Reply to referee #2: Interactive comment on "A High Speed Particle Phase Discriminator (PPD-HS) for the classification of airborne particles, as tested in a continuous flow diffusion chamber"

By Fabian Mahrt et al.

Reviewer comments are reproduced in **bold** and our responses in normal typeface; extracts from the originally submitted manuscript are presented in *red italic*, and from the revised manuscript in *blue italic*.

We have numbered the reviewer's major comments for ease of cross-reference within the other reviews.

The authors present a new instrument, a High Speed Particle Phase Discriminator (PPD-HS), for phase discrimination of cloud particles. The instrument is an extension to the Small Ice Detector (SID) family but compared to the SID-3 and the Particle Phase Discriminator (PPD) it uses two CMOS arrays instead of a CCD camera. This modification allows recording of two 1D rows of scattering information, which reduces the amount of recorded data and allows higher detection rates of several hundred particle per second. The 1D scattering patterns are analyzed for their symmetry to discriminate between spherical and aspherical particles. The authors present characterization of the new instrument and have developed a supervise machine learning algorithm to automatically classify particles for their phase. The paper is well written and the used approach justified. However, few aspects should be addressed before publication.

We thank the reviewer for carefully reading the manuscript and the overall constructive comments on it. We hope that the responses below satisfactorily address the reviewer concerns.

Major comments:

1. **The random forest model was trained with a test particle dataset of droplets and NaCl particles. However, it is somewhat questionable if NaCl particles would be a good proxy for aspherical ice particles. Why the random forest model was not trained using real ice particles from the experiment where NH4NO3 aerosol was frozen at T = 223 K?**

   The reviewer raises a valid point here, that real ice particles could have been used to train the random forest model. However, there are multiple reasons, why we decided to use VOAG data sets only for calibration purposes, as outlined below.

   The particle classification by the random forest model is purely based on particle shape analysis or more precisely the symmetry of the scattering pattern (P6L25-28, initial manuscript).The NaCl particles produced by the VOAG are aspherical, mimicking the aspherical features similar to that of ice crystals that is needed to test the working principle of PPD-HS. The reason for not using the ice data to train the model is to have independent data sets that can be used to test the performance of the trained algorithm. In fact, the overall good classification of particles in Figs. 7 and 10 provide direct evidence that the trained the random forest using both PEG and NaCl particles produced by the VOAG can successfully be applied to distinguish cloud droplets from ice crystals produced in HINC.

   Next, it should be noted that formation of ice crystals in HINC is limited to crystals with diameters < 10 µm, as we note on P10L3 (initial manuscript, see also comment #3 by reviewer#2). In order to test the performance of PPD-HS over a larger particle size range, for potential future use of PPD-HS in field measurements, where larger crystals can form,

we decided to use the VOAG to produce larger aspherical particles. Production of larger particles within HINC is limited by the residence time within the chamber as well as the gravitational settling of particles owing to the horizontal alignment of the chamber, which is circumvented in the VOAG setup shown in Fig. 3. A unique feature of the VOAG is the production of almost monomodal particle populations, which cannot be achieved to that extent in a CFDC setup as shown by Garimella et al. (2017), so that the random forest model would be mis-trained by the presence of e.g. small spherical ice particles (see Fig. 7). Finally, using the HINC data for training the random forest model would further be complicated by ice crystals and cloud droplets forming at different temperatures and relative humidity conditions that could influence the particle shape and hence bias the random forest model. By using the VOAG to produce both sets of spherical and aspherical particles we minimize such instrumental bias from the particle production mechanism, which is essentially the same for both particle types.

2. **Alternatively, would it have been possible to use SID-3 and PPD 3D scattering patterns of ice particles as training sets?**
   The diffraction patterns captured e.g. by PPD-2K (see Fig. 1 in Vochezer et al. (2016)) or those shown in our Fig. 2a-c are post-processed to determine particle shape and are not *directly* comparable to the scattering intensities captures by our linear CMOS arrays. One would need to transform the PPD-2K diffraction patterns captured by the CCD camera to scattering data along the two linear array stripes, as schematically depicted by our Fig. 2a. Since the goal of the present work was to characterize particle sphericity based on the linear scattering output of the newly presented PPD-HS (linear arrays allow for the high sampling rate), and the optical geometry used within the new instrument, the usage of ice particle scattering patterns of PPD-2K or SID-3 would not contribute to the overall goal of the presented work and was therefore not done.

3. **The authors do not discuss what is the upper size limit of the PPD-HS.**
   Please note that the largest particle size we have experimentally tested is 32 µm. It should further be noted that with the current experimental setup, we can only experimentally verify and cross-check particle sizing by PPD-HS for particles < 20 µm, i.e. those that lie within the size range of the APS (see P12L32-P13L1, initial manuscript) and for larger particles can only compare the instrument response of PPD-HS to theoretical calculations (see Fig. 5).
   However, the limiting factor determining the upper size limit of PPD-HS is the focus size of the trigger laser beam and the response of the photo diode. We state that the trigger laser beam is focused to a depth of 100 µm at the plane, where particle and trigger laser beam intersect (see P4L29-21 in initial manuscript). Any particle needs to be contained within the beam in order to cause a trigger and become recorded later. Hence, the maximum particle size that can be detected is < 100 µm.
   We have added the following statement to P5L4 (revised manuscript):
   "*The focal depth of the trigger beam of 100 µm constrains the size of particles that can be detected by PPD-HS to this diameter, as each particle needs to be contained within the focus of the trigger laser beam in order to cause a trigger and become recorded later.*"

   Furthermore, the upper detection limit of PPD-HS is constrained by saturation of the detector for particles > 70 µm. Please see our answer to comment P6L11-12 of reviever#1.

4. **Although the method is calibrated up to 32 µm using the test particles, the HINC experiments do not produce particles >10 µm. It is unclear how well the PPD-HS discriminates phase in the size range of 10-30 µm.**
   It is true, that for the classification results depicted in Figs. 7, 10 and 11, we are limited to the particle sizes formed within HINC. However, we believe that classification in the range 10-30 µm can be achieved quantifiably and with sufficient accuracy. As shown in Fig. 3,

using the VOAG setup, we generated both spherical and aspherical particles up to 32 µm in size, as stated on P10L1-4 (initial manuscript). This data is also shown in Fig. 4 and a detailed overview of the total number of particles sampled for each type and size is given in our Tab. S1 in the SI. It is further stated in the initial manuscript (P12L21-25), that out of this entire pool of particle data and covering the entire size range, 400'000 particles were randomly chosen for training of the random forest model, with the remainder data being used for assessing classification model performance. Therefore, the confusion matrix presented in Fig. 6 can directly be used to assess the shape (and phase) discrimination capability of the random forest model for PPD-HS particle data covering the entire size range 3-32 µm. In order to make this more explicit, we have adapted the statement on P12L21-25 (initial manuscript) from:
"Using these four predictors, we trained a random forest model on 400,000 randomly selected (training part), constituting of equal fractions of spherical and aspherical particles and using a total number of 200 trees (see SI Sect. S7). Classification performance was then tested on the remaining particle data (test data set; 4,371,162 particles), and subsequently applied to simulated hydrometeors from our HINC experiments."
To read P13L16 (revised manuscript):
"Using these four predictors, we trained a random forest model on 400,000 randomly selected (training part), constituting of equal fractions of spherical and aspherical particles and using a total number of 200 trees (see SI Sect. S7). These particles were randomly selected from the entire VOAG data set (see Tab. S1) covering the entire particle size range of 3-32 µm. Classification performance was then tested on the remaining particle data (test data set; 4,371,162 particles; same size range), and subsequently applied to simulated hydrometeors from our HINC experiments, where particle size is usually constrained to diameters < 10 µm."

We have further changed the sentence P13L11 (initial manuscript) from:
"In Fig. 6 we provide the classification results, when the trained random forest model is applied to the test data."
To P14L12 (revised manuscript):
"In Fig. 6 we provide the classification results, when the trained random forest model is applied to the test data, encompassing both PEG and NaCl particles of the sizes between 3-32 µm in diameter (see Tab S1)."

A similar statement was added to the caption of Fig. 6 to read:
"Confusion matrix of the random forest model applied to test data, i.e. the fraction of particles not used for model training, encompassing both spherical and aspherical particles of diameters between approximately 3-32 µm."

5. **For example, how would 1D patterns of large droplets with multiple rings be classified? Also, larger complex ice particles have 3D scattering patterns showing frequent speckles (as seen in Fig. 2b). How would the symmetry of such particles look like in the PPD-HS?**
The 2D scattering pattern of a droplet with multiple diffraction rings, would result in a 1D scattering pattern captured by our linear CMOS arrays, similar to the one shown in e.g. Fig. S15 #3, hence still show symmetry in terms of TBC and/or AIC. Thus, as long as the diffraction pattern of a spherical particle yields a symmetric scattering pattern along the CMOS arrays, it would be correctly classified by our random forest model as spherical particle, since the random forest model just assesses the symmetry in terms of TBC and AIC, which are independent of the number of diffraction fringes. A similar reasoning applies to speckled ice particles and an example of such a particle when detected by PPD-HS is given in Fig. 2e.

6. **The dead time of the instrument (177 to 267 µs) is high and its consequences to sampling statistics are only discussed quantitatively in the supplementary materials. However, this discussion should be incorporated into the main text. In chapter**

5 the issue is mentioned but there is no discussion of the implications for future ice spectrometer and field measurements. If at a typical mixed-phase concentration of 100 cm^{-3} 100% or more particles are missed, how representative are the retrieved PPD-HS ice concentrations, especially if looking for the first ice?
We agree with the comment of the reviewer, that the dead time of the CMOS arrays used within PPD-HS, which is the detection rate limiting factor, is high (T > 177 µs) compared to other optical particle instruments, such as SID-2H or PPD-2K with electronic dead times of T = 50.0 µs and T = 8.25 µs, respectively, (see e.g. Vochezer et al., 2016).
In fact, incorporation of this discussion into the main text has been part of extensive discussions among the authors during manuscript preparation. However, we believe that such discussion in the main text removes the focus from our main point, which is that reduced light scattering data captured by the two linear CMOS arrays can successfully be used for robust particle shape analysis and phase discrimination. It is true, though; that an upgrade to faster CMOS arrays with reduced dead times in future instruments should be considered when aiming at sampling atmospheric MPC. Certainly, the usage of the current CMOS arrays in PPD-HS with relatively large dead time is a tradeoff between instrument costs and frame rate. We hope to sufficiently address this aspect in the main text through the following changes.

We have added the following statement to P7L13 (revised manuscript):
"We note that the minimum dead time of PPD-HS (T = 177 µs) is relatively high, compared to that of SID-2H (T = 50.0 µs, Johnson et al. 2014) and PPD-2K (T = 8.25 µs, Vochezer et al., 2016)and consequently the fraction of missed particles at high flow rates and particle number concentrations. However, we note that adjustment of the total flow rate as well as of the integration delay and duration parameters determining the overall CMOS dead time is a unique property of PPD-HS, which can be used to empirically optimize the sampling conditions."

We have also added the following statement to P25L10 (revised manuscript):
"Furthermore, we have noted in Sect. 2.4 that the CMOS dead time of PPD-HS is relatively high, compared to similar devices. This causes the fraction of missed particles to be relatively high when sampling with PPD-HS at high particle number concentrations and using high total flow rates with consequences for sampling atmospheric MPC, where the ice fraction is (initially) low. Hence, upgrades to CMOS arrays with reduced dead time would be meaningful in view of potential future field applications of similar devices. Nevertheless, such changes do not affect the capability of using the reduced CMOS array scattering data to successfully determine particle shape."

7. **Consequently, the last sentence in the abstract should be modified accordingly.**
We have changed the sentence from P1L18-20 (initial manuscript):
"We conclude that PPD-HS constitutes a powerful new instrument to size and discriminate phase of cloud hydrometeors and thus study microphysical properties of mixed-phase clouds, that represent a major source of uncertainty in aerosol indirect effect for future climate projections."
To P1L18 (revised manuscript):
"From our laboratory experiments we conclude that PPD-HS constitutes a powerful new instrument to size and discriminate phase of cloud hydrometeors. The working principle of PPD-HS forms a basis for future instruments to study microphysical properties of atmospheric mixed-phase clouds, that represent a major source of uncertainty in aerosol indirect effect for future climate projections."

Minor comments
p.2, line 18: "This is despite the knowledge that cloud particle size distributions comprised of a mixture of cloud droplets and ice crystals are affected by the presence of small ice particles". Please provide references.

We have now added the following references: Lawson, Baker, Schmitt, and Jensen (2001), Korolev, Isaac, Cober, Strapp, and Hallett (2003).

p.2, line 34: Spherical particles can change the incident polarization state. The incident polarization state remains unchanged in the case it is linearly polarized (horizontal or vertical). This is why depolarization techniques use linearly polarized light.
Thank you for spotting this. We agree with the reviewer, that when considering an incident *linearly* polarized light, as in the case of PPD-HS, spherical particles do not cause depolarization of the *scattered* light. We have formulated this more carefully now:
Changed from P2L34 (initial manuscript)
"[…] making use of the fact that aspherical particles change the polarization of incident light, whereas spherical particles do not."
To P2L34 (revised manuscript)
"[…] making use of the fact that incident linearly polarized light is depolarized by aspherical particles, whereas spherical particles do not cause depolarization in the scattered intensity (Bohren & Huffman, 1983; Liou & Lahore, 1974)."

p.3, line 1: Depolarization techniques are sensitive to even small changes from a spherical shape, e.g. spheres with surface roughness or oblate/prolate particles do cause a measurable depolarization signal. However, commercial polarization sensors, like the one used in Zenker et al., 2017, can have difficulties discriminating small ice particles from droplets. These two techniques should not be mixed.
We agree with the reviewer and have adapted the text accordingly.
From P3L1 (initial manuscript)
"In the cases where small near-spherical ice crystals can form, depolarization techniques might be limited in the discrimination between spherical liquid drops and ice crystals."
To P3L1 (revised manuscript)
"In the cases where small near-spherical ice crystals form, commercial depolarization sensors frequently used for cloud composition analysis, might be limited in detecting the depolarization and thus the discrimination between liquid cloud droplets (causing no depolarization) and ice crystals (causing depolarization)."

p.3, line 3: The ability to discriminate phase based on particle shape depends on the used method. Shadow imaging and optical microscopy in best case have an optical resolution around 2 µm and, thus, cannot be used to discriminate the phase of small (<50 µm) cloud particles. 2D diffraction patterns can reveal more details that cannot be resolved by traditional imaging methods.
We acknowledge the concern raised be the reviewer. Certainly, particle type discrimination based on particle shape is ultimately controlled by the optical resolution of the instrument. We have tried to clarify this aspect and reformulated our statement more carefully, which is intended to cover instruments using 2D diffraction patterns of cloud particles.
Changed from P3L3 (initial manuscript)
"Particle shape, for instance, constitutes a powerful parameter that can be used to discriminate hydrometeor types (Hirst and Kaye, 1996)."
To P3L5 (revised manuscript)
"Particle shape, for instance, constitutes a powerful parameter that can be used to discriminate hydrometeor types, given a sufficiently high optical resolution of the instrument used for imaging. For instance, Hirst and Kaye (1996) have shown that analysis of 2D scattering profiles can be used to determine particle shape, which can then be related to particle phase."

p.3, line 9: small-scale complexity, please add the following references: Ulanowski et al., 2014; Schnaiter et al., 2016
Thank you, we have added the suggested references.

p.7, line 4: The discussion of the electronic dead time is crucial for understanding the instrument performance and should be added to the main text. The dead time of 177 to

267 µs is high compared to other cloud instrumentation, which will lead to reduced sampling volume. Fig. S11 shows that at a typical mixed-phase concentration of 100 cm^{-3} 100% or more particles are missed, which will have severe consequences for detecting ice in mixed-phase conditions.
Please see our answer and changes to comment #6.

Fig. 2: The example droplet (a) is a larger droplet with multiple visible rings that does not correspond with the scattering data from CMOS array (d) that shows 1 or possibly 2 maximums corresponding to a small droplet. How would scattering data from a larger droplet with multiple rings look like?
A droplet with multiple rings would result in multiple peaks along each CMOS array at the location of these diffraction fringes (rings) overlap with the CMOS pixels. Example scattering patterns of such spherical particles can be found in our Figs. S20 and S21 and we have made a note of that in the caption of Fig. 2 in the revised manuscript.

p.11, line 20: Are the calibration datasets from the VOAG experiments representative for cloud particles? First, the particle sizes are limited to 32 µm, whereas ice crystals can be significantly larger. More importantly, are salt particles a good proxy for ice crystal shapes? Why not use HINC experiments in cirrus conditions as training data sets?
We agree that atmospheric cloud particles and ice crystals in particular can be much larger than the largest size tested here. However, the goal of the present study is to test and characterize the performance of PPD-HS for use as detector in a CFDC setup (as carefully noted in the title of the manuscript), where particle size is usually constrained to < 10 µm (P10L1-3 in the initial manuscirpt). We agree that it would be useful to test PPD-HS on larger particles of both spherical and aspherical shape prior to using it in e.g. field measurements, which is beyond the scope of this study. Furthermore, please see our answer to comment #1 above.
In order to clarify why the VOAG datasets were used for training of the random forest model, we have added the following statement to the text on P12L5 (revised manuscript):
"Training of the random forest model was constrained to the PEG and NaCl particles generated by the VOAG, which we assume to have similar shape properties to spherical cloud droplets and aspherical ice crystals, respectively. Moreover, the vertical alignment of the VOAG setup allowed us to cover a larger size range without changing the experimental conditions, than would have been possible within the horizontal CFDC setup. Finally, by excluding the HINC-PPD-HS data from model training, these data sets provide independent data for testing classification model performance."

p.13, lines 4-5: The scattering cross section of aspherical particles between 10.6° to 101.0° can be very different to NaCl. Therefore, it cannot be stated that PPD-HS correctly sizes all aspherical particles <20 µm.
The reviewer is right in pointing out that a randomly oriented particle of any shape might not have the same scattering cross section as the NaCl particles investigated here. However, the wide collection angle of the elliptical mirror results in only minor differences in sizing spherical and aspherical particles, such as hexagonal ice crystals (J. Ulanowski, personal communication). Furthermore, it should be noted the assumption of an idealized particle geometry is a limitation of any optical particle counter/instrument. Moreover, changes in detector sensitivity or variation of the trigger laser beam intensity are other parameters that can cause erroneous particle sizing. However, discussion of this is beyond the scope of the presented manuscript.
We have therefore tuned down our formulation on P13L4-5 (initial manuscript) from:
"From these measurements we conclude that PPD-HS correctly sizes particles in the APS size range up to approximately 20 µm."
To now read P14L2 (revised manuscript):
"From these measurements we conclude that PPD-HS sizes particles in the range up to approximately 20 µm, where we can compare to our APS measurements, with reasonable accuracy."

Fig. 4: Please explain do,g and da,g in the figure caption.

The caption of Fig. 4 already explains the parameter space in words. However, we adapted the caption slightly to make this more explicit:
"[…] showing the geometric mean of the optical diameter ($d_{o,g}$) determined by PPD-HS as a function of the geometric mean of the aerodynamic diameter ($d_{a,g}$) obtained from the APS, […]"

Fig. 7: All the particle concentrations are given as counts and not as number concentrations within a volume. Since the sensitive area of the PPD-HS is known the instrument counts can be converted to concentrations. Also, it would be illustrative if the mean modal diameter would be marked in panel e.
We feel that reporting particle detection in terms of counts per time is a more useful and intuitive quantity to assess the framerate of PPD-HS, so that volume-based number concentration would not be an improvement for our purpose. Hence, no change has been made.
It is true that a volume-based number concentration can be calculated under the consideration of the sensitive volume of PPD-HS and should be done when for instance comparing the ambient particle concentration to that detected by PPD-HS and/or comparing the detection rates of multiple instruments.
Similarly, we decided not to include the mean modal diameter as the mode will evolve as it is a function of RH (at a fixed T), as can be seen from our Fig. S19 and the discussion thereof.

P. 24, line 8: What would be the maximum data acquisition rate of the RT-electronics if using priori specified parameters?
In case the RT-electronics are operated independently from the RAW-electronics particle detection rates could reach approximately 3'000 particles per second. We have changed the statement on P24L8 in the initial manuscript from:
"[…] whereas the RT-electronics have the benefit that they could (theoretically) achieve higher particle detection rates than presented here, but […]"
To P25L8 (revised manuscript):
"[…] whereas the RT-electronics have the benefit that they could (theoretically) achieve higher particle detection rates than presented here of approximately 3000 particles per second, but […]"

P. 25, line 12: Please define small.
We note on P17L20-23 (initial manuscript), that the majority of the particles becomes correctly classified as aspherical ice crystals for sizes > 3.25 µm (see also SI Fig. S19) and further note d = 3 µm to be approximately the lower size limit for shape discrimination by PPD-HS (P24L14, initial manuscript).

"Thus, small ice crystals with diameters below approximately 3 µm still remain a challenge for optical instruments such as PPD-HS."

Bohren, C. F., & Huffman, D. H. (1983). *Absorption and scattering of light by small particles*. New York: Wiley Interscience.

Garimella, S., Rothenberg, D. A., Wolf, M. J., David, R. O., Kanji, Z. A., Wang, C., . . . Cziczo, D. J. (2017). Uncertainty in counting ice nucleating particles with continuous diffusion flow chambers. *Atmos. Chem. Phys. Discuss., 2017*, 1-28. doi:10.5194/acp-2016-1180

Johnson, A., Lasher-Trapp, S., Bansemer, A., Ulanowski, Z., & Heymsfield, A. J. (2014). Difficulties in Early Ice Detection with the Small Ice Detector-2 HIAPER (SID-2H) in Maritime Cumuli. *Journal of Atmospheric and Oceanic Technology, 31*(6), 1263-1275. doi:doi:10.1175/JTECH-D-13-00079.1

Korolev, A. V., Isaac, G. A., Cober, S. G., Strapp, J. W., & Hallett, J. (2003). Microphysical characterization of mixed-phase clouds. *Quarterly Journal of the Royal Meteorological Society, 129*(587), 39-65. doi:10.1256/gj.01.204

Lawson, R. P., Baker, B. A., Schmitt, C. G., & Jensen, T. L. (2001). An overview of microphysical properties of Arctic clouds observed in May and July 1998 during FIRE ACE. *106*(D14), 14989-15014. doi:10.1029/2000jd900789

Liou, K.-n., & Lahore, H. (1974). Laser Sensing of Cloud Composition: A Backscattered Depolarization Technique. *13*(2), 257-263. doi:10.1175/1520-0450(1974)013<0257:Lsocca>2.0.Co;2

Vochezer, P., Järvinen, E., Wagner, R., Kupiszewski, P., Leisner, T., & Schnaiter, M. (2016). In situ characterization of mixed phase clouds using the Small Ice Detector and the Particle Phase Discriminator. *Atmos. Meas. Tech., 9*(1), 159-177. doi:10.5194/amt-9-159-2016