Answers to Manuscript Review of Referee 1

In the following, we give the answers to the comments of Referee 1. Our answers are marked in blue font whereas the original comments are written in normal font.

The study that is presented in this manuscript describes an application of polarized light measurements for discriminating water droplets from ice crystals for the purpose of ice nucleation experiments. The abstract was intriguing when it suggests a new approach for interpreting the measured, polarized component of scattered light, i.e. the pulse width. Unfortunately, this is not presented with a convincing argument and, in general, the overall paper is a disappointment, in my opinion. Had I been given the option of reviewing this manuscript prior to its publication as a discussion paper, I would have recommended that it be returned to the authors for further work. In my opinion it is much too premature to be published in its present form and I cannot recommend its publication in AMT in anything close to its present form.

There are multiple issues, that I will address in detail below, that prevent this study from making a useful contribution to measurement science. Broad statements are made with no concrete evidence or references about various aspects of the measurements system that are critical to the reader's understanding of its underlying performance. The numerical simulations are performed, and conclusions drawn assuming an ensemble of randomly oriented spheroids yet the instrument described does not measure ensembles of particles but measures particle by particle. Hence, the whole theoretical basis is suspect from the very beginning. The system is designed to differentiate water droplets from ice crystals on a particle by particle basis, not from an ensemble like lidar systems do. This means that the authors are not taking into account the fraction of ice crystals that will have an orientation such that they appear as spheroids, i.e. they never discuss the uncertainty in their derived ice fraction due to orientation.

The technique that they propose to differentiate water from ice based upon the pulse width was, to this reviewer, incomprehensible with respect to why ice crystals would have a distinctively different height to width relationship than water droplets. They show one set of frequency histograms that are supposed to convince the reader that ice crystals have a pulse width distribution different than water droplets with no theoretical basis. They show a highly simplified Gaussian curve that is supposed to represent how the pulse shape looks as a particle passes through the beam; however, even though they are digitizing at a high rate, and could show actual pulse shapes for water droplets and ice crystals, they never do. In addition, their theoretical curve of pulse height versus pulse width is far from what is actually measured, yet they use this relationship as a definitive way to separate water from ice.

In my opinion, the geometry of the optics offers a really substantive way to separate water droplets from ice crystals given the two planes of detection, yet this is not even considered. Other studies have clearly shown that a comparison of two components of the polarized, scattered light can differentiate spherical from aspherical particles, e.g. Nicolet et al., 2012 and Glen and Brooks, 2012; however, not only do the authors not look at these relationships, they don't even reference the studies that have shown that it works.

It is quite possible that the technique has something unique to offer the measurement community but the results that have been presented here are unconvincing and I would ver wary of any ice fractions that are reported from this instrument using the proposed technique.

As a preamble to our answer to Dr. Baumgardner's general remarks, we would like to express our regret that the manuscript produced such a strong negative impression on Dr. Baumgardner. To our opinion, however, Dr. Baumgardner misunderstood the working principle of the instrument described in the presented manuscript and was thus misled in his judging. Specifically, Dr. Baumgardner is overseeing the fact that the presented instrument does not measure the *ratio* of the cross-polarized components of the scattered light, how it is done in the instrument developed by DMT Company and presented in Glen and Brooks, 2012. What the TOPS-Ice instrument does measure, is the *amplitude* of the scattered component of light polarized perpendicular to the plane of polarization of the incident light. The differentiation between spherical droplets and ice particles is done exploiting the fact that the intensity of cross polarized light is, on-average, higher if scattered by "distorted" ice particles than by spherical droplets of the same volume equivalent diameter (we deliberately avoid the notion of non-sphericity here because it is also the internal structure of ice crystals and their surface properties that causes the change in the polarization state). We also make use of the specific experimental conditions in LACIS, where due to the monodispersity of the seed particles and homogeneity of thermodynamic conditions the droplets (and the ice particles, to this effect) have a very narrow (geometrical) size distribution. These conditions also stay constant with time, so that the measurements averaged over time are identical with the measurements averaged over particle ensembles. The TOPS-Ice instrument was designed to match these laminar diffusion channel conditions and is not intended to operate, for example, on an aircraft.

Further on, we make use of the difference in the amplitude by measuring the pulse width instead of pulse amplitude. Because the amplitude of light scattered by non-spherical particle is higher, the pulse width is larger, and the non-linear relationship between the pulse width and pulse height allows us to improve the differentiation algorithm. There is, of course, NO reason why the pulse produced by an ice particle should be longer compared to the pulse of the same amplitude originated from spherical particles. However, due to the variability of the scattering cross section depending on the orientation of individual ice particles, the *pulse width distribution* for the ice particle ensemble is broader for the non-spherical particles.

We also think that Dr. Baumgardner overestimates the importance of the numerical analysis done for the rotationally symmetrical particles for the interpretation of real ice optical measurements. One does not have to perform highly complex and time consuming T-Matrix calculations for all possible orientations and sizes to arrive at conclusion that the scattered light does not always possess a different polarization. To realize that, it is enough to consider the physical reason of depolarization of the scattered light by non-spherical rotationally symmetrical particles, the way it is done by Chylek (1977). As Dr. Baumgardner is certainly aware, the zero change in polarization (within the scattering plane) is related to the diagonal form of the amplitude scattering (Jones formalism) matrix. The diagonality of the scattering matrix is clearly related to the reflection symmetry of the particle geometry with respect to the scattering plane. Therefore, any rotational symmetrical particle in certain orientation would have a diagonal scattering matrix and therefore produce no change in the polarization of the scattered light. The same is valid for backward direction too.

On the other hand, even the perfect spherical particles will cause the light scattered in a finite solid angle in any scattering direction to be depolarized, simply because the scattering plane is inclined with respect to the polarization plane of the incident light if the azimuthal angle is not zero. Combining this argument with the orientation issue it becomes clear that for the realistic instrument with the finite numerical aperture of a scattering detector no particle would produce zero change in the polarization state. However, the intensity of the cross-polarized component can be used for discrimination of spherical and non-spherical particles. This is the essence of the function principle of TOPS-Ice.

Furthermore, Dr. Baumgardner suggests that we estimate the fraction of non-spherical particles wrongly classified as spherical ones on the basis of theoretical calculations carried out for ellipsoid particles (as it is done in Nicolet, 2010) or for cylinders (Nicolet, 2012). However, we have little information concerning the preferential shape of the frozen droplets in LACIS. We provide the modeling results to show that the scattering geometry of TOPS-Ice enables the differentiation of scattering amplitudes from spherical and non-spherical particles (ellipsoids) of the same volume equivalent diameter *in statistical sense*. Unless no information about the shape and surface properties of the frozen droplets is available, the attempt to characterize the fraction of non-depolarizing orientations from the calculations carried out for rotationally symmetrical particles is meaningless.

Concerning Dr. Baumgardner's mistrust in the TOPS-Ice measurements, it should be mentioned, that the manuscript contains a comparison of results gained with the TOPS-Ice instrument with results from two other instruments which feature completely different measurement principles. The agreement between the different results is very good indicating the applicability of the TOPS-Ice instrument for determining ice fractions at LACIS.

Specifics

Abstract: Line 4. "…measurements of the depolarized component of light…". No, single particles do not depolarize incident light. They change the state of depolarization. The scattered light remains polarized but instead of a single plane of polarization there are two at right angles so that the light is now elliptically polarized. See Harris-Hobbs and Cooper, 1987, in their appendix they explain the inappropriate use of the term "depolarization". Only when light is scattered by a large ensemble of particles will the resulting scattered light have random polarizations, i.e. depolarized.

At this point, Dr. Baumgardner is contradicting himself. We don't see why "...change the state of DEpolarization" is better than the term "depolarized light". As Dr. Baumgardner correctly states, the scattered light in general is elliptically polarized, so that the relationship between the elements of Stokes vector ($Q^2+U^2+V^2=I^2$) still holds. In the literature, however, two approaches exist of interpreting the term "depolarization". One of them, originally used by Kerker and Van de Hulst, defines depolarization as the ratio of the randomly polarized (that is, depolarized) light to the total intensity, whereas multiple works including those of Mishchenko and Hovenier define "linear depolarization ratio" as the ratio of cross polarized components or the"circular depolarization ratio" as the ratio of the components of the opposite helicity. Thus defined, "depolarization" describes how the state of the polarization of the scattered light is changed relative to the original state of polarization of the incident light. This definition is mostly used for the description of the single-particle scattering phenomena, since a single scattering event of the coherent EM wave cannot reduce the degree of polarization (in the van de Hulst sense). We think that both ways are possible as long as the definition is provided. Realizing that in fact we have not defined the term "depolarization" we thank Dr. Baumgardner for his remark and correct this mistake.
We define depolarization within the manuscript now by: "In this manuscript, depolarization means the change of the polarization state of the scattered light compared to the polarization state of the incident light."

Page 5747: Line 5. Some of the most important references on measurements of polarized light are missing here, i.e. Fukutaamd Kramer (1968) who were the first to use polarized light to detect ice crystals in an IN chamber, Nicolet et al. l 2012 who did extensive simulations of light scattering from spheroids in different orientations (more on that below) and Glen and Brooks, 2012 who show that not only can aspherical particles be clearly differentiated from spherical, but that the backscattering signal is quite strong enough to get useful information, contrary to what the authors in the current paper state several times with no justification whatsoever.

We respect the Dr. Baumgardner's opinion to what the "most important references on measurements of polarized light" are, but prefer to keep the references we cited already in the manuscript. We can't cite the paper of Fukuta and Kramer (1968) because it is not available through the online databases. Glen and Brooks (2012) submitted their paper *after* we have submit our paper, but we include the reference in the revised manuscript. The reference to the theoretical study of Nicolet et al. (2012) will is also added to the introduction part.

Page 5747: Line 11. "….to avoid complication…". This is the first of several occasion swhere it is stated that backscattering measurements are complicated. Baumgardner et al, 2001 first showed that backscatter as one component of the CAS measurement system works quite well and the many CAS units that are currently in operation belie the assertion in this paper that backscattering is difficult and that such signals are too weak to be useful. Glen and Brooks, 2012 also show the error in statements of this type.

With all due respect to Dr. Baumgardner, we do not understand why we should build an optical instrument copying the CAS design (which actually uses the total backscattered light), design of CASPOL instrument described by Glen and Brooks, 2012, or design of CAS-DPOL (DMT) instrument. We have been through different designs in the stage of prototyping. The measurements in backward direction are not only complicated because of the low scattering intensity. Actually, during the prototype stage we tested the backscattering geometry and were able to record the signal, even by adding a polarizer in front of the backscatter signal detector. In our prototype instrument the complication with the backscattered signal was the difference in the fields of view of the detectors in forward and backward directions, therefore we redesigned the geometry of the scattering channels. A statement to this effect is added to the introduction part of the manuscript: "From different prototype stages of the TOPS-Ice development, we decided not to use the scattering in backward direction, first, to avoid the different fields of view of the three detectors and, second, to increase the scattering intensity."

Page 5748: Line 14. "…operated in the immersion freezing mode..". Although it is not the intent of the manuscript to discuss the LACIS, per se, I am curious how the immersion mode of nucleation can be differentiated from contact mode nucleation in this flow tube configuration with its miniature cross section.

For the details, we can only readdress Dr. Baumgardner to the references in the manuscript. In the described LACIS experiments, every single aerosol particle is activated to a droplet. These droplets may freeze by further cooling, namely immersion freezing. In other words, there are no particles available for contact freezing. No changes made.

Page 5759: Line 25. The actual sample volume is never shown, i.e. it is supposedly defined by the intersecting fields of view, but this means that particles will be passing through regions of varying intensity, given the Gaussian distribution of the laser beam. This region can be defined precisely using ray tracing and validated with a droplet stream, such as shown by Lance et al. (2010) who validated the theoretical sample area of an optical particle spectrometer by mapping with a droplet stream. The pin hole does not replicate scattering by single particles as it diffracts and does not refract light. In addition, even though it is an approximation, the results are never shown of this mapping so the reader is never aware of just how serious the edge effect issue is.

We agree that the phase function of light diffracted by a pinhole is different from the scattering from a spherical droplet. However, it has been shown by Schmidt (2004) that the sample volume of FSSP can be reliably characterized with the set of pinholes of different diameter. The sample volume characterization with a stream of droplets is only possible with the droplets larger than approx. 10 um (although the size of the droplets is not known a priori and must be measured independently), and Lance et al. (2010) reports positioning accuracy of the droplet stream below 20 um (!). Sampling volume of TOPS-Ice is 25 um (see Section 2.2). We had to do the characterization of the sample volume for the droplets and ice particles in the range from 1 to 10 um.

Furthermore, the pinhole scan through the sample volume was primarily used to align the smaller sample volume of the channel B within the larger sample volume of the channel A (a necessary condition to avoid the edge effect). We do not show the exact characterization of the sample volume because we do not address the issue of measuring the particles number concentration, given that the counting efficiency of the droplets and the ice particles is the same. No changes in the manuscript.

Page 5760: Line 9: To what sizes was the TOPS calibrated? Over the entire size range? And what is the size range of the TOPS?

The calibration was done with PSL particles within a size range between 250 nm and 1600 nm. The differentiation of water droplets and ice particles was tested for sizes larger than 1000 nm. These numbers will be added to Section 2.2 in the manuscript.

Page 5760: Figure 4 and all other figure showing Size Parameter. The actual optical diameter should be used, not the size parameter so that the reader can actually see the regions of uncertainty in terms of actual size. The authors state maximum uncertainty of 0.5 μm but I think that in some regions that uncertainty is much larger.

There are actually two different x-axes, one on top and one on bottom. The one on top shows the actual volume equivalent diameter. Unfortunately, the volume equivalent diameter axis was missing in Fig. 3 and was added in the revised version.

Page 5760: Theory Section. In my opinion, this section is much too large. Most of the theoretical derivations and equations are straight out of light scattering text books or have already been recently described in detail by Nicolet et al., (2012) or Schnaiter et al., (2012). Not only is this amount of detail not needed, in the end isn't actually relevant to the way that the TOPS is eventually used try and differentiate liquid from ice. In addition, as discussed in my opening comments, the T-matrix code is used incorrectly to calculate average values of randomly oriented spheroid, unlike the calculations by Nicolet et al (2012) who show quite clearly why you have to do the calculations for many different orientations to underscore how important orientation is when measuring particle by particle and estimating uncertainties in estimated ice fractions.

The purpose of the theoretical section is to show the calculation of the scattering intensity for the exact detector geometry of TOPS-Ice, therefore, we would like to keep it as it is. As we have admitted in the manuscript, the discrimination of water from ice for a single particle is, of course, possible only with a certain probability, **not only because of particle orientation but also because of the finite size of the acceptance angle in the azimuthal direction**. This is true for all single particle instruments developed and tested so far (CAS-DPOL, CASPOL, FINCH). TOPS-Ice is designed to work with the particle ensembles; therefore we interpret the measurements on the statistical basis, operating with signal distributions instead of particle for particles analysis. In LACIS, properties of the ice nuclei and thermodynamic conditions do not change over time, so that ergodic principle can be applied. From this point of view, the average amplitude of the cross polarized scattered component can be calculated for the ensemble of orientation averaged non-spherical particles. Obviously, such approach does not increase the inaccuracy coming from the unknown particle shape. Therefore we suggest interpreting the presented calculations as a demonstration of TOPS-Ice measuring principle for the specific LACIS conditions. To clarify the purpose of the theory section, we included into Section 3: "The purpose of the following calculations is to show the scattered intensity for spheres and for ellipsoids, as an example of non-spherical particles, for the exact detector geometry of TOPS-Ice. For the calculations, we used orientation averaged ellipsoids. This is possible as the conditions in LACIS do not change over time and allow therefore an interpretation of many single particles with different orientation measured one by one as a statistical ensemble of particles in random orientation. Therefore, the average amplitude of the cross polarized scattering component can be calculated for an ensemble of orientation averaged non-spherical particles and can be considered as the expected value."

Page 5763: Line 27. "Calculations of the response functions of spheroid particles in all fixed orientations are too time consuming…" This is a very weak and unconvincing argument given that Nicolet et al. have done this, as have others. It is exactly this type of simulation that is needed to understand the system performance.

We agree with Dr. Baumgardner that in the modern times of supercomputers and motivated PhD students the argument of anything being "time consuming" is not really appropriate and therefore we remove it from the manuscript. The rest of the argumentation preserves its validity. Nicolet et al. (2012) has done simulations for 5 different types of oriented cylinders with the size parameter not exceeding 8.We will cite this work appropriately.

Page 5765: Line 1. Why are pulse shapes never actually shown from the three detectors under conditions of droplets or ice crystals? With no comparisons with the idealized wave forms that are shown, the reader cannot be convinced that this is anything other than an intellectual exercise, especially since the following derived equation does not fit the actual measurements very well at all, given that the Y-axis is logarithmic.

We do not say that the shape of the pulses look different for the droplets and the ice crystals. The imaging of real measured pulses can be made, but will not give more information to the reader. For more precise answer on this issue, see below.

Page 5765: Line 17. As previously mentioned, the pin hole does not simulate particle scattering. A spherical droplet would have isotropic scattering so that all three detectors should be viewing the same solid angle and intensity of scattered light. The scattered light from the pin hole does not scatter isotropically. Furthermore, the results from this mapping are not shown. Why not?

We don't understand the argument. Neither a pinhole nor a droplet scatters light "isotropically", that is, independently of the scattering direction. Indeed, the angular dependence of the scattering intensity is different. If it is the orientation of the pinhole with respect to the laser beam that Dr. Baumgardner is relating to, we assure that the pinhole was always oriented perpendicular to the beam axis; the rest was already discussed above.

Page 5766: Line 9. The problem with this derivation is that it does not account for the fact that a larger particle exceeds the threshold sooner than the smaller because when it is only partially into the beam it scatters enough light to exceed the threshold whereas the smaller particle must penetrate farther. In addition, this derivation assumes that the particles are passing directly through the center of the Gaussian beam; however, there is a distribution of shapes as particles with the same size pass off center and hence have a difference height to width relationship. This is clearly shown in Fig. 7 where the pulse heights of the smaller particles with shorter pulse widths are overestimated by the theory while larger particles are underestimated.

This is an interesting issue and we thank Dr. Baumgardner for touching on it.

First of all, the measurement shown in Figure 7 was obtained with the water droplets continuously growing with time as LACIS was cooled from ambient down to the working temperature of -40°C. This actually implies that the thermodynamic conditions in LACIS were slowly (within 30 min) changing with time during the measurement, causing, for example, slow perturbations of the flow profile inside the channel. What looks like apparent size dependent deviation of the measured values from the theory might be a simple time dependent variation of particle beam position with respect to the sampling volume.

However, the theoretical curve was fitted to the measured values with a single free parameter - sigma_p, which was found to be equal to 28 us. Almost the same value can be also obtained if one takes the vertical cross section of the laser beam intensity (which has a natural Gaussian shape), and divides the standard deviation of this shape curve by the average linear velocity of the gas flow long the central axis of the tube. The good quality of this agreement (within few percent) indicates that on average all droplets were passing through the center of the laser beam and that observed deviations were caused by the temporal variation of the flow stability as discussed above.

We will add this contribution to the discussion of Fig. 7.

Page 5767: Line 1. "The measured curve shows a general agreement…". This is overstating the fidelity of the agreement. A error calculation would show that the agreement is actually not that good.

We are not actually interested in the perfect agreement between the calculated curve and the measured values. The presented analysis was intended to show where this behavior is coming from and how it can be used. The deviations are due to the point discussed above. We will add to the discussion of Fig. 7: "This figure is showing the principle relation between the pulse width and pulse height which can be found in theory and experiment."

Page 5767: Line 10. "…broadening of the droplet mode and a more compact ice mode". This is stated with no obvious explanation or evidence. I could not understand what this meant. Furthermore, nowhere in this section is an explanation give for why an ice crystal, i.e.aspherical particle, should have a pulse width different than a droplet. I had fully expected that this section would give a detailed pulse shape analysis since I certainly could make an argument why the pulse SHAPE would look different for an aspherical particle, but I can't find any physical reason why the width should differ.

We never say that the ice crystals produce broader pulse due to the non-sphericity. This can not even be expected considering that the ice crystals in LACIS are still much smaller than the width of the sample volume. The higher amplitude of the pulse, and therefore a *broader* pulse, arises from the larger optical size which is *induced* by using the polarization component which is parallel to the scattering plane. What we state, however, is that the variability of the pulse amplitude, and hence variability of the pulse width is due to the non sphericity, and this variability shows as the enhanced width of the pulse width distribution for ice particles. To avoid confusion, we change sentence in the following way: "… broadening of the droplet mode and a more compact ice mode in PWD_C."

Page 5767: Line 18. "…the PHDC in the panel (a) is poorly…". There is no justification for saying that the PHD is poorly resolved and in fact, given that these are more or less mono-dispersed droplets, of 2 µm diameter, I would call this a remarkably well-defined PHD given the nature of the Gaussian beam. The main difference between the water droplet PHD and PWD and the ice crystal PHD and PWD is that the ice crystals are larger, having grown in ice saturated conditions. I can see nothing different between the two distributions that distinguish the ice from the water other than their size. This is exactly how most IN counters already differentiate the water from ice, so it seems that this proposed technique is not really unique.
How is the polarization feature being used? The title advertises polarization yet this is all about pulse width. Something got lost between the start of the paper and the end.
Page 5768: Line 9. "This mode of separation suggests an advantage of using the PWDC for the retrieval of ice fraction in the mixed-phase experiments." This was not convincing to this reviewer. It was neither shown theoretically why there should be a separation other than size, nor experimentally.

Depending on the moment of freezing within the last section of the LACIS channel and the humidity profile along the channel axis the ice particles may slightly grow. The longest growth time possible is 1.6 second (absolute maximum, most ice has much less time to grow). Since the relative humidity the last stage is always larger than 100% with respect to ice and always lower than 100% relative to water, the ice particles are never smaller than the droplets. Therefore there is no contradiction to our measurement technique. At the outlet of the laminar diffusion tube where the measurements are taken both populations, the droplets and the ice crystals, have similar optical diameters. If measured without analyzer in front of the detector, the signal distributions would overlap. Placing a linear analyzer in the optical path of PMT C we reduce the amplitude of the light scattered by droplets stronger than the amplitude of the light scattered by frozen droplets. The rest is pulse amplitude analysis.
We add this discussion to the manuscript.

Page 5768: Section 4.2 The evaluation presented in this section is suspect due to the unconvincing nature of the pulse width approach. Figure 10 just illustrates that you can separate water from ice with the Welas just as easily as the PWD technique that is actually just separating by size (growth rate), as well.

No, this is not true. As it was clearly stated in the manuscript, LACIS was run in two different regimes when using WELAS or TOPS, respectively. As for WELAS, the droplets were evaporated and a differentiation by size between the small dry particles and the ice particles could be made (in fact, the detection limit of WELAS is in the order of the optical sizes of the dry aerosol particles) (Niedermeier, 2010).With TOPS, a differentiation between the liquid water droplets and the ice particles of similar size is possible. Therefore, LACIS was run in a mode where the freshly frozen droplets and the liquid droplets were exiting the flow tube. The differentiation between frozen droplets and liquid droplets uses the difference in optical size measured by PMT C (see above).

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