Response to Reviewers

The authors greatly acknowledge the anonymous reviews for carefully reading the manuscript and providing constructive comments. Each comment is discussed separately with the following typesetting:

Reviewer's comments

Authors response

Changes in the manuscript

I congratulate the authors for this nice work. Nevertheless, I am a bit surprised that the authors has not considered our previous works on this subject that could help them to improve their paper and to correct some flaws.

We thank Dr. Jean-Baptiste Renard for his time reviewing our manuscript. It is great to have comments in the discussion phase because it is the way to improve our manuscript. We feel grateful for his comments and the acknowledge of our work. At the same time, we are sorry for not being cautious in acknowledging Dr. Renard's team experience in Saharan dust measurements. We have learnt a lot after reading some of their works that have served to enrich our manuscript. We believe that more interactions are needed between atmospheric scientists and the teams that perform laboratory measurements of particle phase matrixes.

Line 63: For the size distribution, the authors could consider the measurements from balloon-borne aerosol counter inside a Saharan dust plume : Renard, J.-B.; Dulac, F.; Durand, P.; Bourgeois, Q.; Denjean, C.; Vignelles, D.; Couté, B.; Jeannot, M.; Verdier, N.; Mallet, M. In situ measurements of desert dust particles above the western Mediterranean Sea with the balloon-borne Light Optical Aerosol Counter/sizer (LOAC) during the ChArMEx campaign of summer 2013. Atmos. Chem. Phys. 2018, 18, 3677-3699.

-We are grateful for this reference. After reading that manuscript, we believe that it is very important to remark on the possible presence of the super-coarse mode in the transported Saharan dust. This has been included in the new introduction section (L66-67):

"Mineral dust particles are typically considered as large particles in the coarse (1-10 µm) and super-coarse (> 10 µm) modes (Renard et al., 2018)"

And the reference Renard et al., 2018 has been added to the reference list

Renard, J. B., Dulac, F., Durand, P., Bourgeois, Q., Denjean, C., Vignelles, D., Couté, B., Jeannot, M., Verdier, N., & Mallet, M. (2018). In situ measurements of desert dust particles above the western Mediterranean Sea with the balloon-borne Light Optical Aerosol Counter/sizer (LOAC) during the ChArMEx campaign of summer 2013.

Atmospheric Chemistry and Physics, *18*(5), 3677–3699. https://doi.org/10.5194/acp-18-3677-2018

Line 74: The authors have forgotten to consider the effect of the size distribution of the particles. Scattering properties (including polarization) are sensitive to the size of the particles, even for mineral dust, as shown by our team :

-We agree with the referee, and we have re-written this part of the introduction section. Now it is given as (L119-122):

"… the usual study of scattering matrix elements of dust particles is done in the laboratory for synthetic samples minerals that compose dust particles (Curtis et al., 2008; Huang et al., 2020; Meland et al., 2010; Muñoz et al., 2010a; J. B. Renard et al., 2014; J.-B. Renard et al., 2010) or with collected dust samples (Muñoz et al., 2007a; J. B. Renard et al., 2014; J.-B. Renard et al., 2010, 2024)"

We have added these references to the list of references.

Renard, J. B., Hadamcik, E., Couté, B., Jeannot, M., & Levasseur-Regourd, A. C. (2014). Wavelength dependence of linear polarization in the visible and near infrared domain for large levitating grains (PROGRA2 instruments). *Journal of Quantitative Spectroscopy and Radiative Transfer*, *146*, 424–430. https://doi.org/10.1016/j.jqsrt.2014.02.024

Renard, J.-B., Francis, M., Hadamcik, E., Daugeron, D., Couté, B., Gaubicher, B., & Jeannot, M. (2010). Scattering properties of sands. 2. Results for sands from different origins. *Applied Optics*, *40*(18), 3352–3559. http://www.icare.univ-lille1.fr/progra2/

Renard, J.-B., Hadamcik, E., Worms, J.-C., & Hadamcik, E. (2024). The laboratory PROGRA2 database to interpret the linear polarization and brightness phase curves of light scattered by solid particles in clouds and layers. *Journal of Quantitative Spectroscopy and Radiative Transfer*, *320*, 108980. https://doi.org/10.1016/j.jqsrt.2024.108980ï

After receiving comments from all referees, we figured out that we might have written some inconsistent conclusions in our analysis. In the revised manuscript we have insisted that F_{11} and $-F_{12}/F_{11}$ depend on several factors, particularly particle size distribution, shape and refractive index. It was not possible to perform inversions to retrieve size distribution and refractive index, and also unfortunately we did not have correlative measurements of size distributions with other instruments. Because of these reasons, we avoid speculations in the interpretation of the results, and we just highlight that the different patterns can be explained by different reasons.

We plan to continuously acquire measurements but with correlative measurements of aerosol size distributions and chemical compositions. We are also planning several field campaigns to do closure studies with lidar systems (see comments to referee 2). Moreover, we are collaborating with the GRASP team in the optimization of the inversion to retrieve several modes of aerosol optical and microphysical properties from our PI-Neph measurements. These issues are now clearer in the new conclusions section

Lines 78-82: Why the large database of PROGRA2 (https://www.icare.univ-lille.fr/progra2-en/?noredirect=en_US) that contains such laboratory measurements is not cited?

-We thank the referee for remarking on this point. We are impressed by the work done in PROGRA2 and it has been cited. Particularly, we have added reference Renard et al., (2024) to this part in the introduction section.

Part 2.2.1. If I well understand, the instrument retrieves the light scattering parameter from a cloud of particles, without size selection. Thus, if the authors want to make comparison between different sessions of measurement in different conditions, they must be sure that the size distributions are the same. Otherwise, they will observe a combination of size distributions, refractive indexes, and even porosities of the grains and the agglomerates. The authors must explain how these different parameters influence their measurements and their analysis.

-Yes, the instrument measured light scattered by an ensemble of particles that was sampled from a total air inlet, so there is no size selection or cutoff. We agree with the referee that for comparisons size distribution needs to be monitored and ideally controlled. But the main difference between PI-Neph measurements and those at laboratory is that the PI-Neph is acquiring ambient air and thus it implies changeable aerosol conditions. Our objective is to study phase matrix parameters, particularly F_{11} and $-F_{12}/F_{11}$, in these changing conditions. This has been highlighted in the introduction section of the revised manuscript (L129-146):

“The latest developments use imaging techniques (Bian et al., 2017; Curtis et al., 2007; Dolgos & Martins, 2014a) to determine phase matrix with single detector and relatively compact design that does not require moveable parts. The Polarized Imaging Nephelometer (PI-Neph) was one of the first designs of a polar nephelometer that used imaging techniques, developed by the University of Maryland, Baltimore County (UMBC). This first prototype of the PI-Neph could acquire aerosol phase matrix at 473, 532 and 671 nm with 0.5° resolution. The instrument was deployed on the NASA DC8 aircraft and operated during special field (Espinosa et al., 2018; Reed Espinosa et al., 2017). Other PI-Neph instruments based on the first UMBC design are operated by NOAA (Ahern et al., 2022; Manfred et al., 2018). The main novelty of these prototypes is that they measure phase matrix elements of ambient air, when conditions can be very different to laboratory measurements. However, to date none of these instruments have been operating continuously and report any multiwavelength measurements of Saharan dust. The imaging technique is being expanded worldwide with further designs although limited to laboratory operation yet (Moallemi et al., 2023). All designs in polar nephelometry present physical limitations that limit the measurements to the range 3°-178°, but synthetic tests have revealed that multi-wavelength polarimetric PI-Neph measurements improve the information content for the retrieval of aerosol optical and microphysical properties (Moallemi et al., 2022) . Therefore, measurements of dust phase matrix elements for ambient aerosol samples in the atmosphere will serve to advance in the understanding of mineral dust absorption properties and chemical composition (Di Biagio et al., 2017, 2019).”

More specifically, our study focuses on how phase matrix elements for transported Saharan dust varies between two extreme events and other more moderate that reach our station in the southeast of the Iberian Peninsula. Polarization measurements seem to be very valuable when the aerosol sampled is equivalent to a mixture of particles of anthropogenic origin and those of mineral dust. We have clarified this point in the conclusions section, but adding that more measurements are required particularly at other locations (see comments to referee 2).

L772-789: "This work has focused on the analyses of aerosol phase matrix elements and other optical properties during Saharan dust outbreaks that were registered in the UGR station (Southeastern Spain) in the year 2022. The main novelty of the analyses are the measurements by the multiwavelength Polarized Imaging Nephelometer (PI-Neph) developed by GRASP-Earth and capable of providing two aerosol scattering matrix elements (F_{11} and $-F_{12}/F_{11}$) for three different wavelengths (405, 515 and 660 nm). The uniqueness of PI-Neph is that it allows to measure phase matrix elements of ambient aerosol. The instrument can provide F_{11} and $-F_{12}/F_{11}$ with 10% and 20% uncertainty, respectively, under laboratory conditions. The optimization of the instrument and the use of appropriate data quality check approach served to continuously measure F_{11} and $-F_{12}/F_{11}$ for ambient air, but in these cases the natural variability of the air sampled typically imply large uncertainties, being the typical standard deviations of ~20% for F_{11} and between 0.1- 0.2 for $-F_{12}/F_{11}$ and therefore larger than the uncertainties of the instrument. The multiwavelength F_{11} and $-F_{12}/F_{11}$ measurements for different Saharan dust outbreaks are some of the first carried out for ambient aerosols and serve to complement laboratory measurements of mineral dust particles and of synthetic samples minerals that compose dust particles. The novel measurements of F_{11} and $-F_{12}/F_{11}$ can also complement other optical and microphysical properties of Saharan dust already known from in-situ instrumentation and by active and passive remote sensing instruments, both from the ground and the space. Nevertheless, more F_{11} and $-F_{12}/F_{11}$ measurements are needed at other experimental sites to have a more complete vision of mineral dust role on climate."

The authors must consider some aerodynamic effects that can orient the particles during their motion in the instrument, and also the particles' speed. Such parameter can change the polarization results, as shown by our team : Daugeron, D.; Renard, J.-B.; Gaubicher, B.; Couté, B.; Hadamcik, E.; Gensdarmes, F.; Basso, G.; Fournier, C. Scattering properties of sands, 1. Comparison between different techniques of measurements, Applied Optics, 45, 8331-8337, 2006.

-This is a very interesting effect that we have not considered in our measurements. However, considering the flowrate at which we measure, the speed of the particles is in the lower limit of the speeds explored in Daugeron et al., (2006), which led to less changes in the polarization results. Also, the particles we sample are not as large as the one measured in this work, so the effect will be even less evident. Thus, in our results we assume that particles are randomly oriented. Nevertheless, we think it is appropriate to clarify this point in the revised manuscript (L183-185):

"Given the flowrate used in the measurements we can assume that particles are randomly oriented, avoiding the limitations in polarization results of super-coarse particles with particle speed that can orient the particle in particular orientations (Daugeron et al., 2006)."

And the reference Daugeron et al., (2006) have been added to the reference list

Daugeron, D., Renard, J.-B., Gaubicher, B., Couté, B., Hadamcik, E., Gensdarmes, F., Basso, G., & Fournier, C. (2006). Scattering properties of sands. 1. Comparison between different techniques of measurements. *Applied Optics*, *45*(32), 8331–8337.

Figures 6, 7 and 9, and in text : The negative polarization at the large angles for the blue domain only is strange, and perhaps suspicious. Such high negative values were never observed in laboratory (and even in space) for randomly oriented compact irregular particles. More strangely, such phenomenon does not occur at the other wavelengths. The authors must explain the origin off such negative values (that in fact are almost impossible for such large dust particles). I suspect an instrumental error like a stray light contamination not accurately removed, a too log signal to noise ratio ....

We understand the referee concerns about the pattern observed in 405 nm. That was exhaustively checked by our team, and we are confident in our measurements. The possible influence of straight light was evaluated through the validation of the instrument using monodisperse aerosol (PSL) measurements in laboratory. Results are in Bazo et al., (2024). In particular, we used measurements of PSL of 0.75 µm and 1 µm. Figures below (Figure 10 in Bazo et al., (2024)) show experimental data and fits with the GRASP algorithm. Very good agreements between measured and theoretical/retrieved scattering matrix were obtained. Generally, the agreements in F_{11} are excellent, reproducing all resonances, while for $-F_{12}/F_{11}$ there are some deviations. If we assume that errors in the instruments under controlled laboratory conditions are of 5% and 10% for F_{11} and $-F_{12}/F_{11}$ (associated with issues of calibration and data acquisition of the CMOS plus other errors associated with the imperfections and preparations of the PSL sample), then the differences observed in the Figure can be explained by errors propagation. The agreement between measured and retrieved values is supported by high correlation coefficients and low residuals. GRASP retrievals were able to reproduce particle diameters and refractive index. Similar results were obtained when using Mie code instead of GRASP algorithm.

All these issues were remarked in the revised section 2.1 related to instrument description (L213-215):

"The evaluation of the instrument versus known scattered (monodisperse polystyrene spheres - PSL) showed good agreements with RMSE around 0.10 for both F_{11} and $-F_{12}/F_{11}$."

The other point that makes us believe that our measurements are right is the temporal evolutions observe during the extreme Saharan dust outbreaks – Figures are given in the supplementary material. During the peaks of these intrusions the 405 nm pattern in $-F_{12}/F_{11}$ is the same as observed for the other two channels. However, negative values in $-F_{12}/F_{11}$ are observed in the moments prior and posterior to the Saharan dust advection. If the negative value is an artifact, then it should be present at any moment.

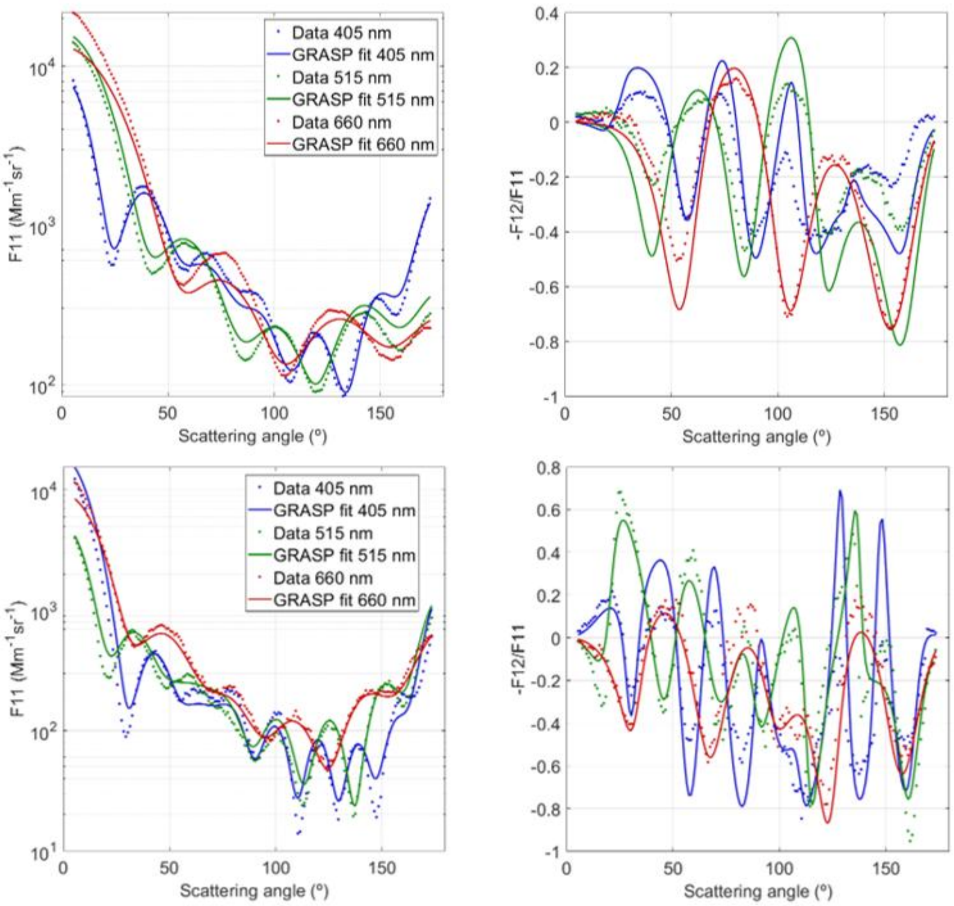

Figure 10 of Bazo et al., (2024). PSL PI-Neph measurements of F_{11} (left) and $-F_{12}/F_{11}$ (right) and GRASP fits for 0.75 µm (top) and 1 µm (bottom).

Finally, we performed computations of F_{11} and $-F_{12}/F_{11}$ using GRASP for a bimodal size distribution with different complex refractive indexes between the two modes. Particularly, we used bi-lognormal size distribution, one representative of fine mode particles with modal radius of 0.15 µm and 0.25 µm of standard deviation and the other representative of coarse mode particles with modal radius of 2.5 µm and 1 µm of standard deviation. Real refractive indexes were assumed non-spectrally dependent with values of 1.6 for fine mode and 1.55 for coarse mode. Imaginary refractive indexes were 0.0015 for fine mode and with no spectral dependency, while for coarse mode they were of 0.007, 0.005 and 0.005 for 405, 515 and 660 nm. The sphere fraction was also fixed for each mode, being 0.7 for fine mode and 0.05 for coarse mode. Three different scenarios were generated giving different weights to each mode: The first is for volume concentrations of 0.3 $\mu m^3/\mu m^3$ for each mode and can be considered as representative of a mixed case where both modes have a similar weight. The second presents more predominance of coarse mode (volume concentrations of 0.5 $\mu m^3/\mu m^3$) but with non-negligible contribution of anthropogenic particles (volume concentrations of 0.1 $\mu m^3/\mu m^3$) in the fine mode. The last scenario is representative of pure dust mode (volume concentrations of 0.5 $\mu m^3/\mu m^3$) with negligible fine mode contribution (volume concentrations of 0.01 $\mu m^3/\mu m^3$).

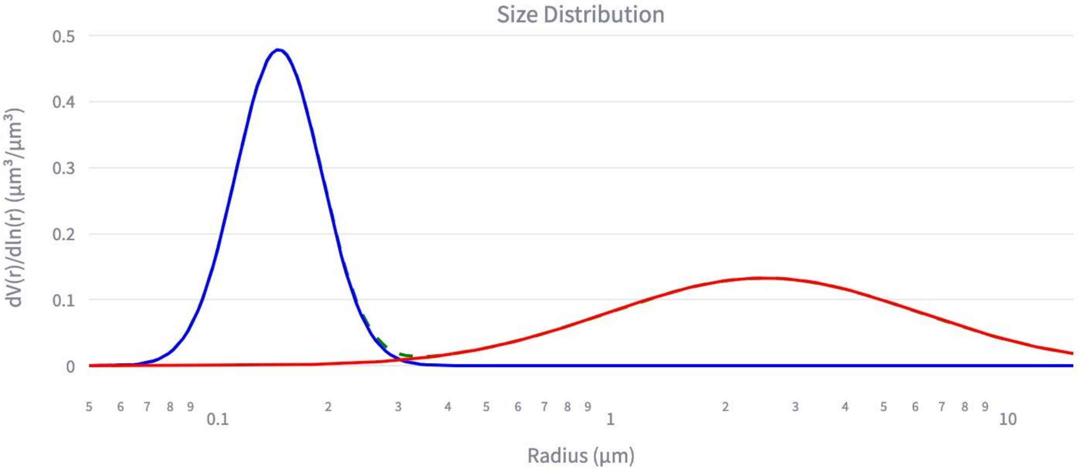

Figure R1. Volume size distribution used for the GRASP simulations of the ‘Urban mixture’ case.

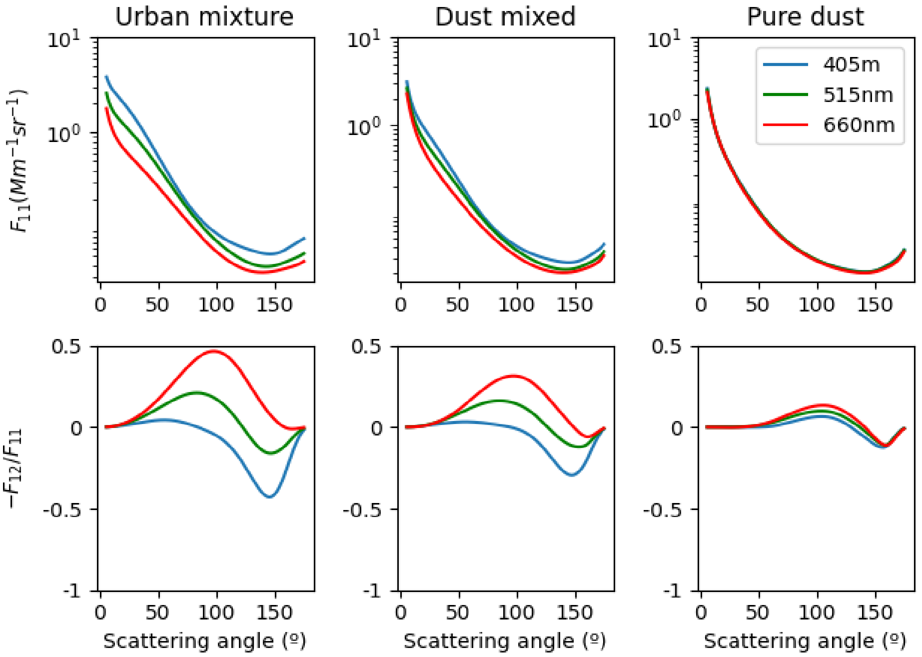

Figure 12 in the manuscript. Simulations of phase function (F_{11}) and polarized phase function ($-F_{12}/F_{11}$) using GRASP forward for three different combinations of bi-lognormal size distributions. *Urban Mixture* with approximately the same weight of fine and coarse mode, *Dust mixed* with predominance of coarse mode but with a non-negligible influence of the fine mode, and *Pure Dust* with strong predominance of coarse mode and negligible fine mode. Note that refractive indexes and sphericity of each mode are different.

The feature of negative values in $-F_{12}/F_{11}$ for the 405 nm channel is also observed in the GRASP simulations. We are aware that the negative polarization is not as high as in our measurements, but further optimizations in GRASP are still needed (i.e. implementation of super-coarse mode). Nevertheless, GRASP simulations support the possible existence of negative values in $-F_{12}/F_{11}$ in the 405 nm for a mixture of dust and anthropogenic particles. These points have been added to the conclusions section (L849-860):

“Simulations performed by the GRASP code for different mixtures of fine mode (anthropogenic particles) and coarse mode (dust particles) revealed that F_{11} and $-F_{12}/F_{11}$ are sensitive to the different contribution of each mode in the mixture, being especially critical for $-F_{12}/F_{11}$ on the 405 nm channel. The negative values for $-F_{12}/F_{11}$ in 405 nm were observed more clearly for the mixture of fine and coarse particles. Thus, these simulations have served to understand the experimental negative values in $-F_{12}/F_{11}$ not observed in laboratory measurements for collected dust. Retrievals of bimodal size distribution with separate refractive indexes for each mode would have shown clarity to this problem. Hower, such retrieval with GRASP using F_{11} and $-F_{12}/F_{11}$ as inputs need to be optimized. Another additional optimization in GRASP will imply the possibility of implementing the retrieval of super-coarse mode particles. Nevertheless, the possibility of explaining the spectral differences in F_{11} and $-F_{12}/F_{11}$ with wavelength has served to understand the temporal evolution of the extreme dust events and the difference and similitudes when comparing versus laboratory measurements and versus other more moderate events of Saharan dust transport.”

Line 389: The wavelength effect was largely studied in the PROGRA2 data base and largely published by our team.

-Thanks for the suggestion. We have added the reference Renard et al., (2014) in this part of the result discussion.

Line 465: Aging can produce more compact particles, but it is difficult to call them “spherical”.

-Thanks for pointing this out. Referee 2 raised several issues in this paragraph, and thus we have re-written the paragraph. The sentence ‘*The values of g suggest the higher predominance of spherical particles when compared to the other cases, so it is likely that the urban background particles suffered from aging*’ has been removed. Now, we only remark that the values of g suggest the lower contribution of dust particles.

Line 485: This analysis could be inaccurate. Other effects than the sphericity of the particles must be considered (size, refractive index, porosity). The presence of “spherical particles” do not change significantly the shape of the light scattering curves (of course, only a medium composed of perfect spheres can change the shape of the curve).

-We agree with the referee and we have removed the second part of this sentence.

Figure 11: Yellow dots are difficult to see.

-Thanks for the suggestion. We have changed the colors of the dots to light blue for better visualization.

Line 620: Do they authors have also considered the super-coarse mode of particles ? Keep in mind that the largest particles are the most luminous, and thus can dominate the scattered (polarized) intensities.

-We have not considered super-coarse particles for the simulations performed with GRASP. To our knowledge, the super-coarse model is not available in the latest version of GRASP. The objective of the simulations is to show the variability in F_{11} and $-F_{12}/F_{11}$ with the mixtures of dust and anthropogenic pollution.

We agree with the referee that the super-coarse mode can be presented in the long-range Saharan dust transport. For the extreme event on 15th-16th March 2022 we measured the size distribution for the deposited dust (Figure R2) and the presence of super-coarse particles in the sample is low compared to the fine and coarse modes. Additionally, the modal radius of the coarse mode agrees with the aerosol size distribution used in GRASP simulations. This has been added to the discussion in the revised manuscript (L730-731).

"The modal radii selected are close to those observed for the particle size distribution of the deposited particles (not shown for clarity)."

However, as the referee suggests, the residual super-coarse mode can have an impact on the phase matrix. This has been remarked in the revised manuscript. This super-coarse mode needs to be also implemented in GRASP. We have added the following sentence to the manuscript (L768-770):

"It is important to mention that the super-coarse mode can also affect the behavior of F_{11} and $-F_{12}/F_{11}$ and the presence of this mode is also observed for long-range transport (i.e. Renard et al., 2010). Future GRASP developments also need the consideration of this super-coarse mode."

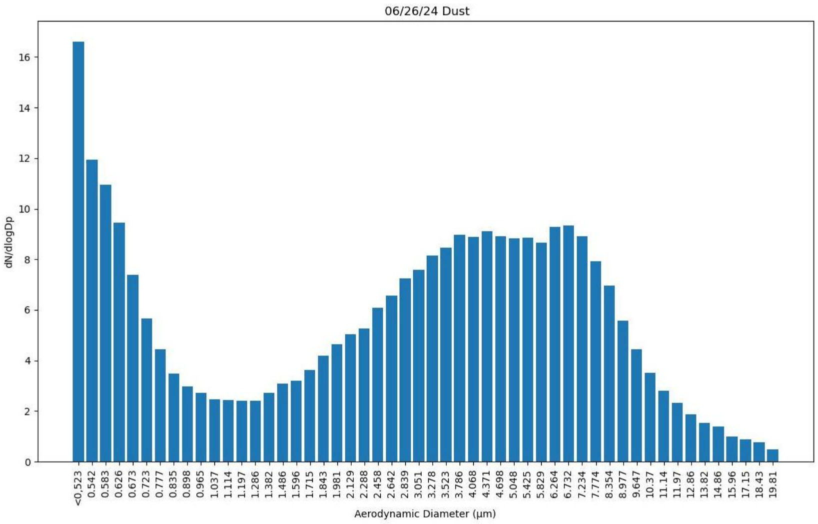

Figure R2. Number size distribution measured for the deposited dust for the extreme dust event on 15th – 16th March 2022.

Figure 12: Negative polarization down to -0.4 are unrealistic for dust sample, unless the size distribution is dominated by submicron particles without large ones.

-Figure 12 shows the simulations with GRASP using a bi-modal size distribution with different refractive indexes (see previous comments). The objective was to show F_{11} and $-F_{12}/F_{11}$ for different mixtures of dust with anthropogenic pollution. In our station, background conditions are dominated by anthropogenic particles. During a Saharan dust intrusion, the dust particles mixture with these background particles lead to different mixtures of particles that ultimately were measured by the PI-Neph. The simulations show that the presence of anthropogenic particles affect F_{11} and $-F_{12}/F_{11}$ when compared with the case of only dust particles. This last point is more relevant for -F_{12}/F_{11} in the 405 nm channel and serves to understand the behaviors observed in Figure 11 and in the temporal evolutions during the extreme dust event given in the Supplementary Material. The text has been accordingly modified (L758-762):

"Figure 12 results show how the presence of anthropogenic particles (fine mode) can alter the spectral dependencies in -F_{12}/F_{11} when compared with only dust particles (coarse mode) in the sample, particularly in the blue channels. However, changes in the F_{11} patterns were not so evident. These results help to understand the different phase matrix elements discussed in this manuscript, and their temporal evolutions during the extreme dust events (Supplementary Material)."