Response to Darrel Baumgardner.

We thank the reviewer for his thorough examination of our manuscript. The reviewer's suggestions have led to a significant expansion of our work.

Below are the responses to the reviewer's comments, these latter partially reported (in bold) and indented for ease of reading. We have also reported, in italics and indented, the relevant additions/modifications to the manuscript.

A)...I think that how they applied the code was also too simplistic, e.g. assuming that all the particles had the same AR rather than trying combinations of size-dependent AR.

We have used a simplified assumption, given our limited knowledge of NAT crystallization. There has been speculation of anisotropic growth, favoring large asphericities when particles grow to large sizes, due to less depleted vapor mixing ratios close to the extremity of the crystals (Grothe et al., 2006). Also, various particle shapes and habits might coexist due to different nucleation and growth histories. Thus, it is certainly possible that a choice of particle size-dependent AR exists, which may improve the agreement with backscattering/depolarization measurements. However, such an agreement, if found, would be easily open to further criticism since there is no basis to make complicated assumptions about the size dependent AR. By suitably adjusting the AR, we would regard such a result as a selection of the most desirable outcome. For these reason, in looking for the AR intervals that best match the backscattering measurements, we prefer to consider only an average AR, as the most conservative assumption. Nevertheless, we have commented on such option in the paragraph <u>4 Discussion</u>:

In the T-matrix theory, for fixed AR, the depolarization depends on the particle size and maximizes for particular sizes. There is certainly a way to assume a particle size-dependent AR in our PSDs so as to reconcile the computations with the observed values. However, such an approach would have little physical basis and could only be justified to maximize the agreement of calculations. Therefore, we have not explored this possibility further, although it is possible that our simplified hypothesis of a common AR for every particle may be the cause of the bad agreement between data and calculations in some case.

In the extension of our work to meet the reviewer's suggestion, we have explored the possibility of deriving the AR that best matches computation and experiment, on a case by case basis. This is the main upgrade of our work, stimulated by the revision process. In the new version, we considered that the previously presented β or δ analysis with respect to (R_{th}, AR) can only constrain a range of R_{th}. In fact, from figures 2 and 3, you can see that R_{th} must be about 0.5-0.8 μ m, while the compatible ARs are all those between 0.3-0.5 and 1.5-3. Hence, at that stage, we acknowledge that a particular choice of AR is somewhat arbitrary.

We decided then to perform an additional simulation. We kept R_{th} fixed at values in the range 0.5-0.8 μm and, for each of the experimental delta we looked for the AR that best matches the T-matrix calculations, in the ranges $\,$ 0.3-0.55 and 1.5-3. Then we selected the AR which gave the best match as R_{th} varied within 0.5-0.8 μm .

Once found these AR, the same were used to compute β and compare it with its measured values, on a case-by-case basis.

This procedure has led to a net improvement in the agreement between experimental and calculated δs , while it has not changed appreciably the agreement between the βs .

We have described this new procedure in the Abstract:

The parameters R_{th} and AR of our model have been varied between 0.1 and 2 μ m and between 0.3 and 3, respectively, and the calculated backscattering coefficient and depolarization were compared with the observed ones.

The best agreement was found for R_{th} between 0.5 and 0.8 μ m, and for AR less than 0.55 and greater than 1.25.

To further constrain the variability of AR within the identified intervals we have sought an agreement with the experimental data by varying AR on a case-by-case basis, and further optimizing the agreement by a proper choice of AR smaller than 0.55 and greater than 1.5, and R_{th} within the interval 0.5 and 0.8 μ m. The ARs identified in this way cluster around the values 0.5 and 2.5.

In paragraph 2.3 Variability with the threshold radius R_{th} and Aspect Ratio AR:

The result of this study allows to identify only the best R_{th} , resulting around 0.5-0.8 μ m, while the ARs compatible with the measurements are all those between 0.3-0.55 and 1.5-3. To further constrain AR we have kept R_{th} at a fixed value, chosen between 0.5 and 0.8 μ m and changed this value with a 0.1 μ m step. For each of these fixed R_{th} , and separately for each PSD, we identified in the intervals (0.3-0.55), (1.5,3) the value of AR which best matched the observed δ with its computed value. Finally, for each PSD we selected the pair R_{th} and AR which provided the best match. Once the ARs and R_{th} have been selected by forcing the agreement between the δ_A , the same ones have been used for the calculation of the β_A .

Figures 4 and 5 have been upgraded with the result of this new approach:



Figure 4. Scatterplot of computed vs measured particle backscattering coefficients β_{A} . The ARs used for the computations have been selected, case by case, to produce the best

agreement between the β computed and measured, and are here represented in color coding. Only ARs in the intervals between 0.3 and 0.55, and between 1.5 and 3, have been considered. R_{th} was also selected within the interval 0.5-0.8 μ m to provide the best match. We report data points with BR greater than 1.2, $\beta_{cross A}$ greater than 5 1026km21sr21 and temperature at the observation below 200 K.



Figure 5. Scatterplot of computed vs measured particle depolarization δ_A . The ARs used for the computations are those that provided the best match between the δ_A computed and measured, and are here represented in color coding. Only ARs in the intervals between 0.3 and 0.55, and between 1.5 and 3, have been considered. R_{th} was also selected within the interval 0.5-0.8 μ m to provide the best match.. We report data points with BR greater than 1.2, β_{cross} A greater than 5 10⁻⁶km⁻¹sr⁻¹ and temperature at the observation below 200 K.

We have also discussed the physical relevance of our results, in the par. <u>5. Discussion</u>, which has been extensively rewritten. See our answer to "2)Diagnose their model results to understand why they are producing unacceptable comparisons."

B) ... the authors never explain how their study will benefit the science, not in the introduction nor in the concluding remarks; hence, the questions are 1) What purpose does constraining the model parameters serve?

Good point. We have not sufficiently placed our study in the broader perspective of PSC research. We have broadened the <u>Introduction</u> to include there the fact that the subsidence of large NAT particles is considered one of the main causes of denitrification of the polar winter stratosphere. Their settling time influences this process, which is in turn dependent on NAT particle shape and size, both of which determine their settling speed and lifetime, hence their denitrification efficacy. Of particular interest is the shape, given that non-spherical particles may fall significantly slower than volume equivalent spheres. A positive result of our study, which maybe we did not adequately underline, is that it strongly suggests avoiding ARs too close to 1.0, preferring ARs below 0.55 or above 1.5. This result has been underlined and referred to previous estimates on the asphericity of NAT particles.

We have reported such considerations in the Introduction:

The aims of this effort are both to verify the ability of the T-matrix approach to reproduce the observations from lidar/backscattersonde, once the PSDs are supposed known, and to provide a contribution to the estimation of the shape and size limits of the NAT PSC particles. The question of the shape of NAT particles is in fact far from being clarified, and has important implications for the denitrification mechanisms of the polar stratosphere, an important step in the process that lead to the destruction of stratospheric ozone. In fact, large PSC NAT particles settling down are considered one of the main causes of denitrification of the polar winter stratosphere (Di Liberto et al., 2015). Their settling time influences this process, and it is in turn dependent on NAT particles shape and size, both of which determining their settling speed and lifetime, hence their denitrification efficacy. Woiwode et al. (2014) assumed significantly non-spherical NAT particles to simulate the NAT settling speed leading to a the vertical redistribution of HNO₃ observed between two companion flights during the RECONCILE airborne field campaign in the Arctic (von Hobe et al., 2013). Woiwode et al. (2016, 2019) have also suggested that NAT particles may be highly aspherical based on the infrared spectrometer MIPAS-STR limb observations exhibiting a spectral signature around 820 cm⁻¹ and an overall spectral pattern compatible with large highly aspherical NAT particles. T-Matrix calculations assuming randomly oriented highly aspherical NAT particles (aspect ratios 0.1 or 10 for elongated or disk-like spheroids, respectively) were able to reproduce the MIPAS-STR observations to a large degree. Molleker et al. (2014) hypothesized strongly aspherical NAT particles to reconcile the amount of the condensed HNO3 resulting from PSC cloud spectrometer measurements with the expected stratospheric values, and to provide consistency between particles settling velocities and growth times with back trajectories. Moreover, Grothe et al. (2006) observed highly aspherical NAT in laboratory experiments. This is in contrast with earlier studies that assumed an AR = 0.9 for the NAT spheroids to match microphysical model simulation with airborne (Carslaw et al., 1998) or satellite borne (Hoyle et al., 2013; Engel et al., 2013) lidar observations.

C) ...2)On what basis do they declare that these ranges are reasonable?

As quoted above concerning NAT asphericities, Molleker et al. (2014) and Woiwode et al. (2014; 2016; 2019) have suggested that NAT particles may be highly aspherical.

Similarly, our study concludes that the best agreement between measurements and optical modeling occurs for strongly aspherical NAT.

This is in contrast with earlier studies that assumed an AR = 0.9 for NAT spheroids (Carslaw et al., 1998, Hoyle et al., 2013, Engel et al. 2013).

Concerning the R_{Th} , the hypothesis of dividing the PSD into a liquid part and a solid part on the basis of size is a hypothesis supported by what we know about PSC particle formation and

measurements (Deshler et al., 2003), leads to sensible results, and is in agreement with the depolarization-large particle correlation qualitatively presented in fig.1

D) ...3) Since they conclude that using prolate and oblate spheroids to model the scattering did not lead to useful results, that constraining the AR range is meaningless.

Evidently we failed to convey our conclusion correctly. The study leads to three useful results: i. in an externally mixed PSC, it is reasonable to place a threshold radius R_{Th} around 0.6 µm, which divides the liquid part from the solid part of the particulate; ii. It is sensible to expect strongly aspherical shapes for the solid part of the particulate; iii. the observed depolarization is difficult to reproduce by a T-matrix approach. The latter can be considered a negative result, but it is a result nonetheless.

These considerations are reported in the Conclusion.

... our analysis has provided the range of optimal R_{th} and AR parameters that best match the observations. To sum up: i. in an externally mixed PSC, it is reasonable to place a threshold radius R_{th} between 0.5 and 0.8 μ m which divides the liquid part from the solid part of the particulate; ii. It is sensible to expect strongly aspherical shapes for the solid part of the cloud; iii. There are cases, in particular those related to high depolarization observations, in which, within our assumptions (i.e. a single form for the solid particulate, a fixed threshold radius for all PSD) prevents to reproduce the observed depolarization with a T-matrix approach.

In his suggestion to resubmit the manuscript, the reviewer asked to:

1) Explain the importance of knowing the particle sizes and shapes in mixed phase PSCs.

As outlined above, (see point A)) we have expanded the <u>Introduction</u> by placing our study in the broader perspective of PSC research.

2) Diagnose their model results to understand why they are producing unacceptable comparisons.

The novel approach we have pursued in the revision of our manuscript has led to new results. This has led us to a major revision of paragraph <u>4. Discussion</u>. We report here the full text:

The identification of the best R_{th} in the range 0.5 -0.8 μ m supports what we already know from the theoretical understanding of NAT particle formation in PSC and from measurements (Deshler et al., 2003b). Concerning particle shape, in our model all solid particles in a single PSD share the same AR, but different PSDs can have different ARs. This approach could suggest that the choice of the AR which, case by case, optimizes the agreement between calculations and measurements, may be the result of chance rather than physics. There are two facts that counter this criticism.

First, it appears that the selected ARs may be related to the shape of the PSD. Figure 6 shows the 2D-histogram by occurrence of ARs and of $N(r > 0.7 \mu m)/N_{tot}$, the ratio between particles with radius greater than 0.7 μ m and total particles, which is a parameter related to the PSD shape. In Figure 6 the AR are not distributed randomly. Conversely, there is a tendency for the AR to grow as the percentage of large particles increases. In fact AR values tend to peak around 0.5 in the lower $N(r > 0.7 \mu m)=N_{tot}$ range, while tend to cluster around 2.5 when N(r > $0.7 \mu m$)= N_{tot} is higher. The shape of the PSD mirrors particle formation conditions and history, is linked to the presence of solid particles, as already highlighted in the discussion of Figure 1, and is likely linked to the average particle shape as well.

Second, if we consider the sequences of measurements acquired in individual balloon flights, the corresponding sequences of selected ARs do not evolve randomly but, conversely, are autocorrelated. An example of this behavior is shown in Figure 7, where the time series of β and δ are reported respectively with red and blue dots. The ARs that provide the best agreement between experiment and simulation are shown with black dots. It can be seen that temporally contiguous observations often result in the selection of the same AR. Contiguous observations of PSD are likely to have similar characteristics in terms of microphysics, and this seems to be correctly reflected in the constancy of AR. We are therefore confident that our method produces results with a physics-based content.

In general, our model leads to good correlations between measured and modeled bs. For the ds the measurements are well reproduced by the calculations in many instances, as is the case for many of the selected ARs in the range 0.3-0.55. However, there are other cases in which the agreement is worse (when the best ARs have been selected in the range 1.5-3), or does not occur at all, as in the cases of observed depolarizations greater than 30%. In these latter cases, the impossibility of reproducing the observed values even under the hypothesis of a completely solid particles implies that, for those PSDs, our model is not able to produce the observed depolarizations. In these particular cases in which the model performs particularly badly, there may be problems of inhomogeneities of the cloud. These cases come from Antarctic observations, for which the microphysical observations from the balloon and the optical ones from ground-based lidar are separated geometrically, so that the two instruments sample air masses separated by several tens of kilometres, and it may be the case that some clouds were not homogeneous on such spatial scales.

Different shapes produce different polarization, according to T-Matrix. This has also been proven experimentally since the early work of Sassen and Hsueh (1998) and Freudenthaler et al. (1996) that showed how lidar depolarization ratios in persisting contrails ranged from 10% to 70%, depending on the stage of their growth and on temperature. In the T-matrix theory, for fixed AR, the depolarization depends on the particle size and maximizes for particular sizes. There is certainly a way to assume a particle size-dependent AR in our PSDs so as to reconcile the computations with the observed values. However, such an approach would have little physical basis and could only be justified to maximize the agreement of calculations. Therefore, we have not explored this possibility further, although it is possible that our simplified hypothesis of a common AR for every particle may be the cause of the bad agreement between data and calculations in some case.

To further investigate the causes of the mismatch we turn to the study of the climatology of PSC observations collected from McMurdo's lidar. The measurement of a PSC composed exclusively of solid particles is a rare and uncertain event. The absence of liquid aerosols is difficult to determine for certain. However, Adachi et al. (2001) demonstrated that in a plot of the Total Volume Depolarization δ_T versus 1-1/BR, the experimental points of solid, liquid or variously mixed PSCs are distributed within a triangle whose vertices are (1, 0), (1, $\partial^{\text{usph}}_{TA}$) and (0, δ_{mol}). These vertexes represent respectively the value of δ_T in the case of pure liquid clouds and pure solid clouds for BR =1, when the δ_T coincides with δ_{TA} , and in the case when no particles are present the d_T attains its molecular value δ_{mol} (Young, 1980). Hence the extrapolated intercept on the y axis at BR =1 is precisely $\partial^{\text{usph}}_{TA}$. This procedure allows us to estimate this asymptotic value. This requires the assumption that the experimental points that fill the triangle of vertices defined above represent PSC observations in mixed phase in which all

solid particles share the same aerosol depolarization. Alternatively, one can interpret differently the presence of the data points filling the triangle. These points may as well represent single phase PSC of solid particles but with different shapes, hence producing various depolarizations.

Figure 8 reports a 2D-histogram of d_{TA} towards 1-1/BR from twelwe years of lidar observations from 1990 to 2002 in the antarctic station of McMurdo (Adriani et al., 2004). Despite the dispersion of the experimental points, a value close to 40%, as the highest vertex of the triangle, on the 1-1/BR=1 axis, for ∂^{asph}_{TA} can be assumed. The corresponding value for δ_A is close to 70% according to eqns. (3), (5) and (6).

If we assume that the difficulty of our model to reproduce the observed depolarization in some case is due to the incorrect assumption of a common AR for all solid particles, we are led to interpret Figure 8 admitting that the experimental points filling the triangle of vertices (1, 0), (1, $\partial^{\text{asph}}_{TA}$) and (0, δ_{mol}) represent both PSC in various degrees of mixed phase, and PSC in purely solid phase but composed of particles of various shapes. These various shapes give rise to different δ_{TA} between 0 and $\partial^{\text{asph}}_{TA}$ at the vertex of the triangle.

For completeness we report also new Figure 7.



Figure 7: Sequences of β (red dots) and δ (blue dots) measured on a balloon flight on December 9th 2001, from Kiruna, Sweden. Each data point represents an average over 60s. Black dots represents the ARs providing the best match between the δ and those computed from concomitant measurements of PSD.

3) Run a sensitivity analysis using simulated PSCs and measurements to quantify the observed difference.

In a sense, the sensitivity of the method could have been estimated from the results already presented. Specifically, the sensitivity of our method can be obtained from the range of variability of the RMSEs according to the variability of AR and R_{Th} , as shown in figures 2 and 3. We note that

those results are relative to the analysis of real particle size distributions (PSDs), from measurements. We don't see how a similar study, done on simulated PSDs, would provide additional information.

However, to pursue the reviewer's requests in a different sense, we have estimated the uncertainties to be attributed to both computed and measured β s and δ s (see new figures 4 and 5); We have added in the revised par. <u>3 Results</u>:

The uncertainties associated with the measured β_A and δ_A derive from the error analysis for the single lidar data, which can be found in Adriani et al. (2004) or from the standard deviation for the averaged data, depending on which is greater. The uncertainties on the calculated β_A and δ_A , are 40% as determined by Deshler et al. (2003a) for any moment of a PSD derived from the OPC measurements. Deshler et al. determined this through a Monte Carlo simulation which used the uncertainties of the OPC size and concentration measurements to quantify the uncertainties in the PSD parameters and their subsequent moments.

4) Correct a large number of typographical and grammatical errors that made the current manuscript distractive to read.

We apologize for the poor quality of the written English, responsibility of the first author only. This has been corrected in the revised manuscript.

Other comments, questions and suggestions:

1) From the abstract onward the authors erroneously talk about comparing "microphysical and optical" measurements. This makes no sense since all of the measurements are microphysical and optical, i.e. the OPC uses an optical technique to derive size distributions that help describe the microphysical properties of the PSCs. Likewise, the remote sensing techniques are optical and are also used to derive microphysical properties of PSCs.

This seems to be splitting hairs. If we are not mistaken, almost every cloud probe available today uses an optical technique to measure cloud and aerosol size distributions. The days of impactors has long since faded into history. Yet the results of the aircraft optical probes are used to discuss cloud and aerosol microphysics. It is not clear to us that we are doing anything different, as long as the uncertainties inherent in the instrument, due to its use of optics to make the measurement, is clearly described, as it is here and in the referencing literature. Generally, when an instrument provides size distributions it is discussed as a microphysical measurement, not an optical measurement, even though the fundamental principle on which the measurements is made is optical. The lidar and backscattersonde provide less detailed microphysical measurements, but there is still microphysical information contained therein, such as the extent of aspherical particles which is a microphysical property measured optically. Airborne lidars are routinely used to measure cloud base, cloud top, and the extent of ice in the cloud, all microphysical as well as optical properties.

2) The authors never explain the relative importance of mixture of particle types in mixed phase PSCs. Had the modeling exercise been successful, who would benefit?

There are mainly three goals. To test the ability of the T-matrix code to reproduce the observations from lidar/backscattersonde, once the PSDs are known. To provide an estimate of the AR parameters and of the smallest dimensions of the solid part of the PSC particulate mixture. These goals are now reported more clearly in the Introduction.

3) Nothing is discussed about the contribution to the backscattering of other types of stratospheric particles, e.g., meteoritic dust, sulfate particles, etc. How does that impact the measurements and modeling?

The study exploits measurements taken within PSCs. The contribution to backscattering and depolarization of the background atmospheric particulate matter (SSA, meteoric dust, etc.), possibly observable in isolation outside the cloud, is negligible within a cloud in the majority of cases under exam. However, it is possible that unaccounted background aerosol led to unaccurate lidar calibration. We acknowledge this fact in the 4 <u>Result</u> paragraph.

Despite the dispersion in Figure 4 the points cluster around the straight line $\beta_{calc} = \beta_{meas}$, indicating the agreement between computation and measurements can be considered fine for β with the exception of β values below 4 10^{-5} km⁻¹sr⁻¹ where β_{calc} underestimate the measurements. Such underestimation seems to be of the order of 10^{-5} km⁻¹sr⁻¹, of the same order of the backscattering from the background atmospheric particulate matter in volcanic quiescent conditions, a magnitude compatible with possible inaccuracies in the calibration of the lidar data.

4) The backscatter instrument described by Adriani (1999) had multiple wavelengths. Why is only the 532 being used? Wouldn't modeling multiple wavelengths have improved the retrievals?

We compare PSD mainly with measurements from a lidar which does not have the second wavelength. The backscattersonde has also been used on a part of the dataset and it provided measurements at a second wavelength, but they have not been judged accurate enough to be published.

5) If this was a true modeling study, an iterative methodology should have been used to vary the mixtures of shapes and sizes until most closely matched by the measurements.

In our study both the AR and the R_{Th} were independently varied in order to simulate different mixtures of shapes and sizes, thus the parameter ranges that best matches the measures were identified. Figures 2 and 3 in our manuscript provide what the reviewer is asking here. This analysis was effective in delimiting the variability of R_{th} , but not of AR, therefore in the revision of the manuscript we proceeded to look for the best match on a case-by-case basis, identifying for each PSD the best AR within the wide ranges of variability previously identified. The result is, for R_{th} within the limits 0.5-0.8 μ m, a distribution of AR within these intervals, which clusters around the values 0.5 and 2.5, approximately, as reported in the new figure 6 where the AR distribution is reported in terms of a parameter characterizing the shape of the PSD.



Figure 6. 2D-histogram of occurrence of ARs and of $N(r < 0.7 \mu m)=N_{tot}$, the ratio between particles with radius greater than 0.7 μm and total particles. Only ARs in the intervals between 0.3 and 0.55, and between 1.5 and 3, have been considered.

6) How homogeneous are these clouds and what do the PSDs look like derived from the OPC? The reader never sees the actual shapes of PSD or what the number concentrations are. This is important because it will impact the backscattering and depolarization. It is stated in the results section that apparently the larger particles are biasing the depolarization but this depends on the total concentration of particles and how homogeneous the mixture is. I could not find in the Adriani (1999) paper what beam volume is at each measurement gate.

Answering the last question first. The backscattersonde laser beam cross section is approximately 20 mm², and 90% of the backscattered signal comes for 2 to 6 meters from the backscattersonde, so the sampled volume is of the order of 50-100 cm³.

The question of homogeneity of PSCs is relevant. Our database is composed of two groups of measurements. In the first, the OPC on board a balloon is compared with Antarctic ground-based lidar measurements. In the second, the OPC is compared with Arctic measurements from a backscattersonde on board the same balloon. It is clear that the second class of measurements is less affected by cloud inhomogeneity problems, given that both instruments measure in-situ the same cloud.

Opposed to this is the first group where measurements taken in cloud regions are by their nature separated in space and time as the balloon drifts down range from the lidar, so cloud inhomogeneity must be considered. This issue was addressed in Snels et al., (2021), a work based on the same Antarctic measurements. Snels et al. compared the lidar profiles with backscattering computations from the OPC data. Of the 18 coincident lidar-balloon flights, only 15 profiles were

used for the analysis. The choice was based on a visual inspection of the coincidence of the main cloud features in the lidar and balloon flight altitude profiles.

As outlined above, the present work adds to the Snels et al. (2021) Antarctic dataset, measurements from Arctic balloonborne OPC and backscattersonde flights. This addition does not alter significantly the goodness of the computed vs measured backscattering regression line. This gives us confidence that cloud inhomogeneity did not play a significant role in causing the dispersion of the points in the regression line.

However, for some data points for which the computed vs measured do not match at all, we have invoked a possible inhomogeneity of the cloud as a possible explaination. This has been reported in the last lines of the 4. Discussion paragraph (see answer to point 2) above).

Concerning the shape of the PSDs which is suggested to display, our study is based on 473 data points (i.e. 473 triplets of PSD, backscattering coefficient, and depolarization). Given the range of the observations, it is difficult to provide a representation of the actual shapes of PSD or what the number concentrations are. In any case we proceeded to use the ratio $N(r>6\mu m)/N_{tot}$ in the new figure 6 as a parameter to characterize the shape of the PSD. We have also checked a lack of clear correlation of N_{tot} with the result of the model-measurement comparison.

7) In Figs. 4 and 5, there is no noticeable difference between AR-0.5 and AR=1.5. This does not surprise me because if you have an ensemble of randomly oriented spheroid, an oblate spheroid will look like a prolate spheroid, depending on their relative orientations; hence why even use ARs < 1?</p>

It is certainly true that there are particular orientations for which prolate spheroids can appear like oblate spheroids, and vice versa. However, two PSDs with identical distribution parameters, one composed of oblate and the other of prolate spheroids, with reverse ARs, randomly oriented, need not necessarily have the same backscattering properties. That this is the case can be deduced, for instance, from Figure 1 in the work of Liu and Mishchenko (2001). However, old Figures 4 and 5 have been discarded.

In the new approach pursued following the reviewer' remarks, the difference in behavior between ARs less than or greater than 1 is more apparent.

8) There is no quantification of the comparisons, i.e. no correlation coefficients, curve fits or other statistical tests applied to justify comments like "fine" or "reasonable. In fact, the authors' conclusions that the backscattering comparison is "fine", does not agree with what we see in the figures where the dispersion is hidden by the logarithmic scales on the figures.

Good point. We have provided quantitative data on the goodness of the fit for the β comparison (the Pearson correlation coefficient) and added 1:1 lines to the data in Figs. 4 and 5. We did not perform goodness-of-fit tests for comparison of δ s. In this case it is clear that there is a set of well-aligned points along the 1:1 line, and sets of points that significantly deviate from it in a non-random way. We have discussed the different characteristics of these sets in the <u>3 Result</u> paragraph, totally rewritten, which we report here in its entirety:

3 Results

Figure 4 reports the scatterplot of measured vs computed β_{A} , colour coded in terms of AR. The figure represents the analogue of figure 4 in Snels et al. (2021), where in the present case we have used a larger dataset, including now four Arctic balloon flights, and used T-Matrix instead of a factor 0.5 reduction in the Mie backscattering. Figure 5 reports the scatterplot of measured vs computed δ_A similarly color coded in terms of AR. The uncertainties associated with the measured β_A and δ_A derive from the error analysis for the single lidar data, which can be found in Adriani et al. (2004) or from the standard deviation for the averaged data, depending on which is greater. The uncertainties associated with the measured β_A and δ_A derive from the error analysis for the single lidar data, which can be found in Adriani et al. (2004) or from the standard deviation for the averaged data, depending on which is greater. The uncertainties on the calculated β_A and δ_A , are 40% as determined by Deshler et al. (2003a) for any moment of a PSD derived from the OPC measurements. Deshler et al. determined this through a Monte Carlo simulation which used the uncertainties of the OPC size and concentration measurements to quantify the uncertainties in the PSD parameters and their subsequent moments.

Despite the dispersion in Figure 4 the points cluster around the straight line $\beta_{calc}=\beta_{meas}$, indicating the agreement between computation and measurements can be considered fine for β_A with the exception of β values below 4 10^{-5} km⁻¹sr⁻¹ where the β_{calc} underestimate the measurements. Such underestimation seems to be of the order of 10^{-5} km⁻¹sr⁻¹, a magnitude compatible with possible inaccuracies in the calibration of the lidar data. The Pearson correlation coefficient for the entire dataset is 0.56, and increases if the lower values of β are neglected.

The δ_A scatterplot shows the presence of a good number of points that align along the $\delta_{calc} = \delta_{meas}$ correlation line, with AR selected mainly around the value 0.5. However, for depolarization values greater than 30% there is no AR that will reproduce the measurements. These points correspond to those presented in Figure 1, with low values of BR and high values of the concentration ratio of large to total particles. They mainly come from three single observational periods of about one minute each, characterized by air temperatures between 184-188 K. Given the magnitude of the depolarization, it is possible that those observations are not referable to clouds in mixed phase, but rather to clouds of predominantly solid particles. For that particular set of points, we also explored the possibility that all particles were solid, but even under this assumption the comparison with the experimental data did not improve appreciably.

In Figure 5 for depolarizations lower than 15%, the points which deviate, by excess or defect, from the 1:1 straight line have predominantly AR greater than 1.5. So it seems that selected ARs greater than 1.5 generally produce a worse correlation. From Figure 4 we observe that AR values in the range (0.3-0.55) tend to be associated with medium-low β values, while AR values in the range (1.5-3) are mainly associated with medium-high β .

To conclude, the choice of R_{th} in a range between 0.5 and 0.8 μ m leads to a reasonably good agreement between the β 's, but there seems to be a discrepancy between the calculated value and the measurements in their lower range of variability.

From Figure 4 such mismatch, which makes the measurements larger than the calculations, seems to be of the order of 10^{-5} km⁻¹sr⁻¹. The selection of the AR that produces the best agreement with the observed δ 's leads to three results: i. The ARs in the range 0.3-0.55 tend to be selected in correspondence with medium-low β 's, the ARs in the range 1.5-3 in correspondence with medium-high β 's. ii. ARs in the 0.3-0-5 range reproduce the measurements well, except for some observations where the depolarizations are greater than 30%; iii. the ARs in the 1.5-3 range reproduce the measurements less well; iv. There is no AR that will allow the calculations to reproduce the measurements for depolarizations greater than 30%.

9) I recommend that the analysis of the OPC data to derive backscattering should use the actual scattering measured by the OPC, rather than converting scattering to equivalent optical diameters and then computing scattering. This adds additional uncertainty because there are large errors in size derivation because of Mie oscillations and unknown shape. If the authors derived backscatter from the measured forward scattering, as was done by Baumgardner and Clark (1998), this removes much of the inherent error.

In Baumgardner and Clarke (1998) the authors infer the total single particle scattering coefficient from the forward scattering coefficient measured between 4 and 12 degrees. This inference is made by calculating, with the aid of Mie's theory, the relationship between the scattering, calculated in the above angle interval, and the total scattering. The inference is then that the total particle scattering can be inferred from the FSSP measured scattering, seemingly on a particle by particle basis. The OPC we employ is not a single particle scatterer, but rather discriminator levels are used to collect all photo multiplier pulses larger than a preset level. Thus the number concentration in any discriminator bracket is the result of all particles which provide a light signal above the lower level and less than the next discriminator level. These OPCs measure a maximum of 12 sizes between 0.19 and 10.0 μ m, much less than the FSSP, and thus require fitting of size distributions to obtain estimates of backscatter. Estimating backscatter from 12 sizes would be insufficient to compare with the lidar measurements which are inherently ensemble measurements. In addition, the OPC uses white light to measure scattering at 40 degrees, an optical signal not directly comparable to the lidar. Furthermore, instead of using Mie theory, we should use T-matrix calculations and replicate them for many different AR and R_{Th}.

10) I was disappointed by the excessive typographical and grammatical errors since the second author is a native English speaker. "Author contributions. FC was responsible for most of the writing, review and editing process, supported by all co-authors." This appears to be inaccurate.

The first author assumes responsibility for typographical and grammatical errors and we are now taking greater care in revising the English used.

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