### Responses to the Referee #2 Comments on

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

by Irene Bartolomé García et al.

The authors would like to thank both anonymous referees for their comments, that helped improved the manuscript and make it more comprehensible. In the following, the comments and questions of referee #2 are addressed one by one. A reviewed version of the manuscript is attached at the end with the deleted parts in red and the new additions in blue.

## Answer to Referee#2 (RC2)

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

Review of Tecnical note: Bimodal Parameterizations of in-situ Ice Clouds Particle Size Distributions Authors: Irene Bartolomé Garcia et al.

The authors are proposing a new technic for the parameterization of ice particle size distributions with gamma normalized size distributions as in Delanoë et al 2014. But, they are using two normalized distributions, one for Diameters smaller than 50 µm and one for Diameters larger than 50 µm, instead of one for all spectrum of size of measured ice crystals. They are comparing their retrieved ice PSD with the ones of retrieved with the former methods i.e Delanoë et al., (2014 and 2005) and applied to their dataset. Globally, overall their dataset (Figure 4 and 6) the new method seems to be more accurate to retrieve small ice crystals concentration. They motivate their study, on the fact that concentrations of small ice crystals are too often neglected or not considered, impacting accuracy of retrieval methods for clouds properties. The main reason being the measurment uncertainty of small ice crystals.

This is not the first study that offers a parameterization of ice PSD with two modes (two gamma distributions cf. Field et al., 2007). However, this is the first in my knowledge that includes ice particles since  $3 \mu m$ .

## **Major Comments:**

#### 1. **Bimodality:**

(a) Are you assuming that all ice PSD in your ice clouds are bimodal?

No, we do not assume that all PSDs are bimodal, but that bimodality can be observed in ice crystal PSDs. Futhermore, Sourdeval et al., (2018) showed using mean PSD from in situ aircraft observations compared with the retrieved mean PSD from satellite measurement that the occurence of bimodality impacts the capability of single-mode parameterization to predict the PSD shape and leads to major retrieval issues in these warmer clouds. The deviation between the mean of the PSDs is indeed clear for temperatures  $T > -50^{\circ}$ C where the bimodality is present (Fig. 1, where aggregation and possibly secondary ice production processes can occur. This is in agreement with other studies that analyzed in situ data, for example Jackson et al. (2015)

- (b) Line 53: You are introducing frequencies of bimodality in the discussion, would it be consistent to divide the distribution in two modes if there is only one mode? Since the modes of the distribution correspond one to the small particles and the other one to the large ones, even if the PSD is monomodal, it would be covered by one of the two modes or by both. In Fig.3, the rmse (root mean squared error) of the correlation between the parameterized and the observed N<sub>ice</sub> is compared for each of the parameterizations. It is shown, that for the monomodal ones the warmer the temperature interval, the larger the rmse is, whereas for the bimodal parameterization it remains approximately constant. Therefore, the impact of using one mode when bimodality is present (warmer temperatures) seems stronger than the use of two modes when one mode is present (colder temperatures).
- (c) Hu et al 2022 have developed a method to estimate the number of modes in ice PSD. They, showed that at coldest temperature (-50° C to -40° C) ice PSD are 60% of the time one mode; except for IWC> 1.5  $gm^{-3}$ . Why there should be bimodality? Please, see answers to (a) and (b).
- (d) In the introduction you are linking the shape of the ice PSD and the growth process. Then, it is shortly discussed in section 4.3. You are assuming that it is the difference of newly formed ice particle against sedimenting sizes. I encourage you to improve the discussion on this topic. Because, if the evolution of the size of the hydrometeors is linked to the growth rate: vapor diffusion, aggregation and riming. Then, If there is more than one growth process (without counting secondary ice production) there should be more than one mode in ice PSD!?

The following has been added to the manuscript in Sect. 4.3:

In cirrus clouds, riming and secondary ice production play no role and aggregation is nearly negligible. These processes are of importance for mixed-phase clouds, which, as mentioned in Sect. 4.1 entail 9.8 % of the analyzed data. In Jackson et al. (2015) it was discussed that at temperatures lower than  $-45\,^{\circ}C$  the growth of the ice crystalls is likely due to depositional growth and sedimentation and aggregation are less significant. For warmer temperatures, smaller particles grow by vapor deposition and aggregation, being sedimention from above another possible source for the large particles, which together with heterogenous nucleation taking place at the same time would explain the bimodality (Zhao et al., 2018).

(e) Then, you choose a cutting diameter of  $50 \mu m$ , do you mean that the division of the growth processes such growth by vapor diffusion against growth by aggregation (or sedimentation) is here. Can you give a reference or an argument, assumption maybe, for this cutting diameter? If I observe one column of few hundreds of micron wasn't it a monocrystal of few microns in its past?

The diameter of  $50 \mu m$  was first tested because it is the smallest diameter in D14, but also because it seems to agree well with the division between two modes when computing the median PSD (Fig. 2). The following has been added in Sect. 4.3: This cutting diameter agrees well with the division between the small and large modes when plotting the median PSD of all data (not shown).

A diameter of 20  $\mu$ m and 100  $\mu$ m have been tested to see if there are major differences with the current results. 20  $\mu$ m was chosen as a division between smaller particles being mainly dominated by nucleation / evaporation and larger by growth / coa-

lescence / aggregation processess (Krämer et al., 2022).  $100 \, \mu m$  was selected as one of the cutting diameters from Hu et al. (2022). Figure 4 compares the rmse of the correlation between retrieved  $N_{ice}$  and the observed  $N_{ice}$  for 20, 50 and  $100 \, \mu m$  using the parameters specified in the manuscript (obtained using a diameter of  $50 \, \mu m$ ). There is a slight decrease for the coldest temperatures and a slight increase for the warmer ones when using a cutting diameter of  $20 \, \mu m$  with respect  $50 \, \mu m$ . For  $100 \, \mu m$  there is a slight increase for all temperature intervals.

Additionally, the alpha and beta pairs have been computed using  $20 \,\mu\text{m}$  and  $100 \,\mu\text{m}$ . Figure 5 shows the comparison of the rmse of the correlation of parameterized and observed N<sub>ice</sub>. It is shown that for colder temperatures the results for 20 and 50  $\mu$ m are close, but the warmer the temperature, the greater the difference, being the rmse for  $20 \,\mu\text{m}$  higher. For  $100 \,\mu\text{m}$ , the rmse is for all temperature intervals above the rmse for  $50 \,\mu\text{m}$ . Considering the results shown inf Fig. 4 and Fig. 5, we consider a cutting diameter of  $50 \,\mu\text{m}$  is an adequate choice.

(f) You are citing Field et al., (2007) that also proposed a bimodal normalized parameterization, but as function of optical maximum length and effective radius. However, they did not use concentrations of small ice under 100 microns. What would be the impact by taking the concentration from 3  $\mu$ m (this would be maybe to consider for a second part publication).

The parameterization by Field et al (2007), hereafter F07, is technically bimodal but only one mode was constrained with in-situ observations. The second mode, for crystals with sizes smaller than 100  $\mu$ m, correspond to an exponential extrapolation. Following the Sourdeval et al (2018) study, the authors performed a similar investigation of the performance of the F07 parameterization as part of an internal evaluation for the MetOffice. Fig. 11 shows one such comparison done for the SPARTICUS campaign. It can be seen that F07 (in green; here their mid-latitude parameterization) does not perform as well as D05 for the colder temperature bins but especially that D05 and F07 perform equally poorly when bi-modality occurs. This makes the present study relevant for even parameterizations such as F07. If the study of Field et al., (2007) was updated using a database that includes ice particle size down to 3  $\mu$ m (like the JULIA database used in our study), we consider that the resulting parameterization could deliver better results.

## 2. Melting diameter and mass-size relations:

(a) To retrieve the melting diameter you are using a mass-dimension relationship used in Krämer et al., (2016). In this later study, it is justified for temperature less than -38° C (235.15K) in cirrus cloud and based on former studies. Is it consistent to use it for T > 235K, knowing that few studies with direct measurment of IWC have shown an impact of the temperature on the m(D) coefficients in ice clouds.

The reason that we used the same m(D) relation at warmer temperatures is from the comparison of various m(D) relations in Afchine et al. (2018) (see Figure 8 here). There it can be seen that the difference between all relations is small, even when looking at relations derived for warmer temperatures. However, we are aware that the uncertainties of the derived IWCs are larger at warmer than at colder temperatures. This explanation is now included in the manuscript in Section 2.2.

(b) I would like to see IWC retrieved with this m(D) and original ice PSD, compared with the measured IWC available in your dataset; and also as function of temperature. Why not use, your own retrieved m(D) from the dataset you are using and see the impact on the Nice. And also with Brown and Francis as in the original version (see first review comment).

Figure 6 shows correlation plots between the retrieved IWC using the modified m(D) of Mitchell et al. (2010) together with the bimodal parameterization and the same m(D) but with the observed PSDs. Each correlation plot corresponds to a temperature range of  $10\,^{\circ}$ C. The agreement between parameterized IWC and observed IWC is overall high, especially for temperatures between  $-90\,^{\circ}$ C and  $-60\,^{\circ}$ C.

A comparison between measured IWC (with a hygrometer) and IWC from the observed PSDs and the modified m(D) of Mitchell et al. (2010) was done by Afchine et al. (2018) for two campaings (Fig. 7) showing satisfactory results. However, the comparison was made only for the colder temperature range, since the measured IWC was only available in this range. Direct measurements of IWC together with measurements of PSD are only available for one campaign, therefore it is not possible to derive our own m(D) for each single campaign as in Delanoë et al. (2014).

Regarding Brown and Francis m(D), Afchine et al. (2018) did a comparison between several m(D) relations (Fig.8). The differences were not significant, except for diameters around  $100\,\mu\text{m}$  where the Brown and Francis m(D) presents a higher mass. Additionally, in Delanoë et al. (2014), it is argued that it was obtain primarily at temperatures between -  $20\,^{\circ}\text{C}$  and -  $30\,^{\circ}\text{C}$  and dominated by particles between 200 and  $800\,\mu\text{m}$ . Therefore, we consider that the Francis and Brown m(D) might not be the most suitable one for our study since we cover colder temperatures and smaller particles.

- (c) Figure 4 and 5, I would consider plotting the error in percent regarding original concentrations instead of pure concentration, with a recall of your measurement uncertainties especially for smaller size. Small crystals and large ones do not have the same order of concentrations; this is important.

  Figure 9 and Fig. 10 in this response show the median percentage error of the PSD for each size bin for each parameterization. The shadowed region correspond to the range between the percentile 25 and the percentile 75. Inside the panels it is
- (d) Figure 5 only, AS your study is questioning the retrieval of small ice concentration, I would summarize it, for small and large ice particles i.e. below and above the cutting diameter, instead of showing it as function of size bins.

have been added to the manuscript replacing the previous figures.

indicated the median percentage error when considering all size bins. This figures

In the new version of Fig. 5 (Fig. 10) for each temperature interval it is included the median error for diameters smaller and larger than  $50 \,\mu m$  and for the complete range of diameters.)

- (e) Line 243: I do not think that IWC and ice PSD can be dissociated. Can you be more clear on your description of the error of IWC, dimension you are using instead of log, , rate of underestimation and overestimation. It does not talk for someone who is not a specialist.
  - The units of the IWC are  $gm^{-3}$  and the parameterized and observed IWC are compared in correlation plots similar as the ones for N<sub>ice</sub>. Line 243 has been modified in the manuscript: There is a slight underestimation (about 2 %) of the IWC for values between about  $1 \times 10^{-7} gm^{-3}$  and  $1 \times 10^{-5} gm^{-3}$  and an overestimation (about 7 %) between about  $1 \times 10^{-3} gm^{-3}$  and  $1 gm^{-3}$ .
- (f) 'IWC is sensitive to large particles': it is more complicated than that. Where do you define large ice particles hundred of microns, millimeter ... The spectrum of all ice crystals goes from few microns to centimeter in some case. Then, C, S and X band radars would be enough to retrieve IWC in cloud. For a fact, W band and Ka band do a better job i.e Delanoë et al., (2005 & 2014) which are less sensitive to very large ice crystals.

We have modified the the sentence to: Since all parameterizations have a similar behaviour for the large particles and IWC is sensitive to large particles ( $\gtrsim 300$  µm), this result was to be expected.

#### 3. Remarks on the conclusion;

- (a) The methods of Delanoë et al., is developed for all ice clouds, while I understand that the dataset used in this study is mainly made with sampling in cirrus clouds (except for ACRIDION campaign). What about the temperature below -20° C? Can we generalize your conclusions to all ice clouds and to all range of temperature? If yes Why?
  - In our study we focus on the retrieval of ice PSDs and for temperatures lower than about -20° C. Therefore, we wouldn't generalize the results of our parameterization for warmer temperatures and we would suggest a specific study.
- (b) Maybe you can recall the definition of cirrus clouds you are using, does it agree with the one in Heymsfield et al., (2017) and the AMS glossary for example?
  - We consider all clouds colder than -38°C to be cirrus (see Krämer et al., 2016), because at warmer temperatures clouds can also be in the mixed-phase state. This physical definition is based on the ice formation mechanism and includes in-situ origin cirrus that form directly as ice, and liquid origin cirrus, which forms at lower altitude as liquid clouds which completely galciate latest at -38°C (included now in the manuscript in Sect. 4.1). This is not entirely in line with Heymsfield et al. (2017) or the AMS glossary, however, as discussed by Heymsfield et al. (2017):
  - 'Classifying cirrus by means of the formation mechanisms leads to cirrus types characterized by physical parameters, besides those embedded in the terminology of the WMO (1956) for all cloud types (see section 2a), which are defined based on morphology derived from observations of visual appearance.'

We are aware that these two cirrus definitions currently exist side by side and a discussion is ongoing which one should be accepted in the future.

I suggest these references to help the discussion:

Korolev, A., Heckman, I., Wolde, M., Ackerman, A.S., Fridlind, A.M., Ladino, L.A., Lawson, R.P., Milbrandt, J., Williams, E., 2020. A new look at the environmental conditions favorable to secondary ice production. Atmospheric Chemistry and Physics 20, 1391-1429. https://doi.org/10.5194/acp-20-1391-2020.

Heymsfield, A.J., Schmitt, C., Bansemer, A., 2013. Ice Cloud Particle Size Distributions and Pressure-Dependent Terminal Velocities from In Situ Observations at Temperatures from 0° to 86°C. J. Atmos. Sci. 70, 4123â-4154. https://doi.org/10.1175/JAS-D-12-0124.1

Schmitt, C.G., Heymsfield, A.J., 2010. The Dimensional Characteristics of Ice Crystal Aggregates from Fractal Geometry. Journal of the Atmospheric Sciences 67, 1605-1616. https://doi.org/10.1175/2009JAS3187.1

Heymsfield, A.J., Krämer, M., Luebke, A., Brown, P., Cziczo, D.J., Franklin, C., Lawson, P., Lohmann, U., McFarquhar, G., Ulanowski, Z., Tricht, K.V., 2017. Cirrus Clouds. Meteorological Monographs 58, 2.1-2.26. https://doi.org/10.1175/AMSMONOGRAPHS-D-16-0010.1

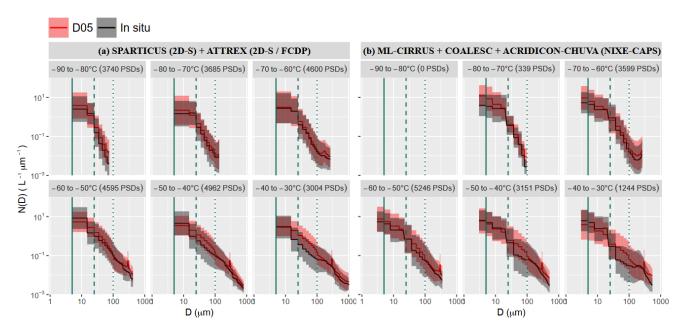


Figure 1. (a) Mean PSDs measured (black lines) during SPARTICUS and ATTREX, averaged per 10 °C temperature bin (from -90 to -30 °C). Black contours indicate one standard deviation around the mean. The mean and spread of one-to-one predictions by the D05 parameterization are similarly indicated in red. The total number of PSDs in each  $T_c$  bin is indicated in the panel heading and the relative contributions from each campaign can be deduced from Fig. S1. Vertical plain, dashed and dotted green lines indicate D = 5, 25 and  $100 \, \mu m$ , respectively. The SPARTICUS data with  $T_c < -60 \, ^{\circ}$ C are ignored here to avoid contaminating FCDP measurements with uncertainties arising from the first size bins of 2D-S. (b) Similar to (a) but for the ML-CIRRUS, COALESC and ACRIDICON-CHUVA campaigns.

Figure 1: Figure 1 from Sourdeval et al., 2018, ACP

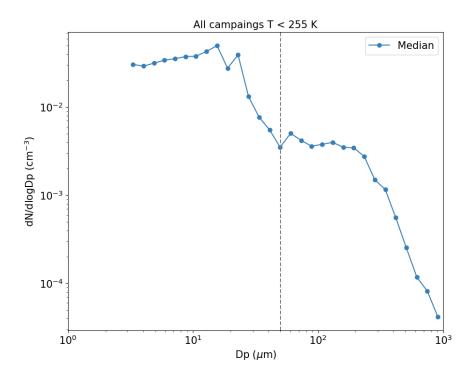


Figure 2: Median PSD of all campaigns considering temperatures lower than 255 K.

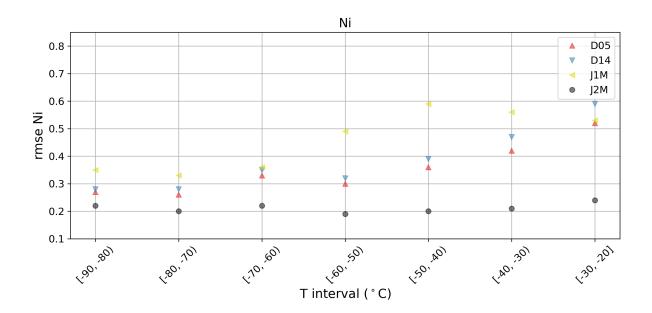


Figure 3: Root mean square error (rmse) of the correlation between the parameterized ice number concentration  $(N_i)$  and the observed  $N_i$  for several temperature intervals and for each of the parameterizations presented in the study.

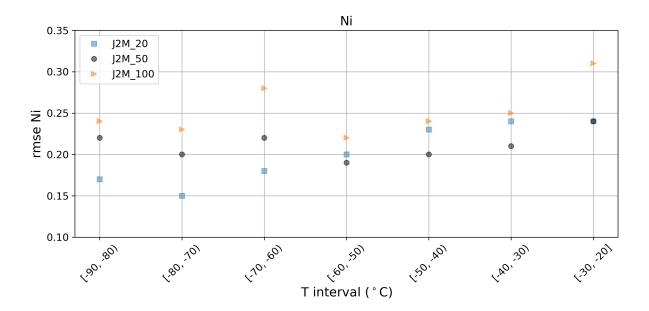


Figure 4: Root mean square error (rmse) of the correlation between the parameterized ice number concentration  $(N_i)$  and the observed  $N_i$  for several temperature intervals for three cutting diameters. The fitting parameters correspond to the ones specified in the manuscript.

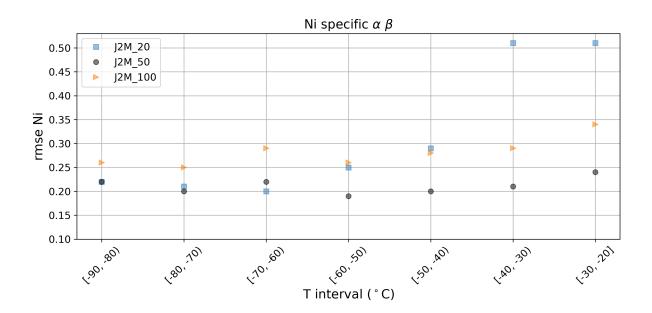


Figure 5: Root mean square error (rmse) of the correlation between the parameterized ice number concentration  $(N_i)$  and the observed  $N_i$  for several temperature intervals for three cutting diameters. The fitting parameters for the gamma function were computed for each cutting diameter.

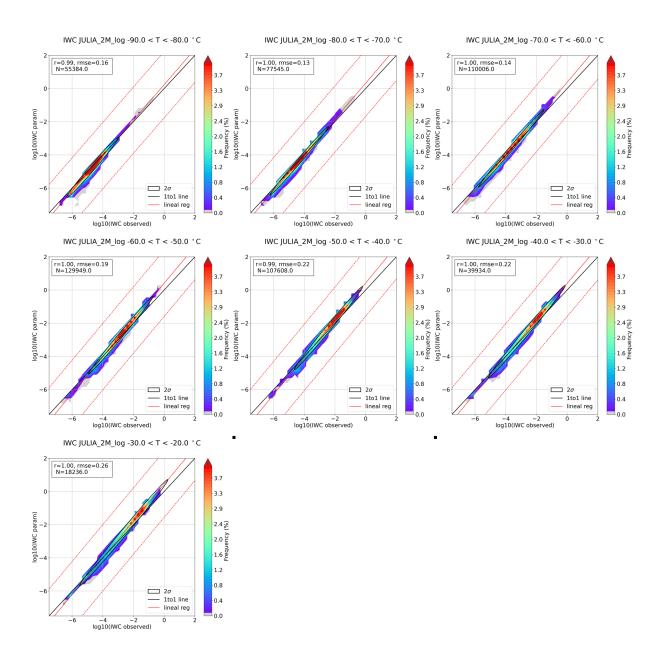


Figure 6: Correlation between the parameterized IWC and the observed IWC for temperatures between -90  $^{\circ}$ C and -20  $^{\circ}$ C in intervals of 10  $^{\circ}$ C, . The parameterized IWC corresponds to the use of the bimodal parameterization. The observed IWC refers to IWC computed using the measured PSDs. The IWC was computed with units of gm $^{-3}$ . Both axis correspond to the logarithm of the IWC.

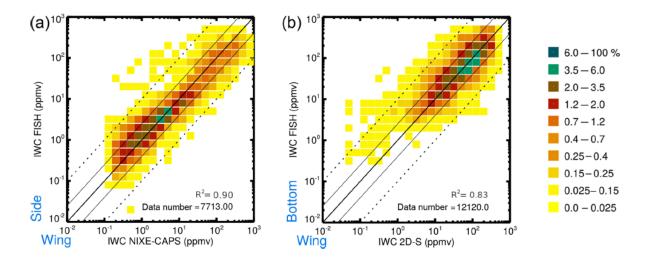


Figure 7: Figure 11 from Afchine et al. (2018). Comparison between IWC measured with a hygrometer (y-axis) and IWC derived from a cloud spectrometer (x-axis).

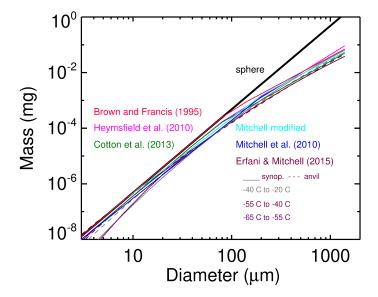


Figure 8: Afchine et al. (2018), Figure 8 (left panel) with the m(D) relation of Brown and Francis (1995) added.

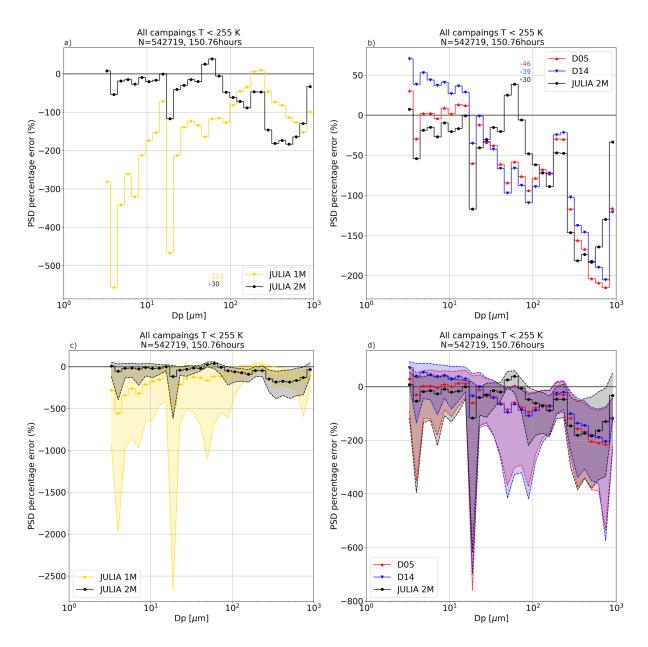


Figure 9: Percentage error of the parameterized PSD. The numbers inside panels (a) and (b) indicate the median error for each parameterization (D05, D14 and bimodal J2M). The shadow region in panels (c) and (d) correspond to the area between percentile 25 and percentile 75.

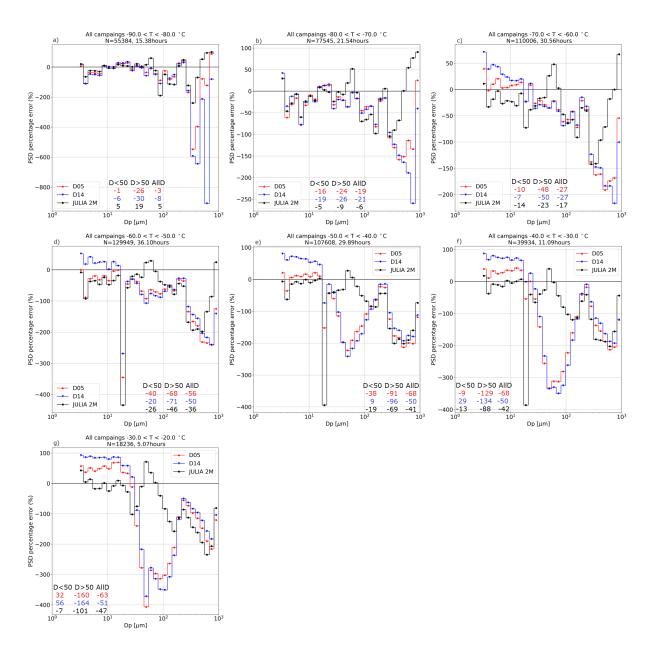


Figure 10: Percentage error of the parameterized PSD in  $10\,^{\circ}$ C temperature intervals. Inside each panel the median error for diameters smaller than  $50\,\mu$ m, larger than  $50\,\mu$ m and the complete range of diameters is indicated for each parameterization (D05, D14 and bimodal J2M).

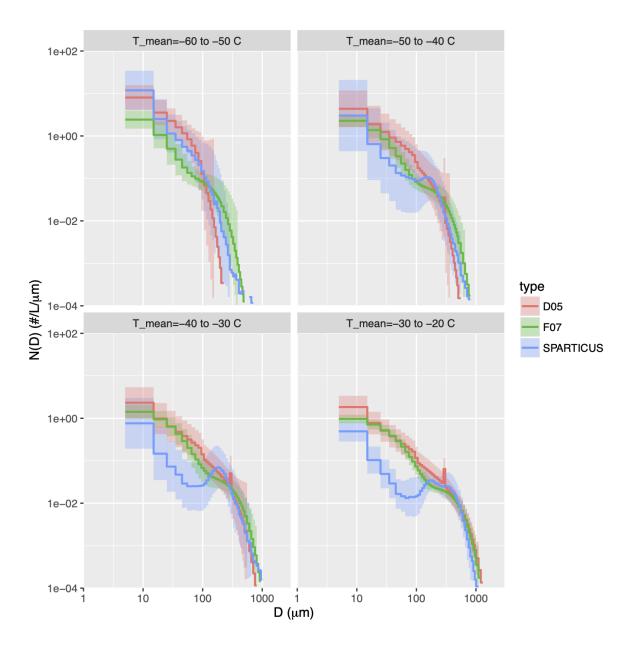


Figure 11: Predictions of D05 and F07 corresponding to observations during the SPARTICUS campaign.

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Afchine, A., Rolf, C., Costa, A., Spelten, N., Riese, M., Buchholz, B., Ebert, V., Heller, R., Kaufmann, S., Minikin, A., Voigt, C., Zöger, M., Smith, J., Lawson, P., Lykov, A., Khaykin, S., and Krämer, M.: Ice particle sampling from aircraft – influence of the probing position on the ice water content, Atmos. Meas. Tech., 11, 4015–4031, https://doi.org/10.5194/amt-11-4015-2018, 2018.

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Jackson, R. C., McFarquhar, G. M., Fridlind, A. M., and Atlas, R.: The dependence of cirrus gamma size distributions expressed as volumes in N0-λ-μ phase space and bulk cloud properties on environmental conditions: Results from the Small Ice Particles in Cirrus Experiment (SPARTICUS), J. Geophys. Res. Atmos., 120, 10,351–10,377, doi:10.1002/2015JD023492, 2015

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