

Interactive comment on “Characterization of long-range transported Saharan dust at the Caribbean by dual-wavelength depolarization Raman lidar measurements” by S. Groß et al.

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We thank Dr. Miffre for his suggestions to help us improve the paper.

The answers are given in a direct response (bold, italic).

The manuscript proposed by S. Groß et al. reports on lidar measurements performed in the Caribbean Island of Barbados. The introduction states the difficulty of studying dust particles after long-range transport and underlines the complexity of involved atmospheric processes such as aging, mixing with other aerosols or nucleation to quote only a few. Facing such a complexity is indeed difficult while ideally lidar measurements should become standard tools for atmospheric studies. In this context, all my comments are intended to improve the science of this manuscript and help potential future readers.

On the methodology

The methodology proposed by S. Groß et al. to analyze two-component particle mixtures is not the only existing one nor the pioneering one. As the manuscript may potentially have a large impact, other methodologies (Shimizu et al. JGR2004, Nishizawa et al. JGR2007, David et al. ACP2013) could be quoted, especially in a context where more than 45 % of the given references are from one of the co-authors.

We do not claim to develop a new methodology for data analyzing, but we use an established procedure which (for our measurements) is described in the referenced literature and apply it, for the first time, to lidar measurements of long-range Saharan dust at Barbados. This method is based on a long history of data analysis. As this history might be of interest for readers we included some more references in the text.

On the measurements and the analysis

How is the PLDR-value and uncertainty of 0.27 ± 0.01 retrieved from the measurements? Looking at the observed vertical profiles, the uncertainty seems larger. Accordingly, how does the “highly accurate $\pm 45^\circ$ calibration method” (page 19331, line 9) relate with other published methods (Alvarez et al. JTECH2006, David et al., APB2012) that rely on a dozen of points ?

We guess that the comment refers to Figure 8 and Table 1. Indeed the error bars are too large in the figure by mistake. The data in Table 1 are correct. We changed Figure 8. As mentioned in the text, “the error calculation of δv and δp was done analogue to Freudenthaler et al. (2009)”. For the determination of the calibration factor V^ , or G , Alvarez et al. (JTECH 2006) and David et al. (APB 2012) use a halve wave plate to rotate the plane of polarization of the light in the receiving optics by angle φ , while POLIS uses a mechanical rotation of the receiving optics. For both methods Eq. 10 in Freudenthaler et al. (Tellus 2009) shows that the signal ratio of the cross and the parallel channels δ^* , from which the calibration factor V^* is determined, depends on φ and on the volume linear depolarization ratio δv , except for $\varphi = \pm 45^\circ$, where the dependence on δv vanishes. Actually only one measurement of δ^* at $\varphi = +45^\circ$ or -45° would be necessary to determine V^* , but usually the plane of polarization of the laser beam and the absolute position of the calibrator rotation are not exactly known, which introduces another parameter y , the offset angle, which is considered in Alvarez (JTECH 2006) but not mentioned in David (APB 2012). That means in general three*

parameters have to be retrieved, which requires at least three measurements at different angles. The measurements of δ^ around 0° and 90° are most sensitive to δv and least around $\pm 45^\circ$, while the sensitivity to γ is highest around $\pm 45^\circ$ and lowest around 0° and 90° . For the latter problem it is shown by Freudenthaler et al. (Tellus 2009) with a numerical simulation (it can also be shown analytically), that the geometric mean of two measurements around $\pm 45^\circ$ exactly 90° apart the influence of δv and γ are decreased by a factor of about 100, which reduces the number of necessary measurements again to two. Another reason for using as few as possible measurements is the signal to noise ratio, which increases with averaging time and vice versa decreases with the number of measurement points for a given time for the whole calibration. This is important because the calibration should be done regularly (we do it every time we switch on the lidar system) until the temporal stability of the calibration constant is verified. With several lidar systems we discovered both a day to day variance above the noise error, maybe caused by thermal influence on the detection electronics or optics, and long-term changes due to degradations of various components. The 90° difference can be achieved quite accurate by mechanical means (POLIS), but also using good actuators, as it is probably the case in David (APB 2012) because there no errors are mentioned for the measurement angles. The possible detector saturation at $\pm 45^\circ$ is an issue, but is solved by means of a polka-dot attenuator on top of the telescope, which doesn't influence the ratio of the polarization measurements. Such an attenuator would also be necessary at smaller angles, albeit with lower attenuation.*

In Figures 6, 8, 10, 12, 13 and 14, could you improve the PLDR-graph so that the reader may see the data points? Why is the PLDR not retrieved below 2 and above 4 km? It may be useful for the reader. In the same way, in Figures 5, 7 and 9, could you modify the δv color scale? I only see two colors while a 8-bins color scale is used.

We changed Figures 6, 8, 10, 12 to show the retrieved PLDR below 2 km. Above 4 km the aerosol load is too low to calculate the PLDR with significant uncertainty. We removed Figure 13 as suggested by Reviewer #1.

In their assumed two-component mixture, S. Groß et al. use "for the aerosol type separation 0.30 at 532 nm for dust and 0.02 for marine aerosols according to the findings for pure Saharan dust and marine aerosols (Freudenthaler, 2009, Groß 2011b)". According to the observed variability in δp in the quoted papers, how is the 0.30 value chosen? Which value is used at 355 nm? To what extent do the corresponding uncertainties (at 355, 532 nm) modify your conclusions?

The chosen value represents the mean value of the measurements presented in the referenced literature. Certainly there is a natural variability of these values, however this variability was found to be smaller than our measurement uncertainties. Uncertainties of the used input values result in uncertainties of 10-30% of the retrieved quantities as stated in the referenced literature. As this comparison is performed for sun-photometer and lidar measurements at 500 nm and 532 nm, respectively, we do not use a value for 355 nm.

On the interpretation of the measurements

I disagree with the interpretation proposed by S. Groß et al. on several points.

1: To interpret their lidar measurements, S. Groß et al. assume a two-component particle mixture (page 19331, line 20). In the Caribbean, I would rather expect a three- component particle mixture, with water-soluble, sea-salt and dust particles to be more realistic: some back-trajectories (like the blueish) fall very close to sea level. While sea-salt particles can be found up to the tropopause (Ikegami 1994), could you discuss on your assumption of a two-component mixture? Reference literature to three-component particle mixtures (Sugimoto et al. Atm. Res.2010, David et al. ACP2013) are not quoted, while they may interest potential future readers. How do you discriminate

non-spherical dust from sea-salt particles, which are non-spherical below 40 % relative humidity? This remark is important because aerosols are then classified at the end of the manuscript.

Our assumption of a two-component mixture in the lowermost layers is based on co-located in-situ measurements; Publications within this Special Issue are in preparation. The separation of sea-salt particles and dust was performed in the lowermost layers where the relative humidity was always larger 40 %.

2: As detailed in the introduction, reference literature exist showing that δp is modified for an aged Saharan dust plume (Wiegner et al. 2011) compared to values measured in fresh Saharan dust plumes (Groß et al. 2011b). Additionally, aerosol lifetime (Amiridis 2009) modifies the lidar ratio. However, in this manuscript, S. Groß et al. arrive to the opposite conclusion. How do you explain this difference? Is your conclusion a general established fact or a specific particular case? I do not understand the comparison of the SALTRACE experiment in the Caribbean with former experiments performed in Munich and during SAMUM campaigns a few years before. Could you assume that the source regions are the same, and if they are, that the activity of these sources is the same? On this point, I agree with Reviewer #1.

The work of Amiridis et al., 2009 deals with lidar measurements of biomass burning aerosols and is not suited for a comparison with our dust measurements here. In our analysis we found no significant differences in consequence of different activation methods or source regions. For the dust layers observed over Barbados and Munich (Wiegner et al., 2011) we found similar dust source regions.

3: I have some concerns with the classification proposed by S. Groß et al.

- i) In the classification scheme, the plotted quantities (i.e. δp and Sp) are representative of the particles mixture and hence not specific to one type of particles (see Miffre et al. GRL2011 for the difference between δp and δns). δp is not a tracer for nonspherical particles (David et al., ACP2013). A “pure case” is however often reported in the manuscript (in the abstract and in the text). Could you provide evidence of a “pure case” here? Can we find “pure case” in the atmosphere? The coauthors have already used this terminology but it has never been defined in terms of chemistry and particles content. Optical devices are not sensitive enough to claim the existence of pure cases (one compound).

We do not see the point of this comment. The classification scheme shown in Figure 16 and adopted from other peer-reviewed publications is based on a multitude of co-located and synergistic lidar and in-situ measurements, while the stated article shows lidar measurements of a single event of volcanic ash and Saharan dust mixtures. Furthermore the stated paper declares ‘Aerosol UV-depolarization serves as an independent means to discriminate ns from s-atmospheric particles’ while in the next sentence of this comment they say ‘ δp is not a tracer for nonspherical particles (David et al., ACP2013)’. Our assumption of ‘pure dust’ is based on coordinated airborne in-situ measurements of the chemical and microphysical parameters of the observed aerosol layers. These results will be published in further publications within this Special Issue.

- ii) Above all, though δp and Sp are intensive, a great variability can be observed in Sp and δp just by varying the size, the shape or the chemical composition. As published by M. Kahnert (JQSRT, 2015), a prerequisite for a potential classification scheme is that the variation in δp among particles in the same type is small compared to differences in δp among particles of different types and according to the state-of-the art literature, this is not a given. How do you account for this publication?

The publication of Kahnert deals with scattering of single particles in which case the shape and the chemical composition of the particle is of great importance. Lidar measurements represent measurements of a certain volume of air with a composition of different shape, chemical properties and orientation. From all our measurements including SAMUM, SALTRACE and long-range transported Saharan dust to Europe within EARLINET we do not see a strong variability in δp and Sp for Saharan dust plumes which had most properly no influence from other aerosol types. We do not know which state-of-the art literature is meant here showing that the intensive optical properties of one type varying stronger than the intense optical properties of different aerosol mixtures.

4: S. Groß et al. conclude that the intensive properties do not vary over several thousands of kilometers. Literature Reference (Ridley et al. ACP2013) indeed shows that the particle size distribution is modified during transport "Dust particle size showed a weak exponential relationship to dust age. Two cases of freshly uplifted dust showed quite different characteristics of size distribution". Following the link that exists between δp and the particles size, the particle depolarization is then modified. How do you account for this remark in regards to your conclusion that δp remains constant? By looking at Figure 14 in detail, a potential reader may wonder if this conclusion results from the atmosphere or from the lack of precision or/and sensitivity of the experiment. Could make some comment on this?

We guess that not the publication of Ridley et al. (GRL2013, ACP2014) but the publication by Ryder et al. (ACP2014) is meant in this comment. We totally agree that δp is dependent on the particles size. Following the results published by Ryder et al. the difference in δp for long-range transported dust in the Caribbean compared to fresh δp measurements close to the source should be in the range of 0.01 to 0.02. These are about the changes in δp we observed at Barbados and report in our article. Changes of the microphysical properties of long-range transported dust are under investigation and will be published in separate articles within this Special Issue. Referring to the comment on Figure 14 it is clear that the uncertainties in the retrieved optical properties are dependent on the measurement/aerosol conditions. Most of the measurement points shown in Figure 14 clearly show constant values with small uncertainties.

One specific comment

Page 19340, line 13: S. Groß et al. wrote that "It has been shown that the lidar ratio and the particle depolarization ratio are quite different for different types of aerosols" and quoted reference to Sakai et al. (2010) to justify this statement. In their paper, Sakaï et al. only addressed the particle depolarization and nothing is said about the lidar ratio. Could you provide another reference?

We added more references addressing particle depolarization and lidar ratio.