Answer to comment of M. Tesche

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).*

 We thank M. Tesche for the extensive comments on our ACPD-paper. Our point by point response to M. Tesche's questions/comments is given below. To ease the reading, each original comment, in italics, precedes our response.

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M. Tesche

"From this and recent papers, it seems like the group in Lyon is running a well-characterized depolarization lidar instrument. I am sure there will be nice measurements in the future. However, I don't think that this particular study should be published in ACP for the reasons given below».

Authors reply

As authors, we are of course not very pleased with this remark. However, the conclusion drawn by M. Tesche in this introductive part of his comment is opposite to the conclusions drawn by both anonymous reviewers (#1 and #2). In addition, M. Tesche's conclusion is clearly positive about the quality of the data provided by our instrument: "*To conclude, I believe that the polarization measurements with your instrument are indeed sensitive and accurate.*" This is of particular relevance, because our manuscript has been submitted for the ACP-Special Issue on Light Depolarization. Our opinion is hence that the evaluation has been biased by the complexity of the reading of our manuscript (that M. Tesche well-identified). This statement is justified by the fact that most of the questions that he raised were already addressed in our original manuscript, but we agree that the presentation was not as clear as it could. Some reasons for this un-clarity are already addressed in our comments to reviewers #1 and #2. The other comments require only minor revisions to address, what we did. We hope that our answers to this criticism, presented below, will be found useful and help to clarify the scientific contribution of our manuscript, by improving its reading. We hence propose modifications, taking into account both the anonymous referee comments and the additional criticism provided by M. Tesche and other readers.

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M. Tesche

"First of all, the manuscript is hard to read. It is badly structured, lengthily written, contains many redundant parts, and seems overly complicated."

Authors Reply

We agree that the manuscript could have been much clearer. The other referees also pointed this out and, accordingly, we have proposed steps to improve that in our response to Reviewer #2.

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M. Tesche

"In the speculative parts on the other hand, the authors fail to critically discuss their assumptions and findings or to relate their work to the literature. Furthermore, it is hard to estimate the applicability of the presented method, as I will try to explain later."

Authors Reply

− On the assumptions and findings: we agree that some assumptions and details of the methodology were not as clear as they could. We will address these below in each corresponding comment.

 − On the bibliography: as detailed below, most of the references proposed by M. Tesche are out of the scope that we fixed in the introduction. Also, our scientific contribution is not a review paper (we however provided 78 references in our ACPD-paper). Moreover, reviewer #1 estimated that "*The introduction and the referencing to the current literature are well done".*

− On the applicability of the presented method: we note that both anonymous Reviewers (#1 and #2) arrived at opposite conclusions: '*The proposed methodology could arise wide interest in the related atmospheric*

community" (Reviewer #1) and *"The paper is of great interest to the atmospheric aerosol community"* (Reviewer #2).

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M. Tesche

 "During the description of the methodology, the reader is often referred to the instrument, the measurements, and the numerical simulations that are described later in the text. Restructuring of the outline of the manuscript would improve readability significantly. I think of something like this: Instrument, Simulations, Methodology, Results".

Authors Reply

We agree. See our comments to Reviewer #2.

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M. Tesche

*"The review on previous literature on aerosol-type separation in the introduction should be more comprehensive (see the discussion in Tesche et al. (2009)). Also, recent papers on the application to the Eyjafjallajökull ash plume are missing, e.g., Marenco and Hogan (2011), Determining the contribution of volcanic ash and boundary layer aerosol in backscatter lidar returns: A three-component atmosphere approach, J. Geophys. Res., **116**, doi:10.1029/2010JD015415. Ansmann et al. (2011), Ash and fine-mode particle mass profiles from EARLINET AERONET observations over central Europe after the eruptions of the Eyjafjallajökull volcano in 2010, J. Geophys. Res., **116**, doi:10.1029/2010JD015567."*

Authors Reply

− In the introduction, M. Tesche et al. (2009) is already cited. We do not wish to repeat the discussion in our introduction, but rather refer to work where it can be found. Indeed, reviewers #1 and #2 found our current introduction already too long, so we have taken steps to shorten it.
− We have, however, added reference to Marenco and Hogan (2011) as a more recent work not discussed by Tesche et al. (2009) and because it is relevant for our application (see text highlighted in cyan color). Please note that the three-component mixture discussed by Marenco and Hogan includes atmospheric gas molecules as one component. Using Marenco's terminology, our new approach should be regarded as a four-component methodology.
− Reference to Ansmann et al., 2011 has not been added since we already quote A. Ansmann et al., 2012 'Profiling of fine and coarse particle mass…" as an update in which Ansmann et al., 2011 is quoted. We wish to point out that our manuscript, already including 78 references, is not a review.
− Concerning the references quoted by M. Tesche at the end of his comment: in our discussion paper (page 1896, lines 26-28), we stated that "*the coupling of Lidar with inelastic Raman (Muller et al., 2001) as well as with passive remote sensing (Dubovik et al., 2006; Boyouk et al., 2011) has not been considered*".

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M. Tesche

"Furthermore, it seems like the authors misunderstood the method presented in Tesche et al. (2009) or other studies of aerosol-type separation (see the introduction and later on page 1904, lines 8-23). The authors state that some maximum value in δ_p is used as a reference for δ_{ns}. This is an oversimplified statement. The methods usually refer to a value of δ_p that represents a pure case (no contribution of spherical particles) of the respective aerosol type, i.e., mineral dust, volcanic ash, marine aerosol, or biomassburning smoke. These values are obtained from measurements close to a source of this particular aerosol type. The advantage of this assumption is that the method refers to values that are actually observed in the atmosphere. For an aerosol plume of a mixture of the different aerosol types, it is assumed that the intensive optical properties of the respective aerosol types do not vary during transport. This might not be en-tirely true in reality. However, little is known about the aging of non-spherical particles during long-range transport and assuming that the intensive optical properties of the aerosol mixture are the result of a mixing of the pure aerosol types seems closer to the truth than the results of numerical simulations based on some randomly chosen microphysical particle properties."

Authors reply

− As stated in our manuscript (page 1904, line 9), the statement "the maximum value of δ_p is used as a reference for δ_{ns}" originates from A. Shimizu et al.'s work (2004), which we cited as a nice pioneering work.

− The subject of the intensive lidar ratio raises important discussions in the recent literature because several assumptions are made, which may interfere with accuracy. As you note, assuming constant intensive parameters "*may not be entirely true in reality*". Indeed, this assumption should not be made as reported in a very recent ACP-publication by C.L. Ryder et al. (2013) who showed that the particle size distribution is modified during transport, which, in turn, modifies the intensive particle depolarization ratio and the Angström exponent: "*Dust particle size showed a weak exponential relationship to dust age. Two cases of freshly uplifted dust showed quite different characteristics of size distribution*". Hence, in our discussion paper (page 1915, lines 12-17), we chose particle size distributions representative of long-range transport and addressed the role of aging of non-spherical particles by quoting the work of Zhang (2008) and Schumann et al. (2011), which you know as a co-author. Reference to Miffre et al. (2012b), which rely on these two papers, may also eventually be used in addition for choosing a correct PSD after long-range transport, obtained by some other means. As you mentioned above, it is indeed important to discuss assumptions and we agree on that point. Whether the "*pure*" value would be representative further away from the source is, according to the literature, an assumption and we have thus chosen to avoid making such an assumption. We here propose a new methodology, consisting in analyzing (page 1896, lines 12-13) "*what is objectively retrievable when combining these two methodologies* (polarization lidar experiment + numerical light-scattering simulations)*, as opposed to comparing both approaches separately, as is often presented in the literature.*" The assumptions and new findings inherent to this new methodology are then analyzed in details in the discussion section (see page 1922, subsection 5.2.3). In particular, the results of the T-matrix numerical simulations, used to get the needed parameter values, are discussed by testing "*the robustness of our new methodology, using δ_{ss} and $\mathring{A}_{ns,\perp}$ (UV, VIS) exaggerated values*" (page 1922, line 25). Please note that our new methodology has been applied to two/three component particles external mixture after long-range transport. So, measurements close to the source region are out of the scope of our study, as specified in the manuscript (page 1896, line 15; page 1904, line 17) and in the title.

− Moreover, our new methodology – applicable to two/three particles component mixtures – is very general in the sense that it can be applied by using parameter values needed for the inversion, that could be derived by other means (including your measurement close to the source), if somebody wishes to do so. For the sake of clarity, we added this sentence in our conclusion (see text highlighted in cyan). Hence, our work objectively highlights the usefulness of our new methodology, found to be of "*great interest to the atmospheric aerosol community*", as concluded by anonymous Reviewer #2.

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M. Tesche

"*It should be clarified in the introduction that different optical properties of the aerosols are measured with lidar.*"

Authors Reply

As stated in our introduction (page 1897, line 22), "*polarization-resolved particle backscattering profiles are retrieved* » with a lidar. With a lidar, it is indeed possible to measure particles backscattering and particles extinction (using Raman channels). However, as learnt from Mishchenko and Sassen (1998), for an ensemble of particles, extinction is insensitive to the polarization state of the light and introducing this well-known aspect would increase the complexity of the reading. Otherwise, Raman lidar channels are suitable for nighttime measurements only (which is not our case). They can of course however be added to improve the precision, as you did it by applying A. Shimizu et al. (2004)'s methodology in M. Tesche et al. (2009). However, it is not our main concern (page 1896, lines 26-29) and, moreover, our new methodology would remain identical and still applicable. To include your comment, we however added the information on the particles extinction in a sentence of the introduction (see text highlighted in cyan color).

M. Tesche

"Backscatter coefficients depend on particle concentration (extensive parameter) while particle depolarization ratios depend on particle type (intensive parameter). Only intensive parameters can be used to separate the contribution of different aerosol types since they are directly linked to the size, shape, and refractive index of the respective particle types. I don't believe that the use of polarized backscatter coefficients (i.e., extensive parameters) rather than actual aerosol depolarization ratios (i.e., intensive parameter) is applicable for aerosol-type separation."

Authors Reply

– Thank you for the remark. We learnt from Mishchenko that it is a little bit more complicated. As he explained, scattering matrix elements depend on the particles size, shape and refractive index. Hence, not only intensive parameters depend on particles size, shape and refractive index: the lidar backscattering coefficient (equal to (F11-F22)/2 for the co-polarized channel, (F11+F22)/2 for the cross-polarized channel) is also directly linked to the particles size, shape and refractive index. This scattering matrix formalism is well-adapted for aerosol type separation, as explained in Mishchenko's textbook (see page 102 in particular) or as we published in 2011 in GRL, in a paper based on his pioneer work.

– Moreover, please note that the outputs of our numerical simulations, which are given in Table 3, are indeed intensive parameters only. In our discussion paper, only intensive parameters are used to separate the aerosols components, since these computed values are designed for that purpose (indeed, they are specific to each component: ash, dust, sea-salt and do not depend on the particles number concentration). Extensive parameters are only used at the end of our new methodology, to retrieve ns-particles backscattering coefficients, as shown by Equation (11) of our manuscript.

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M. Tesche

"Also, the authors should clearly state that some pre-requisites need to be fulfilled, to achieve aerosol-type separation. The different aerosol types contributing to the mixture should be (1) known and (2) externally mixed, and (3) lidar measurements should yield aerosol-type specific parameters, i.e., the particle depolarization ratio, the particle lidar ratio, or the Ångström exponent. The methods were mainly applied to two-component mixtures, since these provide the experimentalist with conditions that can be controlled - to at least some extent. Such background knowledge should be provided to non-expert readers."

Authors reply

We of course agree that background knowledge must be provided to non-expert readers (in the revised manuscript, we have moved much of this to an appendix). In the discussion paper, the raised pre-requisites points are detailed along the manuscript: for "*known*" and "*externally-mixed*", see the abstract (page 1892, lines 6, 9-10), the body of the text (page 1922, line 17) and the conclusion (page 1924, line 27). For "*aerosol-type specific parameters*", see for example the introduction (page 1896, line 22). It is however true that it is distributed all over the manuscript rather than being concisely summarized in one place. We hence added the following sentence in the introduction of the new Section 4 (Methodology): "*The different particle components should be known and be externally mixed, so that the lidar measurements, when separated for each particle component and combined with the simulated single-scattering properties corresponding to these components, provide backscattering coefficients specific to each ns-component.*" (see text highlighted in cyan color).

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M. Tesche

"The authors present the scattering matrix formalism which is clearly textbook knowledge but don't refer to any book. If not a new finding, such detail is not needed in a scientific publication and complicates the matter."

Authors reply

We indeed referred to Mishchenko's textbook: see the reference list, page 1930, line 5.

We agree that this formalism is textbook stuff, but we also feel that some readers will benefit from seeing how it relates with lidar measurable, hence applying your previous recommendation: "*some background knowledge should be provided to non-expert readers*". We propose to move the formalism into an appendix, where the interested readers can find it. We note that Reviewer #2 found the scattering matrix section useful.

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M. Tesche

"It is good to show how the parameters measured with lidar are related to this formalism, but it again seems futile since the authors end up at the regularly used parameters β and δ. They continue their derivation of the separation method from there and claim that it was done using scattering matrix formalism. This entire theory section leaves the impression that simple things are expressed in an unnecessarily complicated way. The purpose of scientific writing is the exact opposite of that."

Authors reply

As stated in the discussion paper (Equations (11) and (15)), our new methodology combines $\beta_{ns,\perp}$-polarization lidar experiments with T-matrix $A_{ns,\perp}$ and δ_{ns}-numerical retrievals. The scattering matrix formalism is involved in both. Combining single-scattering simulations with polarization lidar can only be performed within this formalism. This combination is useful as it enables us to retrieve range-resolved backscattering profiles specific to each ns-particle component. We insist on including the scattering matrix formalism, because it is essential in establishing the connection between the simulations and measurements, and contain in a concise form all the information about the scattering process. It is hence, in our opinion, not futile, and otherwise may be useful to non-expert readers.

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M. Tesche

"The scattering matrix formalism shows how the model simulations use the particle microphysical properties to end up with lidar parameters but it does not help to understand what the lidar measures or how the separation of different aerosol types based on measurements of their optical properties works. The authors imply that long-range transport produces mixtures of spherical and nonspherical particles. They should mention that measurements of isolated aerosol layers presented in the literature also show intensive optical properties that are similar to what is measured close to the source of different particle types."

Authors reply

– Scattering matrix indeed helps to understand what the lidar measures, as shown by Mishchenko and Sassen for contrails (1998). The scattering matrix formalism can be used to interpret polarization lidar measurements as in I. Veselovskii et al. (JGR, 2010), A. Miffre et al. (GRL, 2011) or J. Gasteiger et al. (ACP, 2011). In the absence of further justifications or literature references, we cannot discuss the first sentence further.

– As shown in I. Veselovskii et al. (JGR, 2010), indeed, after long-range transport, "*a significant part of the particles can be represented by spheres*". As above discussed, it is not our methodology to compare intensive quantities between our remote site and their source region. Please note that our new methodology is hence different from others in general and from M. Tesche et al. (2009) in particular. Otherwise, we quoted in the introduction the literature corresponding to other possible methodologies.

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M. Tesche

"Mixing of different aerosol types is more related to convective processes rather than the laminar flow in which elevated aerosol layers are often imbedded. It is also much more likely to happen during particle emission rather than transport. Aging might change the microphysical and optical properties of the aerosols but this is not a given. Also, little is known about that since Lagrangian observations of individual aerosol plumes would be required to investigate aerosol aging during long-range transport."

Authors reply

As you know as a co-author, in his ACP-2011 paper, U. Schumann showed that aging indeed modifies the particle size distribution after long-range transport, which hence exhibits a cut-off radius due to gravitational settling. Reference to Schumann (2011) for ash aging, and Zhang (2008) for dust and sea-salt aging, are given in our manuscript to support the discussion on the role of aging on nonspherical particles, which is presented in page 1893, lines 21-27. According to the literature, change of microphysical properties during transport is hence indeed a given. Moreover, this given is indeed confirmed by a very recent ACP-publication by C.L. Ryder et al. (2013): "*Dust particle size showed a weak exponential relationship to dust age. Two cases of freshly uplifted dust showed quite different characteristics of size distribution*".

M. Tesche

"The authors end up at Eq. (13), which is (as they state themselves) the same as Eq. (14) in Tesche et al. (2009) under the assumption that $\beta_{s,\perp} = 0$, i.e., $\delta_s = 0$ (only using the factor X instead of $\beta_{nd}=\beta_p$). Note that from the experience with actual measurements the assumption of $\beta_{s,\perp} = 0$, while theoretically justified, is basically never fulfilled. For instance, measurements of marine aerosol presented in Groß et al. (2011) Characterization of the planetary boundary layer during SAMUM-2by means of lidar measurements Tellus 63B, doi:10.1111/j.1600-0889.2011.00557.x. show values of 2-3% and the values obtained by Sakai et al. (2010) (which is cited inthe manuscript) for liquid droplets vary around 1%. The authors need to keep in mind that the atmosphere is not a laboratory and never provides ideal conditions. Also, if you set $\beta_{s,\perp} = 0$, there is nothing that could level the depolarization ratio of the non- spherical particles to end up with a spherical fraction. What I mean is that Eq. (14) in Tesche et al. (2009) can be used to calculate the backscatter coefficient for bothspherical and non-spherical particles. Eq. (13) of this manuscript requires that the contribution of spherical particles is derived as $\beta_s = \beta_p - \beta_{ns}$. Later in the paper the authors assume that the depolarization ratio of spherical particles(now awkwardly referred to as non-non-spherical particles) is assumed as 1%. This contradicts the previous assumption of $\beta_\perp$ being zero (see Eqs. (10b) and (14b))."

Authors reply

− In our discussion paper, the equation $\beta_{s,\perp} = 0$ is not an assumption: indeed, $\beta_{s,\perp}$ is zero all along the manuscript, since homogeneous spherical particles do not depolarize laser light, as we know from Mie theory. We recall what we wrote in our discussion paper (page 1906, lines 19-22), *"to account for possible background depolarization, the particle mixture is preferably partitioned between ns (ash, dust, sea-salt) particles on the one hand, and non-ns particles, i.e. particles that are neither ash, dust nor sea-salt particles, on the other hand."* These non-ns particles may slightly depolarize laser light and this is what we call the background depolarization. This approach is totally compatible with what is measured experimentally. In S. Gross et al. (2011), the background depolarization ratio is measured (hence a 2-3 %-value is found) since, as published by A. Miffre et al. in GRL (2011), the δ_p ratio is not specific to spherical particles, unlike δ_s. In T. Sakaï et al. (2010), the 1%-laboratory measured depolarization ratio is compatible with Mie theory, within error analysis.

− The requirement that the β_s-coefficient be derived from $\beta_p - \beta_{ns}$ is not a main concern, since, as we recently published in PNAS (2012), β_s can be derived independently from β_{ns}. As you noticed above, *"If not a new finding, such detail is not needed in a scientific publication and complicates the matter."* We agree with you, and for this reason, we did not detail points that we already published elsewhere (see Y. Dupart et al., PNAS, 2012). Moreover, both reviewer #1 and reviewer #2 ask for simplifications to improve the readability.

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M. Tesche

"In Section 4, it is stated that: Despite the complexity, it is now well-established that the optical properties of size-shape distributions of such [non-spherical] particles can be well-mimicked by size-shape distributions of homogeneous spheroids. Not a single reference is provided to support this comment. The statement is true in the more general sense that T-matrix simulations are much more reliable than Mie scattering theory. However, recent literature (e.g., Gasteiger et al. (2011), Müller et al.(2010a, 2010b, 2012)) that presents the results of model simulations of light scattering by nons-pherical particles and compares these to actual measurements shows that the models are not able to properly describe what is happening in the 180_ backward direction that is investigated with lidar instruments".

Authors reply

− We referred to M.I. Mishchenko and M. Kahnert's numerical codes who are well-recognized T-matrix specialists. For a literature reference on T-matrix, you may hence refer to M.I. Mishchenko's review (JQRST, 2013). Many reference literatures are indeed provided on that topic in our manuscript (see page 1914, lines 18-21). These references *"have demonstrated that size-shape distributions of randomly-oriented spheroids can reproduce the phase function of real dust particles."* As stated at page 1914 (lines 21-25), please note that our computations only act as a demonstration of our new methodology and that, as detailed in our manuscript, it is not the subject of this paper to address different numerical simulations, as we propose to do so in a forthcoming paper (page 1917, line 28) including improved numerical simulations, as for example using DDA-approximation (see the outlook, page 1925, line 26). The proposed numerical simulations will hence be replaced by something more sophisticated in the future, but again, this point does not affect the applicability

of our new methodology, it just may add precision to our methodology, which is new and has never been published elsewhere.

– Finally, as you know, care should be taken when comparing measurements with numerical simulations since neither are error free. Hence, comparing measurement with theory is meaningful provided that particles' lidar measurements are ns-particles specific.

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M. Tesche

 "Note also that little is known about the transformation of the optical properties and size distribution of non-spherical particles during long-range transport. The authors take some size distributions that are assumed to be representative for aged non-spherical particles (without discussion their choice or the resulting implications on the results), use these as input for model simulations (without critically assessing the limitations of such simulations), and take the output of these model simulations as part of these retrieval scheme (again without critical assessment)."

Authors reply

Again, we wish to point out that this is a demonstration of the methodology, so it is quite reasonable to make some simplifications. Indeed, simple test cases better allow assessing the performance of the method. Moreover, please note that information on the points that you raise were already available in our manuscript, for example the discussion about the ash particle size distribution (page 1917, lines 7-12). Regarding dust and sea-salt particles, please see our response to the Reviewer #2. The impact of the assumed properties is discussed at line 25 of page 1917, lines 22-28 of page 1922, and lines 1-19 of page 1923. Moreover, the robustness of our new methodology is discussed in a dedicated paragraph (subsection 5.2.3). In particular, we state in page 1922 (lines 25): "*To test the robustness of our new methodology, we used exaggerated δ_{ss} and $\dot{A}_{ns,\perp}$ (UV, VIS) values*", as stated again in our conclusion (page 1925, lines 18-20). Hence, the retrieved Angstrom coefficients $\dot{A}_{ns,\perp}$, which are functions of the particle size distribution and numerically derived, are discussed and the consequences of the numerical outputs are indeed analyzed (see for example page 1922, lines 26-28, page 1923, lines 1-5). With the proposed rearrangement of the manuscript, these aspects hopefully become clearer.

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M. Tesche

 "The results presented in Fig. 5 should be discussed in the context of measurement results that are available in the literature. It seems like none of the values for the lidar ratios or particle depolarization ratios at 355 nm or 532 nm is in agreement with what is actually observed in the atmosphere or in laboratory experiments "

Authors reply

As stated in our manuscript, the retrieved lidar ratios agree with what is actually observed in the atmosphere (see page 1918, lines 1-5 and page 1921, line 19): "*The obtained S_{ns}-value agrees with the literature, derived from Raman Lidar measurements. For example, at λ_1 = 355 nm, S_{ash} equals (60 ± 5) sr at Munich (Ansmann et al., 2012), while for dust particles, Veselovskii et al. (2010) numerically computed S_{dust} = 68 sr. Sea-salt particles exhibit S_{ns}-values around 20 sr, in agreement with (Ansmann et al., 2011).*" These literature references include papers in which M. Tesche is a co-author. As quoted in Ansmann et al. (2012), these lidar ratios are also in agreement with Figure 12 of M. Tesche et al., Tellus **63**B, (2011). The retrieved delta-ratios (for dust particles, for example) also agree with the literature (see V. Freudenthaler et al., 2009), that we quoted.

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M. Tesche

"The authors assume that mixtures of specific aerosol types were present over the measurement site. The assumption of a mixture of spherical and non-spherical particles is justified in the case of the observation in the aftermath of the Eyjafjallajökull eruption. This is because these were quite well-constrained atmospheric conditions. Also, further studies arrive at the same conclusion and present similar results as the authors. However, the results of this first case study are already published in several papers by Miffre at al. (Atmospheric Environment 2011, Geophysical Research Letters 2011, Journal of Atmospheric and Oceanic Technology 2011) and I don't see how this manuscript provides any new or additional information."

Authors reply

As published by I. Veselovskii and M.I. Mishchenko (JGR, 2010), after long-range transport, "a significant part of the particles can be represented by spheres". This assumption is hence justified even in dust episodes, different from the Eyjafjallajökull eruption. The case of the three-component particle mixture is new, and can be understood as an extension of the two-particle component case study with which it can be compared. What we try to show here is that two/three component particle mixtures can be treated with the same formalism, which may sometimes be beneficial. Hence, the three-component mixture algorithm may be applied to the two-component mixture but this is beyond the scope of this paper. For the quoted papers, see our response to Reviewer #2 about the same topic.

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M. Tesche

"While the second case study presents new measurements, the authors miss to discuss the underlying assumptions or the results in the critical way you can expect for scientific publications. The nature of the three-component mixture in the second case study is purely speculative and the authors miss to justify this choice of a mixture and the selection of the different contributing aerosol types. Using backward trajectories is common when investigating events of long-range transport, but trajectories only give you an indication of the history of the air mass that is observed. The authors state (for instance in the caption to Fig. 8) that the trajectories show evidence of sea-salt and dust particle over Lyon. The authors should be more careful with what to extract from the information provided by backward trajectories since they do not directly relate to the aerosols in the air parcel."

Authors reply

The only comment on our new three-component methodology is related to the corresponding back-trajectories, which are of course not the main concern of this paper, dedicated to the combined use of polarization lidar and T-matrix, for the ACP-Special Issue on Light Depolarization. That said, back-trajectories are used with care (page 1921, lines 1-12) and as stated in our introduction, performing a precise chemical analysis is not the scope of our paper (page 1896, line 29 up to page 1897, line 4): "*A precise chemical analysis has not been performed during the experiments carried out, because it is not the scope of this contribution. We hence used 7-days air mass back-trajectories to identify the origin of the nonspherical particles at the remote site. Otherwise, the combined use of our Lidar depolarization experiment with laboratory chemical studies has been already published in Dupart et al. (2012).*" To avoid any confusion, we however also rephrased the caption of figure 8. Otherwise, reviewers #1 and #2 did not draw the same conclusions.

Moreover, our retrieval method indeed shows the presence of three distinct aerosol components above Lyon (see figure 10), and the backward trajectories clearly indicate the possible presence of marine aerosol. Note that the lidar retrieval is consistent with this assumption. In addition, in case another aerosol type as marine aerosol would be present, note that, as we wrote (page 1922, line 27), "*the behavior of β_{ns} with altitude is still retraced for a different δ_{ns}-value.*"

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M. Tesche

"In general, it does not seem likely that marine aerosol is lifted to heights of 3 to 4 km and transported over several days. In fact, it can be taken from the literature that airborne insitu measurements over the ocean hardly show any marine aerosol above the marine boundary layer. Hence, the assumption of having sea-salt crystals over your site is not in agreement with what is generally observed, and thus, highly unrealistic. Independent measurements are needed to support such an argumentation and a critical discussion of the circumstances under which sea salt crystals occur in the atmosphere would still be required. This includes a revision of the respective literature."

Authors reply

Airborne in situ measurements have shown that sea-salt particles may be present at 3-4 km altitude: see Ikegami et al. (Tellus B, 1994) whose title is: "*Sea-salt particles in the upper free troposphere*". Since anonymous reviewers #1 and #2 asked for simplifications, to include your comment, we added only this reference to our manuscript (see text highlighted in cyan color). Otherwise, in situ sea-salt measurements are

difficult to achieve, even with an aerosol mass spectrometer. In addition, sea-salt particles are transported by advection as other particles such as dust particles, but of course the chemistry is different.

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M. Tesche

"The authors obtain backscatter coefficients according to Klett's method (without reference) but miss to discuss the errors introduced by the choice of the intensive parameter lidar ratio, which they need to assume to obtain backscatter profiles. It is also not clear whether a height-dependent lidar ratio or a constant value is used in the retrieval. Also, where comes you knowledge of polarized lidar ratios from? If they are the outcome of model simulations, they should also be critically discussed."

Authors reply

Reference to very well-known Klett's algorithm has been added (we quoted your work on extinction retrievals). In addition, both reviewers #1 and #2 ask for simplications (we now have 80 literature references). The asked information is present in our discussion paper:

  – The error on the intensive lidar ratio induces an error bar on δ_p (see page 1912, lines 16-17 and page 1913, lines 4-5).
  – We chose a constant lidar ratio as stated at page 1919, line 17 for case study 1 (see page 1921, line 18 for case study 2). Reference to G. David et al. (2012) is also given at page 1912, line 16 for a discussion on height-dependent lidar ratio.
  – The knowledge of lidar ratios comes from the literature, as stated at page 1921, line 19.

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M. Tesche

 "Besides the fact that the presence of sea-salt crystals at 3-4km height over Lyon is unlikely, I don't think that there is enough independent information in the lidar measurements (even if depolarization profiling is performed at two wavelengths) to separate three aerosol types. One required information comes from the depolarization ratio. Additional information could come either from the lidar ratio or the Ångström exponent. However, lidar ratios are not measured but assumed by the authors and Ångström exponents are always affected with large error bars and probably not sensitive enough to separate between big dust particles and big sea-salt crystals. The model simulations used to overcome this problem in the manuscript would require some critical discussion and - even if found sound - would still only represent the case of a combination of microphysical particle properties that the authors chose quite arbitrarily."

Authors reply

Because the manuscript was difficult to read, as pointed out by the reader, it was not easy to recognize that this manuscript deals with the demonstration of a new methodology. Again, this manuscript should be read as a demonstration of a new methodology which potentially provides additional information retrievable by a lidar, rather than aiming to replace existing methods. We agree with M. Tesche that Lidar inversions will always be under-constrained, and some assumptions will always be needed to retrieve information about the aerosol particle properties. In our methodology, we use predictions offered by exact light-scattering computations, based on assumed particle properties, to aid the retrieval and to allow retrieval of properties that would not be otherwise retrievable. We believe this to be valuable. But as always, when dealing with under-constrained inversions, the results obtained are subject to assumptions made. The light-scattering simulations conducted here do not attempt to be the most sophisticated possible, but rather reasonable examples that should be representative of at least some populations of the intended target particles, and therefore usable for demonstrating the methodology. Please note again that our new methodology relies on combining T-matrix simulations and 2λ-polarization lidar to resolve up to three-component particle external mixtures, and that we indeed discussed our assumptions (see the devoted paragraph 5.2.3).

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M. Tesche

"It should be noted that different aerosol types often show similar values of the intensive optical properties measured with lidar (i.e., similar particle depolarization ratios and Ångström exponents as in case of mineral dust and volcanic ash; similar lidar ratios as in case of biomass-burning smoke and volcanic ash) and that the complete data set of lidar ratios, depolarization ratios, and Ångström exponents of particles needs to be measured with high precision to separate mixtures of more than two aerosol types."

Authors reply

We agree, and mention this in the manuscript (see Table 3 and in the text page 1918, lines 7-11). However, we have made an attempt to improve the clarity of the appropriate sentences in the methodology section (see text highlighted in cyan color).

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M. Tesche

"To conclude, I believe that the polarization measurements with your instrument are indeed sensitive and accurate. However, the authors have to put the same accuracy to interpreting the measurements to come up with results that can stand up to a critical review".

Authors reply

The conclusion that our polarization measurements are sensitive and accurate is of particular relevance, because this manuscript has been submitted for the ACP-Special Issue on Light Depolarization. We are grateful for the comments that have helped us to improve the manuscript.

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Specific comments:

M. Tesche

"Emphasizing the quality of your work is totally fine but there is no need to state over and over again that what you do is accurate. This is a prerequisite for scientific work. The authors use the word experiment in connection to both the lidar measurements and the numerical simulations. It should be stated when measurements or simulations are performed."

Authors reply

This comment has been already addressed when answering to reviewer #2.

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M. Tesche

"Table 1 provides no information besides pictures of non-spherical particles. The authors should refer to the literature to assist their discussion and support their argumentation instead of listing papers without a trace of purpose."

Authors reply

We again recall that our manuscript is a demonstration of a new methodology. Table 1 is aimed at this goal, showing that, indeed, volcanic ashes, desert dust or sea-salt particles are nonspherical particles. The quoted papers support this argumentation as we stated at page 1899 (lines 8-11): "_For each particle component, some literature references are also given in Table 1, on Lidar remote sensing studies as well as numerical simulations and laboratory experiments on their light-scattering properties._"

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M. Tesche

"What do you want to show with Fig. 1? You introduce the factor X_{ns} but Fig. 1 is missing a discussion that explains the reader what you intend to show here."

Authors reply

This discussion is present in the manuscript (page 1906, lines 3-5 and up to line 10): "_since δ_p is sometimes assumed to equal δ_{ns}, we plotted in Fig. 1 the systematic bias between δ_p and δ_{ns} as a function of X_{ns} for each ns-component_". We however agree that we should have been clearer in figure 1 caption, which has hence been modified to clarify that point (see text highlighted in cyan).

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M. Tesche

"If you want the reader to get an impartial view on your data you should present results on the same scales. See Figs. 5, 7, 9, and 10."

Authors reply
The Reviewer #2 also raised this point. See our response there for details.
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M. Tesche
*"Here is some useful literature on: Polarization lidar in general Sassen (2005) Polarization in lidar. In: LIDAR-Range-Resolved Optical Remote Sensing of the Atmosphere (eds C.Weitkamp), Springer, New York, 105-141. Measurements of lidar ratios and depolarization ratios of mineral dust and volcanic ash Groß et al. (2012) Dual-wavelength linear depolarization ratio of volcanic aerosols: Lidar measurements of the Eyjafjallajökull plume over Maisach, Germany. Atmos. Envir., **48**, doi:10.1016/j.atmosenv.2011.06.017. Marenco et al. (2011), Airborne lidar observations of the 2010 Eyjafjallajökull volcanic ash plume, J. Geophys. Res., **116**, doi:10.1029/2011JD016396. Tesche et al. (2009) Vertical profiling of Saharan dust with Raman lidars and airborne HSRL in southern Morocco during SAMUM, Tellus **61B**, doi:10.1111/j.1600-0889.2008.00390.x. Tesche et al. (2011) Profiling of Saharan dust and biomass-burning smoke with multiwavelength polarization Raman lidar at Cape Verde, Tellus **63B**, doi:10.1111/j.1600-0889.2011.00548.x. Modeling of light scattering by non-spherical particles and comparison to measurements Gasteiger et al. (2011) Modeling lidar-relevant optical properties of complex mineral dust aerosols. Tellus **63B**, doi:10.1111/j.1600-0889.2011.00559.x. Johnson et al. (2012), In-situ observations of volcanic ash clouds from the FAAM aircraft during the eruption of Eyjafjallajökull in 2010, J. Geophys. Res., doi:10.1029/2011JD016760. Müller et al. (2010a), Mineral dust observed with AERONET Sun photometer, Raman lidar, and in situ instruments during SAMUM 2006: Shape-independent particle properties, J. Geophys. Res., **115**, doi:10.1029/2009JD012520. Müller et al. (2010b), Mineral dust observed with AERONET Sun photometer, Raman lidar, and in-situ measurements during SAMUM 2006: Shape-dependent particle properties, J. Geophys. Res., **115**, doi:10.1029/2009JD012523. Müller et al. (2012), Comparison of optical and microphysical properties of pure Saharan mineral dust observed with AERONET Sun photometer, Raman lidar, and in situ instruments during SAMUM 2006, J. Geophys. Res., **117**, doi:10.1029/2011JD016825."*

Authors reply
− Reference to Marenco and Hogan has been added, as discussed in the general comment above. Reference to K. Sassen et al. (2005) was not quoted, as in our discussion paper, we preferred to refer to K. Sassen et al. (2007) as an update.
− We recall that our study is not a review (we however have 80 references already), our introduction is already too long in other reviewers' opinion, and the above reference studies cover topics (short-range transport, Raman lidar, sun photometer lidar, etc.) not critical for our new methodology presented here. In our introduction, we indeed stated that (page 1896, lines 26-28): *"The coupling of Lidar with inelastic Raman scattering (Müller et al., 2001) as well as with passive remote sensing (Dubovik et al., 2006, Boyouk et al., 2011), has not been considered in this paper."*