Atmos. Chem. Phys. Discuss., 13, C422–C430, 2013
www.atmos-chem-phys-discuss.net/13/C422/2013/

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.

M. Tesche

matthias.tesche@itm.su.se

Received and published: 8 March 2013

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.

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

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.

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.

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 biomass-burning 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.

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

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.

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. 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.
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 non-spherical 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.
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.

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 $\beta_{\mathrm{s},\perp} = 0$, i.e., δ_{s} = 0 (only using the factor X instead of $\beta_{\mathrm{nd}}/\beta_{\mathrm{p}}$). Note that from the experience with actual measurements the assumption of $\beta_{\mathrm{s},\perp} = 0$, 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-2 by 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 in the 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 $\beta_{\mathrm{s},\perp} = 0$, 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 both spherical and non-spherical particles. Eq. (13) of this manuscript requires that the contribution of spherical particles is derived as $\beta_{\mathrm{s}} = \beta_{\mathrm{p}} - \beta_{\mathrm{ns}}$.
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)).

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 statements 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 non-spherical 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.

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).
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.

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.

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.
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 in-situ 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.

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.

Besides the fact that the presence of sea-salt crystals at 3-4 km 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. 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.

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.

Specific comments:

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.

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.

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.

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.

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.