RESPONSE TO REFEREE#2 COMMENTS AND PEER-REVIEW REPORT

Manuscript Title: **"Observations of Dust Particle Orientation with the SolPol direct sun polarimeter"**

revised per **reviewer#2** comments as:

"Linear Polarization Signatures of Atmospheric Dust with the SolPol direct sun polarimeter"

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Dear respected Editor/Reviewers,

The authors highly appreciate the comprehensive feedback throughout the review process and kindly reply to the reviewer comments, as follows:

<u>REFEREE#2 COMMENTS:</u>

<u>Major comments</u>

"This paper reports direct sun observations of the three Stokes vector components …

1. There is, however, an important point and it is the fact this paper is titled as an investigation to suggest particle orientation whereas the evidence shown is at best consistent with the fact but by no means the only possibility. Alternate possibilities are not discussed and there are several statements in the text seem too biased to confirm that particle orientation is occurring. As it is now the text reads assuming that particles are oriented, and the discussions are biased towards this end. With this regard, I advise to change the title of the paper without mentioning particle orientation but rather emphasize the novelty of the instrument for atmospheric applications and reported observed DLP for atmospheric dust. I consider this aspect as major point as I do not consider it should be published with such title. I believe this change and some changes in the tone of the text should be relatively simple."

Author's Response: The authors are deeply grateful to the respected reviewer for the effort given and the thorough analysis on the paper key points, as the feedback has been invaluable in refining the content and strengthening the overall quality of the paper. Specifically addressing the crucial point on the paper title, we value the insightful comment which indeed would describe the context in a broader manner and will not predispose the reader towards a single strong elucidation of the reported DOLP signatures. For that matter, we have modified the manuscript title as suggested further in the review, to:

"**Linear Polarization Signatures of Atmospheric Dust with the SolPol direct sun polarimeter**"

<u>Minor/Editorial comments</u>

2. "Abstract and Lines 31-32: The concept of orientation is only mentioned in the introduction and in the last line in a suggestive sentence (as opposed to assertive sentence) … with a novel sunphotometer.")"

Author's Response: addressed in previous comment no1.

3. "Line 63: change to "The latter was addressed …"

Author's Response: Thank you, we corrected it accordingly.
Corrected "*The latter is addressed...*" to "***The latter was addressed***..." as indicated.

4. Lines 49 to 65: The contents and source of information of this paragraph is adequate. However… So, it would be very useful to acknowledge this fact with some text tweaking and text additions (probably much less text than what I just wrote here)."

Author's Response: First of, we would like to thank the reviewer for rigorously reading the introduction and providing helpful insight to the paragraph readability and content bridging. As we state in the manuscript (**Lines 82-84**), this work strives to reproduce the observations summed in the two referenced studies for atmospheric dust orientation from Ulanowski et al., (2007) and Bailey et al., (2008), respectively, with the major differentiation and challenge of using direct daylight measurements. We generally agree with the reviewer that the alignment mechanisms of atmospheric dust are not as straightforward as in the case of intergalactic dust, but they are nonetheless theoretically predicted in orientation models. As an example, Mallios et al., (2021) rigorously examine the alignment of charged/uncharged dust particles under the effect of the large scale electric field that is present in the Earth's atmosphere due to the potential difference between the lower part of the ionosphere and the Earth's surface. This field creates an electrical torque that acts upon the particles and can influence their transport dynamics, therefore their orientation which is depended on the particle size (and shape). Other mechanisms that affect particle orientation are the bombardment of gas particles that tend to randomize the orientation (rotational Brownian motion) and the aerodynamic torque that emerges due to the fact that the drag force acts on the center of pressure of the particle which is different than the center of gravity, and tends to orient the particles horizontally. The electrical torque, on the other hand, tends to orient the particles vertically, according to the study, depending on the particle's charge and size and for electric field strengths larger than 10 kV/m where vertical orientation is possible (Mallios et al., 2021). These required fields are orders of magnitude larger than the typical electric field value or the reported values on elevated layers (e.g., Daskalopoulou et al., 2021b), but were previously measured in dust storms close to the dust source (review by Riousset et al., 2020 and references therein). The aforementioned information is stated in the introduction (**Lines 66-81**) in order to provide the theoretical plausibility of atmospheric dust orientation (considering the much smaller size of dust than ice crystals, for example, that exhibit clear orientation signatures), through electrostatic means and to state that also other effects on particle motion, such as turbulence (e.g., Klett J. D., 1995), may result in preferential orientation but remain to be examined both by modelling and accompanied observations.

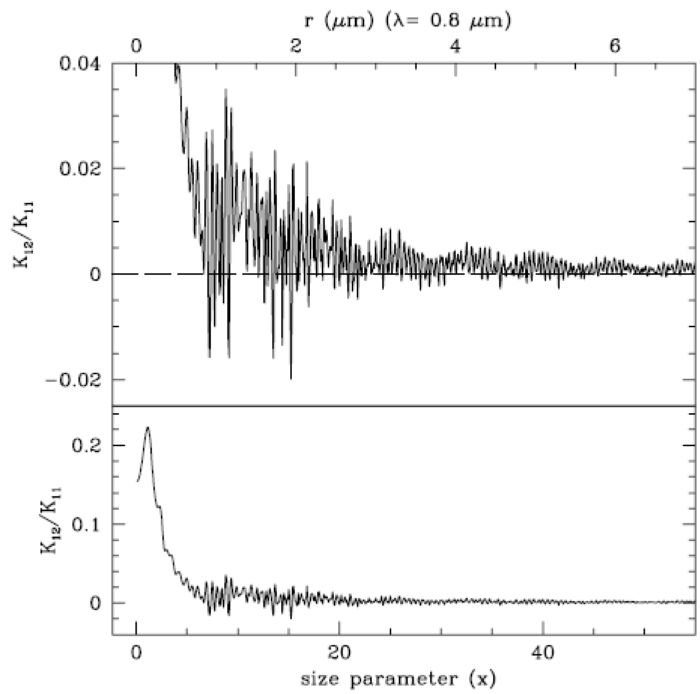

Figure 1: Ratio of extinction matrix elements K_{12}/K_{11} for vertically oriented prolate spheroidal particles of axis ratio 1.8 at a zenith distance of 60◦ as a function of size parameter ($x = 2\pi r/\lambda$). The lower panel shows the full range of variation including the peak corresponding to the small particle regime that causes interstellar polarization. The upper panel is on an expanded scale showing the oscillatory nature of the polarization at larger sizes, but also that the mean level is positive (horizontal polarization). The particle size at a wavelength of 0.8μm is shown on the top scale.

In terms of the differentiation of aligned particle optical properties in contrast to non-aligned particles, we rely on the previous modelling work of Bailey et al., (2008) who investigated the expected polarization for spheroidal particles via T-matrix calculations of the forward scattering (extinction) matrix. A range of particle sizes, aspect ratios and zenith distances were used for a particular particle composition, so although the study focuses on a longer wavelength, for size parameters that correspond to particles larger than 3μm for a 550 nm wavelength, there is an expected excess in linear polarization for vertically or horizontally aligned particles (see adapted Figure 1 from Bailey et al., 2008).

By carefully considering the highlighted distinction between the intergalactic dust and its atmospheric counterpart, we rephrase **Lines 56** to **66** and add the following sentences:

"Dichroism measurements provide information on the magnetic field orientation, which is the dominant alignment mechanism **for these sub-micron particles** (Andersson et al., 2015; Dasgupta Ajou K., 1983; Kolokolova and Nagdimunov, 2014; Lazarian, 2007; Siebenmorgen, 2014; Skalidis and Tassis, 2020). **Concerning the much larger atmospheric dust particles, the geomagnetic field is considered a weak alignment mechanism, since multiple processes such as the bombardment by gas particles, the imposed aerodynamic and electrical torques (Mallios et al., 2021; Ulanowski et al., 2007a and references therein), but also turbulence (e.g. Klett J.D., 1995) compete (or counter-balance) for the most dominant atmospheric particle alignment mechanism. Based on a similar optical approach, though**, atmospheric dust may provide distinct linear polarization (LP) signatures, as vertically oriented particles can lead to dichroic extinction of the transmitted sunlight. This was indicated for starlight observations during nighttime, which showed predominantly horizontally polarized light during a Saharan dust episode in La Palma (Bailey et al., 2008; Ulanowski et al., 2007b). **Also in the same study, modelling of the forward scattering matrix through T-matrix calculations was employed and showed that excess polarization for spheroidal particles of a specific composition and orientation is to be expected for particle sizes larger than 3 μm.** However, the discussed measurements refer to column-integrated values that are not capable of resolving the vertical distribution of the phenomenon throughout the dust layer. The latter was addressed by a novel polarization lidar for detecting dust orientation, nicknamed WALL-E, which is expected to provide valuable information for monitoring the phenomenon of dust polarization in the Earth's atmosphere (Tsekeri et al., 2021).
In order to provide the theoretical feasibility of orientation, a potential mechanism that is capable…"

Regarding the reviewer's suggestion/inquiry on "what is the connection and consequence of not having particle random orientation and the observed optical properties?", the origin of polarization of direct sunlight is already explained in the Introduction (Lines 49-54) and further discussion can be found in the references cited (Ulanowski et al. 2007, Bailey et al. 2008). See also response to Comment 5.

References:

Bailey, J., Ulanowski, Z., Lucas, P. W., Hough, J. H., Hirst, E. and Tamura, M.: The effect of airborne dust on astronomical polarization measurements, Mon. Not. R. Astron. Soc., 386(2), 1016–1022, doi:10.1111/j.1365-2966.2008.13088.x, 2008.

Klett, J. D., 1995: Orientation Model for Particles in Turbulence. J. Atmos. Sci., 52, 2276–2285, https://doi.org/10.1175/1520-0469(1995)052<2276:OMFPIT>2.0.CO;2.

Riousset, J. A., Nag, A., & Palotai, C. (2020). Scaling of conventional breakdown threshold: Impact for predictions of lightning and TLEs on Earth, Venus, and Mars. Icarus, 338, Article 113506. http://dx.doi.org/10.1016/j.icarus.2019.113506.

Ulanowski, Z., Bailey, J., Lucas, P. W., Hough, J. H. and Hirst, E.: Alignment of atmospheric mineral dust due to electric field, Atmos. Chem. Phys., 7(24), 6161–6173, doi:10.5194/acp-7-6161-2007, 2007.

5. "Also, in my search of bibliography … Chapters 5-7 are also very relevant: Coulson, Kinsell L. "Polarization and Intensity of Light in the Atmosphere." (1989). A. Deepak Pub., Hampton, Va.,"

Author's Response: The reviewer correctly states that not many direct sun (i.e., dichroic extinction) polarization measurements do exist, but then mentions some recent papers describing all-sky measurements. Indeed, in contrast to the direct measurements, there is a huge body of past work focused on all-sky (diffuse) polarization. Unfortunately, such measurements have limited relevance to the present study, which is focused on particle alignment. The reason is that the principal cause of dichroic extinction is particle alignment, while the interpretation of diffuse polarization measurements in terms of particle alignment is highly ambiguous, because multiple causes contribute to sky polarization. Hence it is much harder to derive information on particle alignment from sky, as opposed to direct sun, polarization measurements. We thank the reviewer, though, for providing references to bibliography, based on all sky polarization measurements, and on some classic textbook information that will strengthen our knowledge on the subject. We were not aware of the very recently published papers by Li S. et al., (2023), Pan P. et al., (2023) and Guan L. et al., (2018) and we have incorporated them in the main text with the prospect of potentially being used as background information on future studies. Therefore, a paragraph is added as follows:

Line 54 to 64: "Interpreting linear polarization measurements in the direct direction (i.e., measurements taken when the observer looks directly towards the Sun) can be challenging due to the overwhelming intensity of sunlight and secondary sources of linear polarization in the observational line-of-sight. All-sky polarization patterns can serve as a background reference tool to aid in the interpretation of direct polarization measurements, as the direct direction is a subset of the former (Guan L. et al., 2018; Pan et al., 2023). Recent studies that compare radiative transfer simulations for aerosol media of well-defined optical properties with all-sky observations, show that the degree of linear polarization (DOLP) is impacted, and in fact decreases, by the increasing optical depth of desert dust particles (assumed spherical) even in the forward scattered direction (Li S. et al., 2023). However, the interpretation of diffuse polarization measurements in terms of particle alignment is highly ambiguous, because multiple causes contribute to sky polarization. Hence it is much harder to derive information on particle alignment from sky, as opposed to direct sun, polarization measurements."

6. "Line 294 : Gassó et al, reference is wrongly listed. Check the proper reference."

Author's Response: There was an unfortunate reference mix-up here, we have corrected it accordingly in the manuscript, thank you. The proper citation would be:

Gassó, S. and Knobelspiesse, K. D.: Circular polarization in atmospheric aerosols, Atmos. Chem. Phys., 22, 13581–13605, https://doi.org/10.5194/acp-22-13581-2022, 2022

7. "Line 316 : note the Kemp et al, paper also reports a non-zero V component in observed solar flx (of the order ~10-6)"

Author's Response: Indeed, Kemp et al. (1987) report CP values of the order of ~ 3 x 10^{-6} in 550 nm, for the case of targeting specific cardinal co-ordinates in the Sun's disk (Fig. 2 in paper), with half the aperture angular diameter used in SolPol. These measurements could potentially be enhanced by regions of intense magnetic activity and strong area-dependent circular polarization near the solar limb. In contrast, in the whole Sun disk measurements (Fig. 3 in the same paper) CP values drop to the order of ~ 10^{-7} due to mainly the use of a larger aperture which would increase the observed effective diameter and subsequently, if the increase in circularly polarized flux (*V*) is not linear with the increase in total solar flux (*I*), the CP will decrease (V in Kemp's paper denotes the normalized *V/I* quantity).
The latter observations are consistent with what we observe with SolPol for the total solar disk in the forward direction, which is not negligible although near the limits of the instrument sensitivity. In order to extract the proper information for aligned atmospheric particles from CP (please refer to **Lines 293-300** in the preprint manuscript), we should focus on further processing the existing dataset and we tend to in future research, so we thank the reviewer for the remark.

References:

Kemp, J. C., Henson, G. D., Steiner, C. T. and Powell, E. R.: The optical polarization of the Sun measured at a sensitivity of parts in ten million, Nature, 326(6110), 270–273, doi:10.1038/326270a0, 1987.

8. “Line 365-366: Not a clear sentence, I do not understand what you are traying to say. Please rewrite/clarify as needed.”

Author’s Response: We thank the reviewer for the comment and we will try to clarify. The sentence subtly refers to the particle viewing geometry from the instrument’s reference frame and how the angle under which we perceive particle orientation changes with respect to the increasing solar zenith angle. From our understanding, we have attempted a schematic that depicts the viewing angles in SolPol’s line-of-sight for a small collection of dt dust particles that are either i. horizontally oriented with respect to the ground and ii. vertically oriented with respect to the ground (Figure 2). As seen in the following figure, in both orientation cases (either horizontal or vertical) the instrument does not detect preferential alignment directly in the zenith, where SZA equals to zero, as specifically in the horizontal orientation case the azimuthal distribution of the grains is uniform and cannot be ignored. While, in the vertically aligned collection particles gradually appear to become “more” vertically aligned as we progress to larger SZAs. Hence, the sentence refers to either that differentiation in viewing angles and that as we progress to larger SZAs we would expect linear dichroism to be more prominent or to the increase of oriented particles as the instrument targets larger airmasses with the increasing zenith angle.

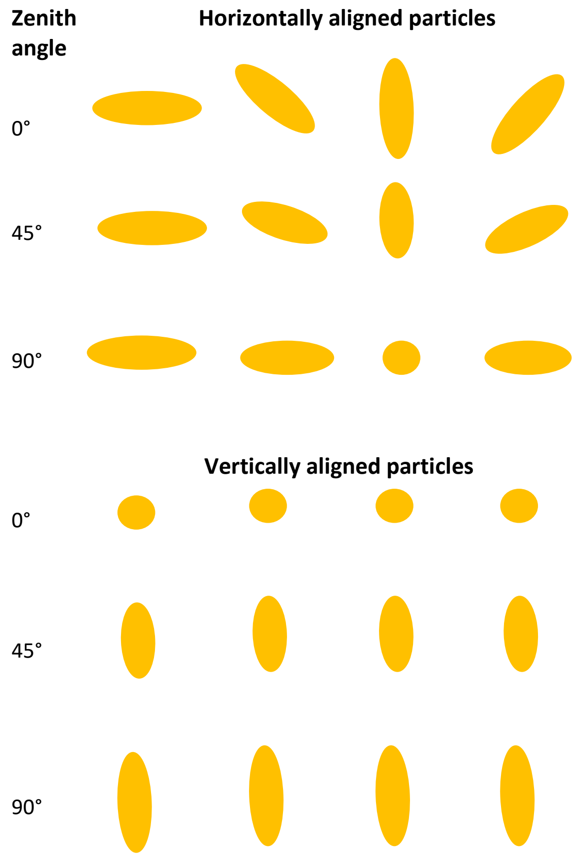

Figure 2: Four representative horizontally (top panel) and vertically (bottom panel) aligned particles for three distinct zenith angles, in the instrument’s frame of reference.

Therefore, we have rephrased the sentence as follows:

Lines 365 - 368: the sentence “As the SZA increases, particles that are preferentially aligned with respect to the vertical axis present themselves in the frame of reference of the instrument as being more strongly aligned. Moreover, the airmass increases, hence the slant optical thickness and therefore dichroic extinction (which is an extensive scattering property – it grows with the number of scattering particles) also increase. Consequently, it is expected that at near-zenith angles, particles that are aligned with their long axis vertically will not influence the polarization and result in near zero values. Conversely, strong polarization is expected for large SZA, similarly to what was previously reported (Bailey et al., 2008).”

9. “Lines 335-337: "This is our first consistent indication ... These two facts warrant additional analyses as MS and molecular polarization can play an unknown role and are a consideration that probably were not necessarily in the cited studies."

Author’s Response: We refer the reviewer to the authors’ answer on the first reviewer’s comment no2. i.e. *“2. The Rayleigh optical depth is ~0.14 for 500nm ... diffuse scattering could be better observed” and the* Lines 446-451 of the submitted manuscript where we state that more tests are needed in order to further intensify our arguments. Taking into account the reviewer’s kind suggestions, we change the tone of the following sentences:

Lines 335 - 337 moved to **Line 341** and rephrased as: “This could potentially indicate that preferentially - vertically or horizontally - aligned dust particles might be present in the observed layers and cause the strong dichroic extinction of sunlight transmitted through the layer.”

Lines 379 - 381: “The segregation between reference days and dust driven days, along with the distinct behaviour of DOLP with increasing SZA is consistent with the findings from stellar polarimetry (Bailey et al., 2008; Ulanowski et al., 2007b), although these studies are targeted on longer wavelengths and potential Rayleigh contamination my vary our results towards probing particle orientation.”

10. “Line 438-440 "The observed excess in LP… (Figure 8b).
Can you reference this? This quite a speculation. How can you be sure that this LP vs AirMass relationship is related to particle orientation? Other considerations maybe be at play here such changes in atmospheric loading and size distribution, variability in dust composition or variability of multiple scattering through the day due to loading changes.”

Author's Response: We, again, deeply appreciate the reviewer for challenging our arguments and therefore strengthen the paper rationale as we incorporate the input**.** The relationship between the excess linear polarization and aerosols is a complex one and can be influenced by several factors other than particle orientation, such as: i. the particle size distribution and particle shape, ii. the dust particle composition as it can greatly affect their polarizing properties, iii. the observation geometry – including solar zenith angle (hence airmass) and iv. the particle concentration and spatial distribution. So, as the reviewer correctly points out there is a level of ambiguity when stating that the linearity can be attributed to particle viewing geometry or particle orientation. Having said that, no other hypotheses currently exist for the origin of dichroic polarization itself, apart from particle alignment. In this context, a linear relationship between airmass and LP is expected (see response to Comment 8) and is fully consistent with the alignment hypothesis. In order to be entirely conclusive, further comparison of the polarization measurements with synergistic retrievals of the particle microphysical properties (co-located CIMEL sunphotometer and lidar system) under the specific dust cases are scheduled in a future study, which again is a challenging topic as inherent biases of these instruments should also be considered. Nonetheless, we might be able to narrow down to the cases where there is not much load variability within the day and the layer is relatively homogeneous (for example, the May 2020 cases shown in the manuscript exhibit such a behavior through preliminary study). In order to properly consider the size distribution daytime variability, sophisticated in-situ measurements are needed to complement SolPol. These were available during the ESA Cal/Val ASKOS 2022 experiment where we also operated the instrument, but due to overcast conditions and extreme winds the co-location of both was difficult.
The scope of this initial research was to present a novel dataset of linear polarization observations of elevated dust layers with an instrument otherwise used in astropolarimetric studies, and attempt to interpret the measurements as indications of particle orientation, considering the previous work. As a future step we plan to perform a more complete testing, by acquiring observations of high-AOD pollution cases, where we know that the particles are spherical and thus produce no dichroic extinction.

Therefore, considering the reviewer's comment, to clarify our argument we have rewritten **Lines 438-441** as:

"We expect that for small AOD values (< 0.1) the linear polarization values will be within the noise threshold as derived from the clean days' behaviour, as is the case here and since there are no observed dust events with such small loads no increasing trends are exhibited (dark blue circles). As the optical depth increases, we observe upward trends in linear polarization for a specific SZA values, mainly above 40°, intensifying in proportion to the AOD, with the highest values of DOLP recorded under heavier dust loads. This indicates that, as expected, dichroic extinction is enhanced due to the presence of a larger concentration of preferentially oriented dust particles for fixed viewing angles and, consequently, for constant airmass, as seen in the linear relation of Figure8b. Furthermore, the observed LP is linearly proportional to the airmass, which could be attributed to aligned particle viewing geometry for fixed AODs (since in the instrument frame of reference particles become more aligned for increasing SZA), or the amount of aligned particles along the line of sight, or both (Fig. 8b). This linear dependence is seen more clearly when we inspect the correlation between DOLP and the AOD corrected with airmass (slant optical depth) in Figure 9. The dependence exhibits a threshold at AOD values of 0.1, higher values corresponding solely to the presence of dust. As we move to larger dust loads this trend contains cases of increasing linear polarization values even for smaller airmasses. The correlation strength could be further tested in future studies with dust loads close to one, as particle orientation could affect the geometric formalism for the derivation of the AOD."

11. "Line 510 : can you add a reference where this statement is explained? I mean why the mean orientation should be 60 degrees?"

Author's Response: The specific statement regarding mean alignment angle for randomly oriented is erroneous and it was an unfortunate remnant of an early, pre-submission manuscript version. The 60° mean alignment angle refers to the mean polar angle and does not imply the existence of any form of preferred/dominant orientation (as expected from random alignment, by definition). The distribution of alignment angles measured around any polar axis, including the line of sight which is the axis relevant here, is uniform for randomly oriented particles, therefore the mean angle has no significance as an alignment angle. Thus, we have erased the sentence in question.