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Atmospheric Chemistry and Physics Discussions

Interactive comment 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.

Anonymous Referee #2

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General comments:

This paper describes the methodology to discriminate the individual optical properties of each of those particle (spherical/no-spherical) components, usually supposed to be present in the atmosphere after long-range transport, of aerosol external mixtures.

The apparent goal of the study is to show as this methodology based on the combined used of Lidar (UV-VIS laser emissions and accurate polarization detection capabilities) measurements together with T-matrix numerical simulations can retrieve the particular

(height-resolved) backscattering coefficient and proportion for each type of particles in the overall ensemble, differentiating between spherical and no-spherical particles. In addition, two case studies corresponding to two- and three-component aerosol mixtures, respectively, are shown as examples of the application of this methodology.

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).

Specific comments:

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. Page 1899, rows 21-22: Please, rephrase, that sentence is confusing. Page 1899, rows 23-26, and page 1900, rows 1-21: Sect. 2.2.1 must be

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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. 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). Page 1901, row 21: Please, replace "... volume backscattering coefficient ..." by "... volume particle backscattering coefficient ...". 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. 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? 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". 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. Page 1912, row 10: Introduce the term "Lidar ratio". Page 1912, row 16: Use the term "parallel backscattering ratio" instead of "parallel Lidar ratio", which is confusing. Page 1913,

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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). Page 1913, row 8: Please, include also the integration time used for lidar measurements. Page 1913, row 24: Replace, please, “Backscattering cross-sections” by “Backscattering coefficients”. 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. Page 1916, row 2: Please, see my previous comment on the meaning of F-matrix and its elements. 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 microm 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. 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. 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? 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. 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 sub-sect. 5.2.3. Page 1921, rows 1-12: FLEXTRA back-trajectory analysis is performed

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

Technical corrections: Several technical corrections are listed below. Check them, please.

Page 1896, rows 5-6: Please, check the reference, there is a misprint. Page 1905, row 7: Eq. (9b) doesn't exist, that is Eq. (9). Page 1908, rows 26-27: Replace "Eq. (15a,b)" by "Eq. (15a,c)", and "Eq. (15c,d)" by "Eq. (15b,d)". Page 1912, row 23: Replace "... by using Eq. (19)" by "... by using Eqs. (18) and (19)". Page 1914, row 7: In "(Nousiainen (2009)", remove the first parenthesis. Page 1917, rows 22-23: Replace, please, the

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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) ...". 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". Page 1919, row 13: Please, check the figure number: there must be Fig. 6 instead of Fig. 7. Page 1919, row 19: Please, check the figure number: there must be Fig. 7 instead of Fig. 8. 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. 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. 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. Page 1939, Fig. 2: Please, enlarge the texts inside the figure. Page 1940, Fig. 3: It is difficult to distinguish numbers and texts. Please, increase the size of numbers and fonts. 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)." 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? Page 1943, Fig. 6: Increase the size of numbers, symbols and fonts as possible. Page 1945, Fig. 8: Increase the size of numbers, symbols and fonts as possible. Provide larger figures, otherwise. Page 1946, Fig. 9: Increase the size of numbers and fonts as possible. For instance, the readability of colour scales is almost impossible. 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,

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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.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 1891, 2013.