

Given that this a manuscript is primarily a review article I approached it as a guide that could be used to justify and drive future developments in the field of polarizations measurements, in general and circular polarization, in particular. Although the majority of my experience is with in situ sensor technology, the measurements of polarized, scattered light is of particular interest to me for all the reasons that the authors highlight.

Reviewer #1 has done a stellar job of identifying a number of areas that need clarification or correction, so there is no point in my reiterating those points, especially since the reviewer caught quite a few that I had missed.

I am mostly writing to encourage the authors to address two questions that I think are relevant to this discussion.

- 1) There is much information that can be extracted from both LP and CP measurements, but such information seems to be predicated on knowing a priori if the particles are spherical with homogeneous composition. In the active sensing realm of measurements, measuring the LP components, the polarization ratio is sensitive to the complex refractive index (RI) as well as the asphericity but changes in the ratio due to asphericity tend to be larger than changes due to the CRI. For measurements of CP in the current application, how can you differentiate changes due to shape compared to changes due to chemistry?

R2 raises an interesting point for which we do not have a clear answer. Active remote sensing is an advantage because the control in the polarization in the incident light. From the viewpoint of observations (specifically of real atmospheric aerosols), we have not encountered much evidence regarding CP. For LP, it is clear that shape and size drives the magnitude of DLP more than composition. For CP, there is not enough evidence to say one way or another. It is possible that in the realm of polarized optical cytology research (detection of biological cells) particularly in the sub-field of stereochemistry, this question has been addressed. We found a large body of knowledge in sophisticated optical modelling, observation and attempts to distinguish non-spherical cells (many are cited in section 3). However, it is difficult to tell to what degree lessons learned in this research area are directly applicable to atmospheric applications, at first sight the mediums (blood plasma in one discipline contrasted with air) and composition (cells and atmospheric aerosols) are just too different.

In section 5, we had the following paragraph :

“Interestingly the distinctive CP found by lidar is in agreement with the theoretical modelling study by Kolokolova & Nagdimunov, (2014) where optically active particles were shown to have non-zero CP and zero linear polarization in the backscattering direction. However, a controlled study (Cao et al., 2011) measuring the degree of LP and CP in pollen backscattering found that both scale with each other following the predictions of Mishchenko & Hovenier, (1995). That

study concluded there is no additional aerosol information by measuring both LP and CP. While these two offer somewhat conflicting conclusions, both highlight that indeed biogenic aerosols do produce circular polarization. Clearly additional studies need to be carried out on the independence of information brought by the fourth Stokes term”

where we attempt to stress that within the little evidence we found, there is seemingly contradictory evidence. But since these studies are so different and there are so few papers focusing on CP, we just can't conclude one way or another. We chose not to add more discussion as it is a bit speculative without additional observational evidence.

An additional point is that passive observations of LP are often made at multiple viewing angles, which provide for sampling of different portions of the aerosol phase function and thus the means to differentiate in a way that cannot be done at backscattered directions alone. While presumably such an approach would be valid for CP as it is for LP, the literature on multiple angle CP measurements is scant.

In summary, we think this is an open question as far as CP and more dedicated studies are needed. We are hopeful that with all the technical advances in the last few years observations of the Full Stokes vector will become more mainstream and thus would inspire more explorations on the information content in CP.

2) Although this review paper specifically discusses CP produced from interactions of unpolarized light with particle ensembles, I think it might have been useful to take some examples from the lidar community who already employ CP to show how these measurements are already highlighting how CP measurements offer complementary information to LP.

It appears the reviewer is aware of CP observations that we were not aware of or have not access to. In the gathering of information for this review, we found that there are just a handful of studies dedicated to measure circular polarization in Earth sciences applications, particularly aerosols. In fact, we found just one observational study (Petjaa et al, 2014, cited) where CP scattered by aerosols was observed and, in this case, an HSRL lidar with an incident polarized laser and CP received was used. This study was singular in that it had independent confirmation of presence of biogenic aerosols. In contrast, there are numerous studies focusing on the measurement aerosol linear depolarization with lidar. In conversations with lidar investigators, we found several reasons why circular polarization observations have not been reported. One of them is that the theoretical study by Mishchenko and Hovenier (1995) (cited) demonstrated that in the backscattering applications (ie. lidar), DLP and DCP are not independent, and one can be derived from the other. We downloaded the lidar data from the Petjaa et al paper and files included both CP and LP backscattering measurements. This seemed contradictory to the

statement in the same paper that only CP was measured. We approached the lidar PI to ask the source of LP data in the lidar files. He confirmed that indeed CP backscattering was measured, and they used the MH equation to derive DLP. He said that because DLP is a more commonly used parameter in the lidar community, it made sense to make it available. This illustrates the attitude in the lidar community of the current usage of polarized observations and why LP seems more favored. Further, because CP is (normally) at least an order of magnitude lower than LP, it has been a technical and operational challenge to attempt CP measurements in the field for many groups. (A version of this explanation has been added prompted by R1 comments. See new text in Section 5 where the data in figure 6 is described)

As result, there have not been many studies focusing on CP associated to atmospheric aerosols at all. From these conversations, it became clear to us that these reasons were part of the general wisdom by which many lidar researchers operate. There has been a few more explorations regarding CP measurements in clouds but we did not dwell much in it. Thus, it is hoped that this work provides an incentive to explore additional polarization properties in atmospheric aerosols.

So, in addressing the R2 question, there are just not enough observations of atmospheric CP (to the extent of our literature research) that can provide a clear an indication one way or another. If the reviewer is aware of such information, we would very much appreciate if he/she could share it so we can acknowledge it.

I think that the authors have done a very comprehensive job of promoting the benefits of CP measurements and this will be a very useful review that will be used for developing future measurement capabilities.

We sincerely thank the reviewer for the kind words.