Responses to suggestions of Franco Marenco

Dear Franco Marenco,

Thank you for reading the manuscript and providing very useful comments and suggestions to improve the paper. The replies to your 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.

The article by Xiaoxia Shang and co-authors describes a new algorithm that allows combining several optical parameters observed by lidar at two wavelengths, to decompose an external mixture of three aerosol types, quantitavely. This new method builds on pre-existing methodologies using a single wavelength to separately quantify two aerosol types. The method assumes prior knowledge of a number of intensive optical properties: the depolarisation ratio of the three pure aerosols at each wavelength, as well as their backscatter Angstrom exponent (Table 1). A system of linear and quadratic equations is derived, which can be resolved in terms of the backscatter fraction of each of the three aerosol components. The equations define the region of the observational space that can be meaningfully populated, and the observational data do confirm the accuracy of this prediction (Figure 5). The method is successfully demonstrated for a number of aerosol mixtures, observed with differing lidar systems, in the context of dust aerosols from the Arabian Peninsula, Asia and the Sahara, mixed with other aerosols. A succinct error analysis is also included.

I definitely think that the method proposed is promising and that this article is worth publishing. I however also feel that the paper can be substantially improved with a little more in-depth analysis of some points: better mathematical analysis of the system of equations, better quantification of the observational bias and uncertainty requirements, better use of the data collected for an update of the assumptions of Table 1, better highlight of a side scientific result as explained below, and stronger conclusions. The abstract needs moreover more work to make it self-understandable. With a little additional work, I believe that the paper will become a highly-cited reference paper for a wide number of applications.

MAJOR POINTS:

1. The second half of the abstract is a little hard to follow. My suggestion is as follows (from line 6): "The mathematical relationship between particle linear depolarization ratios at two wavelengths for a mixture of aerosol components has been derived and expressed as a system of equations. The equations define the region of the observational space that can be meaningfully populated and its boundaries: the latter are determined by the depolarization ratios of the pure aerosol components, and their backscatter-related Ångström exponents. Data collected in the Arabian Peninsula confirmed the predicted region of the observational space, and resolving the system of equations allowed us to quantify the contribution of each aerosol component. The novel algorithm has been applied to synthetic dust mixtures and to actual lidar observations of Arabian dust, Asian dust, and Saharan dust, so as to decompose coarse-mode dust, fine-mode dust, and low-depolarising non-dust. The impact of uncertainties in the prior optical properties of the pure-aerosol components are discussed, together with impact of observational uncertainties and biases. The method that we propose offers a promising role of dual-wavelength depolarisation measurements for the understanding the vertical distribution of fine and coarse dust."

Thank you for your comment and suggestion. We have modified the abstract as follows:

"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."

2. I have not done a full analysis of the system of equations proposed by the authors, but I am not persuaded by their statement that there is always a unique solution (lines 163 and 184). What I see is a system of 6 equations and 4 unknowns (two aerosols) or 7 equations and 6 unknowns (three aerosols). One equation (sum of phis for lambda1) has been omitted and should also be included. Moreover, equation 10 is not independent of equations 5-9, so I don't see how it can reduce the number of constrains. In other words, I think that a full mathematical analysis of the existence and unicity of the solution should be added in the paper. It is possible that the statement that "there is always a unique solution" will be found to be correct, but in the current version this is not explained or demonstrated.

Thank you for the comment, we have revised Sect. 2 to provide a clearer and more comprehensive explanation of the methodology.

For a mixture of two aerosol types, two equations at a single wavelength are sufficient for the decomposition (2 eqs for 2 unknowns), which has been used in many studies. We present this approach in an alternative form, via a system of equations at two wavelengths, specifically to derive the mathematical relationship between lidar-derived particle depolarization ratios at two wavelengths. We have revised the text to make this point clearer in the revised version.

For a mixture of three aerosol types, a system of 7 equations is used (eqs.14-20 in the revised version). Among them, there are only 6 independent equations, because any one of eqs.17-20 can be derived from the other three (4 equations, but only 3 independent ones). This point has been clarified in the revised version.

3. The text on the observational uncertainties (lines 223-231) can definitely benefit from an expanded discussion aiming at quantifying observational requirements. Readers will need to know what observational accuracy they need to achieve so as to be able to apply the method proposed by the authors. I would start from what are the average and best measurement uncertainties on delta_p in the literature and finding how they impact the retrieval of the three components. I would then suggest to exploit the equations developed in the article to assess and quantify what are the maximum bias and uncertainty that can be tolerated for depolarisation measurements, when applying the method (this could be more useful than the generic statement that a "small uncertainty" is necessary (line 331).

Thank you for the comment.

We have conducted 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.

Descriptions about the updated sensitivity analysis on observational uncertainties, as well as discussions on the bias and uncertainty for the depolarization measurements have been added in Sect. 2.4. The results of the sensitivity study depend on the initial values of the measurable particle linear depolarization ratios (PDRs), and vary from case to case. Thus, we present the sensitivity study based on selected cases. We have also added such observational requirements in the conclusion.

4. When the authors encounter conditions that challenge the assumptions from Table 1, such as on lines 245-246 or line 282, I suggest to try sing the values suggested by the data themselves as a-priori, to see if there can be an improvement. I would not agree that the algorithm becomes unsuitable (line 282): it is more a case of improving the inputs, and the data collected do contain the information to do this. Also, the generic statement about the effect of averaging (lines 246-248) could be substantiated with verifying how things really are with and without the averaging (either using the original measured data, or if these are not available anymore, with a simulation).

Thank you for the suggestion. We have modified the text for clarity, and added an experimental region in Fig. 7 as well as the descriptions:

"Alternatively, the measurement data can serve as a prior information for determining particle depolarization ratios of each aerosol type. For instance, to include as many measured pairs as possible inside the characteristic region, we can assume $\delta_{dc}(355)$ and $\delta_{df}(355)$ as 0.31 and 0.23, respectively. At the same time, the curvature of the curves needs to be increased by raising $A_{\beta,nd}(355, 532)$ to 2.5. The resulting characteristic region is indicated by gray dotted lines in Fig. 7. This adjustment appears to yield an improvement; however, it represents only a preliminary attempt and was not adopted in the subsequent analyses."
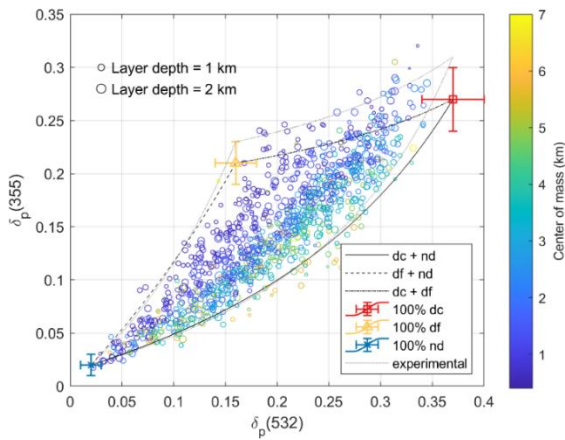
For line 282, we have changed text for clarity:

"Under current assumptions, those δp pairs locate outside the characteristic region, which would lead to some un-physical results (below zero or above one as the fraction). Therefore, the input parameters in Table 1 must be updated."

In the discussion about the effect of averaging (lines 246-248), we refer to the values reported in the literature, which were used to derive the input parameters presented in Table 1.

We made the correction for clarity:

"It may also be that the commonly reported pure values in the literature, based on field measurements, are often layer-mean values derived from smoothed optical parameters, which may underestimate the true

characteristic particle depolarization ratios of dust particles. These values were used as inputs for Table 1 in our calculations."

Thank you for the comment. We have added discussion:

"At higher altitudes, the δp pairs tend to lie closer to the characteristic curve of dc & nd, whereas layers at lower altitudes are located nearer to the characteristic curve of df & nd. This pattern suggests that dust with a higher altitude is generally coarser (mixed with non-dust particles), while dust with a lower altitude is finer."

And in the conclusion, we have added:

"Results of lidar observations from the Arabian Peninsula indicate that dust at higher altitudes tends to be coarser, whereas dust at lower altitudes is generally finer. This finding is consistent with the results of O'Sullivan et al., (2020), which report that operational models often place dust layers too low and underestimate coarse mode dust while overestimating fine mode dust. Height-resolved distributions of coarse and fine dust, retrieved using the proposed approach, can therefore serve as an important constraint for evaluating and improving model performance, offering new perspectives for a better understanding of dust transport mechanisms."

Thank you for your comments and suggestions. We have revised and strengthened the conclusion.

Thank you very much for your detailed suggestions and corrections, we have revised the manuscript accordingly.