RESPONSE TO REFEREE#1 COMMENTS AND PEER-REVIEW REPORT

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

Authors (as declared in the submitted manuscript):

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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:

REFEREE#1 COMMENTS:

"*The paper by Daskalopoulou et al. introduced the very interesting and important observation of polarization in the direct sun transmission measurement which can prove the preferred orientation of dust particles in the atmosphere. The paper provides a clear description on the experimental setup as well as data acquisition and processing. I read this work with much interests and have a few comments for the authors to consider or confirm:*

1. I'm not quite sure about the proof of little contribution of diffuse light scattering to the observed DOLP in the transmitted measurements. DOLP is the ratio of polarized intensity against intensity. So even if the aperture size increases, both intensity and polarized intensity could increase so that the ratio remain not much impacted. Could the author elaborate more on this?"

Author's Response: The authors are grateful to the respected reviewer for the comments about the paper readability in its preprint state. Concerning the first comment on diffuse light contribution to our measurements, we would like to initially state that the testing that we could perform on ambient conditions with SolPol was limited only to empirical measurements by increase/decrease of the incoming diffuse light flux when selecting different aperture sizes. The test procedure comprised of alternating iris sizes from small (at 4.5 mm), to regular (at 5.5 mm, i.e., the one used for regular SolPol measurements) to large (at 7 mm) through consecutive measuring intervals so as to ensure that polarization would not change significantly with the instrument viewing angle. The rationale was that with larger iris, more light is measured by the instrument and by increasing the diffuse light in the solid angle. Hence, if there is contribution of the diffuse light scattering to the measured linear polarization, then the latter should also increase with a larger iris. For a simple adjustable aperture stop as the one used in the current setup, when increasing its diameter, we increase the effective area and therefore the incoming total solar flux. By examining the following figure (Fig. 1, as opposed to Figure 10 - Section 5.3 in the manuscript), we focus on the early morning to noon measurements, when AOD is relatively constant and compare the first 7 mm measurement to the following (second individual) 4.5 mm measurement (both in red squares). As we can see, DOLP increases from larger iris to the smaller iris while we would expect the opposite to happen in the case of strong contribution from the diffuse light to the linearly polarized light or at least that the DOLP ratio would remain constant provided that the decrease in intensity when downsizing is proportionate to the decrease in polarized intensity. Although there is a detailed description in the SolPol manual pg. 13, we have added the following sentence in the manuscript and hope this clarifies the discussion on Section 5.3.

Lines 466 - 467: "**We focus on the early morning to noon measurements, when AOD is relatively constant and compare the first 7 mm measurement to the following (second individual) 4.5 mm measurement.** As seen in **Error! Reference source not found.**,..."

Lines 471 - 472 are rephrased as follows: "**DOLP increases from larger iris to the smaller iris while we would expect the opposite to happen in the case of strong contribution from the diffuse light to the linearly polarized light received by the detector**. This proves that by increasing the…"

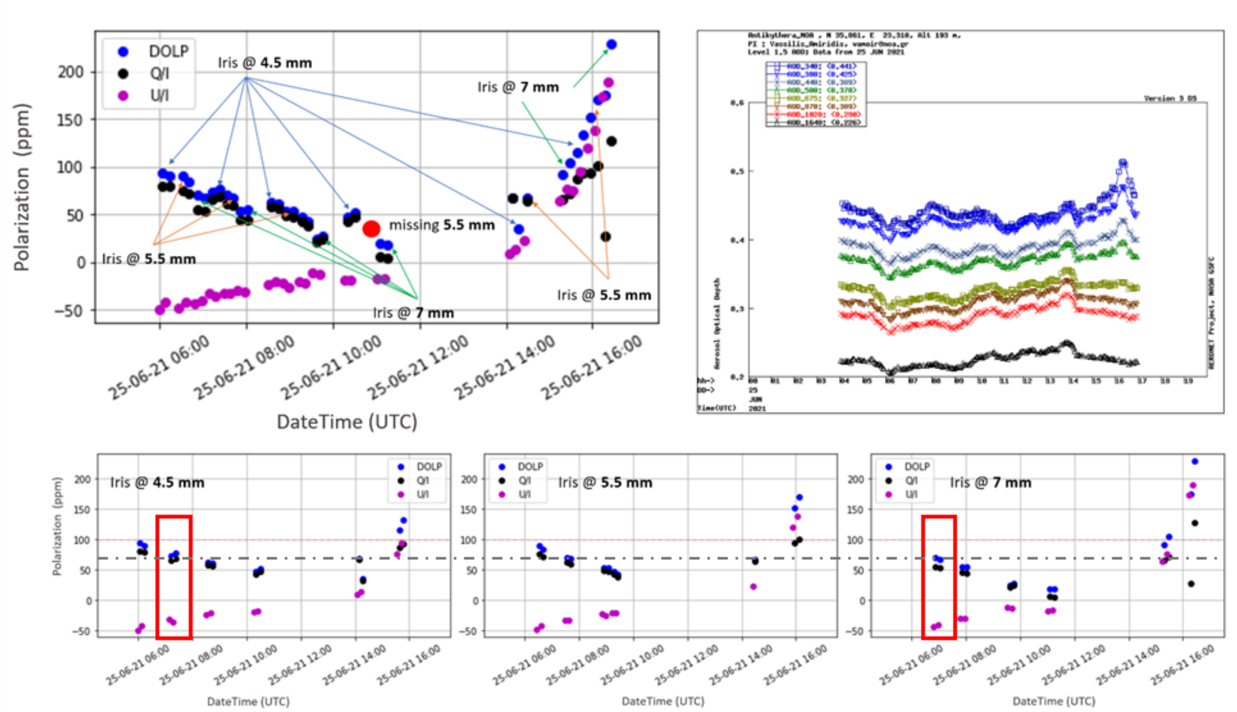

Figure 1: Alternating iris size tests for the quantification of diffuse light contribution to the linear polarization observations under the 25/06/2021 dust layer, with an AOD of 0.370 at 500 nm. Top panels present the normalized Q and U Stokes parameters, DOLP and the different colour arrows denote the respective iris size for each measurement set, along with the AOD progression within the day from AERONET. Bottom panels present the individual measurement sequences for each iris size, while red squares denote the succession from larger iris to the smaller one.

"2. The Rayleigh optical depth is ~0.14 for 500nm. Under clear sky conditions, the diffuse Rayleigh should contribute some signals to DOLP. This will bring minor but potential contamination via multiple scattering which has certain dependence on solar angle. From the Fig. 7a, however, it looks the contribution from Rayleigh to be very small (<50ppm) and there is little dependence on solar angle. Could the authors confirm this is the case? Is it because the direct transmission is very strong so that DOLP is further diluted? To have a better view, the authors may try separating the direct direction of sunlight and scattering contribution given that the solar irradiance is known at 500 nm and the optical depth for both Rayleigh and aerosol (from AERONET) is known. Then the DOLP contributed by diffuse scattering could be better observed."

Author's Response: As is presented in Figure 7a and the zoomed representation in (c), the measured Rayleigh scattering contribution in the forward direction and under clear conditions (no dust particle presence indicated by the lidar retrievals) to DOLP is less than 50 ppms, when we use the default 5.5 mm aperture size. It also appears to have no dependence on the solar zenith angle for the narrowband filter at 550 nm. Compared to the DOLP values we are observing when dust particles are present, the latter are almost one order of magnitude larger than the clean day cases. If we increase the iris diameter, Rayleigh contributions to the polarization fractions at 550 nm can increase by a factor of two, which is consistent with predicted values for Rayleigh scattering at low altitudes (e.g., Mishchenko et al., 1994 and references therein). Since all measurements are direct sun and we ensure a stable tracking of the sun disk without misalignments, incoming diffuse light within our FOV from scattering angles close to 0° (forward scattered light) will not contribute to DOLP and, in fact, linear polarization will decrease with increasing AOD due to further loss of polarization from multiple scattering (see adapted Figure 2 from Hansen and Travis, 1974). Furthermore, these changes due to multiple Rayleigh scattering are found in diffuse light, and for direct solar irradiance only the optical depth affects the beam. Since we are confident that the residual contribution of diffuse light to the measured signal (as explained in the previous question), is very small, the zenith angle independence of DOLP at 50 ppm is explained.

We thank the reviewer for their suggestion concerning the DOLP contribution distinction by utilizing the AERONET data. In order to make these estimations an absolute calibration of the irradiance measurements would be required, in order to separate the two components from theoretical calculations. The setup of this study had a different aim and the constant changes of integration times and apertures would require independent calibrations for each set. We plan for future experiments to include these calibrations in order to make possible this kind of analysis.

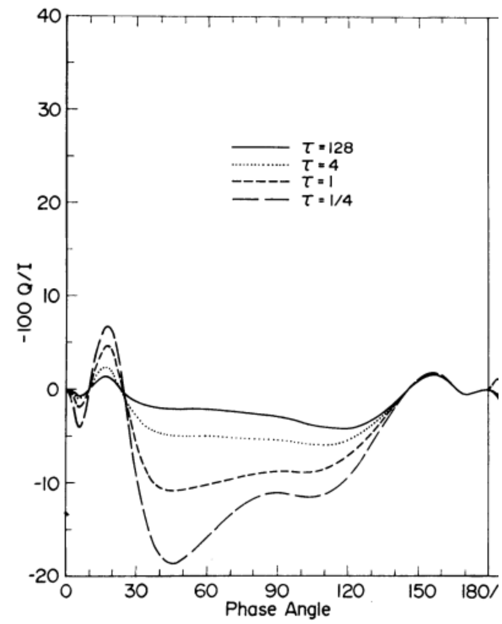

Figure 2: Linear polarization, -100 Q/I, as a function of the phase angle for different aerosol optical depths (figure adapted from Hansen and Travis, 1974).

References:

Hansen, J.E., and L.D. Travis, 1974: Light scattering in planetary atmospheres. Space Sci. Rev., 16, 527-610, doi:10.1007/BF00168069.

Mishchenko, M. I., Lacis, A. A. and Travis, L. D.: Errors induced by the neglect of polarization in radiance calculations for rayleigh-scattering atmospheres, J. Quant. Spectrosc. Radiat. Transf., 51(3), 491–510, doi:10.1016/0022-4073(94)90149-X, 1994.

"3. I wonder whether one likely cause of very small DOLP (<700ppm) to be the short wavelength (500nm) the authors are experimenting whereas the dust particles are coarse (>5 microns). How difficult will it be to switch to a larger wavelength (e.g. 1020nm or even infrared) and repeat the measurement. This way the authors may obtain higher sensitivity of DOLP to the orientation of large particle size."

Author's Response: We thank the reviewer for their insightful comment on considering switching to longer wavelengths, so as to maximize the potential polarization signature for dust. Mineral dust is often described as "white" or "gray" in terms of its wavelength response. As an example, Bailey et al. (2008) have discussed the only logical question if larger dust particles give polarization signatures in the same manner as small dust particle, which are mostly responsible for interstellar polarization. As seen in Figure 3 (adapted Figure 4 from the respective paper of Bailey et al., 2008), where the T-matrix calculated ratio of the total extinction coefficient to the dichroism, K_{12} / K_{11}, is presented as a function of the size parameter χ, for larger particles we are in the plateau regime where the mean polarization is essentially the same as that produced by smaller particles. Since larger particles are the ones that will eventually become preferentially oriented by overcoming randomization due to Brownian motion (e.g., Mallios et al., 2021; Ulanowski et al., 2007), we do not expect to observe a significant contribution to the phenomena by switching to longer wavelengths.

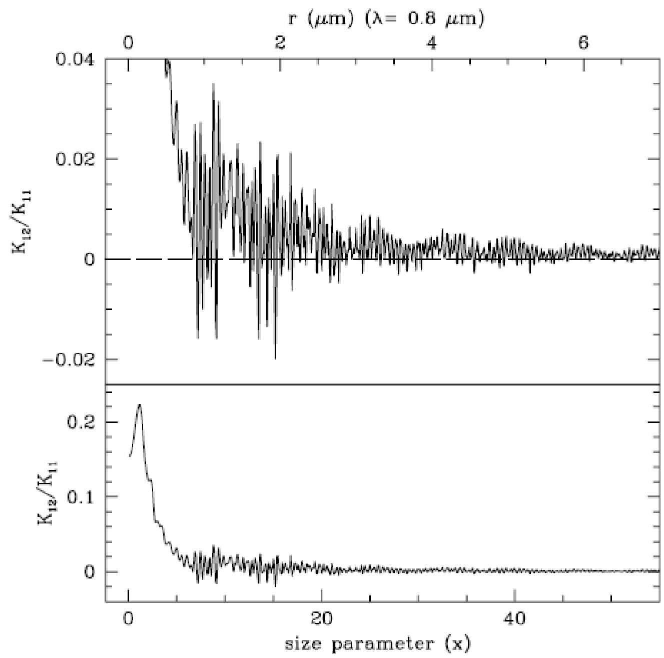

Figure 3: Ratio of extinction matrix elements K_{12}/K_{11} for vertically oriented prolate spheroidal particles of axis ratio 1.8 at a zenith distance of 60◦ as a function of size parameter ($x = 2\pi r/\lambda$). The lower panel shows the full range of variation including the peak corresponding to 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 technical limitations, SolPol is an experimental instrument and its complete characterization has already been painstakingly long. Some of the tasks to be tackled would be the need to consider new communication protocols between instrument peripherals in order to operate at an additional wavelength and, most importantly, integrate a new photodiode detector operating in the NIR with the capability of stably detecting small signals. The responsivity and quantum efficiency of photodiodes decrease with increasing wavelength in the NIR range, meaning that the sensitivity decreases, resulting in lower signal-to-noise ratios and reduced detection efficiency. The limit for reasonable signals from the specific photodiode is about 750-800 nm with a good SNR. When approaching wavelengths beyond 1000 nm it is a very different experiment than what we have setup so far, with encroaching telluric absorption lines beginning to come into play. Going towards the lower wavelengths' direction, i.e., 450 nm might also be interesting in comparison to our previous 550 nm. Instrument filters on the 450 and 750 nm central wavelengths have been set up in the past for lab measurements and Rayleigh contribution field measurements in Hatfield, but definitely the bandwidth of these filters will make a difference in ambient field conditions in Antikythera. Thermal noise is also a significant source of error to be considered in these wavelengths, thus the trade-off led initially to the use of the instrumental configuration as is.

Nonetheless, we again thank the reviewer for the kind suggestion and we will revisit the SolPol assembly configuration in the NIR as a potential upgrade to be completed in the near future and provide complementary data on dust orientation from various wavelengths.

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.

Mallios, S. A., Daskalopoulou, V. and Amiridis, V.: Orientation of non spherical prolate dust particles moving vertically in the Earth’s atmosphere, J. Aerosol Sci., 151(August 2020), 105657, doi:10.1016/j.jaerosci.2020.105657, 2021.

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.