Author Response to Comments on "A new method for measuring optical scattering properties of atmospherically relevant dusts using the Cloud Aerosol Spectrometer Polarization (CASPOL) instrument" by A. Glen and S.D. Brooks.

ACPD 12, C6426-C6429, 2012

Reviewer's Comment:

The authors have chosen to call a parameter (Eqn. 1) that is the fraction of the backscattered light whose polarization has been rotated by 90 degrees from the source laser the "polarization ratio". I understand their argument that the "depolarization ratio" is already a defined parameter used by the LIDAR community, and that this parameter is not the same; and this feels a bit like a "how many angels can dance on the head of a pin" argument, but I think their nomenclature is particularly unhelpful. I would strongly suggest calling the ratio something more descriptive like the "flipped" or "scrambled" or "rotated polarization ratio".

Authors' Response:

Since our instrument does not measure the quantity commonly known to the lidar community as depolarization ratio, we felt that we need to use another term, so as not to mislead. We have debated this nomenclature among ourselves, and have concluded that "polarization ratio" is a reasonable choice. We see the Reviewer's point that rotated is more accurate for our specific case. However, lidar depolarization ratios are also "rotated" in the same way, in that both in lidar and in our in situ instrument, radiation which is polarized perpendicular to the polarization of the incident laser beam is evaluated. Thus we think "rotated polarization ratio" or some rendition of it is has the potential to be even more misleading.

Reviewer's Comment:

On a more substantive note, I am at a loss to explain how a ratio of the intensity that is in a particular unfavorable (perpendicular) polarization to the total intensity of the back-scattered light can be greater than unity. Unless there is something more subtle going on, I have to conclude that the polarized and total backscatter detectors weren't balanced and that the ratio is not what it appears to be. (This is not really important to the central arguments in the paper, since the absolute value of the polarization ratio wasn't used.) I do think the authors should clarify this and also say whether the forward and backscattering detectors were balanced, since that would impact the calculations of back-scatter crosssections reported near the end of the paper.

Authors' Response:

As the referee comments, the polarized and total backscatter detectors are not balanced, this was an artifact of the construction of the prototype instrument and one which will be addressed in revised versions of the instrument by DMT. While normalizing the data is tempting, doing so would essentially hide the fact that the prototype CASPOL has a significant limitation. This limitation, while exciting in its ability to distinguish between aerosol dusts as well as the variations between individual particles within a

single dust type, results in data that is not robustly quantitative in achieving accurate polarization ratios. Based on the Reviewer's comment, we realize we should have mentioned this limitation in the text. On page 12, the text now states, "It should be noted that the polarization ratio shows values greater than unity, this is a result of an instrumental artifact in this prototype unit and will be addressed in revised versions of the instrument by DMT."

Reviewer's Comment:

I would like to see the authors "stretch" a little bit more in the final version of the paper and provide some suggestions with respect to the physical phenomena being probed in these experiments. For those readers who are not used to thinking about scattering and polarization effects, some simple explanation of how back-scattering from a dust particle can result in a 90 degree rotation of the polarization of the light would be very useful. And this explanation could be used later in the paper to try and suggest some level of interpretation of the observations. Admittedly, the polarization ratio is a complicated function of shape, size, composition, etc. but surely there must be some level of rationality of the observations and linkage to the dust particle characteristics that can be used to provide a more fundamental interpretation of the results, or at least some guidance in extrapolating the characterization to "similar" but previously unexplored dust types?

Authors' Response:

According to Sassen (1991), spherical particles do not cause depolarization in light scattered in the back direction. Conversely, irregularly shaped particles such as ice or solid aerosol particles produce high depolarization ratios (Mishchenko, 2009). The following has been added to the text in page 5 of the introduction, "Mishchenko et al. (1997) show that even after applying particle size and orientation averaging, a single spheroidal shaped particle always produces a unique shape specific phase function distinctly different from those produced by other spheroidal particles. Thus depolarization ratios can be used to differentiate non-spherical or rough particles from spheres." has been added to page 5 of the introduction.

In the conclusions on Page 22432 we originally stated "Our results imply that due to differences in aerosol shape and composition, lidar backscattering could vary greatly, even for dust particles of the same particle size". The conclusions will now include the following statement in addition "The dust types used were differing in their composition, refractive indices, shape as observed by the SEM, and measured particle size distributions when produced under identical generation conditions. While the effects of the differences in size distribution can be separated by inspection of particles in a single size bin, i.e. Figure 6, the remaining characteristics could not be isolated in this study. Based on our results, it was not possible to identify the dominant trait which causes the differences in the optical properties as these differences are most likely due to a complex relationship between the shape, size, morphology, composition and refractive index."

Reviewer's Comment:

In Figure 4 there are error bars on the forward-scattering determined particle sizes, but there is no discussion of the precision of the measurement that I can find. Based on the error bars in Fig. 4, it appears that the relative precision is of the order of 20 - 50% but the figure of merit for this is not clear (for example, if the error bars are +/- 1 sigma, the 95% confidence level for a single-particle size could be of the order of 100%). This should be clarified, and the ideal measurements to base it on would be the calibration results (it is my understanding that the VOAG produces a very tight monodisperse aerosol). If possible, the authors should also provide a relative uncertainty estimate for the total and polarized back-scatter measurements (just the optical part).

Authors' Response:

Failing to discuss the error bars in Figure 4 was an oversight on our part. The following discussion is now included on page 12. "The uncertainties shown in Figure 4 for the VOAG diameters are based on the uncertainty in the theoretical calculation of the VOAG generated particle diameters, as described in the instrument manual. The uncertainties in the CASPOL diameters are the standard deviations from the mean diameter of the log normal size distributions fitted to the CASPOL measured calibration particle size generated by the VOAG."

Reviewer's Comment:

I also would find it useful if the authors would explain how the qualification detector determines whether a particle is in the laser beam or not (if these details are not proprietary). Some comment on the "hit rate" – i.e., the fraction of particle signals that were usable or had to be disqualified for both the (spherical) calibration aerosols and the dust particles, which are known to have problems following flow lines – would be useful in constructing a signal estimate, as explained below.

Authors' Response:

We have elaborated on the discussion of the qualifier detector in the manuscript. On page 7, the text now reads, "The qualifier detector has an optical mask which restricts scattered light from particles that are further than 0.75 mm from the center of focus of the laser beam. Particles which are within the depth of field (\pm 0.55 mm either side of the center of focus) are measured using the qualifier detector. The beam splitter which separates the two detectors is split with 70% of the light delivered to the qualifier and 30% delivered to the forward scattering detector. If the qualifier detector signal exceeds the forward scattering detector.

To answer the reviewers comment, it would indeed be interesting to estimate a "hit rate" bit unfortunately it is not possible to compare hit rates between the forward scatter detector and the qualifier detector with the current instrument, as the measured signal from the qualifier detector is not recorded.

Reviewer's Comment:

In looking at Figures 9 and 10, my eyes are trying to suggest that group C is a linear combination of groups A and B. Are there any statistical validations of the assignment of the dust aerosol types to three distinct groups? Is it possible that there is a continuum of types and the authors fortuitously sampled types at either end of that range?

Authors' Response:

The authors would like to disagree with the reviewer's assessment that group C is a linear combination of groups A and B. Focusing on the polarization values shown in Figure 9, Group A has a maximum polarization ratio of 1.0, Group B has a minimum polarization of 1.5. There are no measured polarization ratios in Groups A & B between 1.0 and 1.5. However when looking at Group C, polarization ratios between 1.0 and 1.5 can clearly be seen, with no data for polarization ratios less than ~ 0.75. Therefore the linear combination of Group A & B would not resemble Group C.

Reviewer's Comment:

My last substantive comment is inspired by the beautiful and revealing Figure 6. It appears that a single particle measurement could give virtually any "answer" suggesting that a robust analysis/characterization of the dust aerosols will require a substantial data set. Have the authors attempted a "signal calculation" based on a best case hit rate from the calibration experiments and atmospherically-relevant dust number concentrations to see if that type of data acquisition will be feasible? It seems to me that this estimate would be very useful to include in the final version of this paper to guide potential adopters of the method. (Would it be likely to succeed in an airborne application?)

Authors' Response:

We thank the reviewer for this insightful comment, and have reprocessed several portions of our data in a "signal calculation" as suggested. Figure 6 shows the distribution in total backscattering intensity for single particles of a single size for each dust composition. If we understand this comment correctly, the Reviewer notes that for some dust compositions, the range of backscattering signals observed is very broad. We consider this an advantage, since the observed distribution for an ensemble of particles is characteristic of the dust group to which it belongs, as discussed in the text. However, as the Reviewer points out there is also a serious caveat to this, which is that a single dust particle sampled in the atmosphere may have a backscattering intensity anywhere within the characteristic distribution of backscattering by particles in its group. Thus the CASPOL clearly needs to sample more than one particle for it to provide any information on possible group identification. The question then is just how many particles does the CASPOL need to sample to provide this information, and does that translate to a concentration which might feasibly be sampled in a field application or not?

To answer this question, we employ a thought experiment. We have produced scattering signatures based on an atmospherically relevant number concentration of particles during a high dust event. Iwasaka et al. (1983) report dust concentrations of 50 cm⁻³ (upper atmosphere) and 225 cm⁻³ (lower atmosphere) during an Asian dust event. Similar values have been reported by Gringel and Muhleisen (1977) and Prodi and

Fea (1979). If we assume the conservative value of 50 cm⁻³ and the standard CASPOL flow rate (1.2 L min⁻¹), there would be approximately 60,000 particles within one minute sampling, which seems like a reasonable timescale for sampling within a dust storm. Next we apply the same signature analysis as done throughout this manuscript to a total particle number of 60,000 of each type. The figure below (C1) shows the scattering signatures for total backscatter intensity versus the polarization ratio for the three representative dusts discussed in the manuscript. This analysis uses a total number of particles of 60,000 to represent 1 minutes worth of sampling during a high dust event. Compared to Figure 9 of the manuscript, it can be seen that the shape and position of the signatures here are comparable though not as distinct as in Figure 9. Thus we conclude that the same three Groups may be established, and that such data might be obtained when flying through a dust storm. While speculative, a brief discussion of this thought experiment will be included in the final text.



Figure C1. The total backscatter intensity vs. polarization ratio for representative members of the optical scattering Groups A (hematite), B (white quartz) and C (zeolite) are shown.

Reviewer's Comment:

Reviewer: On page 22419 near the top, the paper by Baumgardner is not currently available, so this section should be rewritten to stand on its own, optimally incorporating a physical basis for the phenomena as mentioned above.

Authors' Response:

Upon more thorough consideration of this complex issue, we feel that the topic warrants a detailed discussion which is interesting in its own right but is beyond the scope of this manuscript. Rather than expanding this section, the section has been removed from the text.

Reviewer's Comment:

Page 22425 line 2 – is the only possible interpretation that bigger particles are more spherical?

Authors' Response:

No. Other factors, such as the scattering properties of various dust types becoming more similar as size increases is a potential factor. The combined factors of variation in size and shape with the refractive index and surface morphology all play a part in determining the scattering response. No changes to the text were made.

Reviewer's Comment:

And line 4 – the absolute differences between the sets are getting smaller, but are the relative differences?

Authors' Response:

The relative differences are also changing at differing rates. No changes to the text were made.

Reviewer's Comment:

Page 22426 line 11 - it is true that size isn't the only dominant factor but the variation in backscatter with size is still bigger than with shape, etc., so this statement seems too strong.

Authors' Response:

The text currently states "Overall, these results indicate that particle size is not the dominant factor in the total backscattering intensity. Other particle properties such as composition, refractive index, morphology, and orientation may also play a significant role in determining the intensity of backscattered light from a particle of given size." This will be changed to "Overall, these results indicate that particle size is influential but not the dominant factor in the total backscattering intensity. Other particle properties such as composition, refractive index, morphology, and orientation may also play a significant role in determining the intensity of backscattered light from a particle properties such as composition, refractive index, morphology, and orientation may also play a significant role in determining the intensity of backscattered light from a particle of given size."

Reviewer's Comment:

There are also some technical corrections that I noted, although this was generally a well-written manuscript: Passages on page 22418 line 9 and 10 and page 22429 line 16 - 18 are not complete sentences and should be fixed.

Authors' Response:

The noted technical corrections have been addressed in the manuscript.

References:

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