Technical note:
Bimodal Parameterizations of
in-situ Ice Clouds Particle Size Distributions

by Irene Bartolomé García et al.

Answer to second review of Referee#1 (RC1)

The comments of the referee are in black,
responses by the authors in blue,
changes in the manuscript text in light blue.

Second review of "Technical note: Bimodal Parameterizations of in situ Ice Cloud Particle Size Distributions", by Irene Garcia and coauthors, submitted to EGUSphere.
Overall, I like your responses to my first review. I have several additional comments, mostly minor, that I would like the authors to consider in their revision of this revised article.

1. As I noted in my first review, are the actual size distributions bimodal? Your Fig. 2 shows normalized PSDs, which assumes the Krämer et al. mass dimensional relationship based on Mitchell. (a) Could you put in supplemental information showing PSDs from the different projects. (b) Could the bimodality be due to shattering? (c) Alternatively, the sample volume of the probes for the small particles is very small compared to the larger sizes, thereby making their concentration artificially large.

   (a) Figures 1 to 4 show a random selection of bimodal PSDs for different campaigns and different temperatures. These figures have been added as Appendix A in the manuscript. Further, PSDs in terms of frequency distributions of ice crystals for the different campaigns are shown in the paper in Figure 2. Also we like to point to the publication of Sourdeval et al. (2018), where PSDs of a number of the campaigns are shown in their Fig. 1 (please see our first answer to your points).

   (b) The data from the JULIA database has been carefully processed to minimize the impact of shattering as we discussed in the answers to the first review. We would like to add here a few more details about why we are confident that shattering is insignificant in the data base (see also Krämer et al., 2016; Luebke et al., 2016, Costa et al., 2017, Krämer et al. 2020): the wall of the CAS inlet entrance is "knife edged" and the inlet tips of the OAPs are modified, which greatly reduces the area susceptible for ice crystal shattering. Further, IAT (InterArrival Time) algorithms are applied to all measurements and the resulting ice concentration frequencies are carefully analyzed for shattering effects as described in Krämer et al. (2020), Appendix A2.2 (see Fig. 5): significant shattering would appear in the frequency distribution of the ice crystals as can be seen in Fig 5. In the respective graphs of the campaigns considered here and shown in the Supplementary Material of Krämer et al., 2020 (https://acp.copernicus.org/articles/20/12569/2020/acp-20-12569-2020-supplement.pdf), no bias in the frequencies is found. Therefore, we do not consider shattering to be the cause of bimodality.

(c) Regarding the different sample volumes, it is true that for small particles it is smaller than for the large ones (50.0 cm^{-3}/sec compared to 2000 - 18000 cm^{-3}/sec -depending on size- for an aircraft speed of 200 m/s), which can increase their concentration. However, this overestimation is reduced when having a large a amount of seconds, as described in the Appendix A2.3 of Krämer et al. (2020). In our study we have around 543000 seconds of measuremensts (see Table 2), so we consider the air volume is large enough to consider realistic concentrations, even for small particles.

2. Eq. 2. The problem I see is that deriving Deq assumes a mass dimensional relationship. If D is used rather than Deq, then the PSD relationships are independent of the assumed mass dimensional relationship and are based on the measurements themselves. Could you comment on this.
   The normalization method used in our study follows the work done by Delanoë et al. (2005), who adapted to ice particles the framework originally developed for rain by Testud et al. (2001), in the following refered as T01. The framework by T01 uses the mean volume diameter, D_m, which is a "volume weighted" mean diameter. Therefore, it is more convenient to use the equivalent diameter Deq and represent the ice particles with their equivalent spherical water particles, since the ice crystals are present in a wide variaty of types and shapes.

3. Line 140. You mean Deq or D, being the physical diameter.
   We mean m(D).

4. Often when cloud tops are close to or somewhat below 255K, the upper parts of the cloud are liquid or mixed-phase. I think the cutoff temperature should be perhaps 265K to completely rule out liquid water. See the article: A global view of midlevel liquid-layer topped stratiform cloud distribution and phase partition from CALIPSO and CloudSat measurement.
   Fig.6 (from Krämer et al. 2023, in preparation, **confidential**). shows that between 255 K and 265 K coexistence of drops and ice crystals is present in the clouds. Since we are looking at pure ice clouds that only appear below 255 K in our data set, we would like to maintain the cut off temperature at 255 K.

5. Legend, Figure 5 d. "paremeterization" fix spelling.
   Fixed.

6. 232. "crystalls" fix spelling
   Fixed.

7. 250. Underestimates
   Fixed.

8. 282. Remove crystals. The larger particles might be aggregates, which wouldn't be ice crystals.
   Fixed.

9. 284. datasets
   Fixed.

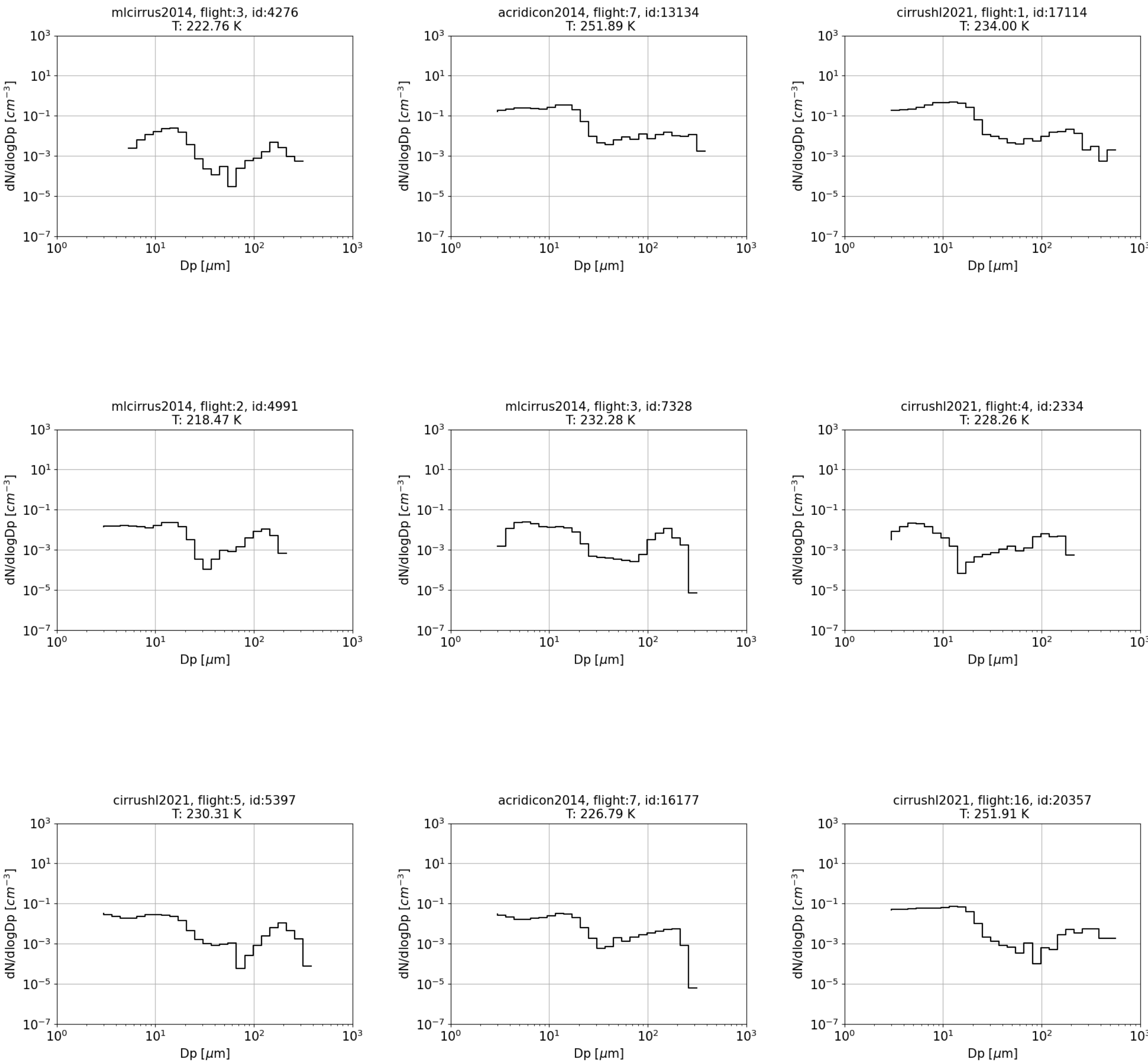

Figure 1: Examples of single PSDs

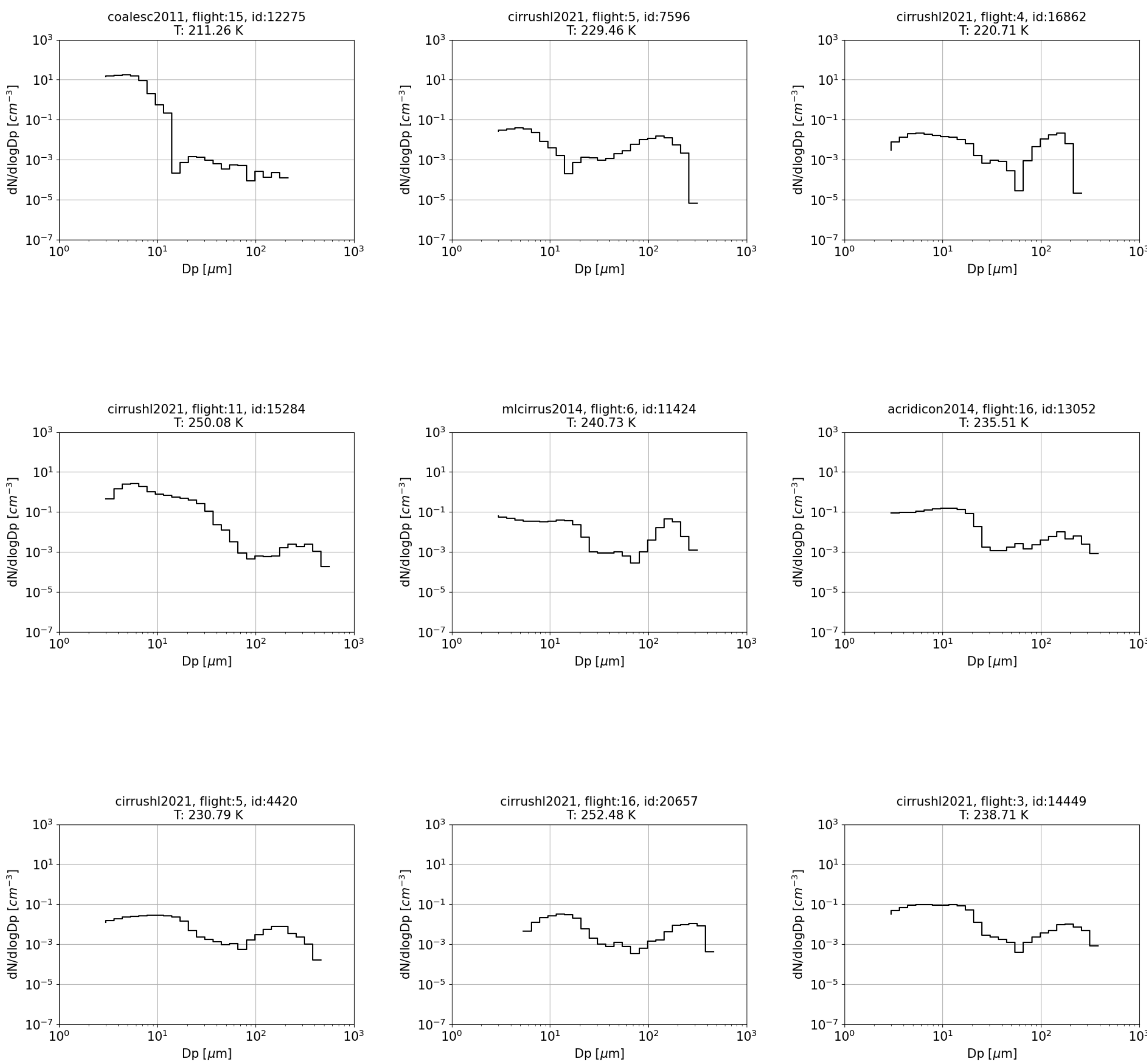

Figure 2: Examples of single PSDs

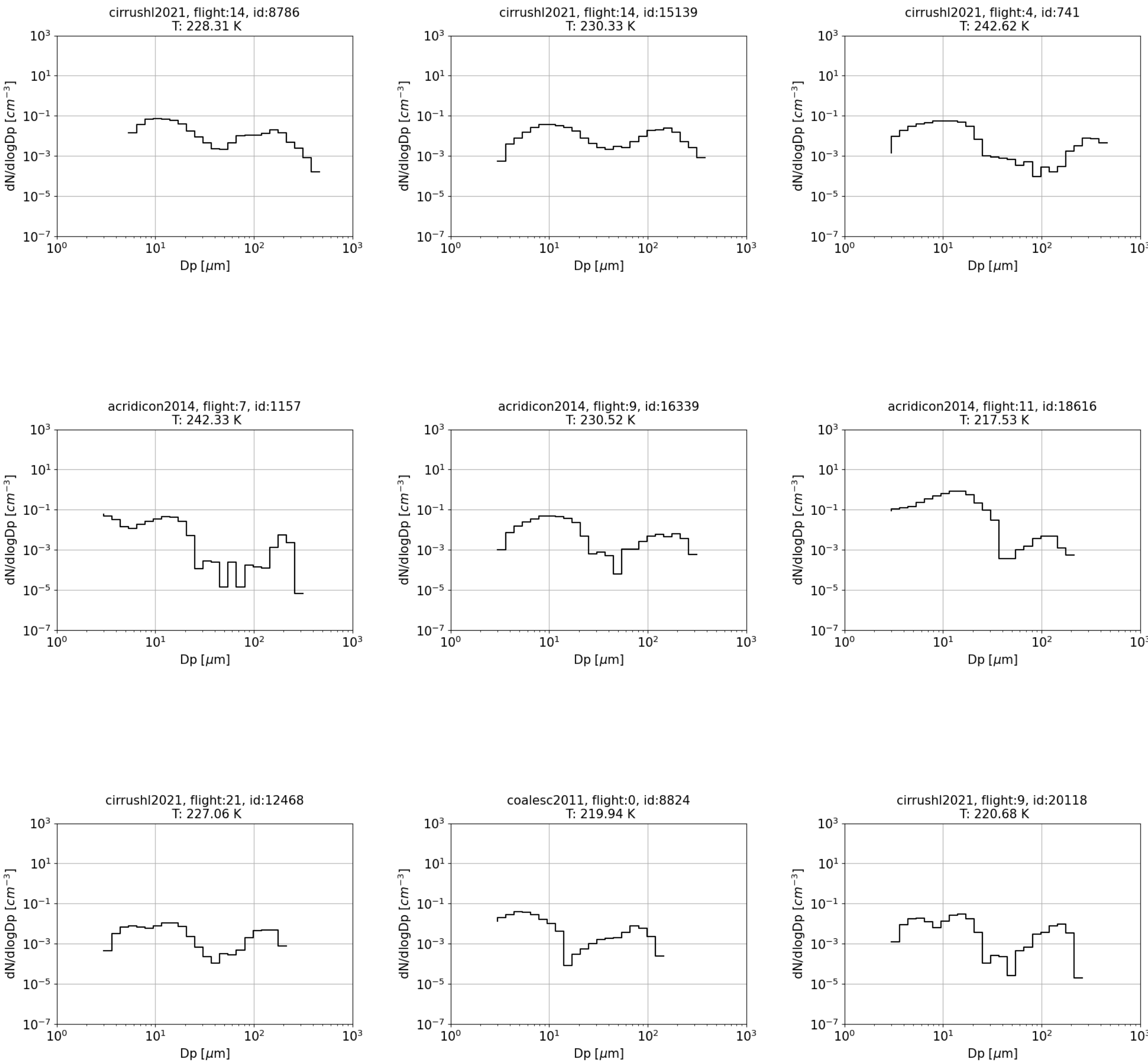

Figure 3: Examples of single PSDs

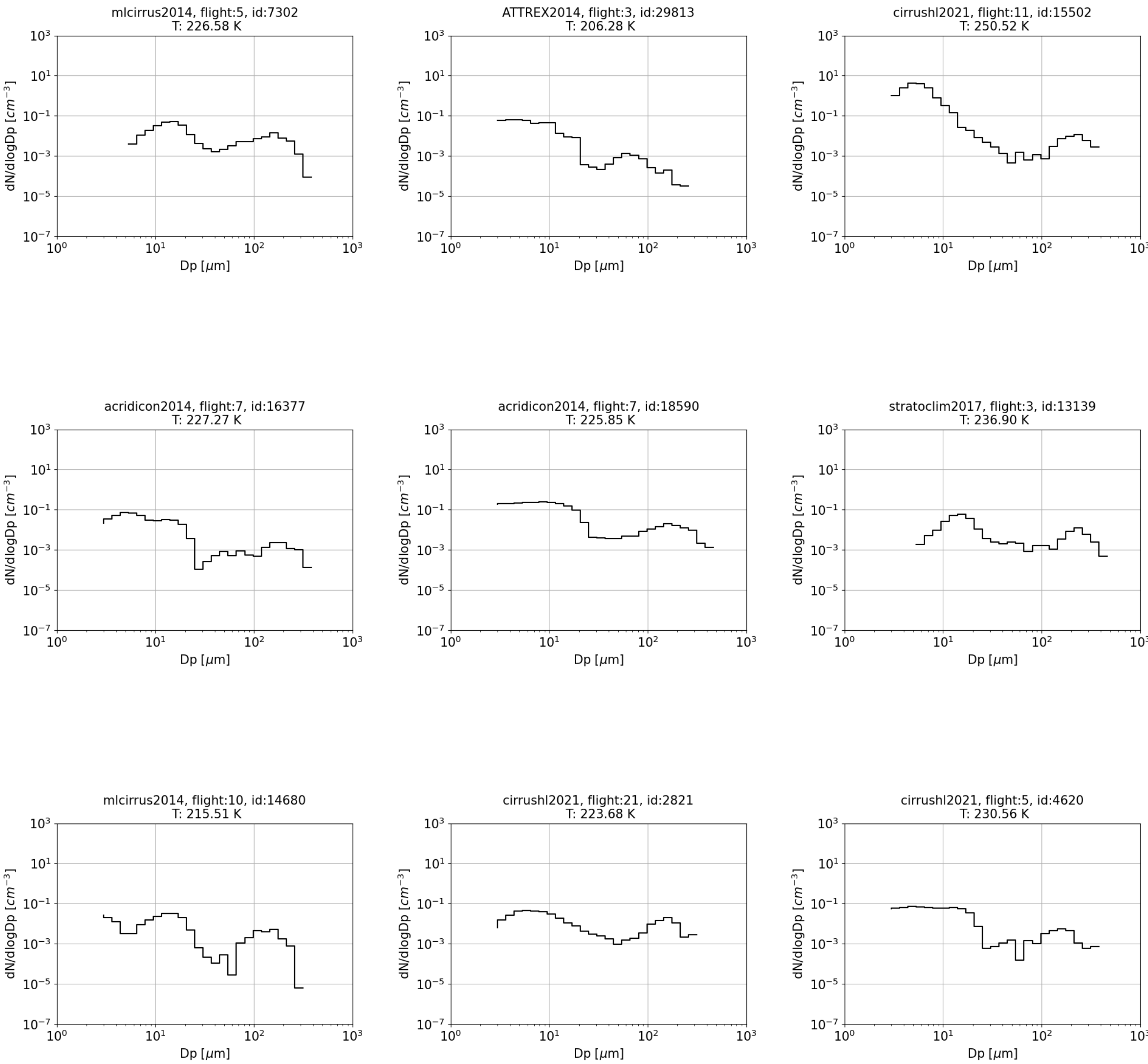

Figure 4: Examples of single PSDs

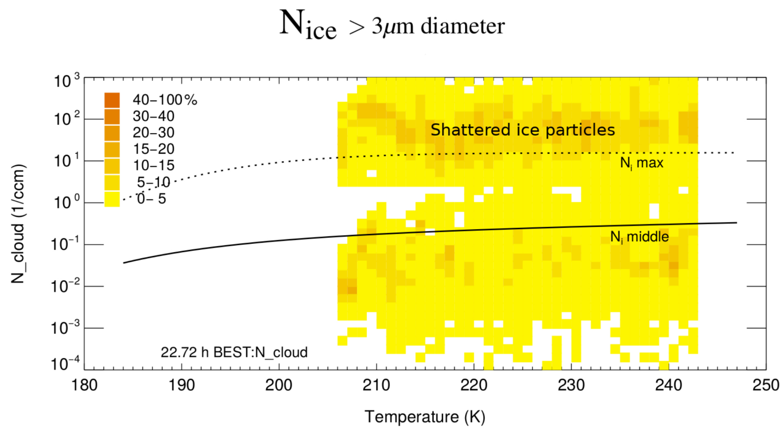

Figure A3. Example of N_{ice} measurements biased by shattering of large ice particles, visible in the frequencies of occurrence: high frequencies appear at N_{ice} concentrations between 10 and 100 cm^{-3} for all temperatures, which are not present in undisturbed measurements (see Fig. A2). The black lines denote the middle and maximum N_{ice} lines from Krämer et al. (2009).

Figure 5: Figure adapted from Krämer et al. (2020).

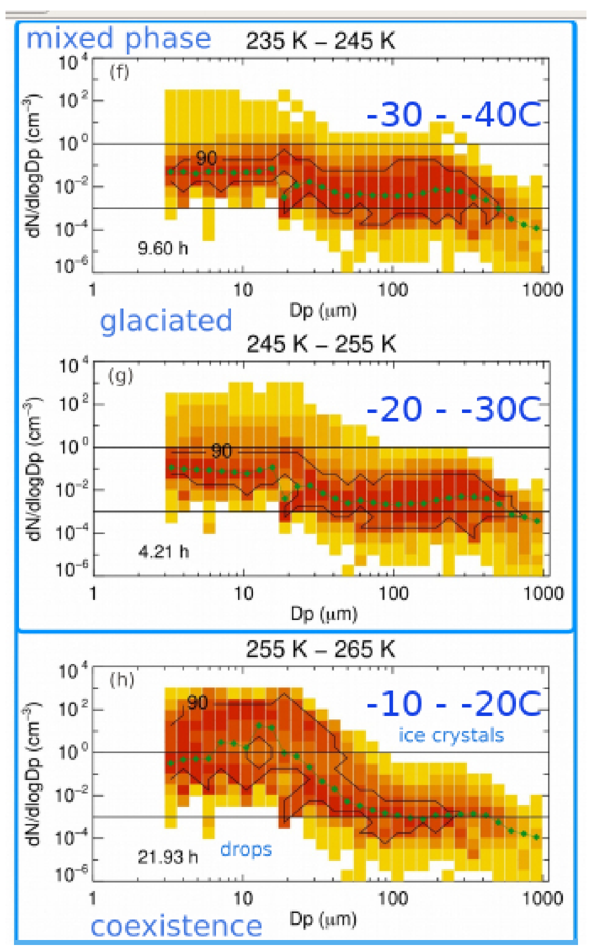

Figure 6: Cloud particle size distributions in 10K temperature intervals, color coded by frequencies of occurence. The green lines show the median PSDs, black/white contour lines enclose 90 / 50% of the data points; Krämer et al. (2023, in preparation), **confidential**.

Answer to second review of Referee#2 (RC2)

The comments of the referee are in black,
responses by the authors in blue,
changes in the manuscript text in light blue.

1. Section 4.2 and 4.3: I found it difficult to understand what is exactly Julia 1M. I understand it is one mode distribution fitting, but what makes it different regards to D05 or D14? Can you clarify please.
D05, D14 and JULIA 1M are in principle computed using the same normalization procedure adapted from the framework of Testud et al. (2001) and described in Delanoë et al. (2005). The differences are in the in situ database used for each of them, the m-D relationship and what parameters of the modified gamma function were used to minimize the cost function (i.e. to predict the in-situ data). D05 and D14 were designed to best fit optical parameters (lidar extinction and radar reflectivity) whereas JULIA 1M aims to better characterise physical parameters (IWC and Ni). Please find more details below:
D05: data from the experiments CLARE98, CARL99, EUCREX, FASTEX, ARM, CEPEX and CRYSTAL FACE; m-D relationship from Brown and Francis (1995), selection of the best parameters for the modified gamma function after analysis of several combinations. Please see Delanoë et al. (2005) for more details.
D14: data described in Heymsfield et al. (2013), m-D relationship used in the DARDAR products (combination of Brown and Francis (1995) for $D > 300\,\mu$m and Mitchell (1995) for hexagonal columns) and lidar extinction coefficient and radar reflectivity to minimize the cost function to chose the α and β parameters of the modified gamma function. For reference, please see Delanoë et al. (2014).
JULIA 1M: data from the JULIA database, modified Mitchell et al. (2010) m-D relationship described in Krämer et al. (2016) and IWC and N_{ice} to minimize the cost function.
The following has beed added in Sect. 4.2, lines 226-228:
To summarize, the parameterizations differ in the data used to compute each of them, the m-D relationship used and how the parameters of the modified gamma function were obtained.
Also in Sect. 4.2 we write in lines 225-226:
In D14 the parameters are proportional to the second and approximately sixth moment of the distribution and in our study to the zeroth moment and between the second and third moment.

2. Section 4.3

- lines 230 : this is not well said. If in cirrus (your definition?) the aggregation processes play no role. Hence, there would not need to use two distribution (or two modes) to fit measured PSD in your dataset. But, later you say that bimodality start to play an important role for T > -60°C. I wonder about the cause of the physic processes that lead to two distributions, if it is not vapor diffusion on one side and aggregation on the other side (riming being impossible if no supercooled water)?
  We added the definition we are using of cirrus at the beginning of Section 4.1: "As cirrus we understand all clouds colder than 235 K (Krämer et al., 2016). In the temperature range directly below, the clouds can also have their origin as mixed-phase clouds that have risen from below and completely glaciated latest at 235 K. This physical definition of cirrus is based on the ice formation mechanism, which is on the one hand the just mentioned complete glaciation of liquid clouds (liquid origin cirrus) and on the other hand cirrus that form directly as ice (in-situ origin cirrus)."

  Regarding the causes for bimodality, we discuss in Section 4.3 that at temperatures lower than $-45\,°C$ the growth of the ice crystals is likely due to depositional growth and sedimentation and aggregation are less significant (Jackson et al., 2015). For warmer temperatures, the ice particles also grow by vapor deposition, but sedimentation from above is a possible source for larger particles that can cause a bimodal particle spectrum (Zhao et al., 2011). Another process that can lead to bimodality is two-step ice nucleation, where there is first heterogeneous nucleation of a few ice crystals that may grow to larger sizes, followed by homogeneous nucleation of more and smaller ice crystals. However, the main reason for the bimodality of cirrus PSDs is the superposition of in-situ origin and liquid origin cirrus. Ice crystals of liquid origin are significantly larger than those of in-situ origin because they stem from lower altitudes where there is more water to allow them to grow to large sizes, especially since only very few drops out of a liquid cloud freeze so the available water vapor is deposited only among them.
  This discussion has been extended between lines 235 and 242 and to avoid confusion, the following sentence in line 233 has been modified:
  In cirrus clouds, riming and secondary ice production play no role and aggregation, at the coldest temperatures, is nearly negligible.

- lines 244 : Indeed, there can be many orders of magnitude between the concentration of small and large ice hydrometeors. Do large errors for large hydrometeors are less important? Maybe it needs more explanation.
  No, they are not, but it is important when analysing the results to take into account that the large hydrometeors are present in lower concentrations.

- Notes about Figure 5, all parameterisations are not accurate for large hydrometeors. However, surprisingly your new parameterisation that is supposed to improve the representation of small ice crystals show more benefits in the modeling of large hdyrometeors. This is well highlighted looking median error for D > 50µm in all range of temperatures. While, median error for D < 50 are similar between D05 and Julia 2M. For the case of small ice crystals, bimodal parameterisations start to pay for the warmer temperature i.e. -30 to -20°C. Also, for temperature ranges warmer than -50°C, the Julia2M improves the representation of ice crystals from 50(20 you said) to 100 microns (the modes of large hydrometeors !?)... So adding a mode of small ice crystals do not benefit only small ice crystals but also larger !!! This is really interesting when taking into account the measurement uncertainties that are commonly admited by the community (Baumgardner 2017) for small and large ice crystals : 100% for D < 100µm and 50% for D > 100µm. Moreover, these former parameterizations do not use concentrations for D < 50µm. Clearly, there is a need to use more than one distributions to model the concentrations of hydrometeors from pristine ice to aggregates.

  We thank the reviewer for this comment that highlights an interesting result of our study. We have added the following sentence in Section 4.3, lines 270-271:

  As indicated by the median of the percentage error for particles smaller and larger than 50 μm, using a bimodal parameterization improves the representation of both the small and large mode, improving the large mode especially for warmer temperatures.

  and also in Section 5, lines 301-302:

  Adding a mode of small ice crystals do not benefit only small ice crystals but also large, despite the large measurement uncertainties associated with the large ice crystals.

3. lines 285 : " it adjusts better to the bimodal shape of the PSDs " I would add when "it occurs".

   Added

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