Answer to comment of Reviewer #1

on "Retrieving volcanic, desert dust, and sea-salt particle properties from two/three-component particle mixtures after long-range transport using UV-VIS polarization Lidar and T-matrix" by G. David et al., *ACPD **13**, 1891-1947, (2013).*

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We thank Reviewer #1 for his review, which adds value to our manuscript. We addressed his comments, as detailed below. To ease the reading, each referee comment is first recalled in italics, then the corresponding authors reply is given. The induced modifications can be followed with the revision tool in the revised version, given as a supplementary material (see also text highlighted in grey color).

General comments

Combining the observations of a UV-VIS polarization LIDAR and T-matrix numerical simulations of different categories of atmospheric aerosols, the authors of this paper show that it is possible to discriminate the optical properties of the single category of particles in the cases of a (vertically layered) mixture of two or three (aerosol) components, when sampling the air masses apart from the sources of the aerosols. The proposed methodology is convincing and, in my opinion, could arise wide interest in the related atmospheric community. The introduction and the referencing to the current literature are well done. The relevant points, addressed by the authors, are well stated, and the conclusions can have a robust background if few points/assumptions are clarified (see Specific comments). The study deserves publication in ACP after considering other minor revisions, mainly: the organization should be improved, as well as the presentation of the analysis:

Authors reply

We agree with Reviewer #1 and improved the organization of our manuscript. To improve its readability, following Reviewer #2 (who also asked for an improved organization), we have modified the outline of our manuscript, while maintaining the same physical content. Here is the new outline: Section 2: UV-VIS polarization lidar remote sensing experiment, Section 3: Simulation of optical properties of nonspherical particles, Section 4: Methodology, Section 5: Application to volcanic ash, desert dust and sea-salt particle mixing.

General comment 1

Paragraph 1, Introduction. It is well done, but it is also quite long. Please consider to insert bulleted lists to summarize part of the discussion, and to improve the readability.

Authors reply on general comment 1

We agree with Reviewer #1 and shortened the introduction (the revised introduction is 25 lines shorter). Following your recommendation, we inserted a bulleted list in the introduction to present the two and the three-component particles mixtures and to give the corresponding examples. To ensure that the bullets effectively brings additional order and structure to the introduction, we focused the bullet list on one aim only (to give examples of different mixtures), leaving the literature review for the next paragraph, to ease the reading. To improve the readability, we also shortened the description of Section 2, which was too long, at the end of the introduction where the organization of the paper is described. Each of these modifications can be seen in the revised version of the manuscript (see text highlighted in grey).

General comment 2

Paragraph 2, Methodology. Too many sub-sub sections, they could be reduced if part of the theoretical expressions concerning the light scattering by atmospheric particles are skipped, most of the relevant references are already cited. In this way, the needed formalism for the depolarization of 2- and 3-component particle mixture results more enlightened.

Authors reply on general comment 2

We agree with Reviewer #1. The new Section 4 "Methodology" (old Section 2) has no sub-subsection left, as we chose to recall the theoretical expressions concerning light scattering by atmospheric particles in an Appendix on "Theoretical considerations on light scattering by atmospheric particles". This considerably shortened the main text to a standard 30-pages manuscript, hence enlightening the needed formalism for the depolarization of 2 and 3- component particle mixtures. To improve the readability, we also deleted old Equation (6) by describing its content in the text, while using the sign $\boldsymbol{\pi} = \{//, \perp\}$ allowed merging old Equations (4) and (5). The appendix has hence five equations left (instead of seven equations for old sub-subsection 2.2). In this way, the new Sections 4.1 (on two-component particle mixtures) and Section 4.2 (on three-component particle mixtures) corresponding to old subsections 2.3 and 2.4 are identically organized with an introduction and then a development on separate retrieval of particle backscattering. This highlights the proposed new methodology and we thank Reviewer #1 for this improvement. In the proposed revised version of the manuscript, the equations numbers have been changed to adapt the manuscript to the removal of two equations.

General comment 3

Paragraph 5. The sub-paragraphs 5.2.1, 5.2.2 and 5.2.3 are redundant, please remove and leave the corresponding texts in paragraph 5.

Authors reply on general comment 3

Thank you for your comment which helps reading the manuscript. In the revised manuscript (text highlighted in grey), we have removed sub-paragraphs 5.2.1, 5.2.2 and 5.2.3 and left the corresponding texts in paragraph 5. Hence, subsections 5.1 and 5.2 are now identically organized, which helps improving the reading of the manuscript.

Specific comments

Specific comment 1

Page 1899, rows 14-16 why "[: : :] when mixing sea-salt and desert dust particles, no internal mixture, [: : :] is assumed"?

Authors reply on specific comment 1

With the model of spheroids used here, the only way to treat internal mixtures is to apply a mixing formula to derive an effective refractive index for the internally mixed constituents and use that in the light-scattering computations. In case of mineral dust, the mixing would occur by dust particles gaining a layer of other substances (e.g. salts) at the surface, and the mixing formulas do not apply well to such situations. Rather than treating the internal mixing badly, we prefer to restrict our treatment here to external mixtures only, and leave the case of an internal mixture for a separate study where it can be done properly. We have, however, rephrased the sentence as follows: "Internal mixtures of dust with other substances are not considered, because they could not be accurately treated using the light-scattering method adapted here (see new Section 3.2), and because they are not critical to the purpose of introducing our new lidar methodology."

Specific comment 2

Page 1904, row 23: explain better because it is possible to assume that there is no interaction between s- and ns-particles.

Author reply on specific comment 2

In this paper, no interaction between s and ns-particles is assumed since the polarization Lidar experiment very efficiently partitions the particles mixture in its s and ns-component. As detailed in Section 3 (page 1911, Line 10), "polarization cross-talks, ensuring efficient s and ns-particles separation, are fully negligible with 10^{-7} accuracy". We hence improved the sentence written in the new Section 4 as follows: "A tracer for ns-particles can be derived from the Lidar formalism by applying the superposition principle

to particle backscattering coefficient β_p, i.e. $\beta_p = \beta_s + \beta_{ns}$, assuming no interaction between s and ns-particles (i.e. no polarization cross-talks between polarization channels, as developed in section 2.1)".

Specific comment 3

Paragraph 4.3: The ns-particle size distributions (PSDs) have to be assumed, choosing different PSDs how does reflect on the calculation of backscatter coefficient and depolarization? Should this be considered in the estimation of the (systematic) indetermination affecting the retrieved parameters for the different classes of aerosol with your methodology ?

Authors reply on specific comment 3

We thank the referee for his remark, because it is clearly an important point of the proposed methodology. We however think that we tackled these issues in the manuscript when computing the δ_{ns}-ratios and β_{ns}-coefficients (page 1917, lines 1-17), before answering to these questions in the discussion (pages 1922-1923) and in the conclusion (page 1925, lines 18-23).

More precisely,

− The chosen PSD does not influence the particles vertical layering, which is still retraced, since the effect of a different δ_{ns}-value (possibly due to another PSD-choice), "*is to shift the corresponding β_{ns}-profile*" as stated in our manuscript at page 1922, line 26.

− Choosing different PSD's may however influence the cross-polarized particle backscattering coefficients (page 1917, lines 15-17): "*with this choice of PSD's, the $\beta_{ns,\perp}$-coefficient is higher in the UV spectral range than in the VIS-spectral range for dust particles, while an opposite behavior is observed for sea-salt particles*". Choosing a different PSD may modify this result and, as a consequence, the numerical values of δ_{ns} and $\dot{A}_{ns,\perp}$(UV, VIS) for dust and ss-particles, and in turn, the retrieved particles backscattering coefficients.

− As a conclusion, indeed, indetermination affecting the retrieved parameters are evaluated and discussed (page 1922, lines 23-25 ; page 1923, lines 11-15) where "*we discuss the possible influence of computed numerical values of δ_{ns} and $\dot{A}_{ns,\perp}$(UV, VIS) for dust and ss-particles on the retrieval results."* Moreover, *"to test the robustness of our new methodology, we used exaggerated δ_{ss} and $\dot{A}_{ns,\perp}$ (UV, VIS) values*". It follows that "*the $\dot{A}_{dust,\perp}$(UV, VIS) and $\dot{A}_{ss,\perp}$(UV, VIS) values can be considered as convergence criteria in our algorithm, since for very different values of $\dot{A}_{dust,\perp}$(UV, VIS) and $\dot{A}_{ss,\perp}$(UV, VIS), negative particle backscattering coefficients were retrieved*". Hence, and as written in the conclusion of our manuscript, "*to apply this new methodology, care should be taken with the ns-PSD assumed in the numerical computations, which must be ns-particle specific and representative of these ns-particles after long-range transport*". We hence chose to deal with Munoz el al. 's PSD (2004) for volcanic ashes, Mallet et al.'s PSD (2004) for desert dust particles, and with Shettle et al. 's PSD (1979) for sea-salt particles, so as to be ns-particles specific.

Specific comment 4

Figures 3, 5 and 10. Some human eyes cannot distinguish between green and blue; although efficiently recalling the LIDAR wavelengths, please consider to use different colour or line style (thicker) for better viewing.

Authors reply on specific comment 4

Thank you for the remark. We chose to use thicker lines to improve the reading of Figures 3 and 5, since we prefer to keep on recalling the LIDAR wavelengths by colours. For Figure 10, we also increased the thickness of the lines, and to ensure that the reader can easily distinguish the different particle contributions, the symbols used in the figure have been enlarged and specified in the figure caption. Hence, in the caption of figure 10, indications for identifying the different particles curves have been added: sea-salt particles (squares), dust particles (triangles), water-soluble particles (spheres).

Technical corrections

Page 1899, rows 16-17: Please rephrase. (page 1924, row 11)

Please use "paper" instead of "contribution"

Author reply on technical corrections
Thank you for the remark. The correction has been done.