Reply to R1

We thank R1 for the editing suggestions and insightful comments. Some of them prompted us to go back and think on aspects of the manuscript that we did not think about. So, we appreciate the intellectually stimulating commentary. We addressed all the points and provided answers or just commentary to some of the points that invited discussion. We did modify the manuscript based on the feedback and we believe the manuscript has been strengthened . We hope the R1 agrees.

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

We made a small change in the abstract to address this concern as we did not want to lengthen the abstract too much.

OLD:
When the same aerosols are found at lower concentrations (but still high enough to be of importance for air quality and other concerns), these methods often produce ambiguous results. Thus, the exploration of additional optical techniques is of interest to improve remote detection of aerosol composition. This study is presented as an overview with a goal to provide a new perspective on an overlooked optical property and to trigger interest in further exploration of this subject.

NEW

When the same aerosols are found at lower concentrations (but still high enough to be of importance for air quality and **cloud formation**), these methods often produce ambiguous results. The circular polarization of aerosols is rarely utilized, and we explore its value for improved determination aerosol composition. This study is presented as an overview with a goal to provide a new perspective on an overlooked optical property and to trigger interest in further exploration of this subject.

Lines 39-42, "As a… Wei et al., 2020)": Please rephrase.

We rephrased the sentence. Now it reads

OLD:
As a result, the total amount of aerosols can now be quantified and thus provides an observational constraint for aerosol transport models and aids in decision-making for different fields such air quality and public health (Duncan et al., 2014; Holloway et al., 2021; Mhawish et al., 2018; Sorek-Hamer et al., 2020; Wei et al., 2020)

NEW:

“With this retrieved quantity, it is now possible to quantify with satellite observations the total amount of atmospheric aerosols globally. Aerosol global transport models now have such observations available and are used to constraint model initializations or validate model outputs. In addition, they contribute to more focused and applied decision-making fields such air quality and public health (Duncan et al., 2014; Holloway et al., 2021; Mhawish et al., 2018; Sorek-Hamer et al., 2020; Wei et al., 2020)”

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”.

We choose to rephrase the sentence and now it reads as less controversial.

OLD:
“While existing methods can identify smoke, dust, volcanic ash, and pollution aerosols (Kahn and Gaitley, 2015; Li et al., 2019b; Mhawish et al., 2018), they fail to identify an important aerosol group: biogenic organic aerosols (BOA).”

NEW:
“Standard aerosol detection techniques can identify several aerosol types (here interpreted as a proxy for aerosol composition) such as smoke, dust, volcanic ash, and pollution aerosols (Kahn and Gaitley, 2015; Li et al., 2019b; Mhawish et al., 2018). However, an important aerosol group is not detected by these techniques: biogenic organic aerosols (BOA).”

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

Done

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

Done

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

Done

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

Replaced with “a large pixel size”

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

We recognize the sentence is a bit too opinionated in that it is more based on personal experience and partial evidence. A brief review of available information does appear to support the statement. Here are a few facts:

Aeronet retrievals of refractive index only occur at AOD(440)>0.4 because this is the minimum threshold advised by the Aeronet team for reliable composition retrievals. When no microphysical properties are available (known as Inversion data in Aeronet terminology), a dedicated study using Aeronet direct retrievals (Sayer et al 2012) recommended to use a screen selection of AOD<0.2 and AE<1.0 for marine aerosol detection. Russell et al (2014) introduced a methodology for aerosol typing identification. They presented an aerosol typing scheme using linear polarization from the satellite sensor POLDER 3 over the ocean and successfully identified aerosol types at lower than 0.2 AOD magnitude. However, the methodology was applied to one region over the ocean and this approach does not appear to be of regular use in the community. For MISR, the recommended threshold of aerosol type identification is 0.2 (Kahn and Gaitley, 2015). Another good example is provided by the Schutgen et al (2021) study where several satellite groups contributed to the analysis. This analysis included data from both intensity only and intensity + linear polarization observations. The goal was to evaluate and compare satellite observations of aerosol type proxies. To have certainty that the satellite obs provided a “truth” against which models could be compared, the authors chose to exclude satellite retrievals with AOD<0.3 . The same authors explore the choice of threshold to find the right value which depended on the sensor, retrievals algorithm used , surface and number of comparison points (See figure 12 in Schutgen et al (2021). Future sensors with LP measurement are expected to perform better at distinguishing aerosols but the uncertainties associate to intense properties to identify aerosol type vary. For example, the Knobelspiesse et al, (2012) modeling studying showed that the retrieval uncertainty in imaginary index refraction is significant for AOD <~0.2 although in the case of single scattering albedo (SSA) appears to be low enough for distinguishing among aerosol groups.

Overall, because there is no single aerosol composition marker in measured radiances, current aerosol operational algorithms identify aerosol groups indirectly. Such labeling arises from spectral and angular variability (and in some cases linear polarization magnitude) observed and an aerosol type is selected following a decision tree or statistical method in the respective algorithms. Generally, aerosol type (or aerosol model selected by the algorithm) is reported in the output file. In those cases where the algorithm developer team describe the product, they recommend using a threshold in AOD for reliable aerosol group identification.

We find that properly backing up this statement may imply to add too much information to support it and since it is not essential, we slightly modified the paragraph to make it less controversial.

Russell, P. B., Kacenelenbogen, M., Livingston, J. M., Hasekamp, O. P., Burton, S. P., Schuster, G. L., et al. (2014). A multiparameter aerosol classification method and its application to retrievals from spaceborne polarimetry. Journal of Geophysical Research: Atmospheres, 119(16), 9838–9863. https://doi.org/10.1002/2013JD021411

Schutgens, N., Dubovik, O., Hasekamp, O., Torres, O., Jethva, H., Leonard, P. J. T., Litvinov, P., Redemann, J., Shinozuka, Y., de Leeuw, G., Kinne, S., Popp, T., Schulz, M., and Stier, P.: AEROCOM and AEROSAT AAOD and SSA study – Part 1: Evaluation and intercomparison of satellite measurements, Atmos. Chem. Phys., 21, 6895–6917, https://doi.org/10.5194/acp-21-6895-2021, 2021.

Sayer, A. M., Smirnov, A., Hsu, N. C., & Holben, B. N. (2012). A pure marine aerosol model, for use in remote sensing applications. Journal of Geophysical Research: Atmospheres, 117(D5), n/a-n/a. https://doi.org/https://doi.org/10.1029/2011JD016689

OLD :

These are features shared by most retrieval schemes using multispectral passive sensor with a single view, such as the algorithms applied to MODIS and VIIRS (Hsu et al., 2013; Levy et al., 2013) . There are, however, more sophisticated sensors that have better aerosol type detection capabilities. For example, angular variability of the spectral radiance (MISR, Diner et al., 2005), active remote sensing (CALIOP, Winker et al., 2010) and polarization (POLDER, Herman et al., 2005; Li et al., 2019) are used to better discriminate between aerosol types, but other technical impediments (limited spatial coverage, adequate pixel size) prevent an effective global coverage. Two sensors (POLDER and CALIOP) are highlighted as the first operational sensors to make observations of linearly polarization (LP) of the Earth surface-atmosphere system. The addition of polarization capabilities demonstrated that aerosols can be better identified by their type. Particle refractive index and particle shape (proxies for aerosol composition) have been successfully used to determine aerosol types where polarization-insensitive methods have fallen short (Dubovik et al., 2011; Omar et al., 2009; Russell et al., 2014; Stamnes et al., 2018). Like the intensity-only passive sensors, these remote sensing technologies are not very effective for aerosol type identification in low-to-moderate loading conditions.

NEW:

These are features shared by most retrieval schemes using multispectral passive sensor with a single view, such as the algorithms applied to MODIS and VIIRS (Hsu et al., 2013; Levy et al., 2013) . There are, however, more sophisticated sensors that have better aerosol type detection capabilities. For example, angular variability of the spectral radiance (MISR, Diner et al., 2005), active remote sensing (CALIOP, Winker et al., 2010) and polarization (POLDER, Herman et al., 2005; Li et al., 2019) are used to better discriminate between aerosol types, but other technical impediments (limited spatial coverage, adequate pixel size) prevent an effective global coverage. Two sensors (POLDER and CALIOP) are highlighted as the first operational sensors to make observations of linearly polarization (LP) of the Earth surface-atmosphere system. **The addition of (linear) polarization capabilities provide for improved determination of aerosol optical properties as a proxy for composition (Knobelspiesse et al., 2012).** Particle complex refractive index and particle shape have been successfully used to determine aerosol types where polarization-insensitive methods have fallen short (Dubovik et al., 2011; Omar et al., 2009; Russell et al., 2014; Stamnes et al., 2018). Like those that use intensity-only passive sensors, the capability of these remote sensing techniques depend on the amount of aerosols, and are less effective for aerosol type identification in low-to-moderate loading conditions.

Knobelspiesse, K., Cairns, B., Mishchenko, M., Chowdhary, J., Tsigaridis, K., van Diedenhoven, B., Martin, W., Ottaviani, M., and Alexandrov, M.: Analysis of fine-mode aerosol retrieval capabilities by different passive remote sensing instrument designs, Opt. Express, 20(19), 21457-21484 , 2012.

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

Done

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."

Done

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

Done. Added Van de Hulst, 1957

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."

Done

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.

Done.

OLD:
"…..it is less clear if there is a potential way to link observed CP with the aerosol type under observation or any identifying observation of the system."
NEW
" …it is less clear the potential to link the observed CP with a specific aerosol type under observation."

Lines 149-151, "A polarized beam… medium as:", Replace with "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:".

Done

Lines 160-161, "The elements… can be used as…": Replace with "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 ….".

Done

Lines 168-171, "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)).

Done, Added the Gateiger et al and Kempinnen et al references.

Lines 189-192, "However,… Section 6).": Rephrase, referring to this light as multiple-scattered light.

Done

Lines 199-201, "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.

Done. We added the matrix for the case of ensemble of chiral particles with an excess of left or right particles. For equal number is the same as Matrix 2.

Lines 238-239, "Exact solutions… (Bohren, 1975).": This is confusing… Can chiral particles have spherical shapes? Please clarify here.

Yes, chiral particles can be spherical as shown by Bohren. The chirality arises because asymmetry in the molecules that make the solid sphere. This asymmetry is expressed in the dielectric constant for the molecule. For non-chiral particles, the dielectric constant is assumed the same for all directions in the solid (ie $\varepsilon_{xx}=\varepsilon_{yy}=\varepsilon_{zz}=\varepsilon_{ij}$). The connection with the bulk optical properties of the solid is through the dielectric constant which is directly related to the square of module of the refractive index. In matrix representation, the dielectric constant is represented by an identity matrix multiplied by ε_o. In the case of a chiral particle, ε_{xx} is not equal to ε_{yy} and the optical property of the solid is represented by a non-identity matrix with at least two different coefficients. Bohren (1975) shows that this directional asymmetry in the solid results in two different indexes of refraction associated to this asymmetry in the dielectric constant representation. In practical terms, when using Bohren scattering code for chiral particles, the user has to input two refractive indexes.

We added the following few words in the respective paragraph that we hope makes it a bit clearer and avoids a lengthy explanation:

OLD:
" Exact solutions for chiral particles were found for the case of homogenous (Bohren, 1974) and layered spheres (Bohren, 1975)."

NEW

"Exact solutions **for solid particles made of chiral molecules** were found for the case of homogenous (Bohren, 1974) and layered spheres (Bohren, 1975)."

Line 263, "Rosenbush et al., 2007": This work is not included in the list of references. Please add.

Added

Line 286, "Two of the main … while airborne.": Include also the marine particles (e.g. Haarig et al., 2019) and pollen.

Done

OLD
Two of the main aerosol types (dust and smoke) in the atmosphere are known to be non-spherical at different stages while airborne. For example, freshly formed smoke contains

abundant chains of coagulated soot particles (Chakrabarty et al., 2014; China et al., 2013; Girotto et al., 2018).
NEW
There are aerosol types known to be non-spherical at different stages while airborne. For example, pollen shape inhomogeneity and dry marine aerosols result in detectable linear polarization (Haarig et al,2019). Also freshly formed smoke contains abundant chains of coagulated soot particles (Chakrabarty et al.,2014; China et al., 2013; Girotto et al., 2018).

Line 297-298, "In this case,… electrical field.": Provide reference.

Done. Added Mallios et al, 2021 reference

Mallios, S. A., Daskalopoulou, V., & Amiridis, V. (2021). Orientation of non spherical prolate dust particles moving vertically in the Earth's atmosphere. Journal of Aerosol Science, 151, 105657. https://doi.org/https://doi.org/10.1016/j.jaerosci.2020.105657

Lines 301-304, "Further… 90 degrees.": Kolokolova and Nagdimunov (2014) also proposed a methodology for differentiating aligned particles from optically-active particles. Please include this info here, as well.

The methodology proposed by KN2014 to differentiate aligned particles from chiral particles is mentioned in a later section 5 where actual observations of chiral particle are discussed.

Lines 308-309, "In these cases… a fire.": Provide reference, or explain more, to support for this statement.

This comment prompted us to search for more evidence on the presence of electrical fields in in smoke from fires. We found a few references (all from Russian origin, we did not find similar studies carried out in the Western hemisphere) reporting ambient electrical field variability associated with smoke from forest fires. So, we rewrote the paragraph as follows:

OLD:
" Also, intense electrical fields are present in large fires so strong that they generate their own lighting (Dowdy et al., 2017; LaRoche and Lang, 2017). In these cases, it is conceivable to consider the presence of alignment of smoke particles immediately downwind from a fire."

NEW:
"Further, smoke from fires have associated variability in the local atmospheric electrical field as it has been observed in Siberia (Ippolitov et al., 2013; Phalagov et al., 2009). This variability was associated to changes in aerosol load, ambient moisture and actinic flux (Nagorskiy et al., 2014). The presence of electrical fields and the fact that fresh smoke particles are non-spherical aggregates suggests the possibility of charge distribution within particles and possible alignment with the ambient electrical field "

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.

We modified the paragraph as suggested:

OLD
“,particles remain charged during long distance transport and persist for significant distances. This includes reports of electrical fields in Saharan dust clouds over the UK (Harrison et al., 2018) and in Greece (Daskalopoulou et al., 2021).”

NEW
“There are additional interesting features to note. While it’s been known that electrical fields are present during dust emission and play a role in the amount of dust lifted (Esposito et al., 2016; Kok and Renno, 2008; Zhang and Zhou, 2020), non-background electrical fields are also found in dust clouds at significant distance from the source, such as dust reaching Greece and the UK (Harrison et al., 2018, Daskalopoulou et al., 2021). These studies suggested triboelectrification (i.e. friction between particles) as mechanism for generation of an electric field within the cloud during transit.”

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.”

DONE

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

Since both this comment and the previous one is similar, we rephrased the sentence hoping that it addresses both points:

OLD
“In summary, circular polarization has been observed originating in interstellar dust. Only by assuming particle alignment in the presence of an electromagnetic field the optical properties can be modelled to match observations. Since similar mechanisms of particle orientation are present in the Earth’s atmosphere, and it may well be possible that these oriented particles scatter circular polarized light as well.”
NEW
“In summary, circular polarization has been observed originating in interstellar dust and in this case only by assuming particle alignment in the presence of an electromagnetic field the optical

properties can be modelled to match observations. Since electrical fields are present in dust clouds in the Earth's atmosphere, it may well be possible that these oriented particles scatter circular polarized light as well."

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

DONE

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.

DONE

Line 350, " …by two different spores as reported by…": Replace with " …by two different spores illuminated with linearly-polarized light, as reported by…".

DONE

Lines 351-352, "Further… matrix": Replace with "Further this is not the only non-zero element S_{4j} or S_{i4} of the scattering matrix.".

DONE

Line 357, "Additional studies…": Do you refer to laboratory studies? Please clarify.

We modified the paragraph slightly to address this point:

OLD:
"Additional studies reported optical properties of hydrosols (a.k.a aerosol in aqueous environments) such as chloroplasts (Gregory and Raps, 1974) and dinoflagellates (Liu & Kattawar, 2013; Shapiro et al., 1990, 1991). They measured CP in commonly found marine aerosols (a.k.a hydrosols). Hydrosols are precursors of atmospheric biogenic organic aerosols (BOA) in the marine environment, and they are not unique in exhibiting chiral signatures. For example,…"

NEW:
"Additional laboratory studies reported optical properties, including CP, of hydrosols (a.k.a aerosol in aqueous environments) such as chloroplasts (Gregory and Raps, 1974) and dinoflagellates (Shapiro et al., 1990, 1991). These particles are also commonly found in the atmosphere and are considered biogenic marine aerosols. For case of dinoflagellates, Liu & Kattawar, (2013) used an ADDA code to simulate the helicoidal shape of the chromosome (a

chiral structure) in the phyoplankton to calculate the 16 Mueller matrix elements. They found a non-zero S_{14} element in backscattering conditions consistent with previously reported observations. They suggested backscattering observations of S_{14} as a possible method to detect dinoflagelates present in red tides. For atmospheric applications, it should be noted that hydrosols are ejected or are precursors of atmospheric biogenic organic aerosols (BOA) in the marine environment, and they exhibit chiral signatures. For example,…

Lines 360-366, "For example, … overlooked": Please rephrase.

We agree the paragraph was confusing , we re-wrote it as follows:

OLD:

"For example, organic films containing amino acids attach to ejected marine aerosols and were detected and classified according to their chiral structure (Kuznetsova et al., 2005; Wedyan and Preston, 2008). Further, they do have spectro-polarimetric signatures.

As far as specific reports of observations of CP in atmospheric aerosols or presence of aerosols with chiral elements, the available evidence does indicate that biogenic aerosols contain chiral materials which suggests they should have distinctive CP signatures. Although polarized light has been proposed as a tool to improve visibility observation in the atmospheric marine boundary layer (Quinby-Hunt et al., 1997), the concept of CP detection was seemingly overlooked."

NEW

"For example, Kuznetsova et al., (2005) and Wedyan and Preston, (2008) measured the chirality of amino acids contained in organic films of ejected marine aerosols.

With regards to reports of observed CP in atmospheric aerosols, the available evidence is limited because there are very few studies specifically focusing on the V Stokes parameter. For example, although polarized light has been proposed as a tool to improve visibility observations in the atmospheric marine boundary layer (Quinby-Hunt et al., 1997), the concept of using CP for the purposes of detection has been overlooked. However, as we show, there is a reasonable amount of evidence that biogenic aerosols do contain chiral materials, and this suggests these aerosols should have distinctive CP signatures."

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.

Caption of Fig 6 does not have such text. We interpret you mean lines 391-394

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.

We agree that the description and explanation provided for figure 6 is not adequate and it can lead to misinterpretation. For example, figure 6 shows low Depolarization , not low polarization as the reviewer points out. So, we added much more information and put more context to interpret the image. We also slightly changed our conclusions in relation to the figure. We also edited the figure caption to clarify how to interpret the observed depolarization CP.

OLD

"Perhaps one of the more compelling observations of aerosol's circular polarization in the Earth's atmosphere is reported by Petäjä et al., (2016) . The BAECC (Biogenic Aerosols—Effects on Clouds and Climate) Campaign was carried out during Spring and Summer, 2014 in the Finish arctic forest, and it focused on characterizing the role of biogenic aerosols in cloud formation. The campaign included several examples of collocated surface lidar observations and aircraft overpasses at a time of the year where pollen and other biogenic aerosols are abundant. Figure 6 shows an example from this study where distinctive CP (here defined as ratio of left circular return divided by right circular return) was measured by ground-based HSRL polarization lidar with in-situ confirmation of high aerosol concentrations and composition consistent with organic aerosols in the boundary layer (Lee et al., 2018). "

NEW

"Perhaps one of the more compelling observations of aerosol's circular polarization in the Earth's atmosphere is reported by Petäjä et al., (2016). The BAECC (Biogenic Aerosols—Effects on Clouds and Climate) Campaign was carried out during Spring and Summer, 2014 in the Finish arctic forest, and it focused on characterizing the role of biogenic aerosols in cloud formation. The aircraft deployed included in-situ composition measurements and it operated near a ground based HSRL lidar that specifically measured circular polarization (personal communication, Robert Holtz , lidar operator) as opposed to linear polarization, the more standard way to observe pollen with lidars (Sassen, 2008; Shang et al., 2022). The campaign included several examples of collocated surface lidar observations and aircraft overpasses at a time of the year where pollen and other biogenic aerosols are abundant. The resulting dataset is unique in that it contains remotely observed CP along with in-situ confirmation of biogenic aerosols, known to contain chiral molecules. Figure 6 shows an example from this study where aircraft in-situ instrumentation data and the corresponding lidar profile are displayed. The left column panels (A and C) show vertical profiles of high CCN and aerosol concentrations in the boundary layer (panel A) and composition measurements demonstrating the high organic carbon content (green line in Panel C) in the boundary layer, consistent with biogenic aerosols (also confirmed by Lee et al., (2018) in this campaign). Panel B shows the lidar profile (from surface) of backscattering with an in situ profile of aerosol concentration and both confirm the presence of high concentrations in the boundary layer. Panel D displays a profile of circular depolarization where a low value implies high circular polarization was observed. In this case, high CP values are reported in the boundary layer where the in-situ data confirms the presences of biogenic aerosol. Petäjä et al., (2016) interprets the high CP as a manifestation of high LP expected if the particles are spherical which may be the case since ambient water content, and thus humidification, was high. Although it is not explicitly mentioned, Petäjä et al., (2016) make use of an expected correspondence between high CP with high LP as expressed by an equation derived by Mishchenko & Hovenier, (1995) (MH95 for short). This is reasonable since there are not many lidar measurements of aerosol CP and the authors use this equation to relate the observed CP with the better known and characterized lidar LP. This expression is frequently used in the lidar community to transform between lidar derived depolarization LP and CP (Robert Holtz, personal communication). While the inference offered by Petäjä et al., (2016) is consistent with the state of knowledge of the time, an alternative interpretation is that CP is the result of the presence of chiral molecules in the biogenic aerosols which, as noted in previous sections, do exhibit CP. "

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.

In the backscattering regime, a deviation from the Mischenko and Hovenier (1995) (or MH95) relationship (equation 9 in MH95) would certainly imply a presence of chiral particles or particle alignment. The latter is what was suggested by Kolokolova and Nagdimunov (2014) , a theoretical study. In our search of experimental proof of the MH95 equation, we only found Cao et al (2011) (https://doi.org/10.1117/1.3657505) as an example of specifically addressing the validity of the equation. As noted , Cao et al (2011) successfully verified the MH95 equation. We do not know why no optical activity was observed. Certainly, their figures 6, 8 and 9 in Cao et al (2011) are convincing (although at the same time there are not that many points in each plot). One physical explanation could be that aging in the pollen used in their experiment did not contain enough chiral material. As noted in the main text (Salma et el 2010), the chiral nature of biogenic aerosol diminishes through oxidation, and it is conceivable that pollen used in the laboratory setting in Cao et al was just too old (they do not report the freshness of the pollen used). While not specifically related to aerosols, Lucas Patty et al, 2017 found that chirality in live leaves immediately diminishes when is detached from the plant until it becomes negligeable. Another possibility is low sensitivity as the R1 notes, but our combined knowledge is not adequate to provide a critical assessment of the conditions and technical features of the observations reported in Cao et al, (2011).

In addition to CP, collocated and simultaneous linear polarization of measurements would be desirable in a lidar setup (perhaps something like Kokhanenko et al, 2021). Inspection of the derivation of MH95 equation reveals that randomly oriented chiral particles assumption is needed to use symmetry arguments that cancel elements in the Mueller matrix, and this obtain such equation. Violation of this rule invalidates the derivation of the equation and suggest the presence of mirror image particles.

It should be noted that the physical setting may be more complicated. Broadly, there are two candidates for chirality. One would be circular dichroism stemming from molecular chirality as already demonstrated by molecular biology and stereochemistry studies cited in the text. The second one may originate from the shape of the pollen and other biogenic particles present . Chiral shapes in pollen are certainly a possibility, but the random orientation of these chiral shapes would probably have result in an ensemble null-S_{41} and S_{42} in the Mueller matrix (unless of course these particles are aligned for some reason). Thus, circular polarization is more likely because of chiral molecules in the aerosol, rather than the aerosol shape itself. Whether this is observable with remote sensors has yet to be determined (other than the case in Fig. 6), and one of the conjectures of this review is that such a signal should be observable.

We changed the respective paragraph and added a bit more discussion on this aspect:

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 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 lack of instrument sensitivity. This result should be confirmed as it appears the only one in the literature found measuring both atmospheric aerosol LP and CP simultaneously. 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.”

Kokhanenko, G. P., Balin, Y. S., Klemasheva, M. G., Nasonov, S. V., Novoselov, M. M., Penner, I. E., and Samoilova, S. V.: Scanning polarization lidar LOSA-M3: opportunity for research of crystalline particle orientation in the ice clouds, Atmos. Meas. Tech., 13, 1113–1127, https://doi.org/10.5194/amt-13-1113-2020, 2020.

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

We rephrased the sentence as follows:

OLD

“However, while these studies stress the fact that atmospheric aerosols do contain OA components, they are aerosol in-situ composition and identification studies and to the extent of this search, there are no studies focusing on the spectral or polarimetric properties and the optical features of bulk chiral aerosols in the atmosphere.”

NEW

“However, while these studies stress the fact that atmospheric aerosols do contain optically active components, most of the studies highlighted here were conducted with in-situ instrumentation in the field deployments or in controlled laboratory settings. To the extent of our research, we have found only one study (Petäjä et al., 2016) were remote sensing instrumentation in combination with in-situ (essential to independently confirmed presence of biogenic aerosols) was utilized to detect circular polarization in aerosols. “

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."

DONE

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

We added more information to make this point clearer.

OLD:
” The simulated case assumes the following conditions: aerosols with very low to high concentrations, homogenously vertical distributed from the surface to 1km, and bi-lognormal size distributions with varying coarse mode concentrations.”

NEW

“ The general aerosol model setup is for a biogenic marine aerosol with a dark background surface. The aerosol models chosen for the simulations are generic in the sense they are consistent with marine aerosol models with bi-lognormal size distributions. The variability of composition and size is studied by considering two sets of simulations: one with fixed bilognormal distribution and variable moderate absorption and another with a variable fine mode fraction (FMF) of the total number of particles and fixed refractive index. The goal is to capture a reasonably wide optical range in aerosol size (as expressed by variable FMF) and different refractive indexes. Instead of explicitly considering parameterizations as a function of wind and ambient relative humidity (both known to impact atmospheric radiances through changes in size and index of refraction), we choose a more practical approach by prescribing the FMF and refractive index with a wide range of values expected to be present in marine biogenic aerosols. Parameters selected here were chosen to be consistent and within ranges of the marine aerosol models reported in Sayer et al., (2012). Thus , the simulated case assumes the following conditions…. “.

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

In the Rayleigh regime (AOD=0 in the plots), DLP peaks at 90 degrees scattering angle and it decreases towards 180 degrees. This is the case when comparing Rayleigh DLPs between Figures 7 and 8 (note that the labeling of the scattering angle was reversed between in both figures in the submitted doc, now corrected). So, the increase in DLP at a scattering angle closer to 90 can be explained by the molecular contribution to total scattering. Further evidence for this is in the increase with shorter wavelength, where Rayleigh scattering dominates. On the other hand, minimal DCP is present in the Rayleigh only case (AOD = 0), and the factor of 2-3 increase in DCP at the larger scattering angle indicates that it is the nature of the aerosol phase function driving this change. The strong viewing geometry dependence of polarization is expected, so measurements at multiple angles are often made in DLP remote sensing to fully characterize the aerosol state (e.g. Deschamps et al., 1994). Ideal remote sensing with DCP would also presumably make use of multi-angle observations.

Deschamps, P. Y., Bréon, F. M., Leroy, M., Podaire, A., Bricaud, A., Buriez, J. C., and Seze, G.: The POLDER mission: Instrument characteristics and scientific objectives, IEEE T. Geosci. Remote, 32(3), 598—615 , 1994.

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.

The global distribution of aerosol concentration or loading as measured by satellite aerosol optical depths is in the 0.05-0.15 range. For example, Levy et al, 2015, figure 8 report such values (https://doi.org/10.5194/amt-8-4083-2015) or figure 1 in Knobelspiesse et al., (2012) (https://doi.org/10.1364/OE.20.021457) or figure 3 in Remer et al, 2006 (https://doi.org/10.1029/2007JD009661) or Anderson et al 2013 with many figures (https://doi.org/10.3402/tellusb.v65i0.20805). There are more modern studies that confirm this and they can be easily verified by looking at the citations of these papers.

We added these references to the respective paragraph to support this point.

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."

DONE

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

DONE

Line 634: Number the equation as Eq. A3.
DONE