Summary:
The paper presents an literature review about circular polarization. It explains how circular polarization is generated and which aerosols in the atmosphere produce circular polarization. A few radiative transfer simulations are carried out to investigate the sensitivity of circular polarization on aerosol optical thickness and on aerosol composition. The conclusion is that measurements of circular polarization from space could provide additional information on aerosol compared to measurements of intensity and linear polarization only. The literature review is very interesting to read but it does not include new scientific results. The simulation part is very short and also seems to be not correct (see comment below). For these reasons I can not recommend to publish the paper in its current status, in my opinion major revisions are required.
General:

- It is claimed that technical advances have demonstrated the feasibility to measure the full Stokes vector from space. However, this is not discussed in the text in more detail. The literature review and also the simulations show, that circular polarization (CP) is several orders of magnitude smaller than linear (LP). The authors should provide more information about planned passive instruments that could measure CP with such a high precision from space.
- The RT simulations are not convincing, because they do not include absorption by trace gases (e.g. ozone in UV). The results show higher linear polarization for a scattering angle of 160.8° than for a scattering angle of 112°, which is not expected because the maximum polarization for Rayleigh scattering is at 90° scattering angle. The relative sensitivity of CP to aerosol composition is large, but since the magnitude is so small it will still be very difficult to measure it.

Thank you for your review. The primary purpose of this manuscript is to review the state of the literature pertaining to CP. Such reviews are within the scope of the journal ACP, and there are many recent examples (e.g. Gao et al., 2021, Bright and Lund, 2020, Korolev and Leisner, 2020). The simulations described in the manuscript were included to augment gaps in the literature and further strengthen the case for additional investigations of CP. In any case, the need for 'new scientific results' is not an appropriate metric for the acceptance or rejection of a manuscript in ACP when it is intended to be a review. That said, you did help us notice a mistake in our simulations that we have since corrected. This is described in more detail below.

Answer to General Comments:

Point 1: We respectfully ~~disrespectfully~~ disagree. As a review that introduces a new subject to the wide aerosol community (as it is the case for ACP), we dedicated an appropriate amount of discussion to measurement feasibility. To some extent, this review is a call for additional measurements as well. Certainly there are more aspects to explore and we could have dedicated additional space to discuss with more detail the detection capabilities of a number of instruments that are currently being planned or in design stages. But it would have lengthened the manuscript in a way that would have distracted the readership from the main point of the paper, which is the inherent potential of CP to reveal the physical nature of aerosols.

Point 2: While we agree that RT computations could have been more thorough and realistic (such as including gas absorption), it would have been beyond the scope of a manuscript, and are only meant to illustrate the general features of circularly polarized radiances in aerosols. This type of study has never been published before. Perhaps the closest paper to this study is the Slonaker et al (2005, cited), a conference paper. In this paper and to make a similar point, they did not use trace gas corrections nor considered sensor bands despite that they do discuss simulations for satellite observations. Most sensor systems operating in the UV / Visible / Near Infrared choose spectral bands to be in windows with minimal or correctable trace gas absorption. Our simulations are meant to replicate this characteristic shared with successful aerosol remote sensing missions such as MODIS, VIIRS and POLDER. The simulation figures are meant to express the changes in intensity, LP and CP as a function of matters such as geometry and aerosol load, changes which are clear even without accounting for minimal trace gas absorption. Again, we emphasize that a more dedicated study would be desirable, but this should be the subject of a specific study and published separately. Regarding the issue of angles, we found an error in the equation used to compute the scattering angle. When corrected, the expressed linear polarization is higher closer to a scattering angle of 90˚ in line with expectations for Rayleigh scattering. Thank you for helping us realize this error.

Slonaker, R. L., Takano, Y., Liou, K.-N., & Ou, S.-C. (2005). Circular polarization signal for aerosols and clouds. In H.-L. A. Huang, H. J. Bloom, X. Xu, & G. J. Dittberner (Eds.), *Proc. SPIE* (Vol. 5890, pp. 58900B-58900B–8). https://doi.org/10.1117/12.619576

Specific comments:
l. 165 ff: "For example, exact solutions can be found for 2pr/l << 1 and |mr/l|<< 1 (Rayleigh scattering) ..."-> you are talking here about the scattering matrix elements S_4j and S_i4, which describe circular polarization. For Rayleigh scattering they are exactly 0, because Rayleigh scattering does not cause any circular polarization.

Yes, it is a bit confusing, these are two sentences describing different points. We added a small correction to separate the concepts expressed.

OLD:

"….. For example, with incident unpolarized light (represented as I_{in}[1,0,0,0]) in Equation 1, the system will exhibit CP if the matrix has non-zero S_{4j} and S_{i4} that result in an the outgoing vector with non-zero V_{out}. From the modelling viewpoint, there are very few analytical solutions of the Maxwell's equations that provide the sixteen S_{ij} coefficients. For example…. "

NEW:

"….For example, with incident unpolarized light (represented as I_{in}[1,0,0,0] in Equation 1) onto aerosols, the system will exhibit CP if the matrix has non-zero S_{4j} and S_{i4} that result in an the outgoing vector with non-zero V_{out}.

From the modelling viewpoint, there are very few analytical solutions of the Maxwell's equations that provide the sixteen S_{ij} coefficients. For example…. "

Fig. 3 (caption): "right panel" should be "left panel" and reference to "right panel" is missing

DONE

l**. 298: "This concept has been applied in astronomy studies where the observations of linear and circular polarization in comets and interstellar dust" -> What is the observed degree of linear/circular polarization in interstellar dust?**

The Rosenbush et al, 2007 reported CP up to 0.8% in comets. In the introduction of Whitnet and Wolff (2002, cited) , several observational papers are cited with observed CP ranging 1-17% in dust associated to protostars.

l. 325: "Lidars with CP detection capabilities have been proposed for cloud phase" -> How can cloud phase be detected with circular polarization? Please explain.

The cited papers in this sentence show modeling studies of linear and circular polarization lidar observing scenarios for incident polarized signals. They show that the different shapes in ice and liquid particles result in both LP and CP. This is explained in the cited references, and we do not think it is relevant to put this additional information here as this is an aerosol focused review.

l. 329: "... (Gilbert and Pernicka, 1967; Lewis et al., 1999) and foggy atmospheres (van der Laan et al., 2017). These studies highlighted the fact that in high- and low-density particle environments, the propagation of circular polarization does not degrade as quickly as linear polarization." -> Why should linear polarization degrade quickly in low-density particle environments? Why does circular polarization degrade slower than linear?

We agree with the reviewer that this is a bit misleading. The papers cited are concerned with very turbid environments and the simulations presented in those studies are for high optical depths (>1) , large particles (fog and marine sediments) and for incident light with a defined polarity (ie incident LP or CP). One paper (Gilbert and Pernicka, 1967, cited) reports laboratory observations. The van der Laan et al (2018) study (not cited) make this point very clear (see respective figures 4 and 12) . They show that the persistence of DCP with respect to DLP as a function of AOD in visible and NIR wavelength ranges. The statement regarding loss of LP in low density environment is in the context of very turbid environments which is not the case of interest here. Also, note the van der Laan et al (2018) computations do not include the range AOD =0 to 1. This is the range considered in our study. The RT simulations shown in section 6 are the first time to our knowledge where this range of aerosol concentrations are considered.

We modified the respective paragraph to make this clearer:

OLD
"...... and foggy atmospheres (van der Laan et al., 2017). These studies highlighted the fact that in high- and low-density particle environments, the propagation of circular polarization does not degrade as quickly as linear polarization. "

NEW
"...... and foggy atmospheres (van der Laan et al., 2018). These studies highlighted the fact that in high-concentration environments (AOD>1), the propagation of circular polarization does not degrade as quickly as linear polarization."

J. D. van der Laan, J. B. Wright, S. A. Kemme, and D. A. Scrymgeour, "Superior signal persistence of circularly polarized light in polydisperse, real-world fog environments," Appl. Opt., vol. 57, no. 19, p. 5464, Jul. 2018.

l. 395: "inagreement" -> "in agreement"
DONE

l. 395: "Interestingly the distinctive CP found by lidar is inagreement 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" -> What are the different assumptions in the studies by Kolokolova and Nagdimunov 2014 and Mishchenko&Hovenier 1995? I assume these are theoretical studies, so there should be a simple explanation for the conflicting conclusions?

There are contrasting differences between both studies, and it is not entirely clear the attribution of the source of those differences. But just to mention a few, the MH95 paper uses simple spherical particles, and it takes advantage of the symmetry in the scattering matrix at the backscattering angle. The KN14 study uses aggregates of small spheres made of chiral material and it does not use symmetry arguments in the calculations. They used a rather sophisticated code (a multi-sphere T-Matrix code, from Mackowski et al., 2011) to calculate the optical properties to report results whereas MH95 only reports calculations from simple equations. We do not think this is the place to elucidate why as we can only speculate without doing a more in-depth study.

However, R1 raised a similar comment regarding this section and the lack of additional explanations. We expanded this section additional text. We refer to the respective answer to R1 for more context. Here we add the new paragraph in this section:

OLD

“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. ”

NEW

“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 the authors did not set out to seek for CP signals in biogenic aerosols, there could be plausible reasons why no positive CP was observed. These include aging of the pollen (chirality disappears with aerosol senescence as shown by Salma et al, 2010 and in decaying leaves Lucas Patty et al, 2017), not enough representative samples and instrument sensitivity. This result should be confirmed as it appears (to the extent of our search) the only study found in the literature measuring atmospheric aerosol LP and CP simultaneously. While these two studies (one theoretical, the other experimental) 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."

Mackowski, D., Kolokolova, L., & Sparks, W. B. (2011). T-matrix approach to calculating circular polarization of aggregates made of optically active materials. Journal of Quantitative Spectroscopy and Radiative Transfer, 112(11), 1726–1732. https://doi.org/10.1016/j.jqsrt.2011.02.003

l. 429: Eq. 4 (circular polarization after 2 scattering events) -> This equation is not very special (included in all VRT codes handling circular polarization). Why is this equation derived and written down here, it is not used at all in the discussion of the results?

We do think that this equation is special in the sense that it provides one of the few analytical ways to illustrate with an equation how CP is linked to incoming radiation. The fact that is included in RT codes does not make it any more accessible or understandable to the scientific community. We included this equation because for much of the intended audience of this paper may not be as versed in the intricacies of RT codes and for whom, polarized radiative transfer computations are generated by complicated modeling tools.

l. 447: "The particle shape is assumed to be spherical." -> In the discussion you highlighted the importance of particle shape and orientation for circular polarization. Then, in the model simulations it is neglected. At least randomly oriented aspherical particles can be handled in most state-of-the-art VRTE models...

We certainly agree that this would be the ideal case, but this option was not available to us (computer power, reliable non-spherical code) at the time of carrying out the simulations.

l. 449: "The atmospheric column only contains air (i.e., no trace gases)" -> Why modelled without trace gases? Calculations without O3-absorptions are very unrealistic in the UV-range. To my understanding "air" normally includes also the trace gases.

We think that as a first attempt to study aerosol CP propagation in the atmosphere at representative wavelengths, this exercise is adequate. Yes, in real observational settings there are absorbing gases and other sources of noise. But as at first of its kind, we wanted to focus on the possible signals. Certainly, additional more realistic scenarios should be tried. We hope these computations are enough to trigger the curiosity to explore this subject in more detail.

Figs. 7/8: What is the definition of the scattering angle? Normally it is given by cos(theta_s)= cos(n_inc * n_sca), where n_inc and n_sca are incoming and scattered directions, respoectively? I doubt that Fig. 7 is for scattering angle of 160.8 degrees. LP by Rayleigh scattering has a maximum around 90° scattering angle, so I would expect much higher LP in Fig. 8 (scattering angle 112°) than in Fig. 7 (160.8°, backscattering direction).

We thank the reviewer for bringing up this point. Verifying computations that have never been reported before can be a challenge. As the reviewer points out DLP at near the 90-degree scattering angle should be the highest compared to other scattering angles. This is expressed by equation

DLP = (Iper - Ipar)/(Iper + Ipar)
= sin(theta)^2 / (1+ cos(theta)*cos(theta))

where Iper stands for radiance measured perpendicular to the scattering plant and Ipara is the parallel component of the radiance. Theta is the scattering angle as defined in Appendix Figure 1. (See Coulson book, Page 179 , Eq 4.25)

So, we rechecked our computations and we found a mistake in the equation we used to compute the scattering angle (a minus signed was misplaced). After correcting the equation and recomputing the angles, the new angles changed to:

SCAT_ANG(old) = 160.8 changed to SCAT_ANG(new) = 103.2
SCAT_ANG(old) = 112.3 changed to SCAT_ANG(new) = 142.5

These new angles are in accordance to what above equation with DLP(103.2) > DLP(142.5) as figures 7 and 8 show at AOD=0 and demonstrate that our computations are in accordance to what is expected.

Coulson, K. L. (1988). Polarization and intensity of light in the atmosphere. Hampton, Va., USA: A. Deepak Pub.

l. 522 "Overall, these plots suggest there is a sensitivity to differences between coarse and fine mode dominated aerosols in both linear and circular polarization." -> Since CP is scaled with 10â´, I assume that even if the relative sensitivity is higher in CP, it can still be better measured in LP? I can not see the benifit of measuring CP.

We disagree with R3 in that we think there is a benefit to explore the measurement of aerosol CP. Again, while we acknowledge that the RT simulations are not as complete as R3 wished, the evidence shown in the first two thirds of the paper plus these limited (but entirely new) RT simulations suggest when looking at all this information together, there are lines of research of interest and unexplored in the subject of aerosol polarization. As far as the magnitude of DCP, it is certainly a low magnitude but not null and it is the thesis of this review that technological advances are improving at a rapid pace such that no only CP measurements can be made in laboratory settings but also remote (as shown in the Petjaa et al study cited in the paper). So it is appropriate to start to explore the benefits of measuring CP as it appears suitable technologies are already available .

Fig.9, labels in right panels: U->V

Corrected

What is the scattering angle of the simulations shown here?

Same as in figure 8. It is indicated in the respective caption.

Author contribution: What is the contribution of KK?

The text corresponding to KK contribution was missing.

OLD

SG carried literature gathering and review, performed the simulations, carried out the analysis, created the figures and wrote the paper.

NEW

SG carried literature gathering and review, performed the simulations, carried out the analysis, created the figures and wrote the paper. KDK reviewed several versions of the manuscript, provided several key suggestions and background material.