Answer to comment of Reviewer #2

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 #2 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 before the authors reply be given. Due to the requested reorganization of the paper, in the revised version of the manuscript (given as a supplementary material), the moved parts of the manuscript are highlighted in yellow. When a whole paragraph has been moved, the title of the subsection is highlighted. In addition to this color marked change, the revision tool has been used for text modifications.

General comment

The paper is of great interest to the atmospheric aerosol community, however a coherent readability is very difficult to carry out, and hence the principal propose of the paper seems to be vanished. The current distribution of the sections of the paper should be improved. As a suggestion for a more clarity in the general reading, Sect. 3 (Lidar instrumentation) and a modified Sect. 4 with the numerical simulations required should be after Sect. 1 (Introduction). Next, Sect. 2 should be modified and re-written accordingly to present the description of the methodology used in the paper. Finally, a section with the main results, including the two case studies as an application of the methodology previously described should be introduced, followed by the Conclusion section.

A revision of the paper should be addressed (see Specific comments below) before it is accepted for publication in ACP. In general, the words "sensitive and accurate" to describe 2lambda-polarization Lidar measurements and "accurate" for numerical simulations appear as a redundancy along the overall paper. Please, modify/remove them accordingly in order to reduce that redundancy and hence improve the reading, as well. Tables and figures, in general, must be improved with larger fonts and numbers and/or by enlarging the size of figures instead (see Technical corrections at the end).

Authors reply to general comment

We agree, have adopted the proposed new structure for the manuscript in the revised version. To fully respect Reviewer #2's comments, we also removed the words "sensitive and accurate" at several times and used larger fonts and numbers in the tables, while increasing the size of the figures.

Specific comment 1 (on the introduction)

Sect. 1. Introduction. It is a bit long. Please reduce it, and include the corresponding reference instead. Page 1897, rows 9-29: After the revision, the structure of sections can be modified. Please, consider the corresponding modifications and renumbering of sections in this part.

Authors reply to specific comment 1

We reduced the introduction and included the corresponding references. The revised introduction is 25 lines shorter. To agree with the corresponding modifications and renumbering of sections, the structure of sections has also been modified.

Specific comment 2 (on our methodology and the F-matrix formalism)

Page 1899, rows 21-22: Please, rephrase, that sentence is confusing.

- The sentence was not necessary, so we decided to delete it altogether.

Page 1899, rows 23-26, and page 1900, rows 1-21: Sect. 2.2.1 must be revised in order to understand the contribution of the T-matrix formalism to that study. Provide more information and details on T-matrix formalism; the meaning of the F-Matrix is missing, add a short interpretation of F-elements; include more references. See the general comments on the new re-distribution of the sections.

- The T-matrix formalism contributes to old Section 2.2.1 (dedicated to the F-matrix formalism) since F-matrix and T-matrix are, in a way, related: they both relate properties of incident and scattered waves, but they are quite different in practice, because the F-matrix relates the waves described by the Stokes parameters, whereas the T-matrix relates waves expressed as spherical function expansions. The T-matrix formalism is just used internally in our methodology to compute the optical properties of nonspherical particles (new Section 3). We have revised the manuscript to add this information (see text highlighted in yellow in the new appendix).

- To provide more information and details on T-matrix, we have modified the manuscript as follows (text highlighted in yellow, in the new Section 3): *"We carried out numerical computations based on the T-matrix method to simulate the single-scattering properties of volcanic ash, desert dust and sea-salt particles for different particle sizes at the wavelengths of interest. Of particular interest were the scattering matrix [F]ns and the scattering and extinction cross sections Csca,ns and Cext,ns, respectively, from which the Lidar observables can be derived.".*

- The meaning of the F-matrix is extensively described in M.I. Mishchenko et al.'s textbook (2002) to which we now refer to in Appendix A. It is also given in old Equation (1) (new Equation (A-1) of Appendix A) which describes the scattering process experienced by an ensemble of particles, whatever their size, shape and chemical composition, by taking into account the polarization state of the light through Stokes parameters. In the old Equation (1), the given F-matrix is the scattering phase matrix, which is non-additive, as stated in the manuscript (page 1903, line 20). Hence, the F-matrices for different particles cannot simply be added up and must be made additive by multiplying by their scattering cross-sections. To include your comment, we modified the manuscript (text highlighted in yellow in Appendix A).

Page 1901, rows 6-8, and 17: In relation with the previous comment, what are F11 and F22 in the matrix formalism? Revise Eqs. (3a) and (3b).

- As stated by M.I. Mishchenko's textbook (2002), $F_{11,p}$ is the scattering phase function, which describes the angular distribution of scattered intensity for an incident unpolarized light. In case the incident light is polarized, the angular distribution of scattered intensity will also depend on other scattering matrix elements, depending on how the radiation is polarized. The quantity $(1 - F_{22,p}/F_{11,p})/ (1 + F_{22,p}/F_{11,p})$ is called the particles linear depolarization ratio. In our manuscript, the subscript p is used to refer to the particle, which can be either spherical (subscript s) or nonspherical (subscript ns). Hence, spherical particles, for which $F_{11,s} = F_{22,s}$, do not depolarize laser light. Also, we checked that Equations (3a) and (3b) were correctly written.

Page 1916, row 2: Please, see my previous comment on the meaning of F-matrix and its elements.

- This point has been addressed in the above comment. To include your comment, in the revised manuscript, we added that the computation of $F_{11,ns}$ and $F_{22,ns}$ has been achieved by using T-matrix numerical code (see text highlighted in yellow).

Specific comment 3 (on the UV-VIS polarization lidar remote sensing experiment)

Page 1901, row 21: Please, replace "...volume backscattering coefficient ..." by "... volume particle backscattering coefficient ..."

- The modification has been done (revision tool highlighted in yellow).

Page 1902, rows 6-7: The term “Lidar ratio” is widely used to define the extinction-to-backscattering coefficients ratio in the Lidar community. Please, introduce that term instead, for the first time, and then use “Lidar ratio” to refer to the variable Sp in the rest of the paper.

- Fixed as suggested. See text highlighted in yellow in the revised version.

Page 1902, rows 9-16: Please, revise the definition of the particle linear depolarization ratio deltap; that Eq. (7), the first equality, is not a definition for deltap, since the molecular contribution is also considered in the relation between both measured intensities Isca,perp. and Isca,paral. (also see Sect. 3). This relation is usually used indeed in the Lidar community, identified as the volume linear depolarization ratio, and denoted for example as DV. For a link between these two depolarization parameters, see the reference: Cairo, F., Di Donfrancesco, G., Adriani, A., Pulvirenti, L., and Fierli, F.: Comparison of various depolarization parameters measured by lidar, Appl. Optics, 38, 4425–4432, 1999. How is this modification affecting to the procedure shown next?

- The old Section 2.2 (or equivalently the new Appendix A) is dedicated to particles only, as denoted by the use of subscript p in the Equation (1) of our manuscript. Hence, in Equation (7), the quantities $I_{sca,\perp}$ and $I_{sca,//}$ refer to particles only. The depolarization ratio δ_m of atmospheric molecules (subscript m) is taken into account in Equation (18) (page 1912, line 7), when distinguishing the volume linear depolarization ratio δ from its molecular (δ_m) or particles (δ_p) contributions. For the sake of clarity, in the revised manuscript, we added reference F. Cairo et al. (1999), as a reference paper for the definition of the depolarization ratio and of its molecular and particles contributions (see text highlighted in yellow in new Section 2.2).

Page 1908, rows 19-21: Sect. 3 is called, hence this is another reason to move it before Sect. 2. In this sense, replace, please, “... at wavelength lambda ...” by “at the two Lidar wavelengths, lambda1-UV and lambda2-VIS”.

- We agree with the proposed rearrangement of the sections and adopted it in the revised manuscript. However, we do not want to indicate UV or VIS in the wavelength symbols because the methodology is generic and might be applied to different spectral bands later (VIS and IR for example).

Page 1912, rows 5-7: Please, use the correct term “parallel backscattering ratio” instead of “parallel Lidar R-ratio” to define that expression. That can be confused with the term “Lidar ratio” (extinction-to-backscattering ratio), also used in the Lidar community. In addition, please, provide a reference for the Eq. (18), unless that is Alvarez et al. (2006), but in this case, it should be also included at that point.

- Changed as suggested. Equation (18) originates from Winker and Osborn (1992), which is now also quoted in the revised version for this equation. We noticed that Equation (21) was similar to Equation (18) and hence removed Equation (21). We thank the referee for this improvement.

Page 1912, row 10: Introduce the term “Lidar ratio”.

- Done.

Page 1912, row 16: Use the term “parallel backscattering ratio” instead of “parallel Lidar ratio”, which is confusing.

- Changed as suggested.

Page 1913, rows 1-8: An explanation is required: as delta is used to obtain deltap, how is delta obtained/calculated then? I guess that you use the Lidar signals, but can you specify it please? See also my previous comment about Eq. (7).

- The depolarization ratio δ is indeed evaluated from our measured lidar signals, after accurate polarization calibration procedure (page 1911, line 25): "*The detector gain electro-optics calibration constant is hence known with better than 2 % uncertainty (David et al., 2012), allowing accurate measurement of the depolarization ratio δ at wavelength λ.*" As demonstrated by G. David et al. (2012), this δ-measurement is accurate provided that polarization cross-talks are fully negligible, which is the case in our experiment (with better than 10^{-7} accuracy). In the revised manuscript, we added supplementary information (see text highlighted in yellow).

Page 1913, row 8: Please, include also the integration time used for lidar measurements.

- The integration time is 7 minutes, which we now mention in the revised manuscript.

Page 1913, row 24: Replace, please, "Backscattering cross-sections" by "Backscattering coefficients".

- We thank the referee for his remark. We computed size-averaged backscattering cross-sections. To be clearer, we modified the text (see text highlighted in yellow) and the corresponding figure (old Figure 5, new Figure 4).

Page 1914, row 4: Consider the combination of Sects. 2.1 and 2.2 with this Sect 4.1 in the new re-distribution of the paper.

- In the revised manuscript, the old Section 2.1 (on nonspherical particle microphysics) has been inserted in the new Section 3 (on the computation of nonspherical particles optical properties).
- Also, the old Section 2.2, which is essentially textbook material (MI Mishchenko et al., 2002), has been moved to Appendix A, where it can be found if needed. We believe that this modification will improve the readability of the paper.

Specific comment 4 (on the computed optical properties of nonspherical particles)

Page 1917, rows 11-17: Why have you chosen the reference of Mallet et al. (2004), which is focused on black carbon, for the selected PSD of dust particles? By observing Fig.4, dust particles with radius higher than 1 micron seem to be absent. Please, provide a more extensive justification for that choice and evidence of that assumption. The selection of a given PSD is crucial in the methodology presented in the paper in order to interpret the results.

- As stated in our manuscript (page 1917, lines 4-7), *"in the absence of complementary measurements, the ns-PSD has been taken from the literature on atmospheric ns-particles after long-range transport, with the criteria of ensuring ns-particle specificity since our numerical simulations are built for that purpose*". To be dust particle specific, we hence chose reference to Mallet et al. (2004), who isolated the dust contribution from their experiments, performed on a mixture of mineral dust, black carbon and water-soluble species, using an eight-stage impactor and an ion chromatograph. Moreover, this PSD seems to be representative of long-range transport episodes occurring at Lyon: particles larger than 1 micrometer have been removed from the dust cloud by gravitational settling and, moreover, their measurements were performed in France, very close to Lyon. Other references are of course very welcomed, provided that they are dust particles specific and representative of long-range transport since, as shown by CL. Ryder et al. in ACP (2013), the ns-PSD changes with the distance from the source. In the revised manuscript, we added this supplementary information to clarify that point for dust particles, as we already did in our manuscript for volcanic ashes: "*For dust particles, we chose Mallet et al.'s PSD, who isolated the dust contribution by performing measurements after long-range transport, close to Lyon (France). We hence ensure dust particles specificity after long-range transport. To apply the methodology proposed in section 4, other literature references are of course possible, provided that the given PSD is representative of long-range transport and dust particles specific.*" The consequences of this PSD-choice have been discussed in our manuscript (page 1925, lines 20-23): it indeed affects the cross-polarized Angstrom exponent value and the "*$\dot{A}_{dust,\perp}$(UV, VIS) and $\dot{A}_{ss,\perp}$(UV, VIS) values were 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*". As stated in our manuscript (page 1925, line 21), "*to obtain accurate retrievals of dust, sea-salt and ws-particles backscattering coefficients, care should be taken with the choice of PSD when applying our new methodology*".

Page 1917, rows 24-28: Although authors stated that a forthcoming paper is expected, more explanation should be introduced after the sentence: "It follows that spheroids may have difficulties in predicting large particle depolarization ratios such as for volcanic ash correctly. Hence, for volcanic ash, we may use O. Muñoz's laboratory measurements deltaash = 40.5 %-value (2004), leaving more appropriate deltaash numerical simulations for a forthcoming paper.", or include corresponding references instead.

- To include your comment, the following explanation has been added to the revised manuscript (see text highlighted in yellow): *"It follows that spheroids may have difficulties in predicting large particle depolarization ratios correctly. A DDA-approach (Lindqvist, 2011) might be a fruitful complementary approach, but extensive DDA simulations are clearly beyond the scope of this paper. Hence, for volcanic ash, we used O. Muñoz's laboratory measurements δ_{ash} = 40.5 %-value (2004), leaving more appropriate δ_{ash}-numerical simulations for a forthcoming paper. Whatever the chosen δ_{ash}-value, the behavior of β_{ns} with altitude is still retraced, as shown in section 4.2 (see Equation (7)) and as discussed in section 5.2".*

Specific comment 5 (on section 5, application to volcanic ash, desert dust, sea-salt particle mixing)

Page 1919, row 17: Why do you use a fixed Lidar ratio of 55+/-5 sr? Provide, please, a reason of the choice of this value. It seems to be between those obtained for volcanic ashes particles from the simulations (see Table 3), but do you have an estimate of the Lidar ratio for sulphate particles?

- To agree with Table 3, at page 1919, row 17, we should have written S = 54 ± 5 sr in the UV spectral range. We hence did the correction. This fixed lidar ratio agrees with Ansmann et al. (2012), who derived constant lidar ratios, after deriving Raman vertical profiles during the same volcanic episode. We also added this literature reference in the revised manuscript (see text highlighted in yellow). In Ansmann et al. (2012), the sulphate lidar ratio is estimated to be 45 sr during the same volcanic ash episode. Using Gross et al. (2011) mixing rule for lidar ratios, we checked that the lidar ratio of the two-component particle mixture kept around 55 sr. Hence, to correct for the particles extinction is not a main concern in our retrieval methodology.

Page 1920, rows 19-20: I guess that the same methodology has been used in Miffre et al. (2011) but for the case of dust (ns-particles) and spherical particles. If that is, the expression "... case study 1 can be extended to a ..." has no sense. That study has been already published. Change, please, accordingly.

- Thank you for the remark. We changed it accordingly as follows: "*case study 1 can be applied to…*"

Pages 1921-1922: Please, revise these sections, which are repetitive making thus difficult the reading. As a suggestion, combine sub-sects. 5.2.1 and 5.2.2, followed by subsect. 5.2.3.

- We did the revisions (see text highlighted in yellow) and hence shortened Section 5.2. Thank you for this improvement of the readability of our manuscript.

Page 1921, rows 1-12: FLEXTRA back-trajectory analysis is performed for potential dust and sea-salt particles occurrence over the site, but what about the water-soluble particles presence? More discussion should be provided for this (spherical) type of particles to be assumed present in that three-component mixture. Are there any available complementary measurements to support the presence of each type of particles?

- In the absence of complementary chemical measurements (page 1896, line 29), we referred to literature references suitable for long-range transport of desert dust particles at Lyon. In the Mallet experiment (2004), performed in France very close to Lyon, dust particles were indeed mixed with sea-salt particles, but also with water-soluble species such as ammonium sulfates and nitrates. Also, our RH-profiles exhibit sufficiently high RH-values to assume that the background aerosol is composed of water-soluble species. A more precise partitioning (involving four or five-component particle mixtures to extract water-soluble specificity) is however clearly beyond the scope of this paper. Please note that, if having complementary measurements will be a nice refinement, it does not affect the main novelty of our contribution, which is relative to our proposed new methodology, based on combining 2λ-polarization lidar experiments with T-matrix numerical simulations for the ACP-Special Issue on Light Depolarization by atmospheric particles.

Page 1921, row 10: In the sentence "For altitudes above 2 km, the relative humidity (RH) was above 40 %, ...", the term "above" must be replaced by "below" by observing Fig. 8, and to have actually the conditions needed for sea-salt particles to be considered cubic-shaped, i.e., as ns-particles. Please, revise.

- Thank you for your remark. We did the correction.

Pages 1921-1922, Sect. 5.2.2: Please, provide more explanation in this section. For instance, deltap is obtained to be lower than deltanc, maybe because of presence of s-particles (e.g., those supposed water-soluble particles), which are lowering the particle depolarization ratio deltap? Hence, the values of deltap obtained (page 1921, rows 25-27) are below those shown in Table 3.

- We agree with the explanation proposed by Reviewer #2. To include his comment, we added this explanation in the revised manuscript (see text highlighted in yellow).

Page 1922, rows 4-10: The choice of the profiles on 18 October 2011 at 16:15 UTC and the RH and potential temperature conditions at that time should be explicitly correlated and examined in more detail, and that discussion consequently introduced.

- We agree with this remark and modified the manuscript as follows: "*At that time, as shown in figure 8, for altitudes above 2 kilometres, the Lyon troposphere is stable as depicted by the potential temperature, while the relative humidity is below the 40 % RH sea-salt crystallization point, allowing sea salt particles to depolarize.* " (see text highlighted in yellow).

Page 1922, row 7: Please, use "plume" instead of "cloud" to avoid confusion.

- Thank you for your remark. We did the correction.

Page 1924, row 5: Please, check the backscattering coefficients beta in opposite phase at around 3.5 km height in Fig. 10 (4th panel): betadust and betass instead of betadust and betaws.

- We agree that β_{dust} is in opposite phase with β_{ss} and did the correction. This may indicate that indeed, sea-salt particles influence dust particles, as pointed out by Zhang (2008), which we already quoted.

Specific comment 6 (on section 6, conclusions and outlook)

Page 1925, row 17: Again, the presence of water-soluble particles besides dust and sea-salt particles is just supposed, but that assumption must be still verified.

- This comment has been addressed in specific comment 5 (20 lines above).

Technical corrections

Technical corrections: Several technical corrections are listed below. Check them, please.
Page 1896, rows 5-6: Please, check the reference, there is a misprint.

- Thank you for your remark. We did the correction.

Page 1905, row 7: Eq. (9b) doesn't exist, that is Eq. (9).

- Thank you for the remark. We should have referred to Equation (10b) and did the correction (with the new numbering of equations due to the reorganization of the paper, it hence becomes Equation (6-b)).

Page 1908, rows 26-27: Replace "Eq. (15a,b)" by "Eq. (15a,c)", and "Eq. (15c,d)" by "Eq. (15b,d)".

- We did the correction.

Page 1912, row 23: Replace "... by using Eq. (19)" by "... by using Eqs. (18) and (19)".

- We should have spoken about Equation (18), and did the correction.

Page 1914, row 7: In "(Nousiainen (2009)", remove the first parenthesis.

- Fixed as suggested.

Page 1917, rows 22-23: Replace, please, the sentence "... the Fig. 3 tendency of deltans to be constant while increasing effective radius ..." by "... the tendency of deltans to be constant while increasing effective radius(see Fig. 3) ...".

- We did the correction.

Page 1919, row 11: Sect. 5.1's caption: use, please, "volcanic ashes" instead of "volcanic dust", unless it can be confusing regarding "desert dust".

- Thank you for your remark. We did the correction.

Page 1919, row 13: Please, check the figure number: there must be Fig. 6 instead of Fig. 7.

- We did the correction.

Page 1919, row 19: Please, check the figure number: there must be Fig. 7 instead of Fig. 8.

- Thank you for your remark. We did the correction.

Page 1935, Table 1: Larger fonts must be used. First image (volcanic ash) must show the size scale as others. Please, list literature references in chronological order.

- We did the proposed corrections.

Page 1936, Table 2: Reorder the columns to put the complex refractive index m together to its literature reference. Include the symbol used for the size parameter range and the aspect ratio (and its step) in the caption.

- Thank you for your remark. We did the corrections.

Page 1937, Table 3: Values of depolarization ratios (UV, VIS) shown for desert dust and sea-salt are correct? They seem to be surprisingly very low values (rather lower than the molecular ones), are they in percentage or not? Please, check them.

- Many thanks for your remark: we made a confusion between percentage values and absolute values and corrected it.

Page 1939, Fig. 2: Please, enlarge the texts inside the figure.

- We did the correction.

Page 1940, Fig. 3: It is difficult to distinguish numbers and texts. Please, increase the size of numbers and fonts.

- Done.

Page 1941, Fig. 4: Please consider the following modified caption for Fig. 4: "Fig. 4. Selected ns-particle size distributions (PSD) as introduced in numerical calculations for volcanic ash (Muñoz et al., 2004), desert dust (Mallet et al., 2004) and sea-salt particles (ss, O'Dowd et al., 1997)."

- Fixed as suggested.

Page 1942, Fig. 5: Increase the size of numbers and fonts. Why are there two curves, green and blue lines, in the four lower figures (bottom panels) corresponding to ss-particles?

- We increased the size of numbers and fonts. In the four panels corresponding to ss-particles, two curves are represented, one for the m-refractive index in the UV, one for the m-refractive index in the VIS spectral range. We have modified the caption of the figure (old Figure 5, new Figure 4) to clear that point.

Page 1943, Fig. 6: Increase the size of numbers, symbols and fonts as possible.

- We did this improvement.

Page 1945, Fig. 8: Increase the size of numbers, symbols and fonts as possible. Provide larger figures, otherwise.

- We did the correction.

Page 1946, Fig. 9: Increase the size of numbers and fonts as possible. For instance, the readability of colour scales is almost impossible.

- We did the correction.

Page 1947, Fig. 10: Increase the size of numbers, symbols and fonts as possible. Provide larger figures, otherwise. Consider the following modification in the caption: ": : : and fraction of ns-particles (dust, ss) and ws-particles (ws, spherical) in the three-component mixture : : :". In addition, symbols in the X-axis must be modified, for instance, betans, betas [Mm-1 sr-1] must appear in the 4th panel, and Xns, Xs [%] in the 5th panel, since the profiles shown in those panels are not only for ns-particles.

- We did this improvement.