 2, the authors conduct an uncertainty analysis for three cases with different depolarization ratios at 355 and 532 nm, comparing reference results with those obtained using Monte Carlo simulations with normal distributions for all input variables. However, the analysis does not address the sensitivity of the results to individual variables. Specifically, which variables have the most significant impact on the decomposition results? For instance, the Ångström exponent is known to vary considerably from different studies, but its specific influence on the results presented in this manuscript remains unclear. While I acknowledge the authors' point that this paper primarily aims to present a method rather than investigate aerosol characteristics (which is beyond the scope of this work), understanding the sensitivity of the results to the input parameters is crucial. Such an analysis would help identify which parameters require more careful consideration in future applications. Furthermore, a sensitivity study could provide a statistical explanation for why certain points in Figure 5 deviate from the curve.

The reviewer raises a valid point. Indeed, the particle depolarization ratios (PDRs) at two wavelengths as well as the relative backscatter-related Ångström exponent (BAE) defines the observational space required by the proposed decomposition method. Therefore, knowledge of the parameters in Table 1 is of paramount importance for maximizing the confidence of the aerosol backscatter fractions in the two- (or three-) type aerosol mixture.

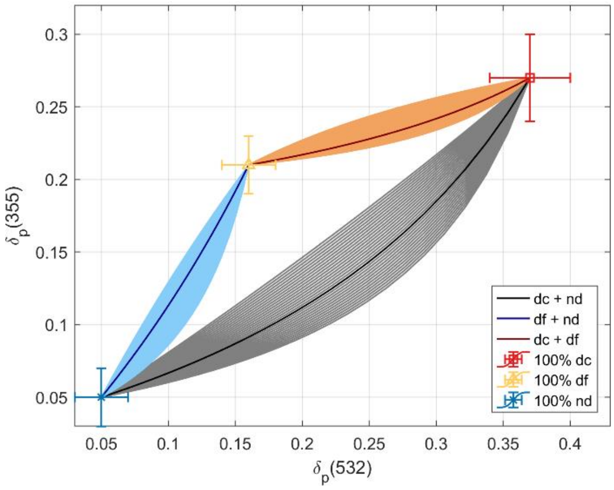
We have performed a more comprehensive sensitivity analysis, and revised the Sect. 2.4 Uncertainty study. In the revised manuscript, we present the sensitivity analysis in 4 parts:

1. A global sensitivity analysis to assess the combined effect of all input variables. Similar as previous version, but adding uncertainties on δp as well, thus considering 11 variables in the revised version.
2. Individual sensitivity analysis using the one-at-time (OAT) method, to assess the influence of each variable independently.
3. We change the uncertainty levels on each variable to investigate the influences.
4. We performed an additional analysis on observational parameters δp, to study their tolerated bias.

From the individual sensitivity analysis, we found that the BAEs have only minor effects on the retrieval compared to other parameters. This finding is important given the considerable variability commonly observed in Ångström exponents. Such results are also added in the conclusion.

Apart from the revised sensitivity analysis present in the revised manuscript, we have performed additional analysis to investigate the effect of PDRs and BAEs.

In Fig. 1d we observe that the characteristic PDRs define the observational space in which the decomposition method is applied while the BAE affects the curvature between these edge points. Equivalent to Fig. 1d, the figure below shows the sensitivity of the BAE for an aerosol mixture of three aerosol types. All possible BAE combinations between -1 and 1, 0 and 2, and 1.5 and 2.5 with an increment of 0.1 for each BAE were considered for coarse, fine, non-dust types, respectively.

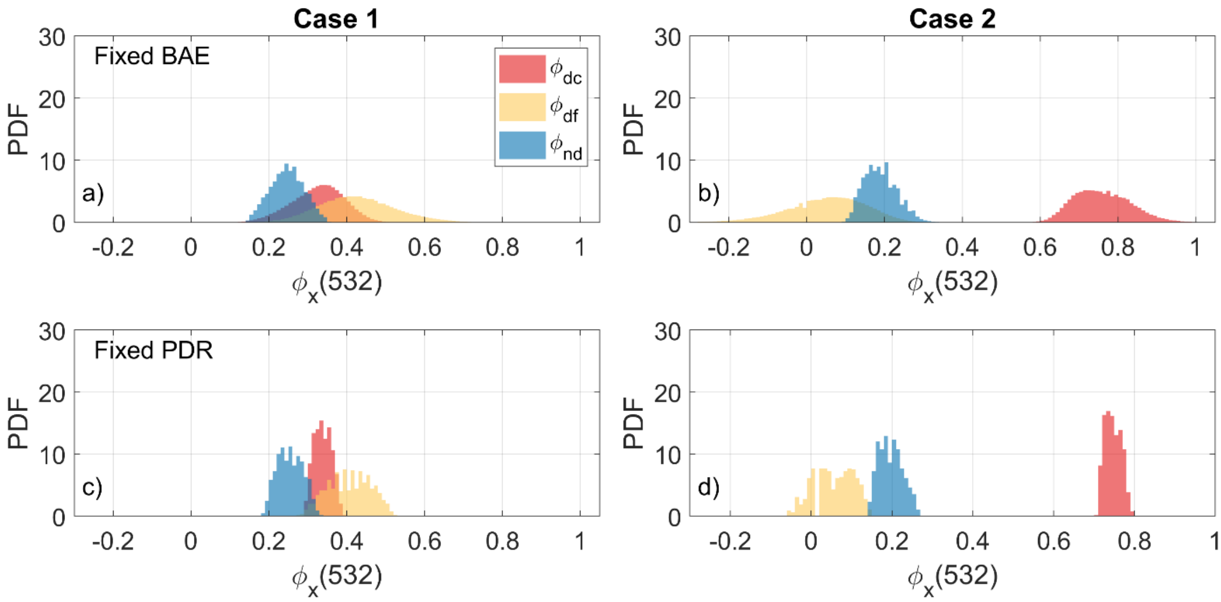


Similar to Figure 1d featuring three aerosol components.

To visualize further the effects, we have made a sensitivity study where one group of parameters is changed at a time from the ones found in Table 1. In Table 1 there are two groups, one that includes all the PDRs and a second one for all the BAEs. Therefore, we have performed two simulations. In each simulation, one group of parameters (e.g., the PDRs) was varied while the second group (e.g., the BAE) was kept at the fixed mean value as marked in Table 1 without accounting for its uncertainty.

Here we assume a 30% uncertainty on PDRs or BAEs as an example. The first simulation includes 77 different combinations where the PDR of coarse, fine and non-dust types was between 0.24 and 0.30 (0.34 and 0.39), 0.19 and 0.23 (0.14 and 0.19), and 0.03 and 0.07 (0.03 and 0.07) with 0.01 increment at 355 nm (532 nm) wavelength, and the fixed BAE coarse, fine and non-dust was -0.2, 1.5 and 2.0 between 355 and 532 nm, respectively. For the second simulation the BAE ranges at -0.2 ± 0.06, 1.5 ± 0.45 and 2.0 ± 0.6 for the coarse, fine and non-dust types, respectively.

For physical meaningfulness, we applied the two simulations to case 1 and case 2 without accounting for the uncertainty of the measured PDR. Cases 1 and 2 are located inside the observational space but at different distances from the edges. Visualizing the effect, we observe that varying the edges of the observational space (PDRs) induces higher uncertainty (wider distributions) than varying the BAEs for the same 30% uncertainty. Therefore, accurate knowledge of the parameters in Table 1 as well as adequate assumption of the aerosol mixture in the atmosphere is important. At the same time, the uncertainty of the measure PDR should be kept within acceptable levels.



Estimated uncertainties varying (top) PDRs or (bottom) BAEs of each aerosol type for cases 1 and 2.

Regarding the points deviating from Fig. 5, we would like to emphasize that different atmospheric studies often use different definitions for the geometrical boundaries of the aerosol layer. These differences, combined with the vertical smoothing applied to optical profiles, can influence the range of particle depolarization ratios reported. Most commonly, the optical properties are reported as mean values with a standard deviation. In Filioglou et al. (2020), the maximum particle depolarization ratios reported were 32% and 35% at 355 and 532 nm, respectively (see Table 1) while mean values amounted to 25 ± 2 and 31 ± 2, respectively. Then, recent laboratory studies focused on the chemical composition of dust particles and their optical effect reporting particle depolarization ratios at 355nm up to 32% depending on the dust mixture (Miffre et al. 2023). A thorough conversation of the selection of the optical properties in Table 1 is already included in the manuscript, and it is out of the scope of this manuscript to define the characteristics of fine and coarse aerosol particles. This manuscript serves as a demonstration of the methodology, and provide an easy-to-apply algorithm. The optical properties of individual aerosol type can be readily updated as more accurate values become available and new observations emerge in the field.

Filioglou, M., Giannakaki, E., Backman, J., Kesti, J., Hirsikko, A., Engelmann, R., O'Connor, E., Leskinen, J. T. T., Shang, X., Korhonen, H., Lihavainen, H., Romakkaniemi, S., and Komppula, M.: Optical and geometrical aerosol particle properties over the United Arab Emirates, Atmos. Chem. Phys., 20, 8909–8922, https://doi.org/10.5194/acp-20-8909-2020, 2020.

Miffre, A., Cholleton, D., Noël, C., and Rairoux, P.: Investigating the dependence of mineral dust depolarization on complex refractive index and size with a laboratory polarimeter at 180.0° lidar backscattering angle, Atmos. Meas. Tech., 16, 403–417, https://doi.org/10.5194/amt-16-403-2023, 2023.

Other comments:

The abstract of this manuscript needs to be improved. Lines 4 - 6: The authors state the advantages of the proposed method, but the logic may confuse readers. Specifically, "And it requires the proper knowledge of characteristic depolarization ratio and the backscatter-related Ångström exponent of each aerosol type" is a prerequisite for the method, not an advantage. Please rephrase these sentences to clarify the distinction between prerequisites and advantages.

Thank you for your comment. We have rewritten the abstract as below:

“Lidar-based algorithms for aerosol-type separation have the potential to improve air-quality assessments, estimates of aerosol direct and indirect radiative forcing, and the detailed characterization of their vertical distribution. In this study, we present an easy-to-apply algorithm that employs lidar-derived particle linear depolarization ratios measured at two wavelengths to separate up to three aerosol-type-specific particle backscatter fractions. These fractions are estimated under the assumptions that the depolarization ratios of each aerosol type in the mixture differ, and that both the depolarization ratios and the backscatter-related Ångström exponents at two wavelengths for each aerosol type are known. The mathematical relationship between particle linear depolarization ratios at two wavelengths for an aerosol mixture has been derived and expressed as a system of equations. These equations define the region of the observational space that can be meaningfully populated, with boundaries determined by the depolarization ratios and backscatter-related Ångström exponents of the pure aerosol types. Data collected in the Arabian Peninsula confirmed the predicted region of the observational space. The proposed algorithm is applied to synthetic dust mixtures as well as to atmospheric lidar observations of Arabian dust, Asian dust, Saharan dust and their mixtures, with the goal of decomposing coarse-mode dust, fine-mode dust, and low-depolarizing non-dust aerosols. We also discuss the impact of uncertainties in the prior optical properties of the pure aerosol types, along with the effects of observational uncertainties and biases. Overall, the method enhances the potential of dual-wavelength depolarization measurements for improving our understanding of the vertical distribution of coarse and fine dust”

Line 12: The claim that the method is “more accurate than the common use of the ratio of the particle linear depolarization ratios” requires statistical support. Please provide evidence or references to substantiate this statement.

Thank you for your comment.

In the literature, the ratio of particle linear depolarization ratios (PDRs) at different wavelengths is often used, and often referred as a constant value. In this study, we derived the mathematical relationship between PDRs at two wavelengths and demonstrated that this relationship is not linear. Hence, the ratio is not constant, but instead follows a curved dependence.

We have reformulated the abstract, the specific sentence in the abstract is now removed.

In Sect. 2.1, we have modified the text as:

“It is concluded that the relationship between δp at two wavelengths is not linear. Thus, the commonly used ratio of δp(λ1) and δp(λ2), which is typically assumed to be constant, is less accurate than the characteristic curved relationship proposed in this study.”

Line 35: The sentence is unclear and should be rephrased for better readability.

The sentence has been rephrased as suggested.

“The POLIPHON method has been applied to many lidar observations across the world, benefiting from the wide availability of these single-wavelength polarization lidars.”

Line 63: The description of Eqs. 1-2 is confusing. The statement, “the calculation involves the aerosol backscatter coefficient (βx) and aerosol-type-specific characteristic depolarization ratio (δx),” implies that βx is not aerosol-type-specific, but I understand βx is also aerosol-type-specific.

Thank you for your comment.

Initially, we intended to state that δx is specific to the aerosol type, while βx depends on the amount of that aerosol type present in the mixture and therefore does not have a fixed value (or characteristic value).

We have modified the sentence for improved clarity.

“In this context, each particle consists of a single aerosol type, and the calculation involves the aerosol backscatter coefficient (βx) and the particle depolarization ratio (δx), where the index x corresponds to each aerosol type (a, or b, or c), or to the mixture (p).”

Line 74: The "Methodology" section requires improvement. The authors list several equations (Eqs. 1-4) but do not systematically introduce the proposed method or explain how these parameters are used to develop the algorithm. The statement, "To apply the novel algorithm for the decomposition of two or three aerosol components," is premature, as the algorithm has not yet been clearly defined. Readers must read the entire manuscript to understand how the parameters are used for aerosol separation. The authors should explicitly introduce the algorithm before discussing its application.

Thank you for your suggestion.

We have revised the structure of Sect. 2 and relocated some paragraphs and sentences in Sect. 2. We have added a new subsection (Sect. 2.3: Synthetic aerosol mixture), which mainly incorporates content previously included in Sects 2.1 and 2.2.

At the beginning of the methodology section, we have added the following introductory paragraph:

"The basic concept is to use lidar-derived particle linear depolarization ratios obtained from a multi-wavelength lidar (or two separate instruments), together with two key optical properties (namely, the particle depolarization ratios and the backscatter-related Ångström exponents for pure aerosol types), to separate the aerosol mixture into its individual aerosol types. In this section, we introduce the algorithm and the corresponding set of equations for decomposing mixtures of two or three aerosol types (Sects. 2.1-2.2). The algorithm is then applied to synthetic aerosol mixtures in Sect. 2.3, followed by a comprehensive sensitivity and uncertainty analysis in Sect. 2.4."

We hope that these modifications have made the methodology section clearer to the reader.

Line 119: The statement, "Assuming the same lidar ratios at 355 and 532 nm, these values can be used for the $Å_{\beta}(355,532)$," requires clarification. Please briefly introduce the lidar ratio, and also provide references supporting the assumption that lidar ratios are the same (or very close) at 355 nm and 532 nm.

Thank you for your comment. We added the following text to the manuscript:

"The lidar ratio, defined as the extinction-to-backscatter ratio, has been widely used in lidar-based aerosol classification algorithms because it provides information on aerosol type. Numerous lidar studies have investigated the spectral dependence of the lidar ratio for different aerosol types (e.g., Haarig et al., 2025). For instance, Floutsi et al., (2023) present a comprehensive collection of depolarization ratios, lidar ratios, and Ångström exponents for various aerosol types and mixtures based on ground-based lidar observations. For most aerosol types, including dust from most regions except Central Asia, the assumption of lidar ratio equality between 355 and 532 nm is generally valid within observational uncertainties. However, for smoke mixtures, this assumption should be applied with caution."

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., and 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, Atmos. Meas. Tech., 16, 2353–2379, https://doi.org/10.5194/amt-16-2353-2023, 2023.

Haarig, M., Engelmann, R., Baars, H., Gast, B., Althausen, D., and Ansmann, A.: Discussion of the spectral slope of the lidar ratio between 355 and 1064 nm from multiwavelength Raman lidar observations, Atmos. Chem. Phys., 25, 7741–7763, https://doi.org/10.5194/acp-25-7741-2025, 2025.

Minor comments:

Line 38: Specify “the 532 nm and 355 nm wavelengths” by adding, for example, “of lidar instruments.”

Thank you for your suggestion. We have rephrased the sentence to:

“Most commonly, the 532 nm and 355 nm wavelengths have been used to perform lidar-derived depolarization ratio measurements.”

Line 46: Replace “at 532 or 355 nm” with “at 532 and 355 nm”.

Replaced according to reviewer’s suggestion.

Line 207: “idea” -> “ideal”?

Corrected according to reviewer’s suggestion.