The work of Santiago Gassó and Kirk David Knobelspiesse on the “Circular Polarization in Atmospheric Aerosols” addresses the not-extensively-researched theme of the remote sensing of aerosols in the atmosphere using circular polarization measurements. The discussion is quite interesting for a remote sensing scientist, involving a literature review on the subject along different scientific fields, and highlighting the importance of circular polarization measurements for monitoring biogenic and oriented non-spherical particles in the atmosphere, as well as monitoring aerosols in multiple-scattering mediums. This work was a pleasure to read and review and it should be published in ACP.

That being said, some parts of the manuscript need explaining and better phrasing, as indicated below.

I should also note that my expertise does not cover the field of radiative transfer calculations, thus my review on Section 6 may miss possible problematic points.

Lines 22-28, “Current techniques... this subject.”: Include also the reasoning for using circular polarization measurements for better typing/characterization at low concentrations.

Lines 39-42, “As a... Wei et al., 2020)”: Please rephrase.
Lines 50-51, “While existing… (BOA)”: Line 49 states that the “remote identification is still inadequate…”. Then you state that the “existing methods can identify smoke etc..”. Please rephrase, or provide a measure of uncertainty for the results of the “existing methods”.

Line 71, “…such biological…”: Replace with “…such as biological…”.

Line 75, “…important in public health…”: Replace with “…important for public health…”.

Lines 73-74, “These are… Levy et al., 2013)”: Move this phrase to the previous paragraph.

Line 82, “adequate pixel size”: Do you mean “non-adequate pixel size”?

Lines 86-88, “Like the intensity-only… loading conditions”: Please provide reference(s) for this statement.

Line 89, “…remote sensing sensors using polarization technologies…”: Replace with “…remote sensing sensors onboard satellite platforms using polarization technologies…”

Lines 139-141, “The difference in… of traversing light.”: Replace with “The difference in propagation speeds in each projection of the travelling wave results in changes in phase and magnitude of each of the observed components of the propagating light. The result of these differences is the elliptical polarization of light, with linear and circular polarization to be particular cases. Polarization thus results from the differential speeds of the wave components of the traversing light.”

Lines 143-144, “…CP can arise …. and non-spherical.”: Provide reference.

Lines 145-146, “In addition… orientation.”: Replace with “In addition, unpolarized light incident upon an ensemble of particles can result in outgoing CP if multiple scattering occurs in the medium regardless of the particle type, shape, and orientation.”

Line 147, “… or any identifying observation of the system.”: What do you mean with the phrase “or any identifying observation of the system”? Please clarify and/or rephrase.
A polarized beam is represented by the Stokes column vector. The Stokes vectors for the incident and outgoing light are related by the 4x4 scattering matrix (S) that represents the scattering medium as:

The elements $S_{4j}$ (bottom row in equation 1) and $S_{i4}$ (right most column) are associated to the circular polarization of the scattered light and can be used as....

For non-spherical particles,... particle shapes.: Include discussion on the numerical solutions as well (e.g. ADDA calculations for dust particles in Gasteiger et al. (2011)).

However,... Section 6).: Rephrase, referring to this light as multiple-scattered light.

Similarly... in matrix 2.: Provide the scattering matrices for the ensemble of chiral particles, and the ensemble of chiral particles that is made up of an equal number of left- and right-handed particles.

This is confusing... Can chiral particles have spherical shapes? Please clarify here.

This work is not included in the list of references. Please add.

Include also the marine particles (e.g. Haarig et al., 2019) and pollen.

Provide reference.

Kolokolova and Nagdimumov (2014) also proposed a methodology for differentiating aligned particles from optically-active particles. Please include this info here, as well.

Provide reference, or explain more, to support
for this statement.

Lines 312-313, “This includes... (Daskalopoulou et al., 2021).” The studies of Harrison et al. (2018) and Daskalopoulou et al. (2021) do not prove that the particles passively "remain charged" far from the sources. Instead, Harrison et al. (2018) refers to "an active charging process" and provides the triboelectrification as a possible cause. Please include more info here from the respective papers.

Lines 320-322, “Since... as well.” Replace with "Since similar mechanisms of particle orientation seem to be present in the Earth’s atmosphere, and it may well be possible that these oriented particles scatter circular polarized light as well.”

Lines 320-321, “Since... atmosphere.” Support this statement with more info.

Lines 325-326, “Lidars... Hu et al., 2003).” Include also the polarization lidars discussed in Paschou et al. (2022) and Tsekeri et al. (2021).

Lines 331-333, "Recent... aerosols." There is no "Martin et al. (2010)" in the reference list. If you mean the work of Martin et al. (2016), they do not provide measurements of mineral aerosol. Please correct.

Line 350, “...by two different spores as reported by...” Replace with “...by two different spores illuminated with linearly-polarized light, as reported by...”.

Lines 351-352, “Further... matrix” Replace with “Further this is not the only non-zero element S_{4j} or S_{44} of the scattering matrix.”.

Line 357, “Additional studies...” Do you refer to laboratory studies? Please clarify.

Lines 360-366, “For example, ... overlooked“ Please rephrase.

Caption of Fig. 6: Provide more details on what we see in the plots. At which heights the “particle concentrations are rather high” and the “aerosol composition is consistent with organic aerosols”? Discuss the effect of particle hydration below 1-1.5km.
Lines 391-394, “Figure 6... (Lee et al., 2018).”: The circular polarization is low in the PBL, probably due the high RH and the hydration of the aerosol particles. Moreover, if I understood correctly form the plots, high values of CP are found for low particle concentrations (higher up). Re-write this part, being more specific on what we see in Fig. 6.

Lines 399-400, "While these two... circular polarization.": Please provide more explanations on why the study of Cao et al. (2011) showed no optical activity for bioaerosols (e.g. due to noisy measurements?). These results contradict a big part of the discussion in the manuscript, thus a thorough explanation should be provided here.

Lines 405-406, “…there are no studies... the atmosphere”: There is the study of Petaja et al. (2016). Please rephrase.

Lines 418-420, “...as well as... matrix.”: Replace with “as well as consideration of the effect of chiral or oriented particles on the extinction matrix, resulting in the dichroic extinction of the propagating light.”

Lines 441-448, “The size distribution... to be spherical.”: Do these aerosol models correspond to specific aerosol types? Please discuss.

Lines 496-497, “However, ... by a factor of 2-3.”: Provide explanation why.

Line 504, “… this is the range of concentrations most globally prevalent...“: I do not think this is necessarily true for all aerosol types. Please provide reference to support this statement.

Line 615, “… in Sij is assumed and omitted for simplicity.”: Replace with “… in Sij is assumed and omitted for simplicity in Eq. A2 and A3.”

Line 632, “… resulting in:” : Replace with “… resulting in Eq. A3.”

Line 634: Number the equation as Eq. A3.