Responses to Reviewer and Editor

Unfortunately, the revised version is not in an appropriate format which creates a lot of additional effort for the reviewer. Therefore, I decided to hand the document back to the authors with (still) major revisions.

Technical Comments
The line numbers in the author's response (especially those in the response to my major comment #1) do not correspond neither to the revised manuscript nor to the track changes document. Therefore, it is hard for me to judge if the discussion of the limitations is really introduced in the manuscript appropriately. Please prepare an updated version of the author's response with the correct line numbers and best copy the additional statements to the author's response.

We have added all line references to the response in red to make it easier for you at the end of this document.

Furthermore, the track changes document is not formatted well. It is hard to follow the small comments on the side. Please prepare a proper track changes document with additions in color and deletions crossed out.

You also mixed up the matrices for the revised version (eq. 4, 5, 7). If you prefer to use Microsoft Word, then, please take all the time to apply the correct formatting before submission. It is not acceptable in the current version.

Corrected.

Additionally, you state that you have updated Fig 6-8 to start with the size parameter of 0. But then I look in the revised manuscript, I see that you have just updated Fig 6 + 8, but not Fig 7.

Corrected.

Similarly, you state that the abbreviation ASL in the author's contribution is corrected. But it is not. ASL is still in and it remains undefined to whom it corresponds. Anthony La Luna? But there is no S.

Changed to AL for your convenience.

Please provide more information on the settings used for ADDA (see comment by referee #3).

For the use of ADDA, we follow the ‘rule of thumb’ described by the user manual of having at least 10|m| (where m is the complex refractive index) dipoles per wavelength in resolution of the particle used in calculations.

“*The rule of thumb for particles in free space with size comparable to the wavelength is: “10 dipoles per wavelength inside the scatterer”, i.e. size of one dipole is*
$d = \lambda/(10|m|)$ *, (1)*
*where* $m$ *is refractive index of the scatterer. That is the default for ADDA (§6.2).*”

The dipole creation and mappings are discussed further in Conny et al., 2019, with greater details regarding how the files were created in Conny and Ortiz-Montalvo, 2017.

Additionally, as described in section 3.3, the other notable ‘setting’ of ADDA is that of the internal averaging setup. For this, the user provides a file of the following format:

```
# Description of the parameters for orientation averaging
#
# this file should be manually modified by user
# Program does not assume any symmetries of the particle. Therefore, possible symmetries should be considered by user
# and this can lead to decrease of integration limits.
# Here zyz-notation (or y-convention) is used for the Euler angles.

alpha:
# calculation for alpha is cheap but only precalculated, therefore Jmax should be rather large.
# Jmin and eps are really not used.
# Completely irrelevant, when only cross sections are calculated ('-scat_matr none').
# Do not change the range from default unless you have a good reason; using only one value of gamma for beta=0,pi is
#   done only when full range is specified here to avoid possible inaccuracies.
# default: min=0;max=360;Jmax=5;equiv=true;periodic=true
min=0
max=360
Jmin=2
Jmax=5
eps=0
equiv=true
periodic=true

beta:
# default: min=0;max=180;Jmin=2;Jmax=4;eps=1e-3;equiv=false;periodic=false
# xy - symmetry plane: max=90;Jmax=3
# Do not use periodic=true since the function is multiplied by sin(beta) before integration.
min=0
max=180
```

Jmin=2
Jmax=4
eps=1e-3
equiv=false
periodic=false

gamma:
# default: min=0;max=360;Jmin=2;Jmax=4;eps=1e-3;equiv=true;periodic=true
# axysymmetrical: max=0
#   more precisely: max=45;Jmax=2;equiv=false
min=0
max=360
Jmin=2
Jmax=4
eps=1e-3
equiv=true
periodic=true

# all angles are specified in degrees
# Jmin,Jmax are minimum and maximum numbers of refinement stages
# Nmax = 2^Jmax + 1
# for those with equiv=true Nmax is effectively less by 1
# total calls of function <= Nmax_theta * Nmax_phi

# equiv means whether it is assumed that max and min values are completely equivalent. If true only one of them is
# calculated.

# periodic means whether function is periodic in the integrated interval. If true trapezoid rule is used; it is possible
# that interval is half of the function period.

# axysymmetrical <=> particle with z - axis of symmetry

This is adjusted in our case from the default provided to reduce the eps to 0 and Jmin=Jmax=n, where n is the level of orientations described in the manuscript. Orientation and details of simulations such as these are often overlooked in studies utilizing DDA computations, simply mentioning they were “randomly oriented” with some comment provided to size and refractive index used is generally seen as sufficient (e.g. Tsekeri et al., 2020, Zhang et al., 2015, Bi et al., 2010). Even more rigorous discussion of DDA often lacks specific reasoning for the number of orientations used (e.g. Carpine et al., 2025). This is why we believe our discussion in section 3.3 is important to provide a method of verification to orientation averaging previously overlooked.

Bi, L., Yang, P., Kattawar, G. W., & Kahn, R. (2010). Modeling optical properties of mineral aerosol particles by using nonsymmetric hexahedra. *Applied Optics*, *49*(3), 334–342. https://doi.org/10.1364/AO.49.000334

Carpine, M.-A., Ysard, N., Maury, A., & Jones, A. (2025). From small dust to micron-sized aggregates: The influence of structure and composition on the dust optical properties. *Astronomy & Astrophysics*, *698*, A200. https://doi.org/10.1051/0004-6361/202554575

Tsekeri, A., Freudenthaler, V., Doxastakis, G., Gasteiger, J., Louridas, A., Georgoussis, G., et al. (2020). Polarization Lidar for Detecting Dust Orientation. EPJ Web of Conferences, 237, 02028. https://doi.org/10.1051/epjconf/202023702028

Zhang, Q., & Thompson, J. E. (2015). A model for absorption of solar radiation by mineral dust within liquid cloud drops. *Journal of Atmospheric and Solar-Terrestrial Physics*, *133*, 121–128. https://doi.org/10.1016/j.jastp.2015.08.013

Scientific Comments:
Even though realistic shapes were applied, the modelled optical properties of these particles are very different from observations. I gave various suggestions in my review, to harmonize the results, e.g. the spectrally dependent CRI, which leads according to your response (Fig. R1) to lower depolarization ratios. But still, there is the large difference compared to the values of CALIPSO in Fig 6 which remains undiscussed. You show the CALIPSO values of Liu Z., et al., 2015, but you don't discuss the differences. It is probably to the polydisperse aerosol in the atmosphere and the limited size range of your studied particles. This large difference to observations makes me skeptical about your DDA results.

The CALIPSO lines we showed in Figure 6 are **global mean** values used by their operational product team, while our results are based on only about a dozen FIB dust samples, which as you have pointed out many times are highly limited in terms of sampling. You are also correct that our results in Figure 6 are single scattering properties for individual particles, while the CALIOP observations are due to volumetric scattering by polydisperse dust with a particle size distribution (PSD). Again we recognize this difference and it is why we extended the single scattering results to PSD averaged results in Section 5. As Figure 11 suggests, the DPR of polydisperse dusts depends on particle size,with the asymptotic value corresponding to the **maximum value rather than the mean value**. Take the case in Figure 12 for example. The

dust DPR at 532 nm is around 0.32 when we use the PSD at Cape Verde observed by AERONET. Note that Cape Verde is close to the dust sources, so dust sizes there are larger than the long-range transported regions where the DPR can be expected to be significantly smaller. We have further added to the discussion on this point again in lines 394-398.

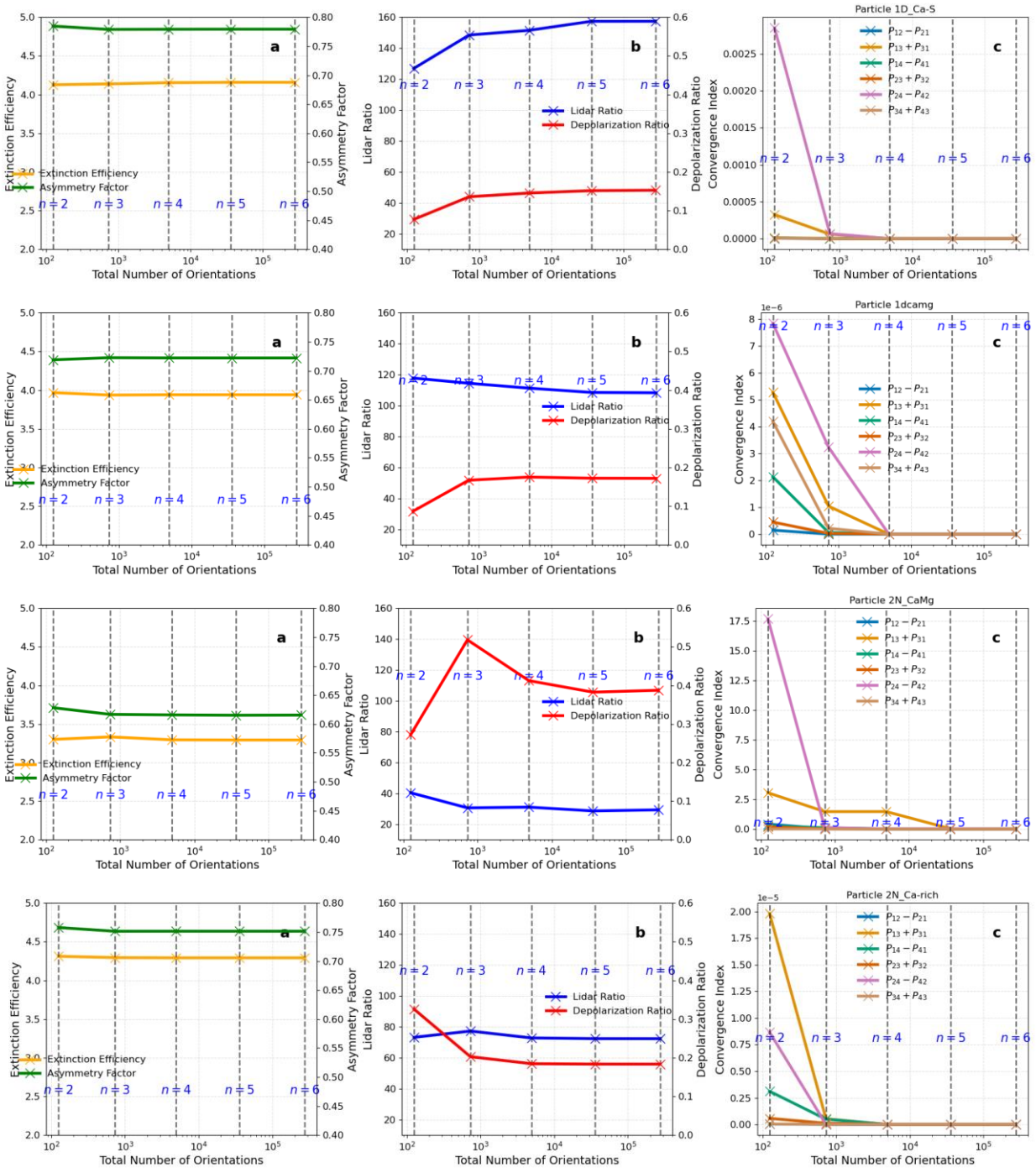
*These asymptotic values differ from the global averages of CALIPSO shown in Figure 6 due to the differences between single scatterers and volumetric measurements produced by a lidar instrument in the atmosphere.. Also note that the asymptotic values correspond to the upper limits while the CALIPSO results are the global mean values used by their operational product team.*

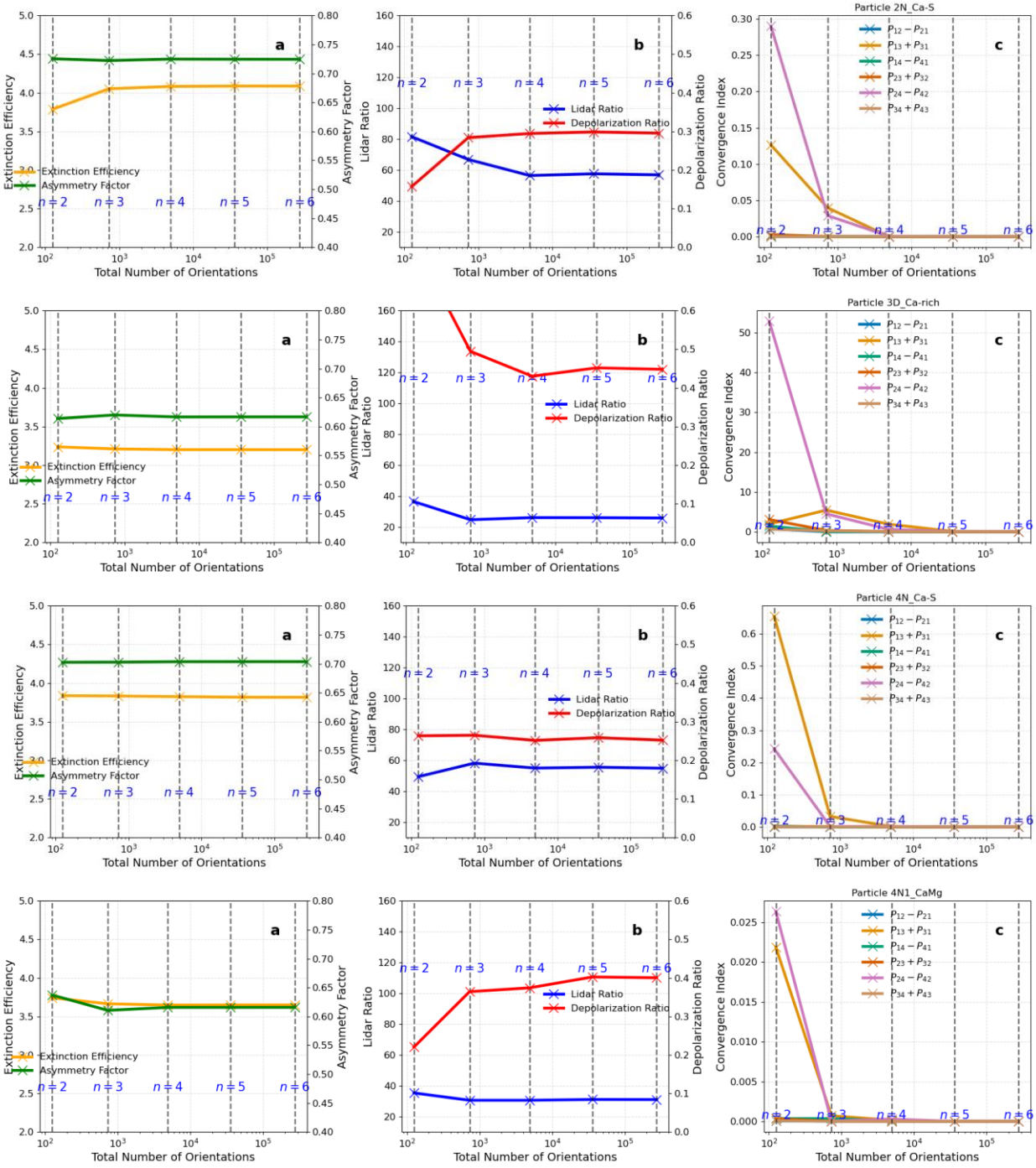
Therefore, I ask for a further proof that you have used enough orientations for your DDA calculations.
The 3D Ca-rich particle has a lower SSA compared to the other particles (Fig 7) and it marks in Fig 4 the lower boundary of P22/P11. The estimate of the number of orientations was done with this specific particle, which is more absorptive than the other samples. Why? How would your convergence plots look like for a with higher SSA? Or in other words: Is the 3D Ca-rich particle really representative to estimate the necessary number of orientations? Especially in the backscatter direction, one has to take care to use enough orientations to describe the scattering properties accurately. E.g., Gasteiger et al., 2011, concludes that orientation averaging is the largest source of error for the ADDA modelled particles. Taking the most absorptive particle from the ensemble might lead to an earlier convergence. Please proof that the convergence at n=5 holds for at least one other, less absorbing particle as well. Probably, you have already done these calculations for your own checks, but please provide this proof to the reader of the paper as well. And please provide a quantitative criterion for the convergence at n=5. In the manuscript it is just stated with a reference to Fig. 3, but no (threshold) value is provided.

The orientation convergence of 3D Ca-rich is shown for continuity across the analysis shown highlighting one such particle from the group as done in other figures in the manuscript (e.g., Figure 2).
"*Especially in the backscatter direction, one has to take care to use enough orientations to describe the scattering properties accurately. E.g., Gasteiger et al., 2011, concludes that orientation averaging is the largest source of error for the ADDA modelled particles.*"
We cannot agree more. It is precisely why we elaborated in great detail in Section 3.3 how we treat the random orientation in ADDA and what criteria are used to assess the convergence. Our method is based on solid physics, e.g., scattering matrix symmetry. We believe that it addresses the concerns raised by Gasteiger et al., 2011, and therefore is a valuable contribution to the literature.

For your request, the convergence test was done for each particle at 532 nm for one refractive index each, shown in Figure R2.1.

The number of orientations used in these simulations are very important for accuracy. We believe the number of orientations used in these results are sufficient to report with minimal error. In fact, particles run with n=5 (35937 orientations) exceed the number of orientations used for Gasteiger et al., 2011 (16832 orientations). As shown in figure R2.1, the variations of lidar properties is generally under 5% from n=4 to n=6, and for those with greater variation (e.g. particle 2N_CaMg) n=6 was used.




Particle 2N_Ca-S
Particle 3D_Ca-rich
Particle 4N_Ca-S
Particle 4N1_CaMg
a
b
c
Extinction Efficiency
Asymmetry Factor
Lidar Ratio
Depolarization Ratio
Convergence Index
Total Number of Orientations
n = 2
n = 3
n = 4
n = 5
n = 6
$P_{12} - P_{21}$
$P_{13} + P_{31}$
$P_{14} - P_{41}$
$P_{23} + P_{32}$
$P_{24} - P_{42}$
$P_{34} + P_{43}$

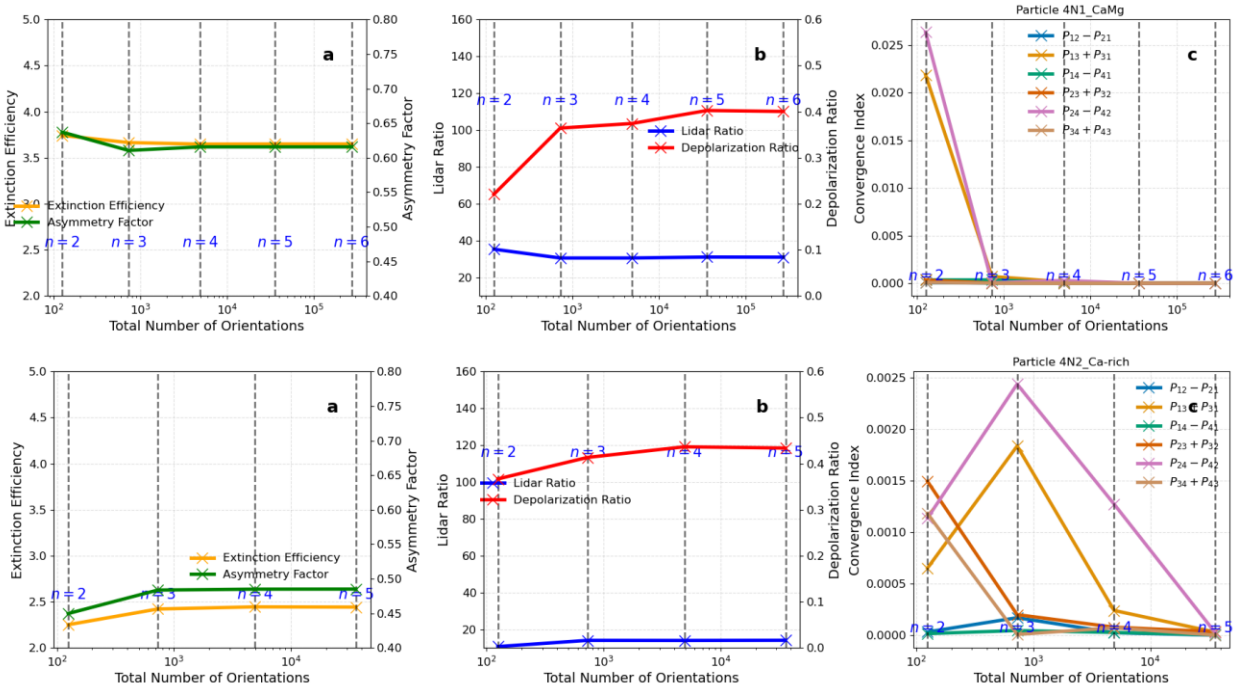



Figure R2.1 (a) Change in extinction efficiency and asymmetry factor with increasing number of orientations for each particle used in the manuscript. (b) $S$ and linear depolarization ratio as function of the number of orientations for dust particle. (c) Convergence Index for each dust particle's Mueller index pairs at 532 nm. Note figures start at $n = 2$.

**Public justification (visible to the public if the article is accepted and published)**:
Dear Anthony,

thank you for addressing the comments of the Referees and for providing your updated files. There are a few items I would like you to consider before I can come to a final decision regarding your submission.

First of all, please have a look at the additional comments by Referee #1 who suggests a few more clarifications.
It seems to me that many of the Referee comments are based on reconciling properties of single particles versus bulk particles. I would therefore like you to carefully screen the text one more time to ensure that the corresponding discussion that puts your findings into the context of earlier work is specific as to whether single or bulk particles are referred to.
Finally, I have some comments regarding your figures:
- Figure 1: There is no need to extend the display up to 30 km. Please consider restricting the height range to 15 km.

We have reduced the height of the CALIPSO plots.

- Figure 3: Everything in these plots appears very small. Also, is it really necessary to use different colours and line styles.

The size of lines and labels were increased, and solid lines were used.

- Figures 6 and 8: I understand that you use the different symbols for consistency with Figures 5, 7, and 10. However, Please consider using colours (355 in blue, 532 in green, 1064 in red) for an even better discrimination of the different wavelengths.

Colors for wavelength were added to the plots.

- Figure 9: Have you considered plotting those points as PLDR over LR with effective sphericity as colour coding? As is, it is not straightforward to connect the individual points in the two panels.

We have added the described plot to Figure 9.

With best regards,
Matthias

Additional private note (visible to authors and reviewers only):
Dear Anthony,

as you have decided to work with the Word template, I would like you to take extra care of

formatting issues when creating your files for upload. This makes it much easier for everybody involved in the assessing your work and preparing your files for publication.

With best regards,
Matthias