

AUTHOR RESPONSES TO REFEREE #1 COMMENTS

We thank the referee for his/her review of this manuscript, and for constructive comments that have significantly improved this paper. We understand that this is a serious undertaking that requires considerable time and effort, and your efforts are appreciated!

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In the pdf supplement, black font is used for the referee comments (RC) and blue font is used for author comments (AC) and new text added to the paper.

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Following our responses is the MS Word version of our revised manuscript with all changes indicated via "Track Changes".

General comments:

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RC G1. A major concern with the current manuscript is its length. The authors cover a very wide range of information and results that are all interesting but their amount is, in my opinion, counterproductive. It should be kept in mind that this study will be of high interest to diverse communities, as it contains are interesting aspects for remote sensing, in situ measurements, modeling or ice microphysics. Based on the current version I suspect that readers will only focus on their section of interest and miss important results. One idea could be to shorten section 2, since a big part of it has already been described in Mitchell et al. [2016]. The results related to the $N(D)_{1=0}$ analyses are very important and should be mentioned in the paper but several of them could be moved to supplementary materials in order to lighten the discussions and the figures. Other analyses that are not focused on N (the novelty of the paper), such as section 5.4, could also be moved to supplementary materials. This would allow to remove some technical discussions and focus the paper on result analyses, which are also better suited for a publication in ACP.

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AC G1. Both referees commented on the paper's length being too long, although the content was appropriate. To remedy this problem, we moved 11 of the original figures and two new figures to a new file titled "Supplementary Materials". Some of the original text was also moved to Supplementary Materials. This shortened the paper's length by about 8 pages.

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RC G2. The authors acknowledge in the manuscript that this new retrieval method is not expected to provide accurate absolute values of N. Only the spatial variability of this parameter is well represented. The accuracy of the absolute N is discussed throughout sections 3 and 4 but would it be possible to summarize a final estimate of the uncertainties on N in the conclusion? By this I mean not only the $\Delta N/N$ but also combining what has been learned from the in situ evaluation. For instance, would it be a factor of 2, 5, one order of magnitude? Also, in what conditions can optimal retrievals be expected? This type of information would be very useful to future users of this dataset, especially since this paper will serve as a reference.

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AC G2. In the 1st paragraph of the new Sect. 3.2 (previously Sect. 3.3), we write that ΔN is computed assuming a negligible error in the relationships.

5 We added the following sentence to clarify that: *“Errors in the relationships create additional systematic uncertainties, as discussed in Sect. 3.1.”*

As suggested by the reviewer, the following text is added in the 2nd paragraph of Summary and Conclusions:

10 *“Four formulations of the retrieval were used, based on either the SPARTICUS or TC4 field campaigns, with the smallest size-bin of the 2D-S probe either assumed valid ($N(D)_1$ unmodified) or by assuming $N(D)_1=0$. The SPARTICUS unmodified $N(D)_1$ assumption gives the highest N values while the TC4 $N(D)_1 = 0$ assumption yielded the lowest N values. The N predicted from these two formulations differed by a factor of two (see Fig. 5), thus defining a possible*
15 *systematic uncertainty in N . The random relative uncertainty, $\Delta N/N$, associated to a N retrieval is most of the time < 0.5 for small crystals ($D_e < 35\text{--}50\text{ }\mu\text{m}$), but increases up to more than 2 at the sensitivity limit”.*

20 In addition, the definition of β_{eff} is now repeated at the beginning of the conclusion and the sensitivity ranges of the retrievals is summarized at the end of the 1st paragraph of the conclusion as:

“Perhaps the most unique aspect of this retrieval method is its sensitivity to small ice crystals via β_{eff} . The sensitivity ranges ($N/\text{IWC} > \sim 10^7\text{ g}^{-1}$ & $D_e < 90\text{--}110\text{ }\mu\text{m}$) are usually compatible with cirrus clouds ($T < -38\text{ }^\circ\text{C}$) since PSDs tend to be narrower at these temperatures, containing
25 *relatively small ice particles.”*

Finally, we note at the end of the 4th paragraph under Summary and Conclusions: “When the sample selection criteria were relaxed to accept samples having cloud OD > 0.1 (instead of OD > 0.3), median N was reduced by a factor of 1.5 on average while $\Delta N/N$ was more than doubled.”
30 This is also mentioned earlier in the paper at the end of Sect. 5.2.

RC G3. A clear limitation of the method is its necessity to filter out a lot of data based on different cloud conditions. This, as shown in table 4, leads to samples that are only
35 representative of up to a 2% frequency of occurrence. Is there a particular reason for not using more data?

AC G3. The rationale for selecting the relevant cloudy scenes for this study is presented in Sect. 2.2.1. We tried to clarify, and Sect. 2.2.1 now reads (changes are in bold):

“Because IIR is a passive instrument, meaningful retrievals are possible for well identified scenes. This study is restricted to the cases where the atmospheric column contains one cirrus cloud layer. We also insure that the background radiance is only due to the surface (see Eq. (3)) allowing a more accurate computation than for cloudy scenes. The retrievals were applied only to single-layered semi-transparent cirrus clouds that do not fully attenuate the CALIOP laser beam, so that the cloud base is detected by the lidar. The cloud base is in the troposphere and its temperature is required to be colder than -38°C (235 K) to ensure that the cloud is entirely composed of ice. This is likely to exclude liquid-origin cirrus clouds from our data set (Luebke et al., 2016). When the column contains also a dense water cloud, the background radiance can be computed assuming that the water cloud is a blackbody. However, because systematic biases were made evident (Garnier et al., 2012), we chose to discard these cases, which reduces the number of selected samples by about 25 %. Because the relative uncertainties in τ_{obs} and in β_{eff} increase very rapidly as cloud emissivity decreases (Garnier et al., 2013), the lidar layer-integrated attenuated backscatter (IAB) was chosen greater than 0.01 sr^{-1} to avoid very large uncertainties at the smallest visible optical depths (ODs). This resulted in an OD range of about 0.3 to 3.0. Similarly, clouds for which the radiative contrast $R_{\text{BG}} - R_{\text{BB}}$ between the surface and the cloud is less than 20 K in brightness temperature units are discarded. IIR observations must be of good quality according to the quality flag reported in the IIR Level 2 product (Vaughan et al., 2017).”

Frequency of occurrence could be increased by relaxing some selection criteria, but the difficulty is that the additional information could be obscured by large additional uncertainties. Following reviewer #2’s suggestion, we reprocessed the data set to include clouds of OD between 0.1 and 0.3. This increased the frequency of occurrence by a factor 2 to 3 (as now shown in Table 4), but this also increased the relative uncertainty in IIR β_{eff} and in CALIPSO N. As noted under AC G2, we have reported how this affects N.

The method seems to be rather straightforward to apply on CALIPSO satellite products, and some of the co-authors should be familiar with such operational treatments. It therefore shouldn't take long to process 10 years of data and greatly improve the statistical significance of the results shown in this study (especially in sections 5 and 6).

The first paragraph of Sect. 2.2 has been shortened and modified to clarify that IIR β_{eff} is based on the CALIPSO IIR Version 3 Level 2 operational product, but that the various improvements implemented for this study required to reprocess the dataset. Processing of other years will be interesting for instance to examine inter-annual variability, and will be carried out with the new version 4 of the operational products. The end of the first paragraph of Sect. 2.2 now reads:

“IIR effective emissivity was reprocessed for this study to reduce possible biases, as described in the following sub-sections. [.....]. These improvements will be implemented in the next version 4 of the IIR Level 2 products. This is why we only focus in this study on a limited dataset

as a proof-of-concept. We will further develop a more statistically relevant analysis using the version 4 products.”

RC G4. Concerning the results shown in section 4 (comparisons to in situ measurements), more direct comparisons between SPARTICUS measurements and CALIPSO could be obtained by looking at the numerous co-incident overpasses between the satellite and the aircraft. This would avoid issues related to non-representative events in the in situ data by comparison to a long-term regional dataset, as noted in page 22. Have you tried looking at these exact satellite overpasses only?

AC G4. Yes, but unfortunately, we found that spatially and temporally coincident measurements are very rare for scenes meeting the IIR cloud selection criteria. We added the following sentence at the end of the 1st paragraph in Section 4.1:

“Data analysis is performed on a statistical basis, as coincident in situ and satellite data only provide a very small dataset due to our data selection.”

RC G5. Perhaps I have missed this information but I am under the impression that the D_e retrieved by IIR is not used in this method. How different would be the results if this operational retrieval was used instead of the in situ relationship? Is there a reason for not doing this?

AC G5. The fundamental parameter retrieved from IIR is β_{eff} . In the CALIPSO retrieval equation of N (Eq. (8), Sect. 2.5), β_{eff} is related to N/IWC , D_e , and $2/Q_{\text{abs,eff}}$ using regression curves derived from aircraft PSD measurements (Sect. 2.3 and Sect. 2.4). Indeed, the three relationships used in Eq. (8) must be derived in a consistent manner from the same in situ PSDs.

IIR D_e reported in the Version 3 data products is derived using the $\beta_{\text{eff}}-D_e$ relationships presented in Garnier et al. (2013) computed using different optical properties, with no size distribution, and by using both the 12.05/10.6 and 12.05/08.65 pairs of channels. Because the technique and the assumptions are different than in this study, we could not use D_e from the Version 3 product for this study. Furthermore, IIR data have been reprocessed for this study in order to implement the improvement described in Sect. 2.2.2 and 2.2.3, yielding different β_{eff} in this study than in the Version 3 products, especially over land.

We removed the comments about IIR D_e in Sect. 2.2 to avoid confusion and added the following text to the end of the 2nd paragraph of Sect. 2.4:

“It is noted that the D_e - β_{eff} relationships used in the IIR Version 3 operational algorithm (Fig. 3a in Garnier et al., 2013) tend to yield smaller D_e than the relationships derived from these PSD measurements, largely because they were computed with no size distribution.”

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Specific comments:

RC S1. Section 2.2: Computing IIR radiances, or converting between optical properties, requires some assumptions on the shape of ice crystals and of the ice particle size distribution.

10 Dubuisson et al. [2008] for instance showed that the IIR brightness temperatures are sensitive to both parameters. Garnier et al. [2012] showed that converting the IIR effective optical depths into absorption optical depths can also depend on ice particle shape assumptions. Do you have an idea if these assumptions have an impact on your N retrievals? Are they accounted for in the uncertainties described in the appendix?

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AC S1. We clarified the presentation of IIR β_{eff} at the beginning of Sect. 2.2, which now reads:

“We use the absorption optical depth $\tau_{abs}(12.05 \mu m)$ and $\tau_{abs}(10.6 \mu m)$ retrieved from the effective emissivity in CALIPSO IIR channels 12.05 μm and 10.6 μm . The retrieved optical depths are not purely due to absorption, but also include the effects of scattering. Thus, their ratio is not exactly β , but the effective β or β_{eff} written as

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$$\beta_{eff} = \tau_{abs}(12.05 \mu m) / \tau_{abs}(10.6 \mu m) .” \quad (1)”$$

The weak dependence of the retrieval on ice particle shape is described in a new paragraph at the end of Sect. 2.3:

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“Cirrus cloud emissivity and τ_{abs} depend on ice particle shape (Mitchell et al., 1996a; Dubuisson et al. 2008). However, this retrieval should not be very sensitive to ice particle shape for several reasons, one being that β_{eff} is directly retrieved from cloud radiances as per (2) and (3). Another reason is that no ice particle shape assumptions are made when calculating β_{eff} from in situ measurements with the exception of the absorption contribution from tunneling (which was not sensitive to realistic shape changes, as described above). That is, the 2D-S probe in situ data include measurements and estimates for ice particle projected area and mass, respectively. MADA optical properties are calculated directly from these in situ area and mass values, thus largely avoiding the need for shape assumptions. Thirdly, this retrieval is most sensitive to the smaller ice particles in a PSD where the variance in ice particle shape is minimal (Baker and Lawson, 2006b; Lawson et al., 2006b; Woods et al., 2018). During the SPARTICUS campaign, many cirrus clouds were sampled so that biases in ice particle shape due to a specific cloud condition are less likely to occur.”

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The uncertainties in N described in the appendix are random errors computed assuming no error in the relationships. Errors in the relationships create additional systematic uncertainty (see AC G2), but the dependence on ice crystal shapes should be very weak, as stated above.

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Also, are the ice crystal shapes used in the MADA method to compute the in situ relationships consistent with the shapes assumed in the treatment of CALIPSO measurements, and if not is there an impact on the N retrieval and their uncertainties?

10 CALIPSO IIR β_{eff} is computed directly from the effective emissivity at 12.05 μm and 10.6 μm . This computation does not involve any assumption about the shapes of ice crystals. As such, CALIPSO IIR β_{eff} can be seen as a direct observation. CALIPSO N is then retrieved from CALIPSO IIR β_{eff} and the in situ relationships presented in this manuscript.

15 RC S2. p. 6 l. 22: By single-layered cloud do you refer to the absence of other ice clouds or also to the absence of a liquid or mixed-phase layer underneath? IIR seems to be able to deal quite well with multi (ice+liquid) layer conditions by adjusting the background radiance. For instance Fig. 10 in Sourdeval et al. [2016] shows that IIR is robust to multi-layer conditions. It would be worth checking if removing this filter makes a big difference in your dataset. Otherwise, not
20 excluding multi-layer scenes would clearly help to increase the global statistical representativity of N retrievals.

AC S2. The rationale for our cloud selection presented in Sect. 2.2.1 has been modified (see AC G3).

25 We confirm that the retrievals are applied when a cirrus cloud is the only cloud layer in the atmospheric column, that is when the background radiance is from the surface. We could have also included scenes for which the cirrus cloud layer is over a low opaque water cloud that fully attenuates the CALIOP laser beam. Indeed, in this second case, the background radiance can be computed assuming that the low water cloud behaves as a blackbody emitting
30 at a temperature inferred from the water cloud altitude and GMAO GEOS temperature profiles. The expected accuracy in this second case was evaluated in Garnier et al. (2012) through comparisons of observed and computed blackbody brightness temperatures of low opaque warm clouds. The observations were colder than the blackbody computations by up to 9 K (their Fig. 10), suggesting significant biases in the cloud temperature inferred from the GMAO
35 GEOS profiles in case of strong temperature inversions at the top of the cloud. We chose to discard these scenes for this study, which reduces the number of samples by about 25 %. These biases will be re-evaluated with the future version 4 of the IIR products which will use MERRA 2 profiles data.

RC S3. Sec. 2.4: Can error bars be added to the points in Fig. 2-4? It seems like these instrumental and computational errors could easily encompass the changes due to the choice of including the first size bin or not.

AC S3. The data points in Figs. 2-4 come from in situ PSD measurements. There is only one PSD per point, and therefore it is not possible to assign an error bar from a statistical basis. One could in principle propagate errors associated with the various stages of instrument data processing (which perhaps is what the referee is referring to), but uncertainties for these data processing stages have not been characterized in most cases. For example, in “Laboratory and Flight Tests of 2D Imaging Probes: Toward a Better Understanding of Instrument Performance and the Impact on Archived Data” by Gurganus and Lawson (2018, JTech), the 2D-S and the CIP probes are intercompared in various ways, but uncertainties are not estimated that could be applied to address this request. This paper has now been referenced to give the reader additional knowledge of 2D-S performance characteristics. As stated at the end of this paper, “The research reported here does not offer quantitative corrections for the instrument errors that have been uncovered. Instead, it describes conditions under which the uncertainties can exist and provides some qualitative estimates of magnitude. Corrections to the data will require more laboratory tests with upgraded facilities, flight tests, improved forward models of the instruments and numerical simulations.”

RC S4. Fig. 9: Could you comment on the very wide spread (75-25% percentiles) in the Krämer et al N/IWC vs T_c figure by comparison to what is noted for the CALIPSO data? Is there no spread plotted for the Krämer dataset on the right plots or is it too small to be seen? Also please indicate in the caption if the CALIPSO retrievals correspond to the entire 2008 and 2013 periods.

AC S4. To shorten the paper, this figure has been moved to Supplementary Materials and is Fig. S6. The caption states that “in situ measurements from Krämer et al. (2009)” are “shown by the grey curves; top and bottom being minimum and maximum values and middle grey solid curve being the middle value.” Thus, these dashed Kramer curves are not the 25 and 75 percentile curves, but rather indicate the max/min values. To reduce confusion, we have identified curves corresponding to the four formulations as colored. We have indicated that percentiles corresponding to the 4 formulations are not shown in the right panel, and that the retrievals shown are from 2013.

The caption now reads: “Left: Comparisons of the median CALIPSO IIR N/IWC (g^{-1}) for the four formulations (colored) with in situ measurements from Krämer et al. (2009) shown by the grey curves; top and bottom being minimum and maximum values and middle grey solid curve being the middle value. Colored solid curves are median values while dashed curves indicate the 25th and 75th percentile values. Right: Comparisons of CALIPSO IIR β_{eff} shown by the black curves (solid curve gives the median value while dashed curves indicate the 25th and 75th percentile

values) with the four (colored) in situ β_{eff} inferred from in situ N/IWC (from Krämer et al. (2009) using the four formulations. Corresponding percentiles are not shown. The navy and light blue curves correspond to the SPARTICUS formulations for the unmodified $N(D)_1$ assumption and the $N(D)_1 = 0$ assumption, respectively. The red and orange curves are using the TC4 formulations for the $N(D)_1$ unmodified and $N(D)_1 = 0$ assumptions, respectively. The CALIPSO IIR retrievals are from 2013 and are for the approximate latitude range (25 °S to 70 °N) of the in situ data, over oceans (top) and over land (bottom).”

RC S5. Fig. 10e: As a remark, the slightly negative relation between N and T (N decreasing towards low temperature) indicated in the Kramer et al. [2009] study is not found anymore in revised version of the dataset (not yet published but seen in recent conference presentations by M. Krämer et al). This should strengthen the statement p. 31 l. 4 that differences are likely to be due to different cloud sampling.

AC S5. We cannot comment on this since the data is not yet published.

RC S6. Sec. 5.4: Is there an explanation to the fact that Re has a very different dependence on T_c and $T_c - T_{\text{top}}$ by comparison to what was previously shown for N ?

AC S6. In the new Fig. 10 (previously Fig. 13), there is a tendency for N to decrease from left-to-right as a function of $T_c - T_{\text{top}}$. In the new Fig. S12 (previously Fig. 16), D_e tends to decrease with decreasing T_c but remains quasi-constant (or less variable) for a given T_c (i.e. less sensitive to $T_c - T_{\text{top}}$). D_e is proportional to IWC/A_{PSD} (as calculated for the in situ data and thus implemented in the retrieval regressions), where IWC = ice water content and A_{PSD} = projected area of the ice particle size distribution (PSD). For a gamma function PSD of the form $N(D) = N_0 D^v \exp(-\lambda D)$ and representing ice particle projected area as $A = \gamma D^\sigma$ and mass as $m = \alpha D^\beta$, $A_{\text{PSD}} = \gamma \Gamma(\sigma + v + 1) N / \Gamma(v + 1) \lambda^\sigma$ and $IWC = \alpha \Gamma(\beta + v + 1) N / \Gamma(v + 1) \lambda^\beta$ where Γ denotes the gamma function. The IWC/A_{PSD} ratio now shows that D_e is independent of N , and D_e is proportional to $\lambda^{\sigma-\beta}$ where λ = PSD slope parameter. The difference $\sigma - \beta$ is slightly more than -1 (Mitchell 1996, JAS), showing that D_e is strongly related to the PSD mean size [note that $\lambda = (v + 1)/D_{\text{mean}}$, where D_{mean} = PSD mean maximum dimension]. The decrease in D_e with decreasing T_c is likely due to a decrease in IWC with decreasing T_c .

New text has been added to the end of the last paragraph in Sect. 5.4 to express these points:

“This weaker dependence on $T_c - T_{\text{top}}$ relative to N is expected since D_e is proportional to IWC/A_{PSD} . For a gamma function PSD and assuming power law relationships for ice particle area and mass, D_e is proportional to $\lambda^{\sigma-\beta}$ where λ = PSD slope parameter and σ and β are power law exponents for area and mass, respectively. The difference $\sigma - \beta$ is slightly more than -1, showing that D_e is strongly related to the PSD mean size (inversely proportional to λ). The derivation also shows that D_e is not dependent on N . The decrease in D_e with decreasing T_c is likely due to a decrease in IWC with decreasing T_c .”

RC S7. Section 6.4: p. 45 l. 11-13: Another possible explanation to the differences in absolute numbers could be that DARDAR-LIM ignores the concentrations of ice particles smaller than 5 μm . p. 45 l. 14-16: As mentioned before, the slight decrease of N towards low T as noted by Krämer et al. [2009] is not found in the most recent version of their dataset, which is not anymore inconsistent with the relation shown in Gryspeerdt et al. [2018]. An increase of N towards low T is also consistent with what is shown in Figures 10ab of this manuscript. Overall, comparisons between the N retrievals presented in this study and DARDAR-LIM are very difficult as both methods are based on different approaches and difference instruments. DARDAR-LIM also retrieves vertical profiles of N whereas IIR retrievals correspond to weighted N average values from cloud top. Nevertheless, it is quite remarkable that despite all these differences the two dataset show such similar results. This clearly strengthens the confidence in both satellite products.

AC S7. Section 6.4 has been changed significantly in view of the referee's suggestions. We agree with the referee that absolute differences in N between the DARDAR-LIM and our CALIPSO IIR retrieval may be partly attributed to the fact that DARDAR-LIM (as per the ACP paper) does not account for ice crystals $< 5 \mu\text{m}$ in maximum dimension D. Fortunately, there are DARDAR-LIM results posted on the ACP website as part of the review process for the DARDAR-LIM paper that show N for $D > 1 \mu\text{m}$. We have cited this work to show that using a $D > 1 \mu\text{m}$ cutoff increases DARDAR-LIM N by a factor of about 1.7, making the CALIPSO IIR N retrievals in general agreement with the DARDAR-LIM N values. New text has been added accordingly:

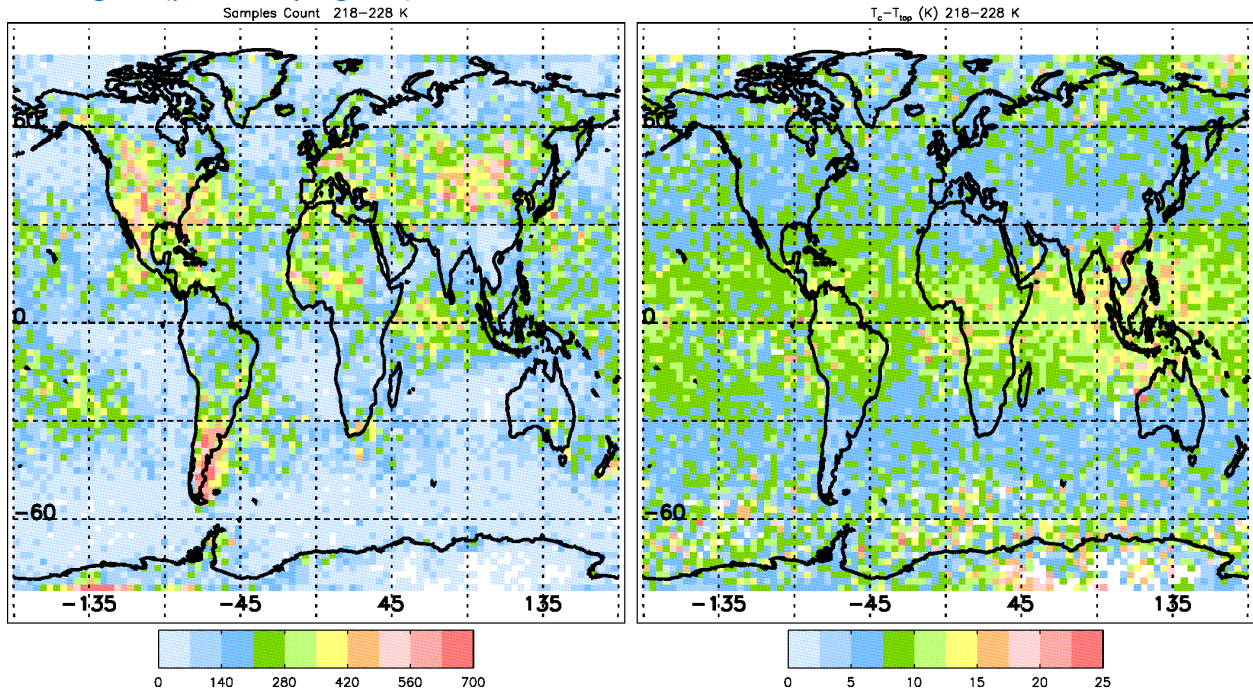
"The highest N values reported in Sourdeval et al. (2018a) and Gryspeerdt et al. (2018) are for ice particles larger than 5 μm . These values would be higher by a factor of about 1.7 if all ice particles larger than 1 μm were considered (Sourdeval et al., 2018b). This is important to note since the CALIPSO IIR retrieval has no cut-off size, and it is very sensitive to ice crystals in the 1 to 5 μm range (Fig. 1). Keeping this in mind, the CALIPSO N retrievals are comparable in value to the DARDAR-LIM N retrievals."

Regarding the temperature dependence of N, although we cannot cite unpublished work, we appreciate this information and have removed the discussion about the temperature dependence of in situ N measurements (in relation to retrievals).

We agree that there is good qualitative agreement between our CALIPSO IIR and the DARDAR-LIM retrievals, and that this "strengthens the confidence in both satellite products."

RC S8. Fig. 17: It would be interesting to see the spatial distribution of the frequency of occurrence of retrievals and of the distance from cloud top (in terms of temperature) corresponding to this figure.

AC S8. Maps of the number of samples (left) and of Median $T_c - T_{top}$ (right) associated with the new Fig. 12 (previously Fig. 17), with T_c between 218 K and 228 K, are shown below:



- 5 These figures have been added to the Supplementary Materials section of the manuscript as Fig. S13. New text has been added in the 1st paragraph in Sect. 6.4:

“The geographical distributions of the number of samples and of median $T_c - T_{top}$ corresponding to Fig. 12 are shown in Fig. S13 under Supplementary Materials. Median $T_c - T_{top}$ is up to 20 °C and is smaller at mid- to high latitudes than in the tropics.”

The 2nd paragraph in Sect. 6.4 starts now as: “Results in Fig. 12 can be compared with the retrieved N values near cloud top at 223 K shown in Fig.1 of Gryspeerd et al. (2018).”

- 15 The reviewer’s question led us to comment briefly on the typical relationship between $T_c - T_{top}$ and $T_{base} - T_{top}$. Thus, isolines of $T_{base} - T_{top}$ have been added to Fig. 10 and Fig. S11, and the text has been modified accordingly in Sect. 5.3 (changes in bold):

“Our retrievals are now examined against both T_c and $T_c - T_{top}$ to estimate the impact of the distance from cloud top. **These** N retrievals are shown in Fig. 10 using the SPARTICUS $N(D)_1$ unmodified assumption in the tropics (0-30°) and at mid- (30-60°) and high (60-82°) latitudes in the winter and summer seasons (using both hemispheres) by distinguishing retrievals over oceans and over land. The associated number of samples is given in Fig. S11. **Added to these figures are isolines of temperature differences between cloud base and cloud top ($T_{base} - T_{top}$).** **For most of the layers, $T_c - T_{top}$ represents 30 to 70% of $T_{base} - T_{top}$ (Fig. S11). Fig. 10 shows a**

strong dependence of N on $T_c - T_{top}$, with large N ($> 500 \text{ L}^{-1}$) seen near the top of the geometrically thin clouds ($T_{base} - T_{top} < 15 \text{ }^\circ\text{C}$), when $T_c - T_{top}$ is smaller than about $5 \text{ }^\circ\text{C}$."

5 RC S9. p. 49 l. 9-10: The "four formulations" have not yet been mentioned in the conclusion. It would be useful to briefly describe again in what they differ.

AC S9. The "four formulations" of the retrieval are now mentioned under "Summary and Conclusions" as follows:

10 *"Four formulations of the retrieval were used, based on either the SPARTICUS or TC4 field campaigns, with the smallest size-bin of the 2D-S probe either assumed valid ($N(D)_1$ unmodified) or by assuming $N(D)_1=0$. The SPARTICUS unmodified $N(D)_1$ assumption gives the highest N values while the TC4 $N(D)_1 = 0$ assumption yielded the lowest N values. The N predicted from these two formulations differed by a factor of two (see Fig. 5), thus defining a possible systematic uncertainty in N ."*

15 Technical corrections:

1. p. 2 l. 13 and 16: it would be better to explicitly refer to "ice clouds" instead of "clouds"

Done

2. p. 11 l. 17: "limit is 1.031"

20 Done

3. p. 43 l. 15: " $\text{m.s}^{-1} \text{ in}$ "

Text removed

AUTHOR RESPONSES TO REFEREE #2 COMMENTS

We thank the referee for his/her comments of this paper, and for constructive comments that have significantly improved this paper. We understand that this is a serious undertaking that requires considerable time and effort, and your efforts are appreciated!

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In the text below, black font is used for the referee comments (RC) and blue font is used for author comments (AC) and new text added to the paper.

10 Following our responses is the MS Word version of our revised manuscript with all changes indicated via "Track Changes".

General comments:

15 RC The manuscript is quite long. If this is an issue, one could consider splitting up the retrieval/radiative aspects and the interpretation of the results.

20 AC We appreciate the referee's recognition of the substantial research effort that produced this manuscript. In regards to the manuscript length, we created another file titled Supplementary Materials that contains 12 figures and some of the text from the original manuscript, which shortened the paper by 8 pages.

Specific comments:

25 RC S1. Section 2.2.0. How do you handle the lapse through the cloud. Cirrus can be extensive in their vertical dimension. Are you errors larger for geometrically thick cloud?

30 AC S1. In Eq. (3), the lapse through the cloud is handled in the determination of the cloud blackbody radiance, R_{BB} . The associated blackbody temperature, T_{BB} , is determined using the approach detailed in Sect. 3 of Garnier et al. (2015) and summarized in Sect. 2.2.3 of this paper. First, we compute the IIR weighting profile in the cloud layer using the CALIOP extinction profile. The IIR weighting profile includes an attenuation term corresponding to the overlying infrared absorption optical depth (see Eq. (11) and Eq. (12) of Garnier et al. (2015)). Then, R_{BB} is the weighted averaged blackbody radiance computed using this IIR weighting profile and the GMAO temperature profile in the layer. We actually forgot to precise that the GMAO
35 temperature profile is used, and the 3rd sentence in Sect. 2.2.3 now reads (changes in bold):

40 *"The CALIOP lidar 532 nm extinction profile in the cloud is used to determine an IIR weighting profile that is used, **together with the GMAO GEOS5 temperature profile**, to compute R_{BB} as the weighted averaged blackbody radiance."*

RC S2. Section 2.2.0: You mentioned you used the 0.55 micron extinction derived mean cloud temperature. I would expect that the cloud weighting functions are a bit different for the 10.6 and 12 micron observations and these may also differ from each other. Does this matter? Or this effect absorbed in the Blackbody radiance calculation in 2.2.3?

AC S2. For this study, T_{BB} is taken identical at 10.6 and 12 μm , following the same approach as in the Version 3 algorithm, but is improved compared to Version 3 by using the corrections detailed in Garnier et al. (2015) and summarized in Sect 2.2.3. Thus, T_{BB} is computed at 12 μm by taking the ratio, r , between visible extinction optical depth and absorption optical depth at 12 μm , $\tau_{\text{abs}}(12.05\mu\text{m})$, equal to 2.

We agree that T_{BB} could have been estimated separately at 12 μm (noted $T_{BB,12}$) and at 10.6 μm (noted $T_{BB,10}$). Furthermore, taking the same ratio, r , for both channels means taking $\tau_{\text{abs}}(12.05\mu\text{m}) = \tau_{\text{abs}}(10.6\mu\text{m})$ or $\beta_{\text{eff}}=1$, which is not consistent with our findings. In order to assess the error resulting from our simplified approach, we re-computed $T_{BB,12}(r_{12})$ and $T_{BB,10}(r_{10})$, with r_{12} and r_{10} not taken equal to 2, but computed respectively using $\tau_{\text{abs}}(12.05\mu\text{m})$ and $\tau_{\text{abs}}(10.6\mu\text{m})$ initially reported in the operational Version 3 products. The analysis was conducted over oceans in JJA 2013 between 82°S and 82°N.

- We find that the difference $T_{BB,12}(r_{12}) - T_{BB,12}(r_{12}=2)$ (see Table 1 below) is smaller than 0.09 K on average with a mean absolute deviation smaller than 0.13 K. The resulting error is negligible compared to the assumed uncertainty of $\pm 2\text{K}$ in $T_{BB,12}(r_{12}=2)$.
- We find that the difference $T_{BB,10}(r_{10}) - T_{BB,12}(r_{12})$ (see Table 2 below) is smaller than 0.12 K on average with a mean absolute deviation smaller than 0.07 K.
- Using $T_{BB,10}(r_{10})$ and $T_{BB,12}(r_{12})$ instead of the same temperature $T_{BB}=T_{BB,12}(r_{12}=2)$ as in this study reduces β_{eff} by less than 0.001 on average, with a mean absolute deviation smaller than 0.0007, which is negligible (see Table 3).

We added the following sentence at the end of the 1st paragraph in Sect. 2.2.3:

“Computing T_{BB} at 10.6 μm and at 12.05 μm yields temperatures that differ by less than 0.15 K on average, which has a negligible impact on β_{eff} for our cloud selection.”

Table 1: Analysis of the difference $T_{BB,12}(r_{12}) - T_{BB,12}(r_{12}=2)$. Oceans, JJA 2013, 82°S-82°N.

Temperature T_c (K)	Samples Count	Min	Max	Median	Mean	Standard deviation	Mean absolute deviation
188	73	0.012	0.082	0.063	0.061	0.017	0.013
192	348	-0.038	0.501	0.056	0.071	0.066	0.044
196	2149	-0.685	0.486	0.076	0.087	0.085	0.056
200	6915	-0.719	1.879	0.083	0.084	0.129	0.081

204	16783	-1.098	3.048	0.063	0.061	0.160	0.100
208	28620	-1.246	2.181	0.048	0.043	0.182	0.118
212	39959	-3.193	3.299	0.022	0.011	0.199	0.123
216	45591	-1.374	3.680	0.004	-0.017	0.199	0.127
220	41081	-1.606	5.351	-0.015	-0.040	0.202	0.123
224	29724	-0.878	1.984	-0.021	-0.044	0.160	0.102
228	10377	-0.644	1.664	-0.018	-0.026	0.124	0.074
232	1084	-0.303	1.087	-0.001	-0.000	0.088	0.040

Table 2: Analysis of the difference $T_{BB,10}(r_{10}) - T_{BB,12}(r_{12})$. Oceans, JJA 2013, 82°S-82°N.

Temperature T_c (K)	Samples Count	Min	Max	Median	Mean	Standard deviation	Mean absolute deviation
188	73	0.016	0.039	0.032	0.031	0.005	0.004
192	348	0.000	0.221	0.040	0.048	0.034	0.028
196	2149	-0.025	0.345	0.051	0.066	0.050	0.038
200	6915	-0.27	0.667	0.083	0.093	0.060	0.046
204	16783	-0.018	0.729	0.095	0.113	0.078	0.059
208	28620	-0.277	1.155	0.098	0.118	0.086	0.066
212	39959	-1.792	0.763	0.091	0.110	0.085	0.063
216	45591	-0.164	0.601	0.080	0.097	0.071	0.055
220	41081	-0.392	0.622	0.064	0.077	0.058	0.044
224	29724	-0.741	0.768	0.044	0.052	0.041	0.030
228	10377	-0.162	0.251	0.025	0.029	0.025	0.018
232	1084	-0.367	0.093	0.010	0.012	0.026	0.010

5

Table 3: Analysis of the difference between β_{eff} computed using $T_{BB,10}(r_{10})$ and $T_{BB,12}(r_{12})$ and β_{eff} from this study. Oceans, JJA 2013, 82°S-82°N.

Temperature T_c (K)	Samples Count	Min	Max	Median	Mean	Standard deviation	Mean absolute deviation
188	73	-0.0001	-0.0000	-0.0000	-0.0000	0.0000	0.0000
192	348	-0.0005	0.0000	-0.0001	-0.0001	0.0001	0.0001
196	2149	-0.0041	0.0001	-0.0001	-0.0002	0.0003	0.0002
200	6915	-0.0048	0.0013	-0.0003	-0.0004	0.0004	0.0003
204	16783	-0.0081	0.0026	-0.0005	-0.0006	0.0006	0.0005
208	28620	-0.0215	0.0080	-0.0006	-0.0008	0.0009	0.0006
212	39959	-0.0220	0.0551	-0.0007	-0.0009	0.001	0.0007
216	45591	-0.0159	0.0075	-0.0007	-0.001	0.001	0.0007
220	41081	-0.0311	0.0129	-0.0007	-0.001	0.001	0.0007
224	29724	-0.0338	0.0319	-0.0006	-0.0008	0.0009	0.0006
228	10377	-0.0109	0.0069	-0.0004	-0.0005	0.0007	0.0004
232	1084	-0.0029	0.0176	-0.0002	-0.0002	0.0008	0.0002

RC S3. Section 2.2.2: What does the bias look like between model and observations before correction?

AC S3. Probability density functions (PDFs) of the differences between clear sky observations and model (BTDoc) at 12.05 μm before (night: light blue; day: orange) and after (night: navy blue; day: red) correction are shown below for six latitude bands over ocean (left) and over land (right) in January 2008. This figure is now Fig. S1a under Supplementary Materials.

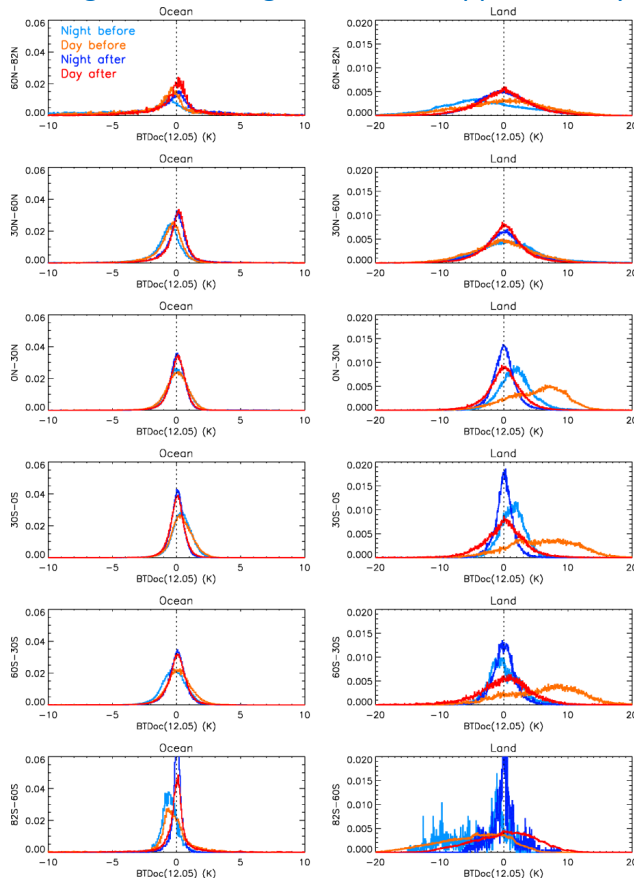


Figure S1a: Probability density functions of the differences between clear sky observations and computations of brightness temperature (BTDoc) at 12.05 μm , before (night: light blue; day: orange) and after (night: navy blue; day: red) correction are shown below for six latitude bands over ocean (left) and over land (right) in January 2008. Note the different scales over ocean and over land.

Similarly, PDFs of the clear sky inter-channel differences [BTDoc(10.6 μm)– BTDoc(12.05 μm)] are shown below, and the figure is now Fig. S1b under Supplementary Materials.

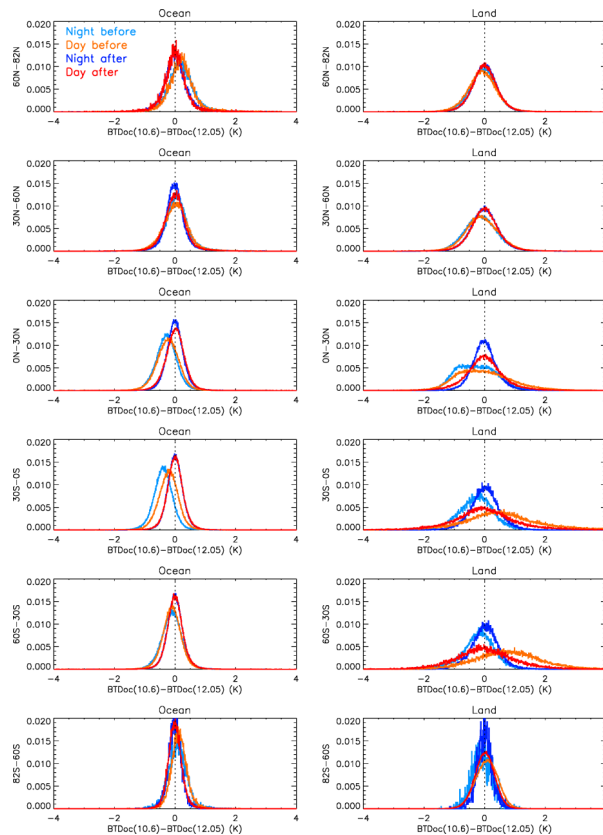


Figure S1b: Same as Fig. S1a, but for the inter-channel difference between observations and computations, $BTDoc(10.6 \mu m) - BTDoc(12.05 \mu m)$.

- 5 The following sentence has been added at the end of Sect. 2.2.2: “Figures S1a and S1b in Supplementary Materials show distributions of $BTDoc$ before and after correction”.

RC S4. Section 2.3: You reference Yang (2005) and make some mention of habits on page 10. Are allowing habit to be a free parameter or are discouraging people from using habits at all in prescribing properties from databases such as Yang’s? This relevant information to the remote sensing community.

AC S4. We have added a paragraph describing the retrieval’s relative insensitivity to ice particle shape at the end of Sect. 2.3. As described in this paragraph (shown below), there is no inferred “ice particle shape recipe” that the retrieval is based on. It would be difficult to apply a database such as Yang’s to in situ data since ideally one would need to know the habit composition of the ice particle size distribution (PSD), where habit varies with size across the PSD. Such information is difficult to extract from in situ probe measurements, although it should be possible using the Cloud Particle Imager (CPI) and suitable image analysis software.

20 The new paragraph reads as follows:

“Cirrus cloud emissivity and τ_{abs} depend on ice particle shape (Mitchell et al., 1996; Dubuisson et al. 2008). However, this retrieval should not be very sensitive to ice particle shape for several reasons, one being that β_{eff} is directly retrieved from cloud radiances as per (2) and (3). Another reason is that no ice particle shape assumptions are made when calculating β_{eff} from in situ measurements with the exception of the absorption contribution from tunneling (which was not sensitive to realistic shape changes, as described above). That is, the 2D-S probe in situ data include measurements and estimates for ice particle projected area and mass, respectively. MADA optical properties are calculated directly from these in situ area and mass values, thus largely avoiding the need for shape assumptions. Thirdly, this retrieval is most sensitive to the smaller ice particles in a PSD where the variance in ice particle shape is minimal (Baker and Lawson, 2006b; Lawson et al., 2006b; Woods et al., 2018). During the SPARTICUS campaign, many cirrus clouds were sampled so that biases in ice particle shape due to a specific cloud condition are less likely to occur.”

RC S5. Figs. 2 and 3 and so on: Why are computed from the TC4 campaign mostly concentrated in regions less than 1.1?

AC S5. As mentioned in Sect. 2.3, PSD sampling times were longer during TC4 (relative to SPARTICUS) with fewer sampling days, resulting in fewer PSD samples. Of these, only aged anvil cirrus sampled on one day by the WB57 were sampled at $T < -60^{\circ}\text{C}$ (Mitchell et al., 2011, JGR). TC4 cirrus sampled at warmer temperatures had substantially broader PSDs that yield lower β_{eff} values, typically < 1.10 . New text has been added to the second-to-last paragraph in Sect. 2.3:

“There are much fewer TC4 points in Figs. 2 and 3 for $\beta_{eff} > 1.1$ since the higher β_{eff} values were obtained only for $T < -60^{\circ}\text{C}$, which only occurred during a single flight.”

RC S6. Figs. 8a and 8b: In these figures, IIR median and in situ compare relatively well. Can the authors show an example scatterplot of the comparisons?

AC S6. Because spatially and temporally coincident measurements are very rare for cloudy scenes meeting the IIR cloud selection criteria, these figures show statistical analyses of in situ data on one hand, and of IIR data on the other hand, and are not one-to-one comparisons. Therefore, we don’t think that we can show a scatterplot of the comparisons. We added the following sentence at the end of the 1st paragraph in Section 4.1:

“Data analysis is performed on a statistical basis, as coincident in situ and satellite data only provide a very small dataset due to our data selection.”

RC S7: Section 5: The selection criteria resulted in less than 2% of qualified pixels. This makes me wonder if the selection requirement is relaxed to include more cirrus clouds, how much do the results in Figs. 11-16 change?

AC S7. The rationale for selecting the relevant cloudy scenes for this study is presented in Sect. 2.2.1. We tried to clarify, and Sect. 2.2.1 now reads (changes are in bold):

“Because IIR is a passive instrument, meaningful retrievals are possible for well identified scenes. This study is restricted to the cases where the atmospheric column contains one cirrus cloud layer. We also insure that the background radiance is only due to the surface (see Eq. (3)) allowing a more accurate computation than for cloudy scenes. The retrievals were applied only to single-layered semi-transparent cirrus clouds that do not fully attenuate the CALIOP laser beam, so that the cloud base is detected by the lidar. The cloud base is in the troposphere and its temperature is required to be colder than -38°C (235 K) to ensure that the cloud is entirely composed of ice. This is likely to exclude liquid-origin cirrus clouds from our data set (Luebke et al., 2016). When the column contains also a dense water cloud, the background radiance can be computed assuming that the water cloud is a blackbody. However, because systematic biases were made evident (Garnier et al., 2012), we chose to discard these cases, which reduces the number of selected samples by about 25 %. Because the relative uncertainties in τ_{abs} and in β_{eff} increase very rapidly as cloud emissivity decreases (Garnier et al., 2013), the lidar layer-integrated attenuated backscatter (IAB) was chosen greater than 0.01 sr^{-1} to avoid very large uncertainties at the smallest visible optical depths (ODs). This resulted in an OD range of about 0.3 to 3.0. Similarly, clouds for which the radiative contrast $R_{\text{BG}} - R_{\text{BB}}$ between the surface and the cloud is less than 20 K in brightness temperature units are discarded. IIR observations must be of good quality according to the quality flag reported in the IIR Level 2 product (Vaughan et al., 2017).”

Frequency of occurrence could indeed be increased by relaxing some selection criteria, but the difficulty is that the additional information could be obscured by large additional uncertainties. As an illustration, we reprocessed the dataset to include clouds of OD between 0.1 and 0.3. Fig. S5 under Supplementary Materials show the interrelationships between β_{eff} , α_{ext} , IWC, and N as well as $\Delta N/N$ for $\text{OD} > 0.1$, and is compared to Fig. S4 (former Fig. 7) obtained using the chosen threshold $\text{OD} > 0.3$. Figure S5 is copied below:

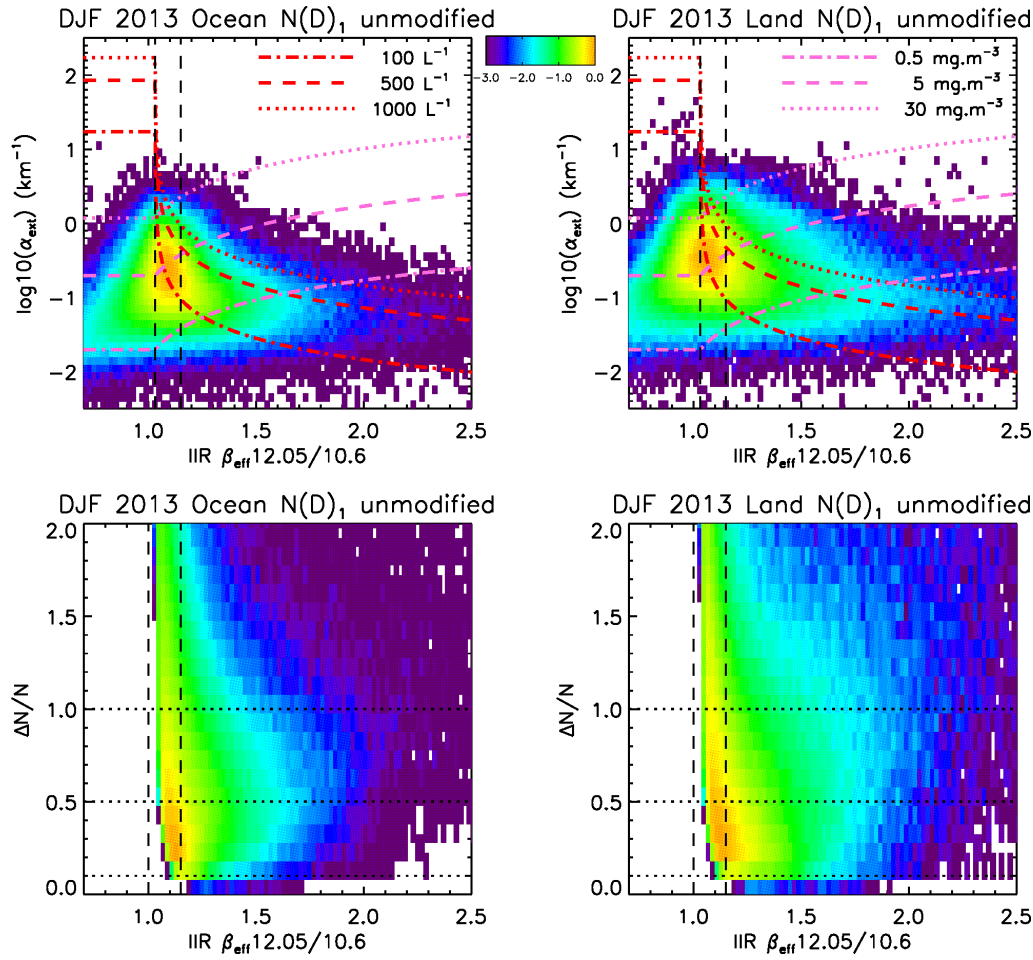


Figure S5: Same as Fig. S4 but sample selection criteria was changed to accept samples having $OD > 0.1$ approximately. Note the larger portion of samples having $\Delta N/N > 1$.

5 The following text has been added under Supplementary Materials:

“This same analysis was repeated in Fig. S5, except the sample selection criteria for minimum OD was changed from 0.3 to 0.1. This increased the sample population considerably. The larger dispersion in β_{eff} and in particular the larger portion of samples with β_{eff} much smaller than 1 (Fig. S5, top row) are due to large uncertainties at OD between 0.1 and 0.3, which also explain the larger portion of samples with $\Delta N/N > 1$ (Fig. S5, bottom row). More samples now correspond to lower values of α_{ext} (down to 0.016 km^{-1}), IWC , and N . “

And at the end of Sect. 3.2, we added:

“We repeated this analysis except using an OD threshold of 0.1 (instead of 0.3; see also Sect. 5.1). Figure S5 shows this same analysis except that the sample selection criteria for minimum OD was changed from 0.30 to 0.10. In the lower row relating $\Delta N/N$ to β_{eff} , the number of

samples having $\Delta N/N > 1.0$ has substantially increased over both ocean and land relative to Fig. S4 due to the lower OD threshold for sample selection, and more samples correspond to lower values of α_{ext} , IWC, and N.”

- 5 Table 4 in Sect. 5.1 has been updated to show frequency of occurrence when $OD > 0.1$. The following new text has been added at the end of the 3rd paragraph in Sect. 5.1:
“Relaxing the lower OD threshold to 0.1 instead of 0.3 would increase the number of samples by about a factor of 2.5.”

- 10 The equivalent of Fig. 9 (previous Fig. 12a) using a OD threshold of 0.1 instead of 0.3 is now Fig. S10 in the Supplementary Materials. This figure is shown below:

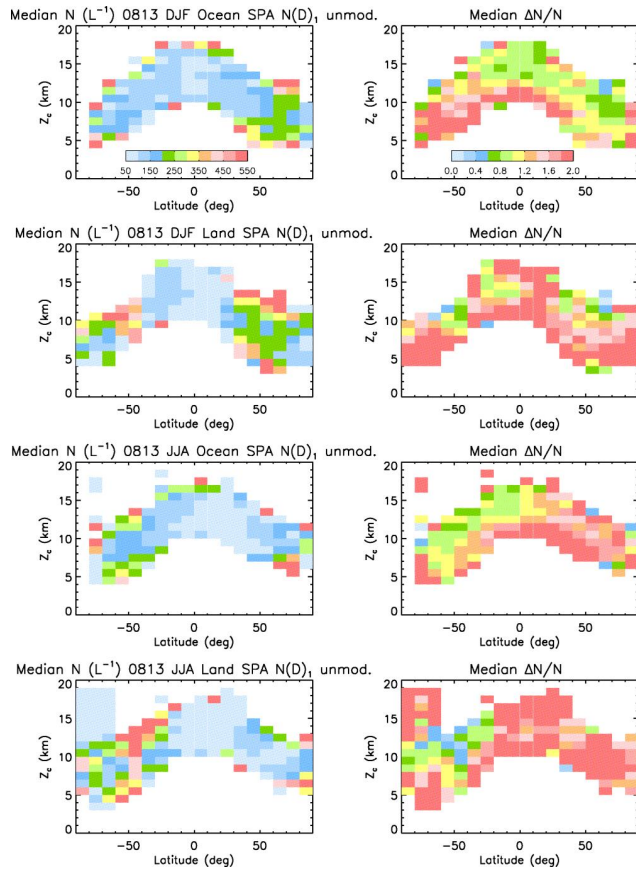


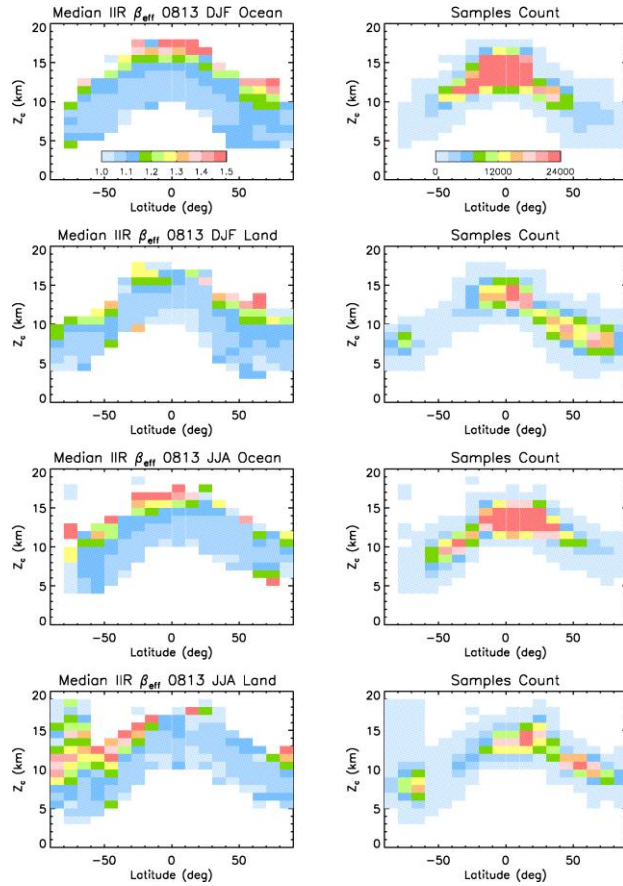
Fig. S10. Same as Fig. 9 in the main paper, but by relaxing the OD threshold to $OD > 0.1$.

- 15 This figure is now introduced at the end of Sect. 5.2 where the following text is added:

“Fig. S10 under Supplementary Materials, shows the same results as in Fig. 9. but by relaxing the OD threshold to $OD > 0.1$. Consistent with Fig. S5, median N is decreased, by a factor 1.5 on average, while $\Delta N/N$ is more than doubled.” A similar statement was added to Summary and

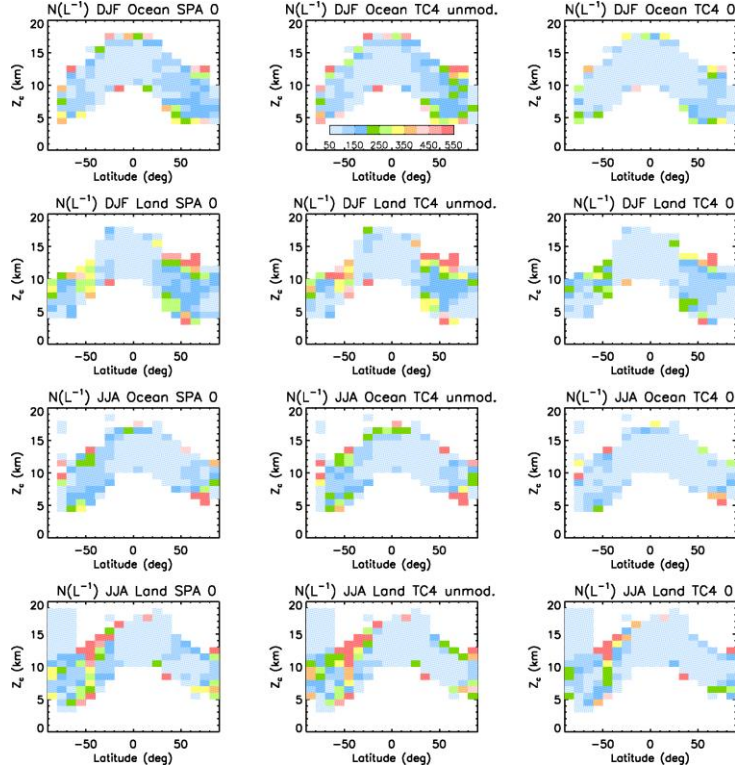
20 Conclusions at the end of the 4th paragraph.

As an illustration, but not included in the paper, below are the equivalent of previous Fig. 11 and Figs. 12b to 16 using an OD threshold of 0.1 instead of 0.3.

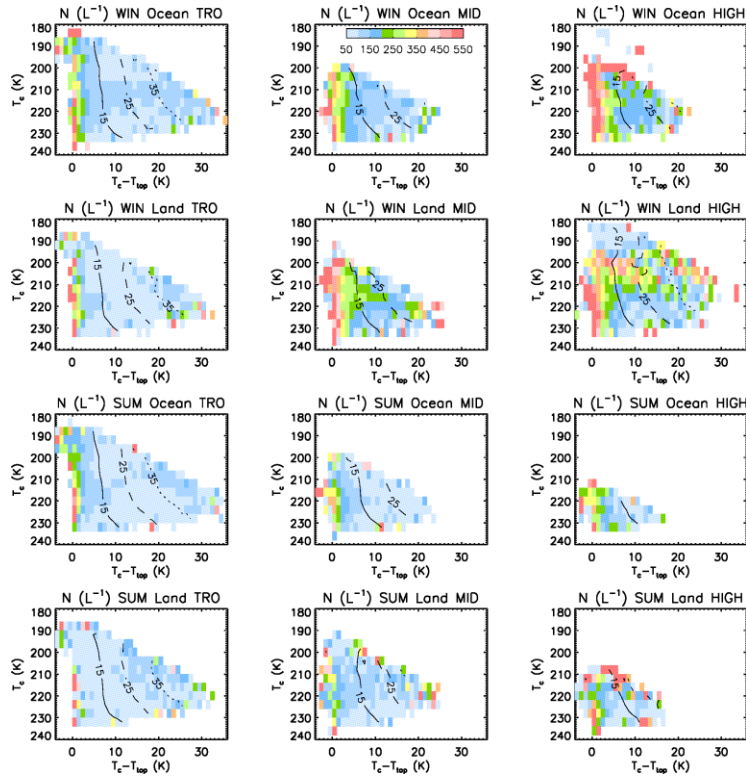


5

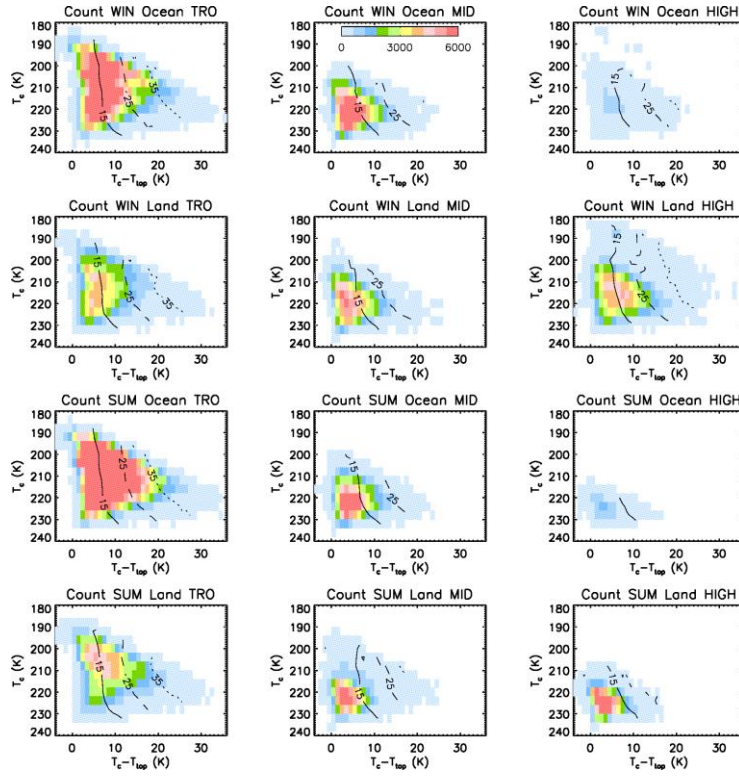
Same as Fig. 8 (previous Fig. 11), but with $\text{OD} > 0.1$. The OD threshold does not change much IIR β_{eff} over ocean. The changes are more notable over land, which could be due in part to larger additional errors than over ocean.



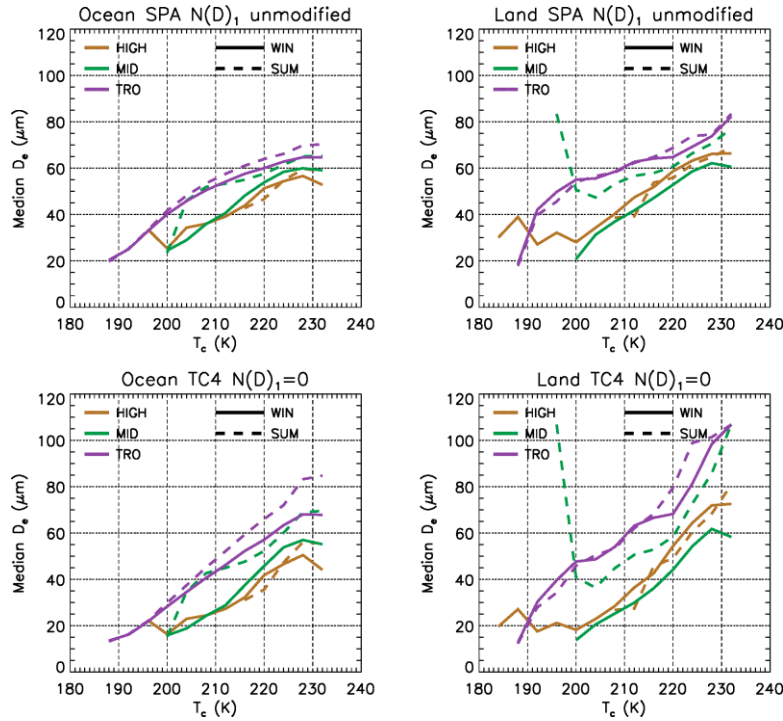
Same as Fig. S9 (previous Fig. 12b), but with $OD > 0.1$.



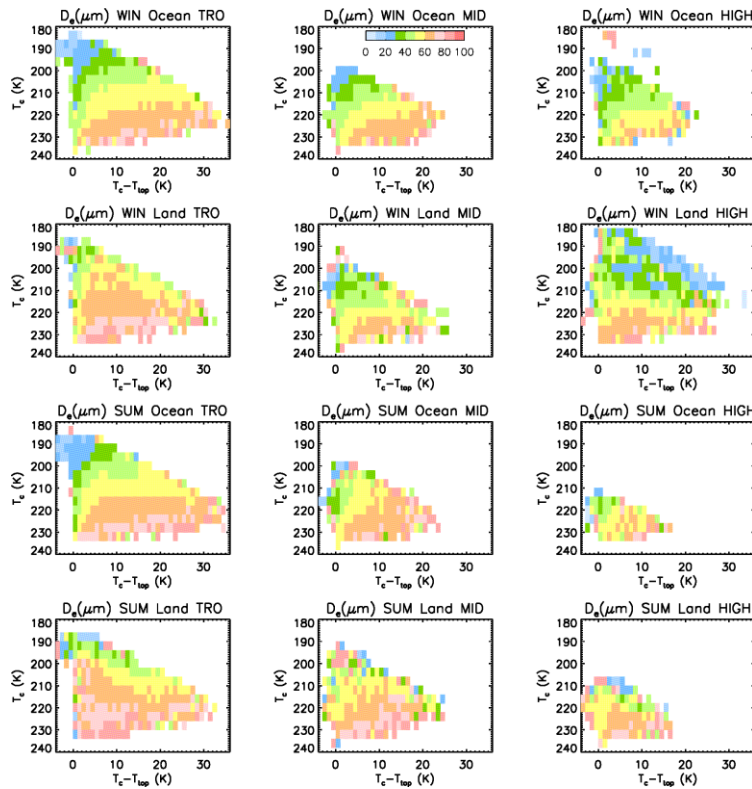
5 Same as Fig. 10 (previous Fig. 13), but with $OD > 0.1$. The relative variations are similar in both figures, with N being smaller when samples of OD between 0.1 and 0.3 are included.



Same as Fig. S11 (previous Fig. 14), but with OD>0.1



5 Same as Fig. 11 (previous Fig. 15), but with OD>0.1. Consistent with Fig. 8, the changes in median D_e are more notable over land. Larger additional errors occur more over land than over oceans.



Same as Fig. S12 (previous Fig. 16), but with $OD > 0.1$. Again, the changes in median D_e are more notable over land than over ocean.

5 RC S8. Figure 11. Your highest Beta_Eff occur at the highest clouds where your sample size is often relatively small. Is that an issue? Or is the “hom” effect.

AC S8. It’s more likely the “hom effect”. That is, we consistently see higher β_{eff} at colder temperatures and higher altitudes with the in situ PSD from field campaigns. This is where hom can produce the highest N, resulting in the smallest crystals that produce the highest β_{eff} . New text has been added at the end of the 3rd paragraph in Sect. 5.2:

“At the coldest temperatures (highest Z_c), hom should be more frequent, resulting in smaller ice crystals and thus higher β_{eff} .”

15

RC S9. Figure 17: Comparing the two figures, can the authors explain why some peak regions remain similar magnitude such as in southwest Southern America, but many weaken significantly for instance over the Arctic Ocean East of Greenland?

20

AC S9. It is important to recognize that while the color legend in Fig. 12 (previously Fig. 17) changes color in increments of 50 L^{-1} , the 1st color bin is from zero to 100 L^{-1} and the last color bin is from 500 L^{-1} to infinity. When comparing the two plots, regions corresponding to the highest N values may not change much due to this legend convention.

New text has been added to the end of the 1st paragraph in Sect. 6.4: *“While the color legend in Fig. 12 changes color in increments of 50 L^{-1} , the 1st color bin is from zero to 100 L^{-1} and the last color bin is from 500 L^{-1} to infinity. When comparing the two plots, regions corresponding to the highest N values may not change much due to this legend convention.”*

CALIPSO (IIR-CALIOP) Retrievals of Cirrus Cloud Ice Particle Concentrations

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10 **Abstract.** A new satellite remote sensing method is described whereby the sensitivity of thermal infrared wave resonance absorption to small ice crystals is exploited to estimate cirrus cloud ice particle number concentration N , effective diameter D_e , and ice water content IWC. This method uses co-located observations from the Infrared Imaging Radiometer (IIR) and from the CALIOP (*Cloud and Aerosol Lidar with Orthogonal Polarization*) lidar aboard the CALIPSO (*Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation*) polar orbiting satellite,
15 employing IIR channels at 10.6 μm and 12.05 μm . Using particle size distributions measured over ~~several~~many flights of the TC4 (Tropical Composition, Cloud and Climate Coupling) and the mid-latitudes SPARTICUS (Small Particles in Cirrus) field campaigns, we show for the first time that N/IWC is tightly related to β_{eff} ; the ratio of effective absorption optical depths at 12.05 μm and 10.6 μm . Relationships developed from in situ aircraft measurements are applied to β_{eff} derived from IIR measurements to retrieve N . This satellite remote sensing method
20 is constrained by measurements of β_{eff} from the IIR and is by essence sensitive to the smallest ice crystals. Retrieval uncertainties are discussed, including uncertainties related to in situ measurement of small ice crystals ($D < 15 \mu\text{m}$), which are studied through comparisons with IIR β_{eff} . The method is applied here to single-layered semi-transparent clouds having a visible optical depth between about 0.3 and 3, where cloud base temperature is $\leq 235 \text{ K}$. Two years of CALIPSO data have been analyzed for the years 2008 and 2013, with the dependence of cirrus cloud N and D_e on
25 altitude, temperature, latitude, season (winter vs. summer) and topography (land vs. ocean) described. The results for the mid-latitudes show a considerable dependence on season. In the high latitudes, N tends to be highest and D_e smallest, whereas the opposite is true for the tropics. The frequency of occurrence of these relatively thick cirrus clouds exhibited a strong seasonal dependence in the high latitudes, with the occurrence frequency during Arctic winter being at least twice that of any other season. Processes that could potentially explain some of these micro-
30 and macroscopic cloud phenomena are discussed.

1 Introduction

The microphysical and radiative properties of ice clouds are functions of the ice particle size distribution or PSD, which is often characterized by the PSD ice water content (IWC), a characteristic ice particle size and the ice particle number concentration N ; all of which can be measured in situ using suitable instruments. To date, satellite remote sensing methods can retrieve two of these properties; the PSD effective diameter D_e and IWC. Most parameterizations of ice cloud optical properties in climate models are based on these parameters (e.g. Fu, 1996; 2007). However, the ice cloud PSD is not fully constrained by D_e and IWC, and ice cloud optical properties at terrestrial wavelengths are not always well defined by D_e and IWC (Mitchell et al., 2011a). Moreover, satellite retrievals of N would be useful for advancing our understanding of ice nucleation in the atmosphere. To realistically predict D_e in climate models, realistic predictions of ice crystal nucleation rates are essential since they determine D_e . Realistic satellite retrievals of N would provide a powerful constraint for parameterizing ice nucleation in climate models.

Retrievals of ice cloud microphysical properties from satellite have evolved considerably since the first developments using passive observations (Inoue, 1985; Parol et al., 1991; Ackerman et al., 1995). The advent of the A-Train has enabled passive and active observations to be combined and more precisely analyzed to study the vertical structure of ice clouds and the atmosphere (Delanoë and Hogan, 2008, 2010; Deng et al., 2010, Garnier et al., 2012, 2013, Deng et al., 2013; Sourdeval et al., 2016). Such retrievals from satellite have been extensively compared to in situ observations (King et al., 2003, Deng et al., 2010, 2013) to validate these retrievals and to estimate representative microphysical parameters such as D_e and IWC (or ice water path, IWP) that can be compared with corresponding large scale model outputs to improve their cloud parametrizations and general climate applications (Eliasson et al., 2011; Stubenrauch et al., 2013; the Global Energy and Exchanges Process Evaluation Studies (GEWEX PROES) <https://gewex-utcc-proes.aeris-data.fr/>). This has advanced a convergence between in situ and satellite studies on ice clouds, where satellite studies do not suffer from certain aircraft probe limitations such as ice particle shattering (e.g. Field et al., 2006; Mitchell et al., 2010; Korolev et al., 2011; Cotton et al., 2013), and in situ studies do not depend on certain relationships relating radiation to cloud properties (e.g. Delanoë and Hogan, 2008).

Recent progress has been made regarding efforts to retrieve N via satellite. The retrieval of N as a function of latitude and topography is of particular importance as it provides insight into specific physical processes controlling N . The satellite remote sensing study by Zhao et al. (2018) has advanced our understanding of the complex relationship between aerosol particles and cirrus clouds, showing the importance of homogeneous ice nucleation ([henceforth hom](#)) under relatively clean (i.e. relatively low aerosol optical depth) conditions. A satellite retrieval for N has been proposed that builds upon the lidar-radar (hence DARDAR) retrieval described in Delanoë and Hogan

(2008, 2010), as described in Gryspeerdt et al. (2018) and Sourdeval et al. (2018a). Relating satellite retrievals of N and D_e to mineral dust observations (Gryspeerdt et al., 2018) and to cirrus cloud-aerosol modeling outcomes (Zhao et al., 2018) have yielded insights into the relative importance of homogeneous and heterogeneous ice nucleation (henceforth *het*) in cirrus clouds as a function of aerosol concentrations.

- 5 This study describes a new approach for estimating cloud layer N , D_e , and IWC in selected semi-transparent cirrus clouds. The technique uses co-located observations from the 10.6 μm and 12.05 μm channels on the Imaging Infrared Radiometer (IIR) aboard the CALIPSO (*Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation*) polar orbiting satellite, augmented by the scene classification and extinction profile from CALIOP (*Cloud and Aerosol Lidar with Orthogonal Polarization*) and by interpolated temperatures from the Global
- 10 Modeling Assimilation Office (GMAO) Goddard Earth Observing System Model, version 5 (GEOS 5) (Garnier et al., 2012, 2013). CALIOP and IIR are assembled in a near-nadir looking configuration. The cross-track swath of IIR is by design centered on the CALIOP track where observations from the two instruments are perfectly temporally co-located. The spatial co-location is nearly perfect, as CALIOP samples the same cloud as the 1-km IIR pixel, but with three laser beam spots per km having a horizontal footprint of 90 m. While the IIR retrieves layer-average
- 15 cloud properties, CALIOP vertically profiles cloud layers, thus providing estimates of representative cloud temperature and the temperature dependence of N , D_e and IWC.

In this paper, we compare IIR retrievals with in situ observations performed during two field campaigns conducted in the tropics and mid-latitudes and develop a method to derive cirrus microphysical parameters from CALIPSO observations. Using different assumptions, several formulations for this retrieval scheme are presented to illustrate

20 the inherent uncertainties associated with the retrieved cloud properties. The objective of this work was not to determine absolute magnitudes for the retrieved quantities, but rather to show how their relative differences vary in terms of temperature, cloud thickness, latitude, season and topography.

Section 2 describes the rationale for developing the retrieval method, along with the retrieval physics and methodology, and discusses several plausible assumptions and formulations. Section 3 describes the retrieval

25 equation that incorporates CALIPSO observations, as well as retrieval uncertainties. In Sect. 4, retrieved layer-average cloud properties are compared with corresponding cloud properties measured in situ. Different retrieval scheme formulations are used to illustrate the inherent uncertainties associated with retrieved and in situ cloud properties. In Sect. 5, IIR-CALIOP retrieval results for N and D_e are reported for 2008 and 2013 during the winter and summer seasons for all latitudes. These results are discussed in Sect. 6, which includes comparisons with

30 previous work using the combined radar-lidar approach (Sourdeval et al., 2018a; Gryspeerdt et al., 2018). The discussion also addresses the radiative significance of these cirrus cloud retrievals and a potential link between

Arctic cirrus and mid-latitude weather. A brief summary of the results and concluding comments are ending the paper in Sect. 7.

2 Developing a satellite remote sensing method sensitive to N

2.1 Satellite retrievals from infrared absorption methods

5 It is widely recognized that the ratio of absorption optical depth from ice clouds, β , based on wavelengths in the thermal infrared domain at 12 μm and 11 μm (or similar wavelengths), is rich in cloud microphysical information (Inoue, 1985; Parol et al., 1991; Cooper et al., 2003; Dubuisson et al., 2008; Heidinger and Pavolonis, 2009; Mishra et al., 2009; Mitchell and d'Entremont, 2009; Pavolonis, 2010; Mitchell et al., 2010; Cooper and Garrett, 2010; Garnier et al., 2012; Mitchell and d'Entremont, 2012; Garnier et al., 2013). These studies have used a retrieved β to
10 estimate the effective diameter D_e , IWP, the mass-weighted ice fall speed (V_m), the average fraction of liquid water in a cloud field, the relative or actual concentration of small ice crystals in ice PSDs and the cloud droplet number concentration in mixed phase clouds. However, the main reason for the emissivity differences in satellite remote sensing channels centered on these two wavelengths was not understood until after the development of the modified anomalous diffraction approximation (MADA) that, to a first approximation, allowed various scattering/absorption
15 processes to be isolated and evaluated independently (Mitchell, 2000; Mitchell et al., 2001; Mitchell, 2002; Mitchell et al., 2006). For wavelengths between 2.7 and 100 μm , the most critical process parameterized was wave resonance, also referred to as photon tunneling (e.g. Nussenzveig, 1977; Guimaraes and Nussenzveig, 1992; Nussenzveig, 2002). This process was found to be primarily responsible for the cloud emissivity difference between these wavelengths (12 μm and 11 μm) in ice clouds, as described in Mitchell et al. (2010).

20 It was originally thought that β resulted from differences in the imaginary index of ice, m_i , at two wavelengths (λ) near 11 μm and 12 μm , but it is actually due to differences in the real index of refraction, m_r . At these λ , m_i is sufficiently large so that most ice particles in the PSD experience area-dependent absorption (i.e. no radiation passes through the particle), and the absorption efficiency Q_{abs} for a given ice particle will be ~ 1.0 for both λ when Q_{abs} is based only on m_i (i.e. the Q_{abs} predicted by Beer's law absorption or anomalous diffraction theory). The observed
25 difference between $Q_{\text{abs}}(12 \mu\text{m})$ and $Q_{\text{abs}}(11 \mu\text{m})$ is due to differences in the photon tunneling contribution to absorption that primarily depends on m_r (Mitchell, 2000). That is, m_r is substantial when $\lambda = 12 \mu\text{m}$ but is relatively low when $\lambda = 11 \mu\text{m}$, producing a substantial difference between $Q_{\text{abs}}(12 \mu\text{m})$ and $Q_{\text{abs}}(11 \mu\text{m})$.

Figure 1 shows the size dependence of the tunneling contribution for hexagonal columns at 12 μm .

30 The greatest tunneling contribution to absorption occurs when the ice particle size and wavelength are comparable and the real refractive index m_r is relatively high relative to m_r for the 11 μm wavelength (Mitchell, 2000). In this

case, β_2 is sensitive to the tunneling process and the relative concentration of small ice crystals in cirrus clouds (i.e. maximum this contribution is becoming smaller than 10% for sizes larger than 60 μm) ice crystals in cirrus clouds. It is thus evident that this contribution is making β well suited for detecting recently nucleated (small) ice crystals that primarily determine N (Krämer et al., 2009).

In this study, we use CALIPSO IIR channels at 12.05 μm and 10.6 μm . Since m_i at 10.6 μm is less than m_i at 11 μm , some Beer's law type absorption may contribute to absorption differences between the 10.6 μm and 12 μm channels when PSD are sufficiently narrow. This acts to slightly extend the dynamic range of sensitivity relative to the 11 μm -12 μm channel combination.

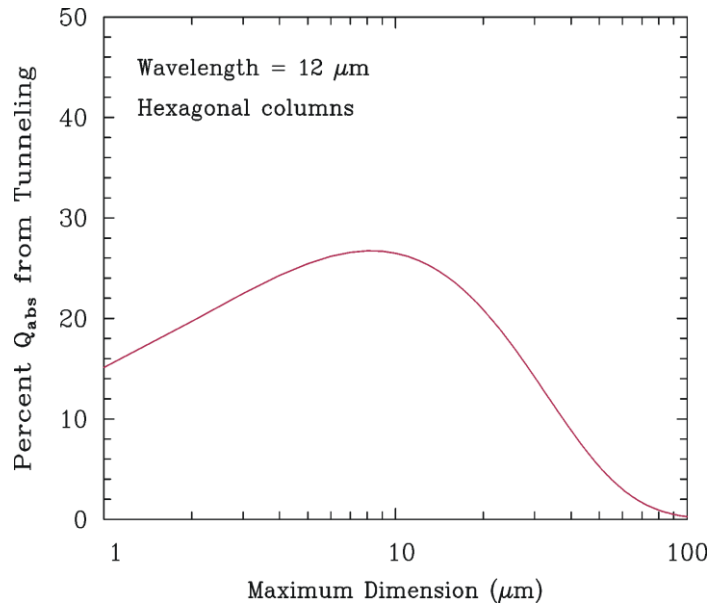


Figure 1: Percent contribution of wave resonance absorption to the overall absorption efficiency at 12 μm wavelength as a function of maximum dimension D for hexagonal columns, as estimated by the MADA. It is decreasing to below 10% of its maximum for sizes larger than about 60 μm .

dimension $D < 60 \mu\text{m}$). It is thus evident that this contribution is making β well suited for detecting recently nucleated (small) ice crystals that primarily determine N (Krämer et al., 2009).

In this study, we use CALIPSO IIR channels at 12.05 μm and 10.6 μm . Since m_i at 10.6 μm is less than m_i at 11 μm , some Beer's law type absorption may contribute to absorption differences between the 10.6 μm and 12 μm channels when PSD are sufficiently narrow. This acts to slightly extend the dynamic range of sensitivity relative to the 11 μm -12 μm channel combination.

2.2 Retrieving β_{eff} from IIR and CALIOP observations

In this study, we use CALIPSO IIR channels at 10.6 μm and 12.05 μm and the absorption optical depth $\tau_{\text{abs}}(12.05 \mu\text{m})$ and $\tau_{\text{abs}}(10.6 \mu\text{m})$ retrieved from the effective emissivity in CALIPSO IIR channels 12.05 μm and 10.6 μm . The retrieved optical depths are not purely due to absorption, but also include the effects of scattering. Thus, their ratio ~~is define~~ not exactly β , but ~~is~~ the effective β or β_{eff} written as

$$\beta_{\text{eff}} = \tau_{\text{abs}}(12.05 \mu\text{m}) / \tau_{\text{abs}}(10.6 \mu\text{m}) \quad (1)$$

where τ_{abs} is the retrieved absorption optical depth for a given λ retrieved from the effective emissivity. However, what is retrieved is not exactly β , but a β that also includes the effects of scattering, defined as the effective β or β_{eff} . β_{eff} is described analytically in Parol et al. (1991). For a given cirrus cloud, retrieved $\tau_{\text{abs}}(10.6 \mu\text{m})$ may be slightly less than $\tau_{\text{abs}}(11 \mu\text{m})$ since m_i at 10.6 μm is less than m_i at 11 μm , meaning that some Beer's law type absorption may contribute to emissivity differences between the 10.6 μm and 12 μm channels when PSD are sufficiently narrow. This acts to slightly extend the dynamic range of retrievals relative to the 11 μm 12 μm channel combination (e.g. the limiting maximum D_e retrieved will be greater using 10.6 and 12 μm relative to 11 and 12 μm). Although it is feasible to retrieve D_e using the IIR 8.65 μm channel (e.g. Dubuisson et al., 2008; Garnier et al., 2013), the same channels are used for both D_e and N to ensure self-consistent retrievals with respect to PSD moments. In addition, the analytical PSD calculations for β_{eff} are more accurate when using the 12.05/10.6 combination relative to the 12/8.65 combination (Figs. 1b and 2b in Garnier et al., 2013, JAMC) since there is less multiple scattering with the 12.05/10.6 combination. IIR retrievals have a resolution of 1 km. ~~Finally, IIR calibrated radiances are from the recently released Version 2 Level 1b products (Garnier et al., 2018).~~ IIR β_{eff} ~~retrievals are~~ is based on the CALIPSO IIR Version 3 Level 2 track product (Vaughan et al., 2017). This product includes a scene typing built from the CALIOP Version 3 5-km cloud and aerosol layer products. The scene classification has been refined for this study to account for additional dense clouds in the planetary boundary layer reported in the CALIOP Version 3 333-m layer product. The methodology for retrieving CALIPSO-IIR effective emissivity and β_{eff} from co-located CALIOP observations and IIR radiances is detailed in Garnier et al. (2012, 2013). IIR ~~effective emissivity retrievals are~~ was further corrected-reprocessed for this study to reduce possible biases, as described in the following sub-sections. Version 3 CALIOP cloud extinction coefficient profiles are used for some of the corrections. ~~Finally, IIR calibrated radiances are from the recently released Version 2 Level 1b products (Garnier et al., 2018).~~ These improvements will be implemented in the next version 4 of the IIR Level 2 products. This is why we only focus in this study on a limited dataset as a proof-of-concept. We will further develop a more statistically relevant analysis using the version 4 products.

For each IIR channel, τ_{abs} is derived from the effective emissivity, ϵ , as

$$\tau_{\text{abs}} = -\ln(1 - \epsilon) \quad (2)$$

with

$$\varepsilon = \frac{R_m - R_{BG}}{R_{BB} - R_{BG}} \quad (3)$$

where R_m is the measured calibrated radiance, R_{BB} is the opaque (i.e. blackbody) cloud radiance, and R_{BG} is the ~~clear sky~~ background radiance that would be observed in the absence of the studied cloud, as described in Garnier et al. (2012).

The cloud ~~effective~~ emissivity ε and the subsequent β_{eff} are retrieved for carefully selected cirrus clouds, as described in Sect. 2.2.1. The retrievals are cloud layer average quantities as seen from space, whose representative altitude and temperature are estimated using additional information from CALIOP vertical profiling in the cloud, as presented in Sect. 2.2.3. As such, they can differ from in situ local measurements in the cloud.

2.2.1 Cloud selection

~~Because IIR is a passive instrument, meaningful retrievals are possible for well identified scenes. This study is restricted to the cases where the atmospheric column contains one cirrus cloud layer. We also ensure that the background radiance is only due to the surface (see Eq. (3)) allowing a more accurate computation than for cloudy cases.~~ ~~A number of selection criteria are defined for the robustness of the retrievals. In this study, t~~ retrievals were applied only to single-layered semi-transparent cirrus clouds ~~(one cloud layer in an atmospheric column)~~ that do not fully attenuate the CALIOP laser beam, so that the cloud base is detected by the lidar. The cloud base is in the troposphere and its temperature is required to be colder than -38°C (235 K) to ensure that the cloud is entirely composed of ice. This is likely to exclude liquid-origin cirrus clouds from our data set (Luebke et al., 2016). ~~When the column contains also a dense water cloud, the background radiance can be computed assuming that the water cloud is a blackbody. However, because systematic biases were made evident (Garnier et al., 2012), we chose to discard these cases, which reduces the number of selected samples by about 25 %.~~ Because the relative uncertainties in τ_{abs} and in β_{eff} increase very rapidly as cloud emissivity decreases (Garnier et al., 2013), the lidar layer-integrated attenuated backscatter (IAB) was chosen greater than 0.01 sr^{-1} to avoid very large uncertainties at the smallest visible optical depths (ODs). This resulted in an OD range of about 0.3 to 3.0. Similarly, clouds for which the radiative contrast $R_{BG} - R_{BB}$ between the surface and the cloud is less than 20 K in brightness temperature units are discarded. IIR observations must be of good quality according to the quality flag reported in the IIR Level 2 product (Vaughan et al., 2017).

2.2.2 Background radiance, R_{BG}

The brightness temperature T_{BG} associated to the [clear sky](#) background radiance R_{BG} is derived from the FASt RADiative (FAStRAD) transfer model (Dubuisson et al., 2005) fed by atmospheric profiles and skin temperatures from GMAO GEOS5 along with pre-defined surface emissivities inferred from the International Geosphere and Biosphere Program (IGBP) surface types and a daily updated snow/ice index (Garnier et al., 2012). For this study, remaining biases at 12.05 μm and 10.6 μm between [observations and FAStRAD model](#) ~~and observations~~ are corrected using monthly maps of mean differences between observed and computed brightness temperatures (called BTDoc) in clear sky conditions. The corrections are applied over ocean and over land with a resolution of 2 degrees in latitude and 4 degrees in longitude, by separating daytime and nighttime data. [Figures S1a and S1b in Supplementary Materials show distributions of BTDoc before and after correction.](#) After correction, BTDoc is equal to zero on average for both channels.

~~2.2.3 Blackbody radiance, R_{BB}~~

2.2.3 Blackbody radiance, R_{BB}

In version 3 IIR products, R_{BB} (and the associated blackbody temperature T_{BB}) is derived from the cloud temperature, T_{caliop} , evaluated at the centroid altitude of the CALIOP 532 nm attenuated backscatter profile using GMAO GEOS5 temperature (Garnier et al, 2012). In this study, a correction is further applied using CALIOP extinction profiles in the cloud layer as described in Sect. 3 of Garnier et al. (2015). The CALIOP lidar 532 nm extinction profile in the cloud is used to determine an IIR weighting profile that is used, [together with the GMAO GEOS5 temperature profile,](#) to compute R_{BB} as the weighted averaged blackbody radiance. The lidar vertical resolution is 60-m, and emissivity is weighted in a similar way with the 532 nm extinction profile. The weight of each 60-m bin is its emissivity at 12.05 μm attenuated by the overlying infrared absorption optical depth, normalized to the cloud emissivity. [Computing \$T_{BB}\$ at 10.6 \$\mu\text{m}\$ and at 12.05 \$\mu\text{m}\$ yields temperatures that differ by less than 0.15 K on average, which has a negligible impact on \$\beta_{\text{eff}}\$ for our cloud selection.](#)

In addition, the altitude and temperature associated to the layer average β_{eff} are characterized using the centroid altitude, Z_c , and centroid temperature, T_c , of the IIR weighting profile. Note that T_c and T_{BB} are not identical, but typically differ by less than a few tenths of Kelvins.

2.2.4 Estimated uncertainties [in \$\beta_{\text{eff}}\$](#)

The uncertainty in β_{eff} , $\Delta\beta_{\text{eff}}$, is computed by propagating the errors in $\tau_{\text{abs}}(12.05 \mu\text{m})$ and $\tau_{\text{abs}}(10.6 \mu\text{m})$. These errors are themselves computed by propagating the errors in i) the measured brightness temperatures T_m associated to R_m , ii) the blackbody brightness temperature T_{BB} , and iii) the background brightness temperatures T_{BG} (Garnier et al., 2015). The uncertainties in T_m at 10.6 μm and 12.05 μm are random errors set to 0.3 K according to the IIR

performance assessment established by the Centre National d'Etudes Spatiales (CNES) assuming no systematic bias in the calibration. They are assumed to be statistically independent. In contrast, the error in T_{BB} is the same for both channels, because the same cloud temperature T_{BB} is used to compute $\tau_{abs}(12.05 \mu m)$ and $\tau_{abs}(10.6 \mu m)$. A random error of $\pm 2K$ in T_{BB} is estimated to include errors in the atmospheric model. After correction for systematic biases based on differences between observations and computations in cloud-free conditions (see Sect. 2.2.2), the error in T_{BG} is considered a random error, which is taken equal to the standard deviation of the corrected distributions of BTDoc (Fig. S1a). As a result, the uncertainty in T_{BG} at $12.05 \mu m$ is set to $\pm 1K$ over ocean, and to $\pm 3K$ over land for both night and day. Standard deviations of the distributions of $[BTDoc(10.6 \mu m) - BTDoc(12.05 \mu m)]$ are generally smaller than $0.5 K$ (Fig. S1b). Accounting for the contribution from the measurements, which is estimated to be $\sqrt{2} \times 0.3 = 0.45 K$, this indicates that the biases in T_{BG} at 10.6 and at $12.05 \mu m$ are nearly canceling out and can therefore be considered identical. More details about the uncertainty analysis can be found in the appendix.

The relative uncertainty $\Delta\beta_{eff}/\beta_{eff}$ is mostly due to random measurement errors, because systematic biases associated with the retrieval of $\tau_{abs}(12.05 \mu m)$ and $\tau_{abs}(10.6 \mu m)$ tend to cancel when these are ratioed to calculate β_{eff} .

2.3. Relating β_{eff} to N/IWC and D_e based on aircraft PSD measurements

Using aircraft data from the DOE ARM supported Small Particles in Cirrus (SPartICus) field campaign in the central United States (Mace et al., 2009) and the NASA supported Tropical Composition, Cloud and Climate Coupling (TC4) field campaign near Costa Rica (Toon et al., 2010), β_{eff} was related to cirrus cloud microphysical properties. The SPARTICUS field campaign, conducted from January through June of 2010 in the central United States (for domain size, see Fig. 2 in Mishra et al., 2014), was designed to better quantify the concentrations of small ($D < 100 \mu m$) ice crystals in cirrus clouds (Mace et al., 2009). Regarding SPARTICUS, the data set described in Mishra et al. (2014) was used, and the TC4 data is described in Mitchell et al. (2011b). Details regarding field measurements, the flights analyzed and the microphysical processing are described in these articles. PSD sampling times during TC4 were often much longer than for SPARTICUS (< 2 minutes), with mean TC4 sampling times for horizontal legs within aged anvils being 10.56 minutes (Lawson et al., 2010; Mitchell et al., 2011b). This, along with more in-cloud flight hours during SPARTICUS, resulted in fewer PSD samples for TC4 relative to SPARTICUS. The PSDs were measured by the 2D-S probe (Lawson et al., 2006) where ice particle concentrations were measured down to $10 \mu m$ ($5-15 \mu m$ size bin) and up to $1280 \mu m$ in ice particle length (when using “all-in” data processing criteria). β_{eff} was calculated from these PSDs using the method described in Parol et al. (1991) and Mitchell et al. (2010). This method was tested in Garnier et al. (2013, Fig. 1b) where β_{eff} calculated from a radiative transfer model (FASDOM; Dubuisson et al., 2005) was compared with β_{eff} calculated analytically via Parol et al. (1991), with good agreement found between these two methods. More specifically, to calculate β_{eff} from PSD in this study, the PSD absorption efficiency \bar{Q}_{abs} is given as $\bar{Q}_{abs} = \beta_{abs} / A_{PSD}$, where β_{abs} is the PSD absorption

coefficient (determined by MADA from measured PSD) and A_{PSD} is the measured PSD projected area. The PSD effective diameter was determined from the measured PSD as described in Mishra et al. (2014), but in essence is given as $D_e = (3/2) \text{ IWC} / (\rho_i A_{\text{PSD}})$, where ρ_i is the density of bulk ice (0.917 g cm^{-3}). The PSD extinction efficiency \bar{Q}_{ext} was determined in a manner analogous to \bar{Q}_{abs} . The single scattering albedo ω_o was calculated as $\omega_o = 1 - \bar{Q}_{\text{abs}} / \bar{Q}_{\text{ext}}$ and the PSD asymmetry parameter g was obtained from D_e using the parameterization of Yang et al. (2005). Knowing \bar{Q}_{abs} , ω_o and g , β_{eff} was calculated from the PSD as:

$$\beta_{\text{eff}} = Q_{\text{abs,eff}}(12.05 \mu\text{m}) / Q_{\text{abs,eff}}(10.6 \mu\text{m}), \quad (4)$$

$$Q_{\text{abs,eff}} = \bar{Q}_{\text{abs}} (1 - \omega_o g) / (1 - \omega_o). \quad (5)$$

Note that β (i.e. β_{eff} without scattering effects) is ~~also~~ equal to $\bar{Q}_{\text{abs}}(12.05 \mu\text{m}) / \bar{Q}_{\text{abs}}(10.6 \mu\text{m})$.

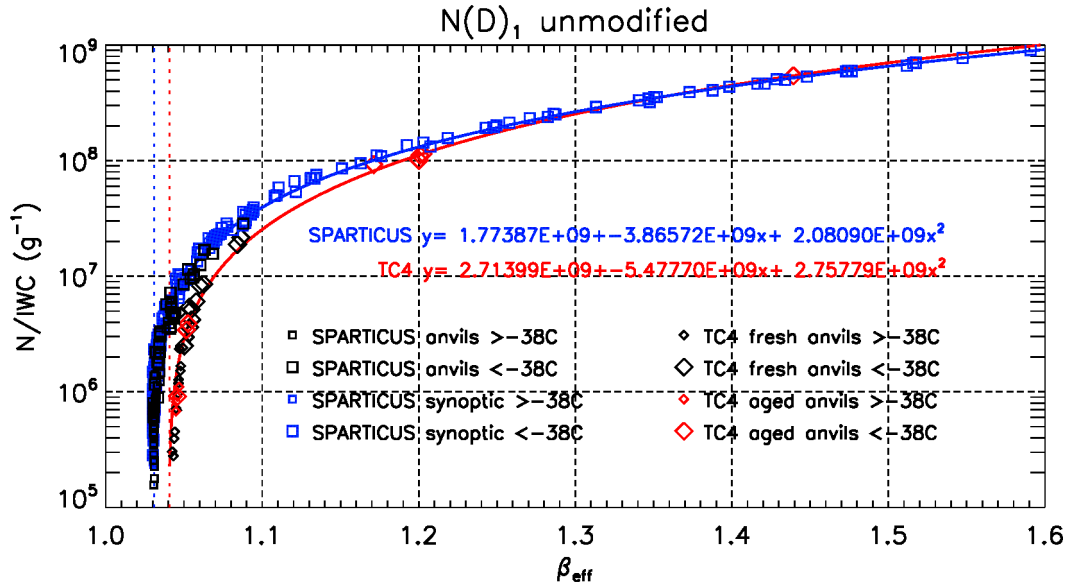


Figure 2: Dependence of $N/IWC \text{ (g}^{-1}\text{)}$ on the effective absorption optical depth ratio β_{eff} as predicted from the method of Parol et al. (1991), based on PSDs from SPARTICUS synoptic cirrus (blue squares) and anvils (black squares), and TC4 aged (red diamonds) and fresh (black diamonds) anvils, where the first size-bin is included. The larger (smaller) symbols denote PSDs measured at a temperature colder (warmer) than -38°C . The curve-fit equations are for SPARTICUS synoptic cirrus (blue) and for TC4 aged and fresh anvils (red).

Figure 2 shows measurements of N/IWC from SPARTICUS flights over the central United States, based on PSD sampled from synoptic (blue squares) and anvil (black squares) cirrus clouds. Cirrus cloud PSD were measured using the 2D-S probe, which produces shadowgraph images with true $10 \mu\text{m}$ pixel resolution at aircraft speeds up to 170 m s^{-1} , measuring ice particles between 10 and 1280 (or more) μm (Lawson et al., 2006). The 2D-S PSD data

was post-processed using an ice particle arrival time algorithm that identifies and removes ice shattering events from the data stream. All size-bins of the 2D-S probe were used here. Two SPARTICUS flights from April 28th were added to this dataset (giving a total of 15 flights) since April 28th was previously mislabeled as an anvil cirrus case study, but was actually a ridge-crest cirrus event (a type of synoptic cirrus) as described in Muhlbauer et al. (2014).

5 This “ridge crest cirrus” had high N (500-2200 L^{-1}) for $T < -60^{\circ}C$. Also shown are N/IWC measurements from the TC4 field campaign for maritime “fresh” (black diamonds) anvil cirrus (during active deep convection where the anvil is linked to the convective column) and for aged (red diamonds) anvil cirrus (anvils detached from convective column). Figure 2 relates β_{eff} to the N/IWC ratio, where β_{eff} was calculated from the same PSD measurements used to calculate N and IWC , based on the MADA method. The PSD measurements include size-resolved estimates of
10 ice particle mass concentration based on Baker and Lawson (2006a), size-resolved measurements of ice projected area concentration, and the size resolved number concentration. This PSD information is the input for the MADA method that yields the coefficients of absorption and extinction. The tunneling efficiency T_e used in MADA was estimated from Table 1 in Mitchell et al. (2006), where for $1 \mu m < D < 30 \mu m$, droxtals and hexagonal columns are assumed and $T_e = 0.90$; for $30 \mu m < D < 100 \mu m$, budding bullet rosettes and hexagonal columns are assumed and
15 $T_e = 0.50$; for $D > 100 \mu m$ bullet rosettes and aggregates are assumed and $T_e = 0.15$. This shape-dependence on ice particle size was guided by the ice particle size-shape observations reported in Lawson et al. (2006), Baker and Lawson (2006b) and Erfani and Mitchell (2016), where small hexagonal columns (for which we can estimate T_e) are substituted for small irregular crystals. These ice particle shape assumptions affect only T_e , and the cloud optical properties are primarily determined through the PSD measurements noted above (i.e. not the value of T_e). Due to
20 β_{eff} ’s sensitivity to tunneling and small ice crystals, a tight and useful relationship is found between N/IWC and β_{eff} for $N/IWC > \sim 10^7 g^{-1}$ for both campaigns. As far as we know, this relationship was not known previously. The associated PSDs were measured at temperatures colder than $-38^{\circ}C$ (large symbols), which is the cloud base temperature of the cirrus clouds targeted for this study. For $N/IWC < \sim 10^6 g^{-1}$, β_{eff} reaches a low limit and is not sensitive to N/IWC , so that N/IWC cannot be estimated from β_{eff} . Much of the data with β_{eff} reaching the “no
25 sensitivity” low limit were derived from ice cloud PSDs at temperatures between -20 and $-38^{\circ}C$ (small symbols) where PSD tend to be relatively broad.

Because the clouds selected in this study are single-layered semi-transparent cirrus clouds, the relationships seen for synoptic cirrus during SPARTICUS could be deemed the most relevant for this study. But interestingly, synoptic and anvil cirrus during SPARTICUS (squares) follow similar relationships, and similarly, aged and fresh cirrus
30 anvils (diamonds) follow similar relationships during TC4. Thus, the fact that anvil and synoptic cirrus during SPARTICUS follow a similar relationship suggests that the relationship established from anvils during TC4 could also be relevant for this study. The blue line shows the curve fit derived from SPARTICUS synoptic cirrus, while the red line is derived from aged and fresh anvils during TC4. The blue and the red lines are similar for the largest

β_{eff} ($\beta_{\text{eff}} > 1.2$) and progressively depart as β_{eff} decreases. The β_{eff} low limit is 1.031 during SPARTICUS, for both synoptic and anvil cirrus, whereas it is 1.041 during TC4. The different β_{eff} low limits during SPARTICUS and TC4 may reflect different PSD shapes measured during these two campaigns. This indicates that as β_{eff} decreases and its sensitivity to N/IWC decreases, the sensitivity of the relationships to the PSD increases. These two curves are indicative of the possible dispersion in the relationships, and therefore will both be considered in the analysis.

Using this same in situ data and methodology, β_{eff} has also been related to D_e as shown in Fig. 3, where all PSD bins were used. D_e is defined as $(3/2) \text{IWC}/(\rho_i A_{\text{PSD}})$ where ρ_i is the density of bulk ice (Mitchell, 2002). Accordingly, D_e was calculated from the measured PSD (see Mishra et al., 2014). The relationships derived from SPARTICUS synoptic cirrus and from TC4 anvils are shown in blue and red, respectively. They are only useful for $D_e < 90$ or $D_e < 110 \mu\text{m}$, respectively, since β_{eff} is only sensitive to the smaller ice particles. The relationships are similar for D_e below $30 \mu\text{m}$. This emphasizes the fact that β_{eff} is a measure of the relative concentration of small ice crystals in a

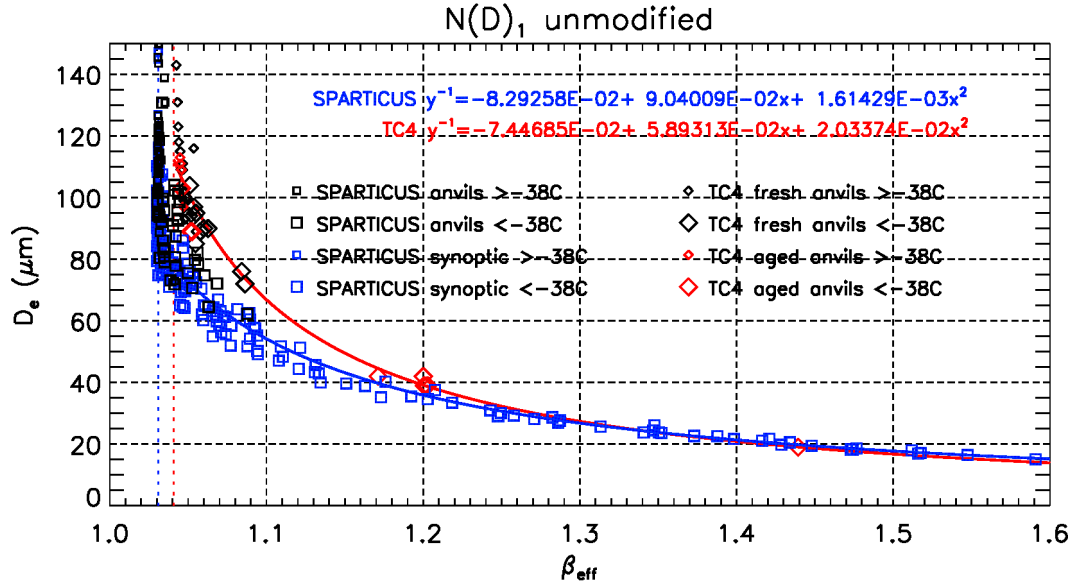


Figure 3: Dependence of the PSD effective diameter D_e (μm) on the effective absorption optical depth ratio β_{eff} as predicted from the method of Parol et al. (1991), based on PSDs from SPARTICUS synoptic cirrus (blue squares) and anvils (black squares), and TC4 aged (red diamonds) and fresh (black diamonds) anvils, where the first size-bin is included ($N(D)_1$ unmodified). The larger (smaller) symbols denote PSDs measured at a temperature colder (warmer) than -38°C . The curve-fit equations give $1/D_e$ in μm^{-1} ; they are for SPARTICUS synoptic cirrus (blue) and for TC4 aged and fresh anvils (red).

PSD (Mitchell et al., 2010). A_{PSD} and β_{eff} (PSD integrated quantities) may be associated with a substantial portion of larger ice particles ($D > 50 \mu\text{m}$) before β_{eff} loses sensitivity to changes in D_e . There are much fewer TC4 points in

Figs. 2 and 3 for $\beta_{\text{eff}} > 1.1$ since the higher β_{eff} values were obtained only for $T < -60^\circ\text{C}$, which only occurred during two SPARTICUS flights on April 28th.

Cirrus cloud emissivity and τ_{abs} depend on ice particle shape (Mitchell et al., 1996a; Dubuisson et al. 2008).

However, this retrieval should not be very sensitive to ice particle shape for several reasons, one being that β_{eff} is directly retrieved from cloud radiances as per (2) and (3). Another reason is that no ice particle shape assumptions are made when calculating β_{eff} from in situ measurements with the exception of the absorption contribution from tunneling (which was not sensitive to realistic shape changes, as described above). That is, the 2D-S probe in situ data include measurements and estimates for ice particle projected area and mass, respectively, as noted above.

MADA optical properties are calculated directly from these in situ area and mass values, thus largely avoiding the need for shape assumptions. Thirdly, this retrieval is most sensitive to the smaller ice particles in a PSD where the variance in ice particle shape is minimal (Baker and Lawson, 2006b; Lawson et al., 2006b; Woods et al., 2018).

These shapes tend to be relatively simple, like quasi-spherical or compact irregular, whereas larger ice particles tend to have more complex shapes. During the SPARTICUS campaign, many cirrus clouds were sampled so that biases in ice particle shape are less likely to occur.

2.4 Impact of the smallest size bin in PSD measurements

The relationships shown in Figs. 2 and 3 were derived by using all the size bins of the 2D-S probe. However, the data in the smallest size bin (5–15 μm) has the greatest uncertainty since the sample volume of the 2D-S probe depends on particle size, and this volume is smallest (with greatest measurement uncertainty) for the smallest size-bin (Gurganus and Lawson, 2018). This motivated us to formulate the retrieval relationships in two ways: (1)

assuming that the PSD first size-bin, $N(D)_1$, is valid and is therefore unmodified (as considered earlier), and (2) assuming that $N(D)_1$ equals zero. Physical reasons to argue in favor of the unmodified $N(D)_1$ assumption or the $N(D)_1 = 0$ assumption are discussed under Supplementary Materials. Jensen et al. (2013a) argues that $N(D)_1$ as measured by the 2D-S probe is anomalously high since it tends to be considerably higher than the adjacent size bins [$N(D)_2$ for example] and that these small ice crystals should rapidly grow or sublimate to larger or smaller sizes ($> 15 \mu\text{m}$ or $< 5 \mu\text{m}$) due to the relative humidity with respect to ice, RH_i , being significantly different than ice saturation ($\text{RH}_i = 100\%$). Therefore, $N(D)_1 > N(D)_2$ would imply frequent ice nucleation events to sustain these higher $N(D)_1$ values, which appears unlikely. This argument provided an additional incentive to formulate this retrieval using assumption (2).

However, there are also physical reasons that argue in favor of the first assumption [$N(D)_1$ is a valid measurement].

For example, if strong competition for water vapor due to a relatively high small ice crystal concentration (e.g. due to a homogeneous ice nucleation event) rapidly reduces the RH_i to $\sim 100\%$, then this relatively high concentration

may last for time periods comparable to the lifetime of the cirrus cloud. High ice crystal concentrations (~ 300 to $10,000 \text{ L}^{-1}$) associated with $\text{RH}_i \sim 100\%$ were documented by aircraft measurements in the tropical tropopause layer (TTL), existing in layers ranging from meters to 0.4 km in depth (Jensen et al., 2013b). These layers were embedded within a deeper cirrus cloud having N typically less than 20 L^{-1} (where RH_i was higher). Evidence that RH_i near 100% is common in cirrus clouds is shown in Figs. 6 and 7 of Krämer et al. (2009), where for the relationships most representative of cirrus clouds, the relaxation time τ for RH_i to develop a quasi-steady state (i.e. dynamical equilibrium denoted RH_{qst}) is on the order of 5–10 minutes. RH_{qst} is where $d(\text{RH}_i)/dt \sim 0$, where the rate of vapor uptake by ice approximately balances the rate of supersaturation development. Between the time of initial in-cloud supersaturation (corresponding to cloud formation) and τ , the cloud updraft w tends to be higher than the w occurring after RH_{qst} is attained. Since cirrus cloud lifetimes tend to be considerably longer than 5–10 minutes, w is relatively low with $\text{RH}_i \sim 100\%$ for time $t > \tau$. This may explain the relatively high frequencies of occurrence of RH_i near 100% in Fig. 7 of Krämer et al. (2009). It can also be argued that cirrus clouds are formed by atmospheric wave activity, and that RH_i near 100% results from averaging transient wave-induced fluctuations of RH_i . However, Fig. 1 in Krämer et al. (2009) shows water vapor concentrations being fairly constant with time over periods of 30 to 50 minutes, while also showing evidence of wave-induced fluctuations in RH_i during another period.

Figure [S2 under Supplementary Materials](#) shows the N/IWC - β_{eff} ([top](#)) and D_e - β_{eff} ([bottom](#)) relationships derived from SPARTICUS and TC4 assuming $N(D)_1=0$. Assuming $N(D)_1=0$ not only reduces N , but it also reduces β_{eff} , especially when β_{eff} is large. For instance, assuming $N(D)_1=0$ reduces the maximum value of in situ β_{eff} from 1.6 to 1.24 during SPARTICUS, and from 1.44 to 1.27 during TC4. As a result, the N/IWC - β_{eff} relationships are fairly close for both assumptions. For example, assuming $N(D)_1=0$ reduces N/IWC by about 20% for the largest β_{eff} , thereby confirming the tight relationship between N/IWC and β_{eff} shown in Fig. 2. Assuming $N(D)_1=0$ has a larger impact on the D_e - β_{eff} relationship, [as shown in Fig. S2](#). For SPARTICUS, the largest relative change in D_e is around $\beta_{\text{eff}}=1.15$, where D_e is reduced by 26% assuming $N(D)_1=0$ instead of $N(D)_1$ unmodified. For TC4, the largest relative reduction in D_e is 38% at $\beta_{\text{eff}}=1.2$. [It is noted that the \$D_e\$ - \$\beta_{\text{eff}}\$ relationships used in the IIR Version 3 operational algorithm \(Fig. 3a in Garnier et al., 2013\) tend to yield smaller \$D_e\$ than the relationships derived from these PSD measurements, largely because they were computed with no size distribution.](#)

The unmodified $N(D)_1$ assumption can be viewed as an upper limit while the $N(D)_1 = 0$ assumption is clearly a lower limit for the actual value of $N(D)_1$, and in this way our relationships are bracketed by these two limiting conditions. The comparison of in situ β_{eff} for each of these assumptions with β_{eff} derived independently from CALIPSO IIR is an additional piece of information, as discussed in Sect. 4.

Uncertainties regarding the photon tunneling efficiency (T_e) values assumed in Sect. 2.3 were evaluated, but these were much smaller than the uncertainties described above. For example, assuming bullet rosettes for all sizes results

in T_e values of 0.70, 0.40 and 0.15 for the three size categories considered (from smallest to largest). Among plausible crystal shape assumptions, this assumption yielded the lowest T_e values, reducing β_{eff} by no more than 2.6%. Note that for $D < 80 \mu\text{m}$, > 85% of the ice particles tend to be irregular (e.g. blocky or quasi-spherical) or spheroidal in shape (Lawson et al., 2006b; Baker and Lawson, 2006b), and these shapes should correspond to relatively high T_e values (Mitchell et al., 2006).

2.5 Retrieving N from CALIPSO β_{eff}

Using cloud layer average β_{eff} derived from IIR and CALIOP observations (Sect. 2.2), the N/IWC- β_{eff} - and D_e - β_{eff} relationships established from in situ measurements (Sect. 2.3 and 2.4) are used to derive the cloud layer N from CALIPSO, as presented in this sub-section. The sensitivity ranges ($N/\text{IWC} > \sim 10^7 \text{ g}^{-1}$ & $D_e < 90\text{-}110 \mu\text{m}$) are usually compatible with cirrus clouds ($T < -38^\circ\text{C}$) since PSDs tend to be narrower at these temperatures, containing relatively small ice particles (e.g. Mishra et al., 2014). ~~We choose to use the relationships established during SPARTICUS and TC4, assuming measured unmodified $N(D)_1$ (Figs. 2 and 3) and $N(D)_1=0$ (Fig. 4), as discussed above. Using these four relationships - Contrasting the SPARTICUS and TC4 relationships for both $N(D)_1$ assumptions~~ provides a means of estimating the uncertainty in N resulting from regional differences in cirrus microphysics.

Using CALIPSO β_{eff} and the N/IWC- β_{eff} and D_e - β_{eff} relationships, the cloud layer N is retrieved as: $N = \text{IWC} \times (N/\text{IWC})$ with IWC computed as:

$$\text{IWC} = \frac{\rho_i}{3} \times \alpha_{\text{ext}} \times D_e \quad (6)$$

where ρ_i is the bulk density of ice (0.917 g cm^{-3}), and α_{ext} is the effective layer-average visible extinction coefficient, which is derived from CALIOP and IIR as:

$$\alpha_{\text{ext}} = \frac{(2 / Q_{\text{abs,eff}}(12 \mu\text{m}))}{\Delta z_{\text{eq}}} \times \tau_{\text{abs}}(12.05 \mu\text{m}). \quad (7)$$

The quantity $2/Q_{\text{abs,eff}}(12 \mu\text{m})$, where 2 is the value of \bar{Q}_{ext} for ice PSDs in the visible spectrum, converts $\tau_{\text{abs}}(12.05 \mu\text{m})$ to an equivalent visible extinction optical depth (OD). It is obtained from β_{eff} as illustrated in Fig. 4 for the unmodified $N(D)_1$ assumption, and for the $N(D)_1=0$ assumption in Fig. [S3 \(under Supplementary Materials\)](#). For a given β_{eff} , $2/Q_{\text{abs,eff}}(12 \mu\text{m})$ is smaller by less than 7 % assuming $N(D)_1=0$, and consequently, IIR α_{ext} is also smaller by less than 7 %. The effective cloud thickness, Δz_{eq} , accounts for the fact that the IIR instrument does not sense equally all of the cloud profile that contributes to thermal emission. This is ~~accounted for~~ ~~taken into account~~

through the IIR weighting profile introduced in Sect. 2.2.3, which gives more weight to large emissivity and therefore to the large extinctions in the cloud profile. Using the IIR weighting profile, the layer absorption coefficient $\alpha_{\text{abs}}(12.05 \mu\text{m})$ for the IIR 12.05 μm channel is computed from the weighted averaged absorption coefficient profile. This yields $\alpha_{\text{abs}}(12.05 \mu\text{m}) > \alpha_{\text{abs,mean}}(12.05 \mu\text{m})$, where $\alpha_{\text{abs,mean}}(12.05 \mu\text{m})$ is the mean absorption coefficient, that is, the ratio of $\tau_{\text{abs}}(12.05 \mu\text{m})$ to the CALIOP layer geometric thickness, Δz . Thus, an equivalent effective thickness is defined as Δz_{eq} where $\alpha_{\text{abs}}(12.05 \mu\text{m}) = \tau_{\text{abs}}(12.05 \mu\text{m})/\Delta z_{\text{eq}}$, or alternatively, $\Delta z_{\text{eq}} = \Delta z (\alpha_{\text{abs,mean}}(12.05 \mu\text{m})/\alpha_{\text{abs}}(12.05 \mu\text{m}))$. In practice, Δz_{eq} is found equal to 30% to 90 % of Δz .

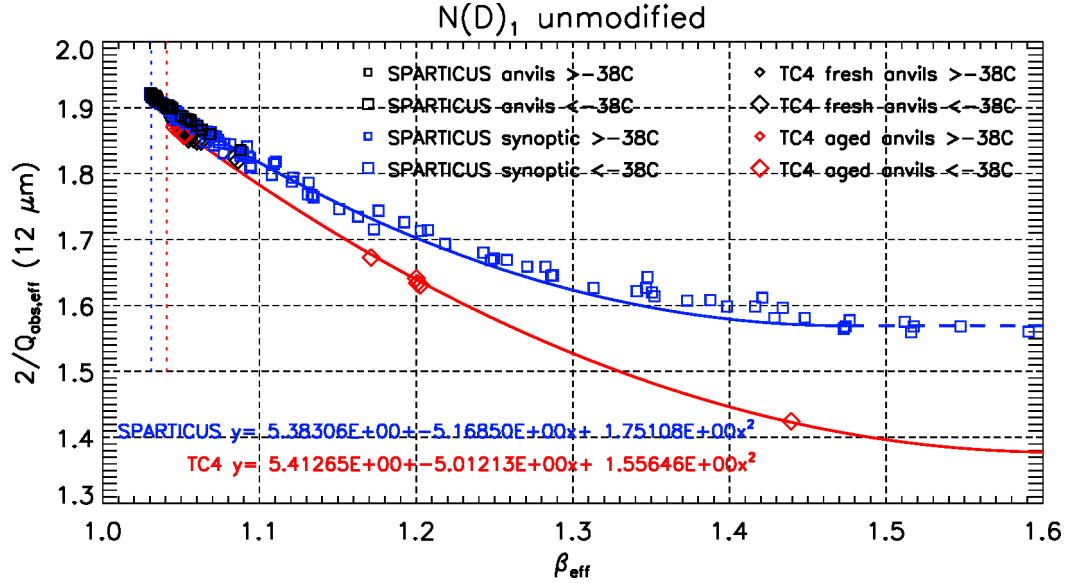


Figure 4: The β_{eff} dependence of the term that converts the absorption optical depth τ_{abs} into visible optical depth in Eq. 7, based on PSDs from SPARTICUS synoptic cirrus (blue squares) and anvils (black squares), and TC4 aged (red diamonds) and fresh (black diamonds) anvils, where the first size-bin is included ($N(D)_1$ unmodified). The larger (smaller) symbols denote PSDs measured at a temperature colder (warmer) than -38°C . The curve-fit equations are for SPARTICUS synoptic cirrus (blue) and for TC4 aged and fresh anvils (red). The dashed lines are where the curve-fit equations are extrapolated (see Table 1).

To summarize, the retrieval equation is:

$$N = \frac{\rho_i}{3} \times \frac{\left[\frac{2}{Q_{\text{abs,eff}}(12\mu\text{m})} \right] \cdot \tau_{\text{abs}}(12.05\mu\text{m})}{\Delta z_{\text{eq}}} \times D_e \times \left(\frac{N}{IWC} \right) \quad (8)$$

The retrieval of $\tau_{\text{abs}}(12.05 \mu\text{m})$ and $\tau_{\text{abs}}(10.6 \mu\text{m})$ combined with the CALIOP extinction profile provides β_{eff} , N/IWC , D_e , α_{ext} , and therefore layer-average IWC and finally layer N . Perhaps the most unique aspect of this retrieval method is its sensitivity to small ice crystals via β_{eff} .

3 Applying the retrieval method

3.1 Regression curves

Regression curves derived from SPARTICUS and TC4 for both assumptions [$N(D)_1$ is unmodified and $N(D)_1 = 0$] for the quantities N/IWC , D_e and $2/Q_{abs,eff}(12 \mu m)$ are given in Table 1, constituting the four formulations of this

retrieval scheme. A number of adjustments of the second-order polynomials were needed to provide retrievals for any value of β_{eff} . They correspond to the dashed lines in Fig. 4 (blue bottom), Fig S2 (bottom) and in Fig. S35. For the $N(D)_1=0$ assumption and $\beta_{eff} > 1.22$, $1/D_e$ is extrapolated linearly from the tangent value at $\beta_{eff}=1.22$. For the $N(D)_1$ unmodified assumption, when $\beta_{eff} > 1.48$, then $2/Q_{abs,eff}(12 \mu m)$ is set to 1.57. For the $N(D)_1=0$ assumption,

When calculating N/IWC , D_e , and $2/Q_{abs,eff}(12 \mu m)$ from β_{eff} , if the retrieved β_{eff} is less than the lower sensitivity limit, then β_{eff} is set to this value. For instance, N/IWC corresponding to this value via the regression curves is about $2.3 \times 10^5 g^{-1}$. As shown in Table 2 for the year 2013, this practice affected 15% and 18% of the N/IWC and D_e retrievals over ocean and land, respectively, when using the SPARTICUS relationships, for which the lower sensitivity limit is $\beta_{eff}=1.031$. Using the TC4 relationships, 20% and 22% of the samples over ocean and land, respectively, had β_{eff} smaller/larger than the sensitivity limit of 1.044. To better estimate the median values and percentiles for N and D_e , N and D_e retrievals calculated using these limiting values are accounted for when determining these statistics.

The retrieved N for the four formulations of the retrieval scheme can be compared through the product of the three β_{eff} -dependent quantities, N/IWC , D_e and $2/Q_{abs,eff}(12 \mu m)$, as shown in Fig. 5. The upper and lower bounds are the SPARTICUS $N(D)_1$ unmodified and TC4 $N(D)_1=0$ formulations, which differ by about a factor 2 in retrieved N for $\beta_{eff} > 1.06$, and by up to a factor 10 when β_{eff} is between the SPARTICUS (1.031) and TC4 (1.044) sensitivity limits.

3.2 Retrieval uncertainties

The inter-relationship between β_{eff} and α_{ext} , IWC , and N is illustrated in the Supplementary Materials under “Relationship between β_{eff} , α_{ext} , IWC and N ”, and in Fig. S4 and S5. After re-writing Eq. (8) as a function of β_{eff} and $\tau_{abs}(12.05 \mu m)$ using the regression curves shown in Fig. 5, the uncertainty ΔN is computed by propagating the errors in β_{eff} (see Sect. 2.2.4) and in $\tau_{abs}(12.05 \mu m)$, assuming a negligible error in ΔZ_{eq} (Eq. (7)) and in the relationships. Errors in the relationships create additional systematic uncertainties, as discussed in Sect. 3.1. More details about the uncertainty analysis and the equations used to compute ΔN can be found in the appendix.

Figure S4 (bottom row) shows $\Delta N/N$ against β_{eff} for the same samples as in the top row of Fig. S4. $\Delta N/N$ decreases as β_{eff} increases, reflecting that the technique is sensitive to small crystals. $\Delta N/N$ is found most of the time < 0.50

Table 1. Regression curve variables and coefficients for second-order polynomials of the form $y = a_0 + a_1 x + a_2 x^2$, used in the CALIPSO retrieval. Units for N/IWC and D_e are in g^{-1} and microns, respectively.

N(D) ₁	y	x	a ₀	a ₁	a ₂
SPARTICUS – Synoptic cirrus					
unmodified	N/IWC	β _{eff}	1.77387×10 ⁹	-3.86572×10 ⁹	2.08090×10 ⁹
unmodified	1/D _e	β _{eff}	-0.0829258	0.0904009	0.00161429
unmodified	2/Q _{abs,eff}	β _{eff} <1.476	5.38306	-5.16850	1.75108
	(12 μm)	β _{eff} >1.476	1.56921	0	0
= 0	N/IWC	β _{eff}	1.22741×10 ⁹	-2.82554×10 ⁹	1.58618×10 ⁹
= 0	1/D _e	β _{eff} <1.22	-0.410624	0.643702	-0.226492
		β _{eff} >1.22	-0.0735133	0.0910615	0
= 0	2/Q _{abs,eff}	β _{eff} <1.293	10.4347	-13.7382	5.31083
	(12 μm)	β _{eff} >1.293	1.55011	0	0
TC4 – Aged and fresh anvils					
unmodified	N/IWC	β _{eff}	2.71399e+09	-5.47770e+09	2.75779e+09
unmodified	1/D _e	β _{eff}	-0.0744685	0.0589313	0.0203374
unmodified	2/Q _{abs,eff}	β _{eff} <1.61	5.41265	-5.01213	1.55646
	(12 μm)	β _{eff} >1.61	1.37763	0	0
= 0	N/IWC	β _{eff}	1.42952e+09	-3.14430e+09	1.70038e+09
= 0	1/D _e	β _{eff} <1.5	-0.396886	0.550041	-0.154148
		β _{eff} >1.5	-0.0500520	0.0875957	0
= 0	2/Q _{abs,eff}	β _{eff} <1.319	11.2409	-14.8504	5.62970
	(12 μm)	β _{eff} >1.319	1.44756	0	0

- 5 $N(D)_1$ unmodified: SPARTICUS: if $\beta_{eff} < 1.031$, then $x=1.031$; TC4: if $\beta_{eff} < 1.04085$, then $x=1.04085$
 $N(D)_1=0$: SPARTICUS: if $\beta_{eff} < 1.03078$, then $x=1.03078$; TC4: if $\beta_{eff} < 1.04410$, then $x=1.04410$

Table 2. Fraction of samples with IIR β_{eff} larger than the N/IWC and D_e sensitivity limit, per season in 2013, over ocean and over land.

Fraction Samples	DJF		MAM		JJA		SON	
	$\beta_{eff} > 1.031$	$\beta_{eff} > 1.044$	$\beta_{eff} > 1.031$	$\beta_{eff} > 1.044$	$\beta_{eff} > 1.031$	$\beta_{eff} > 1.044$	$\beta_{eff} > 1.031$	$\beta_{eff} > 1.044$
Sea	0.86	0.82	0.85	0.80	0.84	0.78	0.85	0.79
Land	0.83	0.79	0.81	0.77	0.79	0.75	0.84	0.80

when $\beta_{\text{eff}} > 1.30$, then $2/Q_{\text{abs,eff}}(12 \mu\text{m})$ is set to 1.55. Furthermore, when calculating N/IWC, D_e , and $2/Q_{\text{abs,eff}}(12 \mu\text{m})$ from β_{eff} , if the retrieved β_{eff} is less than the lower sensitivity limit, then β_{eff} is set to this value. For instance, N/IWC corresponding to this value via the regression curves is about $2.3 \times 10^5 \text{ g}^{-1}$. As shown in Table 2 for the year 2013, this practice affected 15% and 18% of the N/IWC and D_e retrievals over ocean and land, respectively, when using the SPARTICUS relationships, for which the lower sensitivity limit is $\beta_{\text{eff}} = 1.031$. Using the TC4 relationships, 20% and 22% of the samples over ocean and land, respectively, had β_{eff} larger than the sensitivity limit of 1.044. To better estimate the median values and percentiles for N and D_e , N and D_e retrievals calculated using these limiting values are accounted for when determining these statistics.

The retrieved N for the four formulations of the retrieval scheme can be compared through the product of the three β_{eff} -dependent quantities, N/IWC, D_e and $2/Q_{\text{abs,eff}}(12 \mu\text{m})$, as shown in Fig. 6. The upper and lower bounds are the SPARTICUS $N(D)_1$ unmodified and TC4 $N(D)_1 = 0$ formulations, which differ by about a factor 2 in retrieved N for $\beta_{\text{eff}} > 1.068$, and by up to a factor 10 when β_{eff} is between the SPARTICUS (1.031) and TC4 (1.044) sensitivity limits.

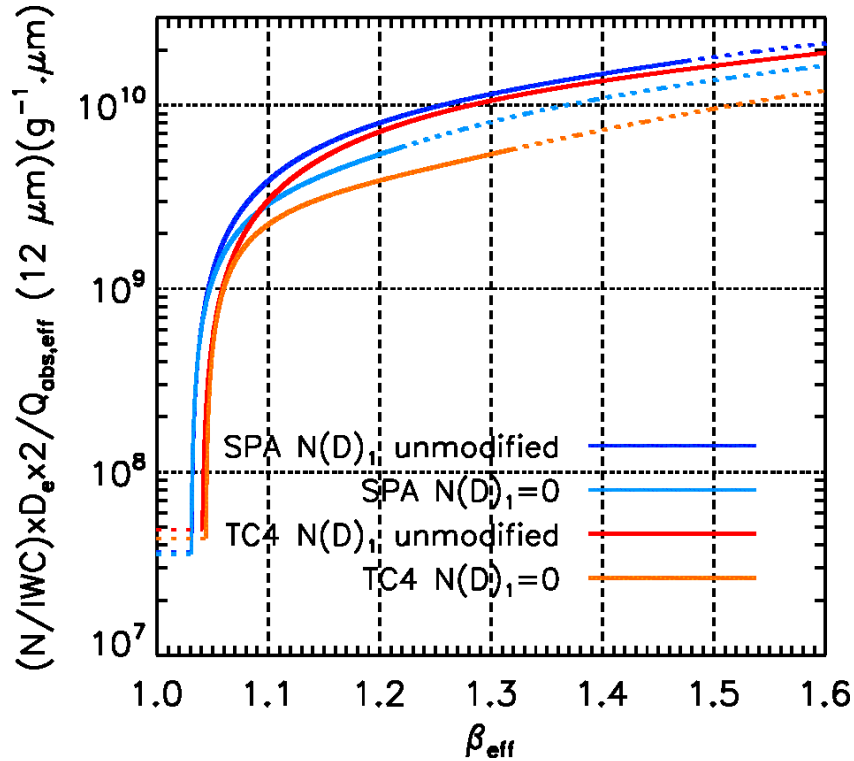


Figure 56: Comparison of ice particle number concentration N derived from CALIPSO β_{eff} using the four formulations of the retrieval scheme, derived from SPARTICUS using the $N(D)_1$ unmodified (navy blue) and $N(D)_1 = 0$ (light blue) assumptions, and from TC4 using the unmodified (red) and $N(D)_1 = 0$ (orange) assumptions. The dashed lines are where the curve-fit equations are extrapolated (see Sect. 3 and Table 1).

3.2—Relationship between β_{eff} , α_{ext} , IWC, and N

As seen from Eq. (6), (7) and (8), β_{eff} and α_{ext} are the two key parameters retrieved from the CALIPSO HR to derive N/IWC, IWC, and finally N. The interrelationship between β_{eff} , α_{ext} , IWC, and N is illustrated in Fig. 7 (top row) for the SPARTICUS relationships using the unmodified $N(D)_1$ assumption, which also shows the range encountered for these properties in the selected cloud population. The red dashed lines are where $N = 100 \text{ L}^{-1}$, 500 L^{-1} and 1000 L^{-1} . The pink dashed lines are where $\text{IWC} = 0.5 \text{ mg g}^{-3}$, 5 mg g^{-3} , or 30 mg g^{-3} . The horizontal red dotted lines for $\beta_{\text{eff}} < 1.031$ (or $D_e > 83 \mu\text{m}$) indicate where the retrieval is not sensitive to N/IWC. For $\beta_{\text{eff}} < 1.031$, N/IWC is set to its limiting (minimum) value so that N is a priori overestimated in these conditions, but typically smaller than 100 L^{-1} . For $\beta_{\text{eff}} < 1.031$, D_e is set to $83 \mu\text{m}$, as denoted by the horizontal pink lines, and IWC is a priori underestimated for these conditions. For our data selection, α_{ext} is mostly between 0.05 km^{-1} and 5 km^{-1} . Large values of N ($> 500 \text{ L}^{-1}$) result from larger values of β_{eff} (yielding smaller D_e and much larger N/IWC) and sufficiently large values of α_{ext} so that IWC is sufficiently large for these small values of D_e . Low values of N ($< 100 \text{ L}^{-1}$) can be retrieved for small values of β_{eff} , yet larger than the low limit of 1.031, only if α_{ext} is sufficiently small. When α_{ext} is smaller than 0.05 km^{-1} , as could be encountered in the case of OD smaller than 0.3 (removed from the cloud sampling), finding N smaller than 100 L^{-1} is very likely.

3.3—Retrieval uncertainties

The inter relationship between β_{eff} and α_{ext} , IWC, and N is illustrated in the Supplementary Materials under “Relationship between β_{eff} , α_{ext} , IWC and N”, and in Fig. S4 and S5. After re-writing Eq. (8) as a function of β_{eff} and $\tau_{\text{abs}}(12.05 \mu\text{m})$ using the regression curves shown in Fig. 6, the uncertainty ΔN is computed by propagating the errors in β_{eff} (see Sect. 2.2.4) and in $\tau_{\text{abs}}(12.05 \mu\text{m})$, assuming a negligible error in ΔZ_{eq} (Eq. (7)) and in the relationships. Errors in the relationships create additional systematic uncertainties, as discussed in Sect. 3.1. More details about the uncertainty analysis and the equations used to compute ΔN can be found in the appendix.

Figure 7 (bottom row) shows $\Delta N/N$ against β_{eff} for the same samples as in Fig. 7 (top row). $\Delta N/N$ decreases as β_{eff} increases, reflecting that the technique is sensitive to small crystals. $\Delta N/N$ is found most of the time < 0.50 for $\beta_{\text{eff}} > 1.15$, but increases up to more than 2.0 as β_{eff} decreases and approaches the sensitivity limit. For a given value of β_{eff} , the variability of $\Delta N/N$ is due to the variability of $\Delta \beta_{\text{eff}}/\beta_{\text{eff}}$ and of $\Delta \alpha_{\text{ext}}/\alpha_{\text{ext}}$. $\Delta N/N$ is larger over land because of a larger estimated uncertainty in T_{BG} and also because the radiative contrast is sometimes relatively weak. $\Delta \beta_{\text{eff}}/\beta_{\text{eff}}$ is mostly due to random measurement errors, because systematic errors associated with the retrieval of $\tau_{\text{abs}}(12.05 \mu\text{m})$ and $\tau_{\text{abs}}(10.6 \mu\text{m})$ tend to cancel when these are ratioed to calculate β_{eff} . The uncertainty in T_{BG} contributes

more importantly to $\Delta\alpha_{\text{ext}}/\alpha_{\text{ext}}$ at the smallest emissivities. Uncertainty in T_{BB} is not a major contributor for semi-transparent clouds of small to medium emissivity.

We repeated this analysis except using an OD threshold of 0.1 (instead of 0.3; see also Sect. 5.1). Figure S5 shows this same analysis except that the sample selection criteria for minimum OD was changed from 0.30 to 0.10. In the lower row relating $\Delta N/N$ to β_{eff} , the number of samples having $\Delta N/N > 1.0$ has substantially increased over both ocean and land relative to Fig. S4 due to the lower OD threshold for sample selection, and more samples correspond to lower values of α_{ext} , IWC, and N.

4. Comparison with in situ cirrus cloud measurements

Because our analysis is applied to IIR β_{eff} using relationships with in situ β_{eff} established from the SPARTICUS and TC4 field campaigns, a quantitative comparison of IIR and in situ β_{eff} from these two campaigns is first discussed. Secondly, retrieved N/IWC are compared against in situ N/IWC measured from many field campaigns -using the dataset of Krämer et al. (2009), where N and IWC are measured independently. The last step of this evaluation is to compare retrieved and in situ D_e , IWC and N.

~~4.1 Comparing IIR and in situ β_{eff} during SPARTICUS and TC4 – Impact of the smallest size bin in PSD measurements~~

~~For SPARTICUS, the CALIPSO retrievals were restricted to the relevant domain (latitude range 31°N–42°N and longitude range 90°W–103°W) and they were obtained from January through April of 2010 during daytime since this period contained only synoptic cirrus based on the SPARTICUS flights. For TC4, the CALIPSO retrievals were restricted to the TC4 spatial and temporal domain (latitude range 5°S–15°N and longitude range 80°W–90°W) over oceans, in July and August 2007. CALIPSO retrievals are cloud layer average properties, and we use T_e (see Sect. 2.2.3) as our best characterization of the representative cloud temperature. Finally, for each campaign, in situ β_{eff} is computed for both $N(D)_1$ assumptions.~~

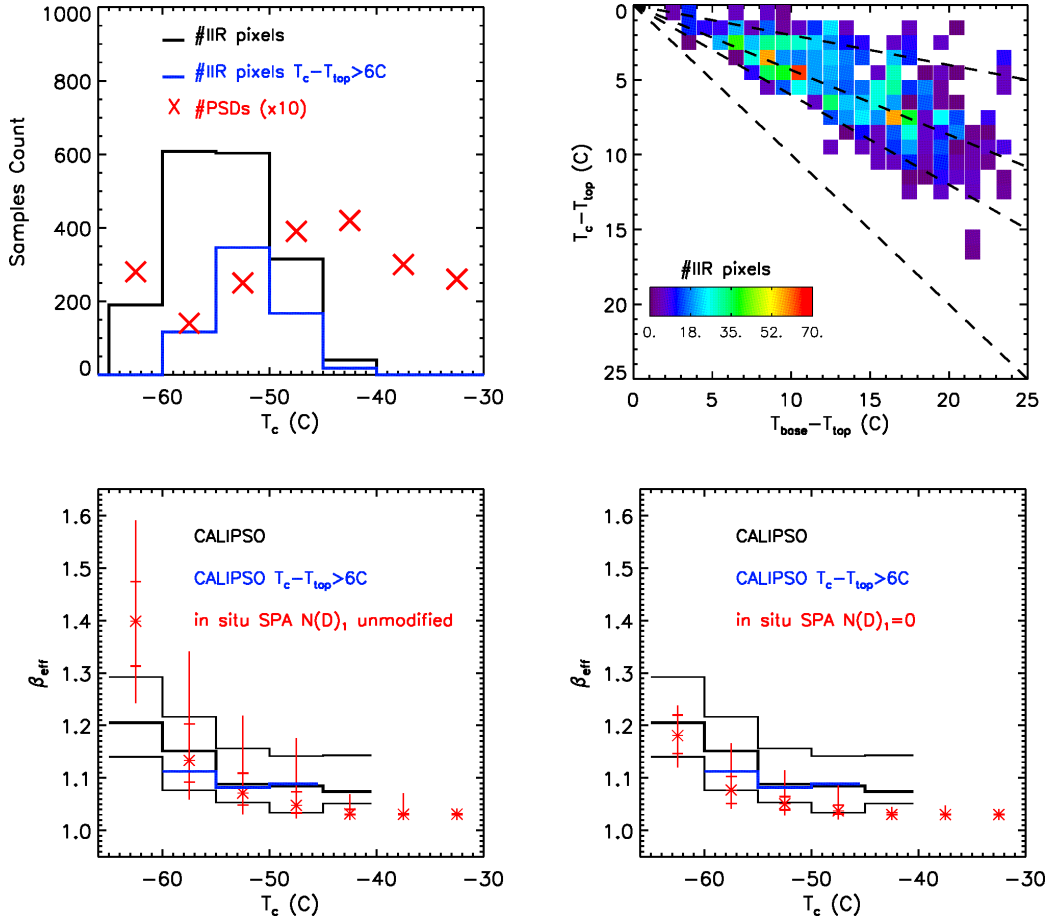


Figure 68a: Statistical comparison of CALIPSO IIR β_{eff} and ~~and~~ in situ β_{eff} (synoptic cirrus) during SPARTICUS (sampled January-April 2010). Top left: The red X symbols indicate the number of PSDs used in the SPARTICUS analysis (multiplied by 10 for clarity of presentation) within 5°C in situ temperature intervals, while the black histogram indicates the number of CALIPSO IIR pixels in each T_c temperature interval, and the blue histogram indicates the number of IIR pixels where $T_c - T_{\text{top}}$ is larger than 6°C . Top right: 2D-histograms of $T_c - T_{\text{top}}$ and $T_{\text{base}} - T_{\text{top}}$ for the IIR pixels. The dashed lines from top to bottom are 20%, 43% (average value) and 60% from cloud top, with $T_c = T_{\text{base}}$ for the lowest curve. Bottom: Temperature dependence of in situ β_{eff} derived assuming $N(D)_1$ unmodified (left) and $N(D)_1=0$ (right). The vertical lines give the measurement range, the horizontal bars give the 25th and 75th percentile values, and the asterisks give the median value. CALIPSO IIR median β_{eff} is given by the thick black histogram, with thin black histograms giving the 25th and 75th percentile values. The thick blue histogram is CALIPSO IIR median β_{eff} where $T_c - T_{\text{top}} > 6^\circ\text{C}$.

4.1 Comparing IIR and in situ β_{eff} during SPARTICUS and TC4 - Impact of the smallest size bin in PSD measurements

For SPARTICUS, the CALIPSO retrievals were restricted to the relevant domain (latitude range 31°N - 42°N and longitude range 90°W - 103°W) and they were obtained from January through April of 2010 during daytime since this period contained only synoptic cirrus based on the SPARTICUS flights. For TC4, the CALIPSO retrievals were restricted to the TC4 spatial and temporal domain (latitude range 5°S - 15°N and longitude range 80°W - 90°W) over

oceans, in July and August 2007. CALIPSO retrievals are cloud layer average properties, and we use T_c (see Sect. 2.2.3) as our best characterization of the representative cloud temperature. Finally, for each campaign, in situ β_{eff} is computed for both $N(D)_1$ assumptions. Data analysis is performed on a statistical basis, as coincident in situ and satellite data only provide a very small dataset due to our data selection.

Comparisons with SPARTICUS are reported in Fig. 68a. The upper left panel shows the number of IIR pixels and in situ PSDs per 5°C temperature-bin. As seen in the upper right panel of Fig. 86a, the difference between the temperatures at cloud base (T_{base}) and cloud top (T_{top}) is smaller than 20°C for most of the clouds selected in the CALIPSO retrievals. Furthermore, T_c tends to be located in the upper part of the cloud layer, at 20 % to 60 % from the top, and ~~in-between~~, on average at 43%, where the maximum number of IIR observations are found (see dashed black lines). This means that IIR β_{eff} is a weighted measure near the middle of the cloud layer, slightly towards the upper part. A 5°C interval is considered in the analysis in accordance with the dispersion observed in the temperature distributions (Fig. 68a top right). Because IIR pixels are required to correspond to clouds of T_{base} less than -38°C, T_c is mostly colder than -45°C, and the number of samples between -45°C and -40°C is relatively small (Fig. 68a, upper left panel). It is also seen that $T_c - T_{\text{top}}$ is smaller than 6°C for the majority of the IIR pixels overall, and for all pixels with $T_c < -60^\circ\text{C}$. The temperature dependence of median IIR β_{eff} and of the 25th and 75th percentile values is given by the black histograms in the lower panels of Fig. 68a. The thick blue histogram shows median β_{eff} when $T_c - T_{\text{top}}$ is larger than 6°C between -60°C and -45°C where the number of samples is sufficient (>20) for a meaningful analysis. This smaller median β_{eff} (in blue) illustrates the sensitivity of β_{eff} to distance from cloud top and cloud depth and indicates a smaller fraction of small ice crystals in the PSD for larger distances.

The temperature dependence of IIR β_{eff} is compared with the dependence of in situ β_{eff} derived by taking $N(D)_1$ unmodified (lower left panel) and $N(D)_1=0$ (lower right panel). The red asterisks denote the median in situ values, the red horizontal bars indicate the 25th and 75th percentile values and the vertical red lines indicate the range of values. Median IIR β_{eff} and in situ β_{eff} exhibit a similar temperature variation. A few issues should be kept in mind when interpreting these comparisons. First, regarding the unmodified version of $N(D)_1$ for $T < -60^\circ\text{C}$, the relatively high median in situ values for β_{eff} and N (ranging from 513 to 2081 L^{-1}) come from two flights on 28 April 2010 that sampled the ridge-crest cirrus mentioned above, raising some concern whether this single cirrus event was representative at these temperatures. A non-representative cirrus event could explain the larger median in situ β_{eff} of 1.4 relative to the median IIR β_{eff} of 1.2 for $T < -60^\circ\text{C}$. Moreover, $N(D)_1$ for these April 28th PSD samples contributed 78% of the total N on average. But this would not violate our understanding of cloud physics if the RH_i was near 100%. That is, high N having very small crystal sizes can exist for long periods when $\text{RH}_i \sim 100\%$ since little ice crystal growth or sublimation can occur then (see Supplementary Materials). Second, aircraft sampling at warmer temperatures may not be representative of satellite layer average retrieval at T_c generally located in the

upper part of the cloud if the cirrus layer is relatively deep with aircraft sampling relatively low in the cloud. For such conditions, the [aircraft](#) sampled ice particles would have relatively long growth times through vapor deposition and aggregation, producing relatively large ice particles and lower β_{eff} and N in the lower cloud. This may have been the case for $T > -50^\circ\text{C}$. This point is illustrated by the in situ measurements and modeling study of Mitchell et al. (1996**b**) where a Lagrangian spiral descent through a cirrus layer was simulated with a steady-state snow growth model for vapor deposition and aggregation. Aggregation in the lower cloud was predicted to reduce N by $\sim 60\%$. The larger dispersion of the IIR β_{eff} distribution seen through the difference between the 25th and the 75th percentile values (thin black histograms) can be explained by the uncertainties reported in Table 3.

Following the same approach as for SPARTICUS, Fig. 6b compares CALIPSO retrievals and TC4 in situ data. The IIR representative temperature is again close to mid-cloud. For temperatures between -69°C and -45°C , where CALIPSO and in situ β_{eff} can be compared, most of the CALIPSO selected cirrus clouds have $T_c - T_{\text{top}}$ larger than 6°C (Fig. 6b, upper left panel) in contrast to SPARTICUS cirrus. The in situ and IIR β_{eff} are in better agreement when in situ β_{eff} is computed without the first size bin ($N(D)_1=0$), especially at the coldest temperatures. The largest in situ β_{eff} at -69°C is in fair agreement with IIR β_{eff} in the neighboring temperature range between -75°C and -70°C .

To conclude, we find that despite the a priori different range of cloud optical depths (because TC4 data are from aged and fresh anvils), CALIPSO and in situ β_{eff} are in better agreement for TC4 when the latter are computed using $N(D)_1 = 0$, most of the time within 0.01-0.02.

Table 3: IIR median β_{eff} (see Fig. 68a) and estimated uncertainty $\Delta\beta_{\text{eff}}$ during SPARTICUS. See Sect. 2.2.4 and appendix for details.

T_c (°C)	Samples Count	Median β_{eff}	Median $\Delta\beta_{\text{eff}}$ Measurement	Median $\Delta\beta_{\text{eff}}$ Background	Median $\Delta\beta_{\text{eff}}$ Blackbody	Median $\Delta\beta_{\text{eff}}$ All
-62.5	190	1.206	0.048	0.062	0.0018	0.079
-57.5	608	1.151	0.039	0.037	0.0020	0.057
-52.5	603	1.088	0.034	0.023	0.0027	0.044
-47.5	315	1.085	0.040	0.026	0.0024	0.050
-42.5	40	1.074	0.045	0.030	0.0029	0.054

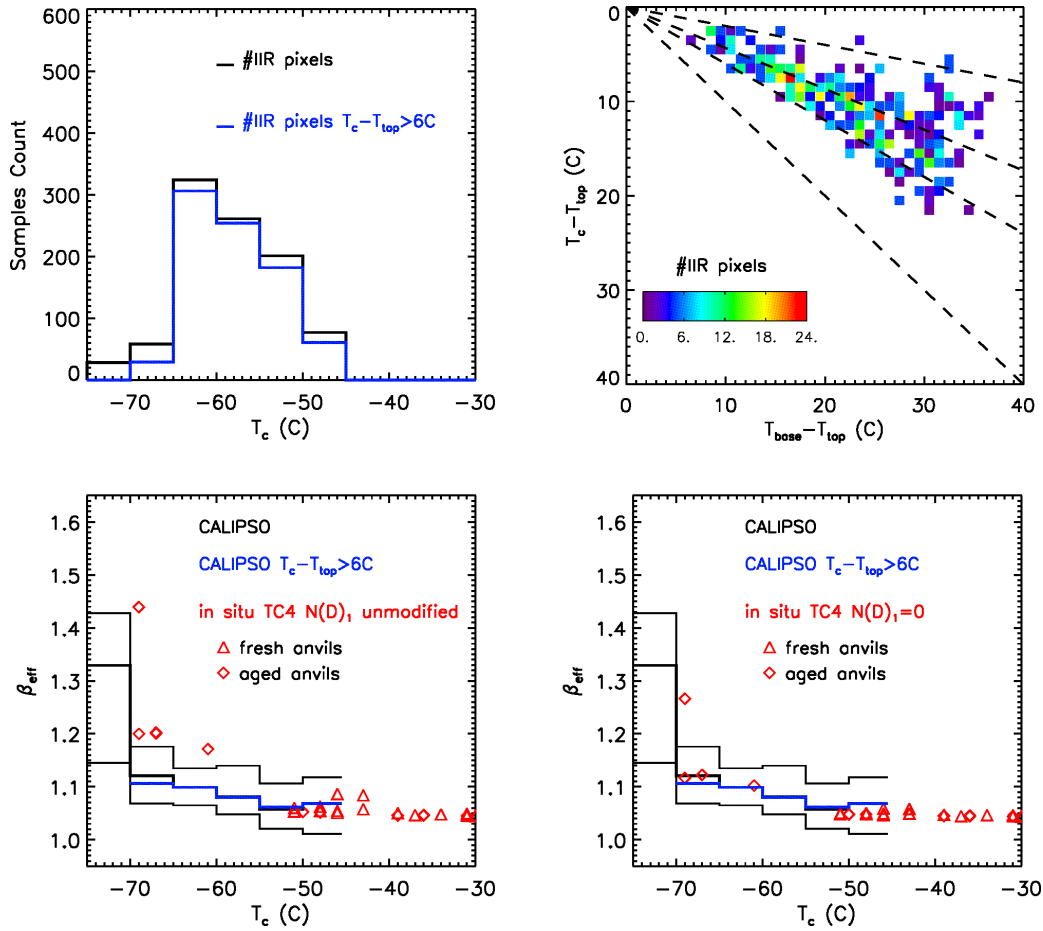


Figure 68b: Statistical comparison of CALIPSO IIR β_{eff} and in situ β_{eff} during TC4. Top left: The black histogram indicates the number of CALIPSO IIR pixels in each T_c temperature interval, and the blue histogram indicates the number of IIR pixels where $T_c - T_{\text{top}}$ is larger than 6°C . Top right: 2D-histograms of $T_c - T_{\text{top}}$ and $T_{\text{base}} - T_{\text{top}}$ for the IIR pixels. The dashed lines from top to bottom are 20%, 43% and 60% from cloud top, with $T_c = T_{\text{base}}$ for the lowest curve, for comparison with SPARTICUS data. Bottom: Temperature dependence of in situ β_{eff} derived assuming $N(D)_1$ unmodified (left) and $N(D)_1=0$ (right) for fresh (red triangles) and aged (red diamonds) anvils. CALIPSO IIR median β_{eff} is given by the thick black histogram, with thin black histograms giving the 25th and 75th percentile values. The thick blue histogram is CALIPSO IIR median β_{eff} where $T_c - T_{\text{top}} > 6^\circ\text{C}$.

Following the same approach as for SPARTICUS, Figs. 8b compares CALIPSO retrievals and TC4 in situ data. The IIR representative temperature is again close to mid cloud. For temperatures between -69°C and -45°C , where CALIPSO and in situ β_{eff} can be compared, most of the CALIPSO selected cirrus clouds have $T_e - T_{\text{top}}$ larger than 6°C (Fig. 8b, upper left panel) in contrast to SPARTICUS cirrus. The in situ and IIR β_{eff} are in better agreement when in situ β_{eff} is computed without the first size bin ($N(D)_1=0$), especially at the coldest temperatures. The largest in situ β_{eff} at -69°C is in fair agreement with IIR β_{eff} in the neighboring temperature range between -75°C and -70°C .

To conclude, we find that despite the a priori different range of cloud geometrical optical depths (because TC4 data are from aged and fresh anvils), CALIPSO and in situ β_{eff} are in better agreement for TC4 when the latter are computed using $N(D)_1=0$, most of the time within 0.01–0.02.

4.2 Comparison of N/IWC with the Krämer cirrus dataset

Krämer et al. (2009) compiled coincident in situ measurements of N and IWC from 5 field campaigns (10 flights) between 68°N and 21°S latitude where N was measured by the FSSP probe and IWC was directly measured by various probes as described in Schiller et al. (2008). They report mass-weighted ice particle size R_{ice} derived from in situ measurements of IWC/N assuming ice spheres at bulk density (0.92 g cm^{-3}). Since R_{ice} can be inverted to yield in situ measurements of N/IWC, this offers the opportunity to evaluate the representativeness of the four N/IWC- β_{eff} relationships derived from the SPARTICUS and TC4 campaigns. Krämer et al. (2009) estimated that the FSSP measurements accounted for at least 80% (but typically $> 90\%$) of the total N in a PSD. These measurements were made at $T < 240 \text{ K}$ where PSD tend to be relatively narrow and ice particle shattering upstream of particle detection (i.e. the sample volume) is less of a problem (de Reus et al., 2009; Lawson et al., 2008). Moreover, the FSSPs used did not use a flow-straightening shroud in front of the inlet; a practice that will reduce the amount of shattering. The complete data set of in situ IWCs reported in Krämer et al. (2009) extends beyond the 10 field campaigns mentioned above, and this complete IWC data set is also described in Schiller et al. (2008).

Since this retrieval is sensitive to the smallest ice crystal sizes, it has the advantage of being sensitive to ice nucleation processes, but this also poses certain challenges. For example, the comparison of retrieved and measured N/IWC and N in cirrus clouds is necessarily ambiguous due to (1) the uncertainty in PSD probe measurements at the smallest sizes in a PSD [assuming the probe is capable of measuring N between roughly $5 \mu\text{m}$ and $50 \mu\text{m}$], (2) the PSD size range used to create the retrieval relationships relative to the PSD size range of the measurements used to test the retrieval, (3) the size range of the retrieved PSD (which is unknown), (4) in situ measurements in optically thin layers below the retrieval limit of the IIR for this study, and (5) the comparison of retrieved layer-averaged quantities to localized aircraft measurements, as discussed earlier. Regarding (2), since this retrieval was developed

from 2D-S probe in situ measurements, ideally it should be validated against 2D-S probe in situ measurements. Comparing with the Krämer et al. (2009) measurements introduces some ambiguity since the smallest size-bin of the 2D-S is from 5-15 μm whereas the Krämer et al. (2009) N measurements are based on the FSSP 100/300 that sampled particles in the size range 3.0–30/0.6–40 μm diameter, and ice crystals larger than this size range were not recorded. Moreover, the amount of additional uncertainty in the FSSP measurements due to the possibility of shattering was not quantified. In order to perform first comparisons, we thus used these in situ measurements of IWC/N and inferred β_{eff} using the four N/IWC vs. β_{eff} relationships previously obtained for SPARTICUS and TC4.

The comparisons between the Krämer et al. (2009) in situ values of N/IWC and the corresponding in situ derived values of β_{eff} (using the four N/IWC vs. β_{eff} relationships) with the CALIPSO IIR N/IWC (using these four relationships) and retrieved β_{eff} values are given in Fig. S6 under Supplementary Materials. In situ and IIR N/IWC values agree rather closely, with IIR N/IWC having only a weak dependence on the relationship used. Similarly, β_{eff} derived from in situ N/IWC (using these four relationships) is very consistent with the direct IIR retrieval of β_{eff} for $T > -82^\circ\text{C}$.

~~Although the cirrus cloud measurements in Krämer et al. (2009) occurred over both land and ocean, no distinction was made in this regard. But since CALIPSO IIR β_{eff} uncertainties are greater over land, Fig. 9 separates in situ and satellite retrievals of N/IWC and β_{eff} over ocean (top) and land (bottom). CALIPSO values are averaged over all seasons for 2013 and over the latitude range roughly corresponding to the field measurements (70°N to 25°S). Temperature intervals are 4°C .~~

~~Shown in the left panels is N/IWC vs. T_e . The N/IWC curve fits describing the in situ measurements of Krämer et al. (2009) are shown by the grey curves, and correspond to the maximum, minimum and middle (i.e. mid-point) value of a cloud property as a function of temperature. They are compared with corresponding retrieved median values, based on our four formulations: SPARTICUS unmodified $N(D)_i$ (solid navy blue), SPARTICUS $N(D)_i=0$ (solid light blue), TC4 unmodified $N(D)_i$ (solid red), and TC4 $N(D)_i=0$ (solid orange), all derived from IIR β_{eff} shown in the right panels (black curves). The dashed curves give the 25th and 75th percentile retrieval values. Using our four formulations, in situ N/IWC is converted into four in situ β_{eff} plotted in the right panels for comparison with IIR β_{eff} in black. Comparing both N/IWC and β_{eff} allows visualizing the non-linear relationship between N/IWC and β_{eff} .~~

~~Note that the Krämer et al. (2009) data used in Fig. 9 contain several non-zero bins between 5 and 15 microns (i.e. the 1st size bin of the 2DS probe). Thus, the in situ PSD do not conform with the $N(D)_i=0$ assumption. However, as discussed above, the retrieved N/IWC (left panels of Fig. 9) is weakly sensitive to the $N(D)_i$ assumption. Given the above ambiguities and uncertainties, the agreement between the median retrieved and in situ N/IWC is noticeable, especially for both SPARTICUS relationships over land. Both CALIPSO IIR and in situ median β_{eff} are~~

smaller than about 1.25 for temperatures greater than 203 K (-70°C), in agreement with CALIPSO IIR β_{eff} retrieved during SPARTICUS (Fig. 8a) and during TC4 (Fig. 8b).

4.3 Comparisons of D_e , IWC and N with the SPARTICUS, TC4 and Krämer datasets

The in situ and retrieved SPARTICUS cirrus cloud properties, namely D_e , α_{ext} , IWC and N, are shown in Figs. 407aa and Fig. S7+0b. They are both based on all size-bins of the 2D-S probe [$N(D)_1$ unmodified] in Fig. 407aa, while these same properties were calculated using the assumption that $N(D)_1 = 0$ in Fig. 40bS7. The data are presented using the same convention as for β_{eff} in Fig. 86a. For in situ data, the $N(D)_1$ assumption changes mostly N and the smallest D_e , but has a weaker impact on α_{ext} and IWC. For IIR, the changes result from the changes in the relationships with β_{eff} (Figs. 2-56). Using $N(D)_1 = 0$ instead of $N(D)_1$ unmodified increases notably the smallest in situ D_e and always decreases IIR D_e . The differences between in situ and IIR D_e increase with temperature as β_{eff} decreases and begins losing sensitivity to D_e at warmer temperatures. Median in situ and IIR layer average α_{ext} , which are both weakly sensitive to the $N(D)_1$ assumption, are within a factor less than 2. The notable larger variability of the in situ data is explained by the cloud local sampling in contrast to IIR average values. Reflecting the changes in D_e , IIR IWC is smaller using $N(D)_1 = 0$. Finally, using $N(D)_1 = 0$ reduces in situ N by a factor of 3 on average, but the change in the various relationships with β_{eff} is such (Fig. 56) that N derived from IIR β_{eff} is reduced by less than 35 %. The comparison between in situ and layer average IIR N, which is partly driven by the comparison between the

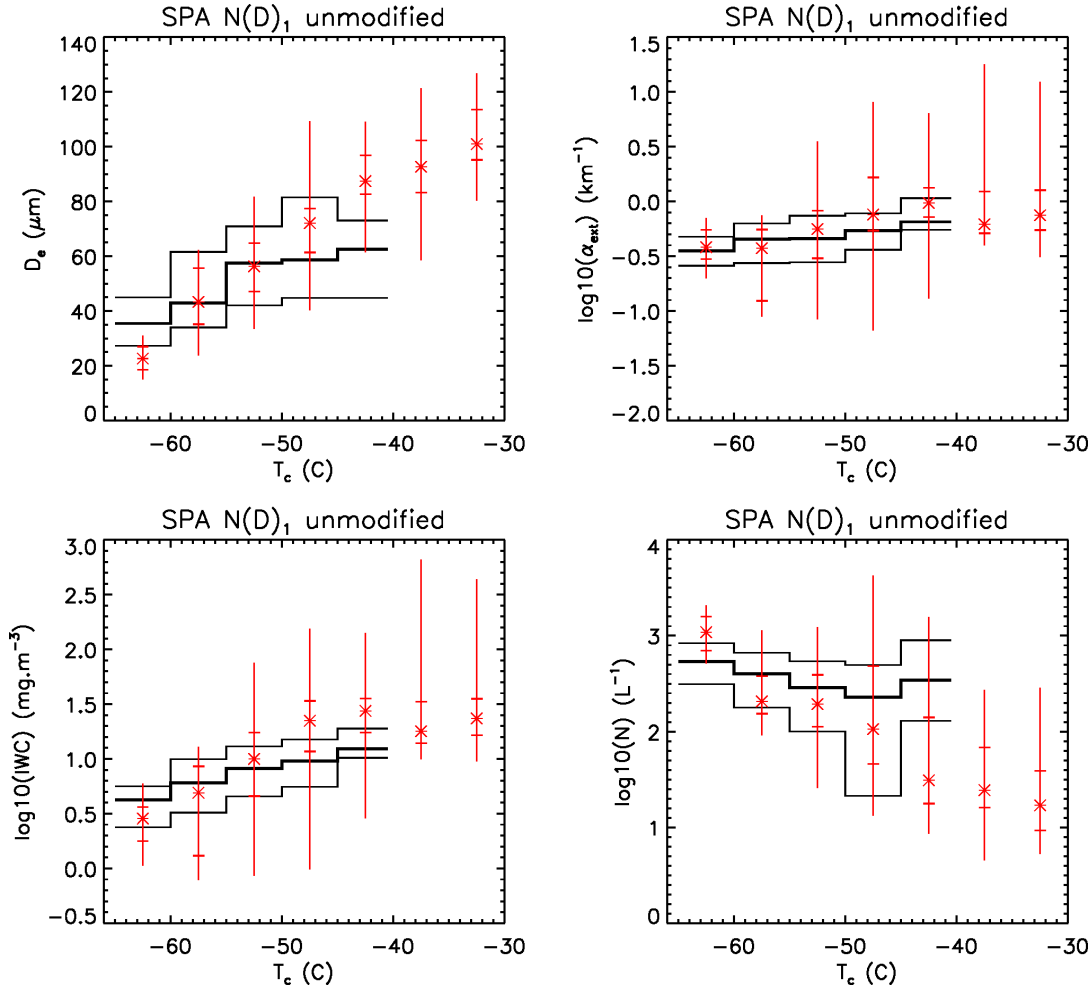


Figure 740a. SPARTICUS in situ measurements for synoptic cirrus (sampled January-April 2010) are in red, showing the temperature dependence of effective diameter D_e (μm), extinction coefficient α_{ext} (km^{-1}), ice water content IWC ($\text{mg} \cdot \text{m}^{-3}$) and ice particle number concentration N (L^{-1}), and correspond to PSD measured within 5°C temperature intervals based on unmodified $N(D)_1$. The vertical lines give the measurement range, the horizontal bars give the 25th and 75th percentile values, and the asterisks give the median value. Corresponding CALIPSO IIR retrieved properties using the SPARTICUS unmodified relationships are given by the thick black histograms, with thin black histograms giving the 25th and 75th percentile values.

respective β_{eff} , is more favorable overall assuming $N(D)_1$ unmodified. The larger median IIR N values at $T_c > -45^\circ\text{C}$ as compared to the in situ median values is partly due to the fact that IIR β_{eff} is larger than the in situ SPARTICUS low limit of 1.03 and possibly to a larger uncertainty ΔN as β_{eff} approaches the sensitivity limit. As discussed earlier, IIR retrievals are layer average quantities with sensitivity close to the middle part of the cloud layers, whereas in situ measurements can be in the lower or upper part of a relatively deep cloud.

For TC4 [comparisons](#), CALIPSO [IIR](#) and in situ D_e ([Figs. 10c and 10d, upper left panels](#)) are in good [show better](#) agreement using the $N(D)_1 = 0$ assumption (Fig. [10d7b](#)) [relative to the unmodified \$N\(D\)_1\$ assumption \(Fig. S8, upper left panel\)](#), consistent with the β_{eff} [comparisons \(Fig. 6b\)](#). The differences in cloud ODs (relative to SPARTICUS) is

made evident when comparing the extinction coefficients. CALIPSO and in situ α_{ext} , are of the same order of magnitude for aged anvils (except at -69°C).

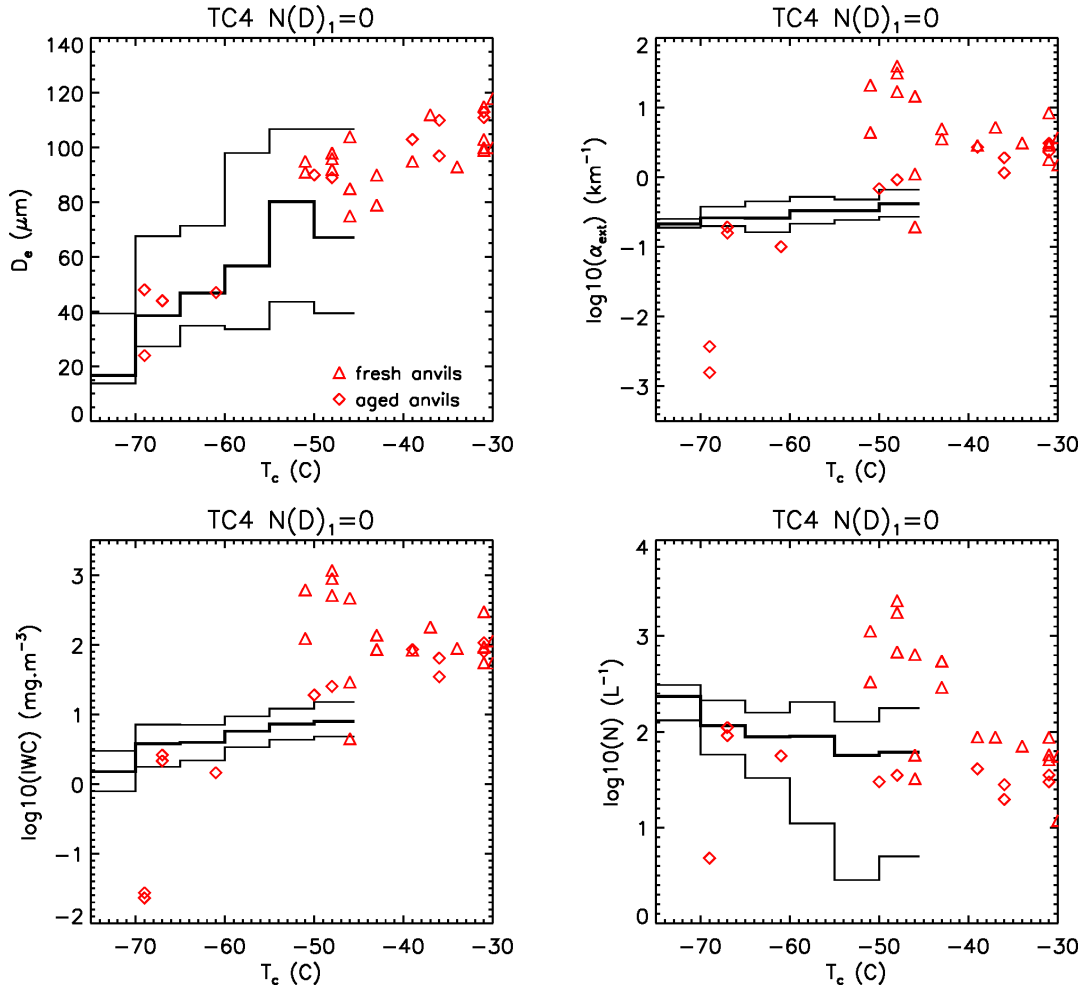


Figure 7b10e. Temperature dependence of effective diameter D_e (μm), extinction coefficient α_{ext} (km^{-1}), ice water content IWC (mg. m^{-3}) and ice particle number concentration N (L^{-1}) during TC4 based on the unmodified $N(D)_1 = 0$ assumption. In situ TC4 measurements in red are for fresh (triangles) and aged (diamonds) anvils. Corresponding CALIPSO IIR retrieved median properties using the TC4 relationships are given by the thick black histograms, with thin black histograms giving the 25th and 75th percentile values.

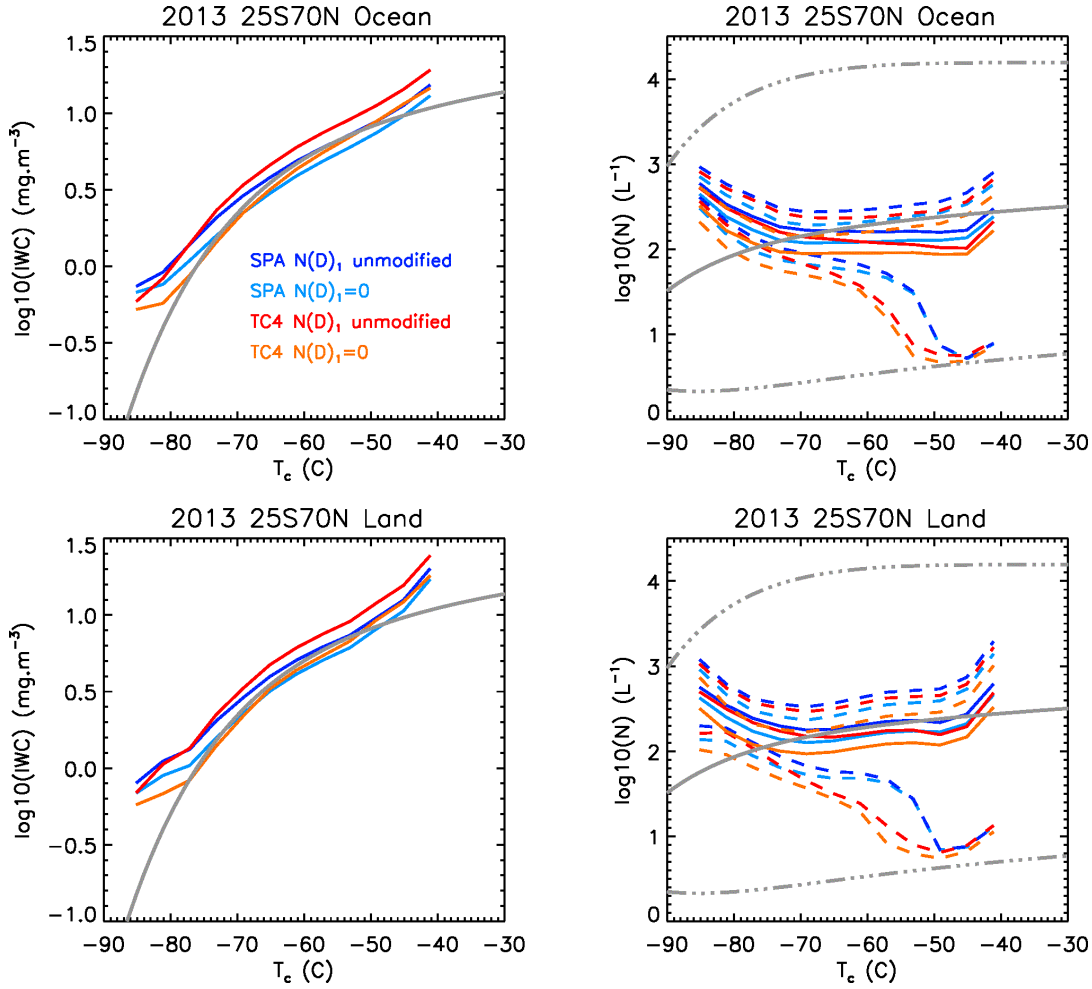


Figure 7c10e : Same as Fig. 9, but for IWC (left, mg m^{-3}) and N (right, L^{-1}). Left: Comparisons of the median CALIPSO IIR IWC (mg m^{-3}) for the four formulations (colored curves) with in situ measurements from Krämer et al. (2009) shown by the grey curve (which is the middle value). Right: Comparisons of CALIPSO IIR N (L^{-1}) shown by the colored curves (solid curves are median values while dashed curves indicate the 25th and 75th percentile values) with the in situ N middle value (solid grey curve) from Krämer et al. (2009). The dot-dashed grey curves give the minimum and maximum in situ values. The navy and light blue curves correspond to the SPARTICUS formulations for the unmodified $N(D)_1$ assumption and the $N(D)_1 = 0$ assumption, respectively. The red and orange curves are using the TC4 formulations for the $N(D)_1$ unmodified and $N(D)_1 = 0$ assumptions, respectively. The CALIPSO IIR retrievals are from 2013 and are for the approximate latitude range (25 °S to 70 °N) of the in situ data, over oceans (top) and over land (bottom).

However, the in situ α_{ext} larger than 10 km^{-1} between -52°C and -46°C are from fresh anvils sampled closer to the convective core and thus likely attenuate the CALIOP laser beam, and are therefore excluded from our cloud selection. For these fresh anvils, in situ N is unambiguously larger than N retrieved from CALIPSO in this temperature range, for both $N(D)_1$ assumptions. Otherwise, CALIPSO and in situ N are typically within a factor 2.

If future research produces convincing evidence that the $N(D)_1 = 0$ assumption is more realistic than the unmodified $N(D)_1$ assumption, then, based solely on the SPARTICUS data, the unmodified assumption may overestimate N by about a factor of 3 and underestimate D_e by up to $\sim 1/3$ for most cirrus clouds. Comparisons with IIR β_{eff} could guide this analysis, keeping in mind that comparing in situ and layer average quantities can be challenging. Overall, it is not yet clear which retrieval assumption yields the best agreement with in situ data.

Figure 4e-7c compares retrievals of IWC and N with corresponding in situ values from the Krämer dataset. Both the retrieved and in situ N exhibit little temperature dependence at $T > 200$ K. Retrieved median N over ocean appear to be slightly lower than middle in situ values by a factor of 1.5 to 3 at -48°C , depending on the formulation used.

Below -75°C , retrieved N can be much larger. The divergence between the retrieved median and in situ middle value for N for $T < -75^\circ\text{C}$ may be due to the in situ sampled cirrus often having mean layer extinction coefficients smaller than the IIR retrieval limit of about 0.05 km^{-1} (see Fig. S47), resulting in the removal of ODs below ~ 0.3 from the sampling statistics. TTL cirrus having $\text{OD} < 0.3$ are extensive in the tropics and have been characterized by lower N (e.g. Jensen et al., 2013b; Spichtinger and Krämer, 2013; Woods et al., 2018). Retrieved median IWC is very consistent with in situ middle values, with the $N(D)_1 = 0$ assumption yielding better agreement for $T < -70^\circ\text{C}$.

In general, comparisons between the retrieved ~~and the in situ and in situ measured~~ cloud properties measured during SPARTICUS and TC4 ~~and the campaigns reported in Krämer et al. (2009)~~ appear favorable despite the uncertainties involved. For any version of the four CALIPSO retrievals (Fig. 56), relative differences in layer average N and D_e in relation to different seasons and latitude zones should be meaningful. From these relative differences, mechanistic inferences can be made and hypotheses explaining these inferences can be postulated, keeping in mind the unique sensitivity of the technique to small crystals through β_{eff} .

5. Retrieval results

5.1 Frequency of occurrence of selected cirrus samples

As presented in Sect. 3, the sampled 1-km^2 IIR pixels are those for which the atmospheric column contains a single semi-transparent cloud layer of OD roughly between 0.3 and 3, of base temperature < 235 K, with a radiative contrast between surface and the cloud of at least 20 K. This greatly limits the percentage of cirrus clouds sampled during this study (relative to all cirrus clouds).

Cirrus clouds of OD between 0.3 and 3 are geographically widespread across all latitudes and are also in an OD range that makes them radiatively important (Hong and Liu, 2015). Frequency of occurrence is defined as the number of cirrus cloud pixels sampled divided by the number of available IIR pixels. To clarify, a cirrus cloud

extending 20 km horizontally along a portion of the lidar track is counted 20 times whereas a cirrus cloud extending 5 km along this track is counted only 5 times.

Two years of CALIPSO IIR data are considered: 2008 (Dec. 2007 to Nov. 2008) and 2013 (March 2013 to Feb. 2014). It is noted that the version of the GMAO Met data used in the CALIPSO products is not the same in 2008 and in 2013. In 2008, it was GMAO GEOS 5.1 until Sept 2008 and GMAO GEOS 5.2 for Oct 2008 and Nov 2008.

Table 4. Sampled cirrus cloud frequency of occurrence for each 30-degree latitude zone, and also for the entire globe (last line) during 2008 and 2013. Two lower OD thresholds were used; OD > 0.3 and OD > 0.1 (separated by a “/”).

Occurrence of selected conditions (%) during 2008 (Dec 2007 to Nov 2008)				
OD>0.3/OD>0.1	DJF	MAM	JJA	SON
60N-82N	0.49/1.08	0.2/0.65	0.21/0.76	0.22/0.64
30N-60N	0.74/1.70	0.96/2.20	0.60/1.52	0.73/1.86
0N-30N	1.58/4.04	1.90/4.76	2.02/4.86	1.70/4.38
30S-0S	1.58/3.86	1.46/3.81	0.75/2.16	1.07/2.81
60S-30S	0.25/0.70	0.39/1.00	0.47/1.07	0.36/0.90
82S-60S	0.16/0.39	0.19/0.45	0.31/0.68	0.72/1.29
Full globe	0.81/1.98	0.86/2.16	0.73/1.85	0.80/1.99

Occurrence of selected conditions (%) during 2013 (March 2013 to Feb 2014)				
OD>0.3/OD>0.1	DJF	MAM	JJA	SON
60N-82N	0.61/1.33	0.31/0.90	0.16/0.57	0.24/0.64
30N-60N	0.90/1.96	0.98/2.14	0.56/1.52	0.65/1.68
0N-30N	1.43/3.70	1.82/4.61	1.86/4.53	1.76/4.47
30S-0S	1.58/3.96	1.47/3.75	0.79/2.32	1.05/2.75
60S-30S	0.30/0.78	0.36/0.93	0.46/1.05	0.35/0.88
82S-60S	0.09/0.29	0.20/0.46	0.42/0.83	0.61/1.16
Full globe	0.82/2.02	0.86/2.15	0.71/1.82	0.78/1.94

In 2013, it is GMAO GEOS FP-IT for the whole period. Retrievals for each month of each year for all latitudes have been analyzed and organized into seasons, with winter as December, January, February (DJF); spring as March, April, May (MAM); summer as June, July, August (JJA); fall as September, October, November (SON). Frequency of occurrence is reported in Table 4 for each season and each 30° latitude zone, and for the entire planet during 2008 and 2013. The selection criteria result in ~~very few sampled pixels relative to the number of available IIR pixels, making the~~ frequency of occurrence generally less than 2%. Relaxing the lower OD threshold to 0.1 instead of 0.3 would increase the number of samples by about a factor of 2.5.

~~Moreover, Thus~~ what is most important in this analysis is not the actual frequency value but the relative differences in these values with respect to season and latitude. It is seen that despite our cloud subsampling, the geographical distribution of the occurrence frequencies is consistent with previous findings for ice clouds ($T < 0^{\circ}\text{C}$) of OD between 0.3 and 3 (Hong and Liu, 2015). The greatest occurrence frequencies are in the tropics (i.e. 30° S-30° N). The occurrence frequency during Arctic (i. e. 60° N-82° N latitude zone) winter is more than twice the frequency of other Arctic seasons for the standard OD threshold and more than 1.4 times this frequency for the 0.1 OD lower limit. In the Antarctic (i.e. the 60° S-82° S latitude zone), frequency of occurrence is greatest in the spring and second-greatest during winter, in agreement with previous studies (Nazaryan et al., 2008; Hong and Liu, 2015). This is important since at high latitudes, the net radiative effect of ice clouds is strongest during the “cold season” where solar zenith angles are relatively low and ice cloud coverage is relatively high (Hong and Liu, 2015). Therefore, the cirrus cloud formation mechanism that governs cirrus microphysical properties will have the greatest net cloud radiative effect (CRE) at high latitudes; during winter and spring for the Antarctic and during winter for the Arctic.

5.2 Latitude, altitude and seasonal dependence of β_{eff} and N

IIR β_{eff} is a measure of the fraction of small ice crystals in the PSD (e.g. how narrow the PSD is) and is an important constraint for our retrievals. Figure ~~844~~ shows the latitude and altitude dependence of median β_{eff} (left) and of the number of selected samples (right), where altitude is the cloud representative altitude, Z_c , defined as the centroid altitude of the IIR weighting profile (Sect. 2.2.3). The plots are for sampling during 2008 and 2013, with from top to bottom DJF over oceans and over land, and JJA over oceans and land.

The majority of the sampled cirrus are in the tropical areas, between 20 °S and 10 °N in DJF and between 10 °S and 30°N in JJA. They are associated with anvil cirrus from deep convection and TTL cirrus. ~~At mid- and high latitude in the Northern Hemisphere (NH), the sampled cirrus clouds tend to form at higher altitudes in summer than in winter.~~ Over Antarctica during winter (JJA), the samples exhibiting centroid altitudes between 10-11 and 18 km correspond to polar stratospheric clouds (PSCs) which are not found in summer, even though their base is in the

troposphere per our data selection. The tropopause here is not well defined, allowing a continuum to exist between tropospheric cirrus and PSCs. Since there is less land in the SH mid-latitudes, the sample counts are fewer there. In the southern oceans, there tends to be more samples during winter (JJA).

- 5 Overall, median β_{eff} decreases with decreasing altitude. It is larger than 1.2 (i.e. $D_e < 25\text{-}40\text{ }\mu\text{m}$, cf Figs. 3 and 4S2) at the top of the sounded atmosphere, prevailing in the winter hemisphere. The lowest values of β_{eff} (< 1.1) tend to be in the lower range of altitudes, and are abundant in the tropics and at mid-latitude in the NH in JJA. At the coldest temperatures (highest Z_c), hom should be more frequent, resulting in smaller ice crystals and thus higher β_{eff} .

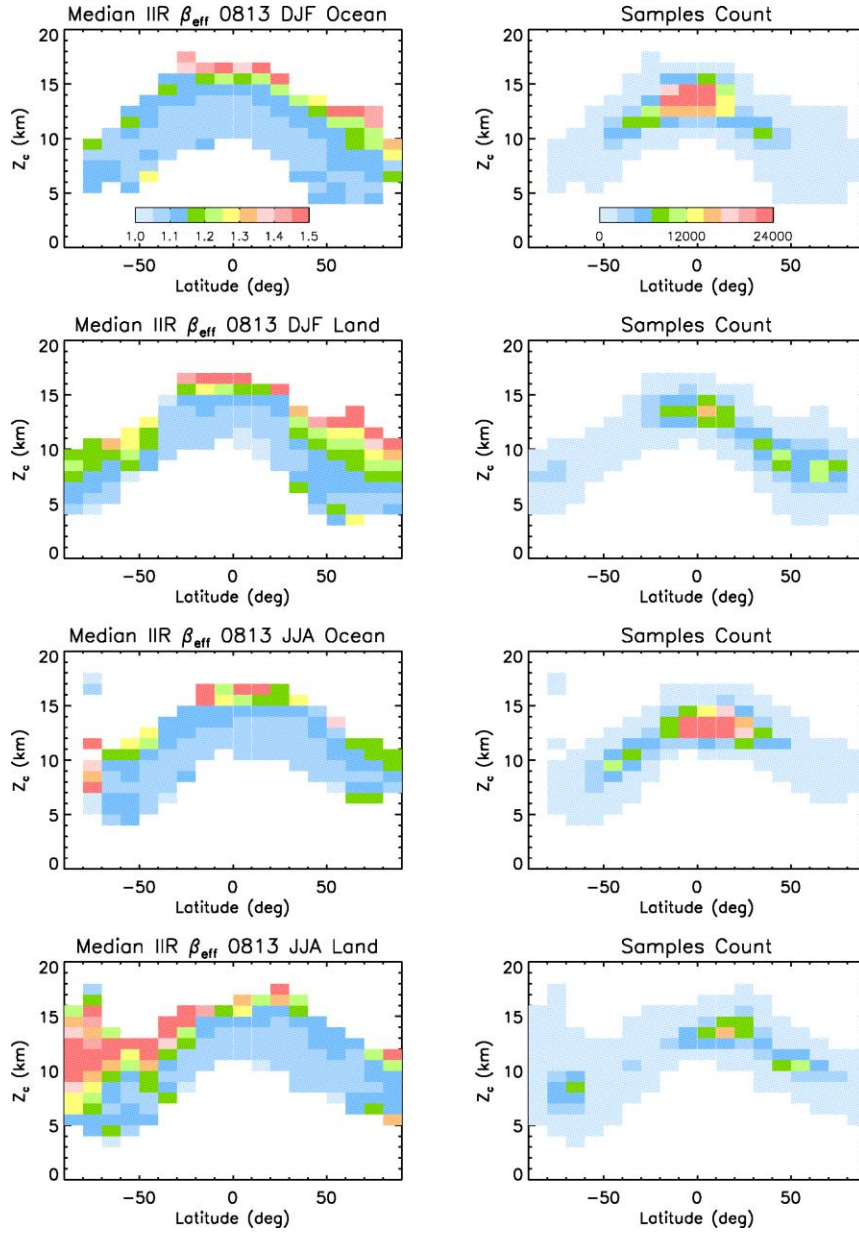


Figure 811. Median IIR β_{eff} (left) and samples count (right) vs. latitude and representative cloud altitude, Z_c , during 2008 and 2013. Panels from top to bottom are for DJF over oceans, DJF over land, JJA over oceans, and JJA over land.

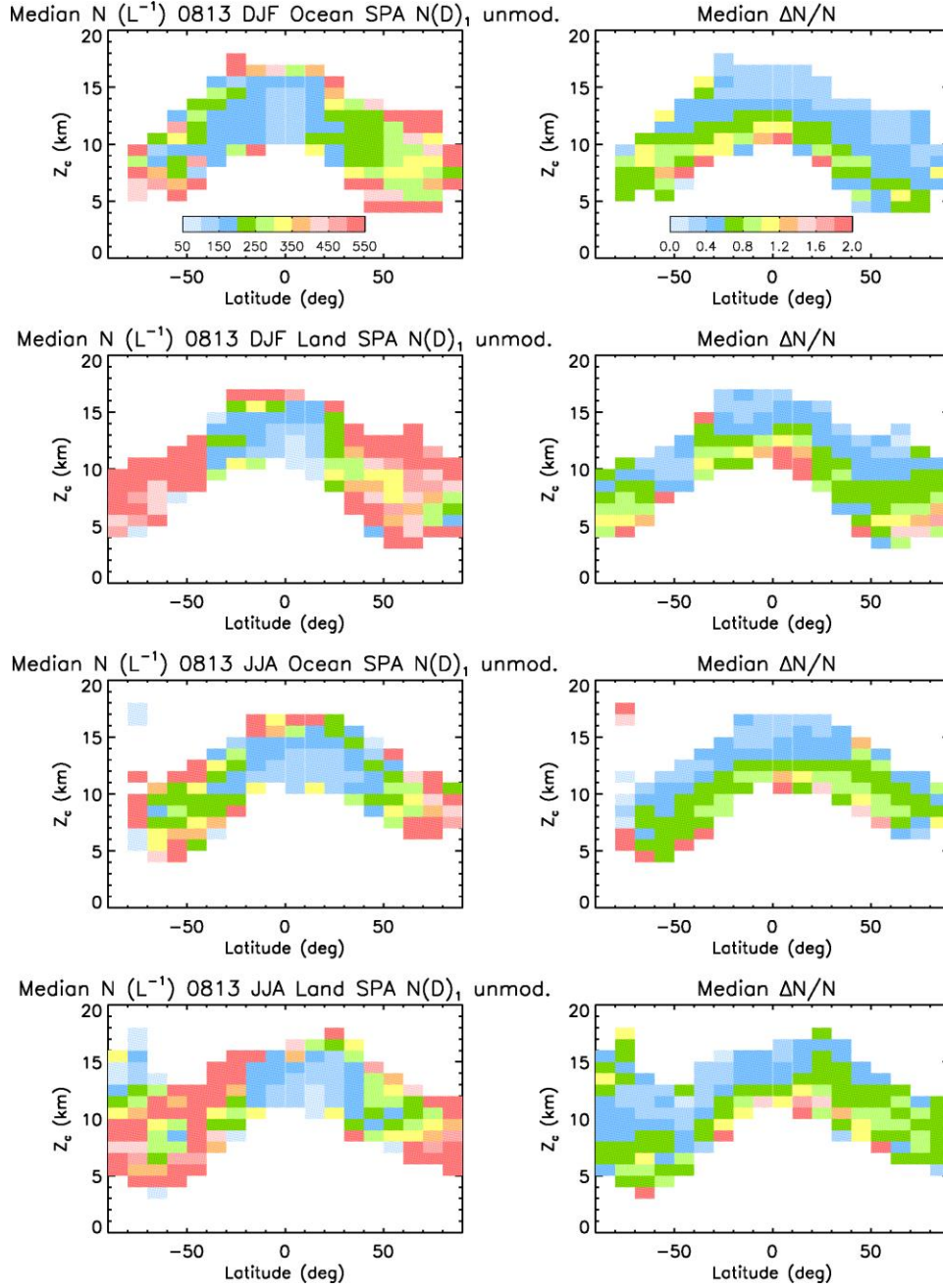


Figure 12a9: Median ice particle number concentration N (L^{-1}) retrieved from the SPARTICUS relationships assuming $N(D)_1$ unmodified (left) and associated relative uncertainty $\Delta N/N$ (right) vs. latitude and representative cloud altitude, Z_c , during 2008 and 2013. Panels from top to bottom are for DJF over oceans, DJF over land, JJA over oceans, and JJA over land.

Fig. 12a-9 shows the latitude and altitude dependence of median N (left) and median $\Delta N/N$ based on the SPARTICUS data with $N(D)_1$ unmodified relationships, and Fig. S942b shows median N based on the three other

formulations, with from left to right SPARTICUS $N(D)_1=0$, TC4 $N(D)_1$ unmodified, and TC4 $N(D)_1=0$. The difference between the four formulations varies with β_{eff} as shown in Fig. 56. As in Fig. 844, the panels from top to bottom are for DJF over oceans and land, and JJA over oceans and land. The number of samples can be seen in Fig. 88. Median $\Delta N/N$, which varies strongly with β_{eff} (Fig. S47), is typically smaller than 650 % when β_{eff} is larger, but often larger than 100 % at the lowest altitudes where β_{eff} is smaller.

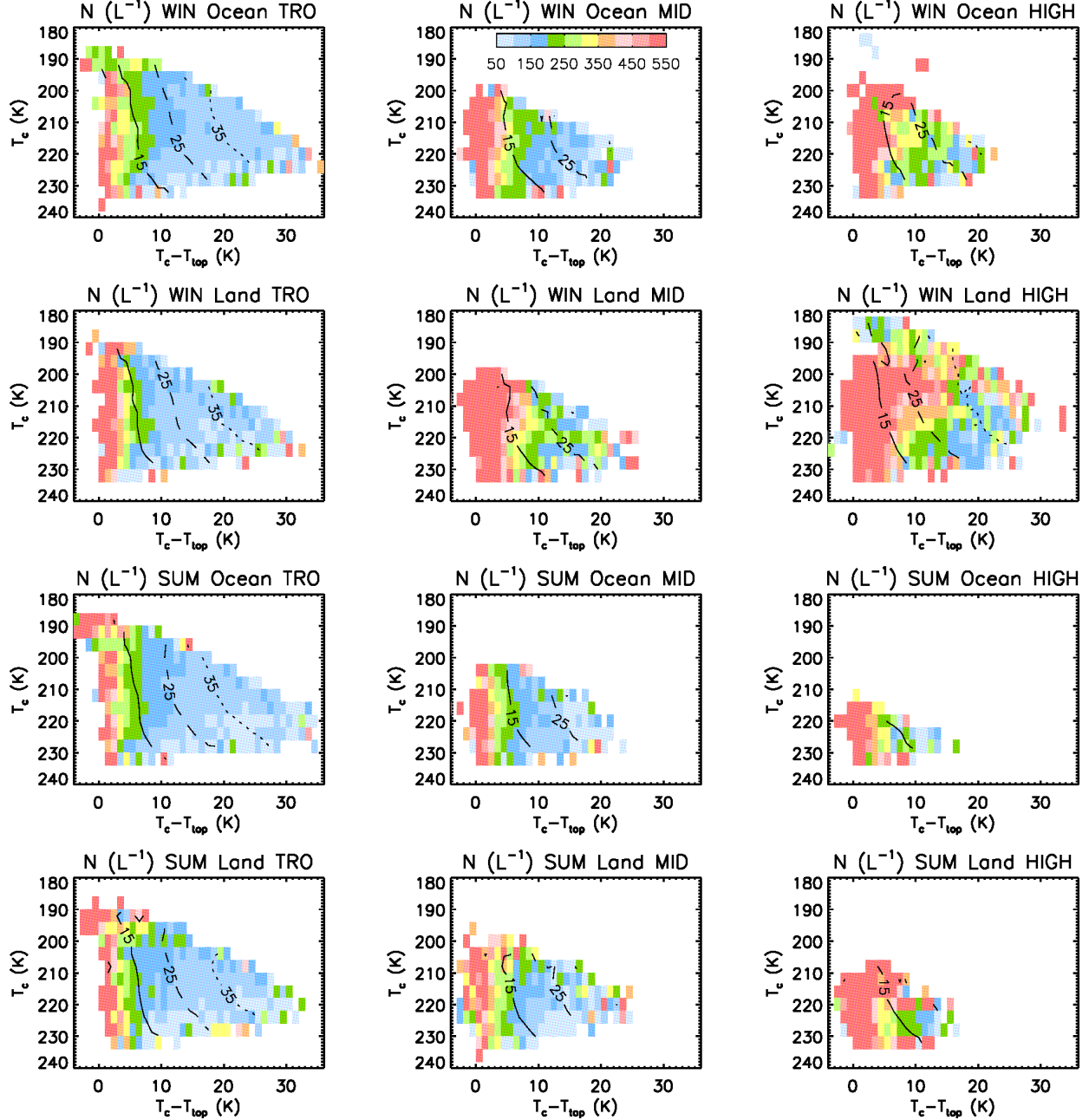


Figure 103. Median retrieved ice particle number concentrations N (L^{-1}) using the SPARTICUS $N(D)_1$ unmodified formulation related to the representative cloud temperature T_c and $T_c - T_{\text{top}}$ at 0° - 30° (TRO, left), 30° - 60° (MID, center), and 60° - 82° (HIGH, right) during 2008 and 2013. Overplotted are isolines of $T_{\text{base}} - T_{\text{top}}$ (solid: 15 K, dashed: 25 K;

dotted: 35 K). Panels from top to bottom are for winter over oceans, winter over land, summer over oceans, and summer over land. TRO, MID and HIGH refer to the tropics, mid- and high latitudes.

Median N is the lowest in the tropics (i.e. 20 °S-20 °N), and retrieved values ~~are depending~~ on the assumptions used, but are generally smaller than about ~~20450~~ particles per liter. Over Antarctica during winter (JJA), the samples exhibiting centroid altitudes between 10-11 km and 18 km corresponding to PSCs have lower N than the cirrus just below them around 9-11 km ~~for both JJA and DJF~~.

Figure S10 Fig. S10 under Supplementary Materials, shows the same results as in Fig. 9, but uses ~~aby relaxed~~ the OD-threshold to $OD > 0.1$. Consistent with Fig. S5, median N is decreased, by a factor 1.5 on average, while $\Delta N/N$ is more than doubled.

~~There is a tendency for N to increase at the lowest altitudes, and this occurs more outside the tropics. This may be due to cirrus cloud layers corresponding to the warmest temperatures to be relatively shallow in geometrical thickness, because cirrus cloud base cannot be warmer than 235 K (as per our selection criteria).~~

~~5.3 Dependence of N on distance below cloud top~~

5.3 Dependence of N on distance below cloud top

Our retrievals are now examined against both T_c and $T_c - T_{top}$ to estimate the impact of the distance from cloud top. These N retrievals are shown in Fig. ~~103~~ using the SPARTICUS $N(D)_1$ unmodified assumption in the tropics (0-30°) and at mid- (30-60°) and high (60-82°) latitudes in the winter and summer seasons (using both hemispheres) by distinguishing retrievals over oceans and over land. The associated number of samples is given in Fig. ~~14~~ S11.

Added to these figures are isolines of temperature differences between cloud base and cloud top ($T_{base} - T_{top}$). For most of the layers, $T_c - T_{top}$ represents 30 to 70% of $T_{base} - T_{top}$ (Fig. S11). Fig. 130 shows a strong dependence of N on $T_c - T_{top}$, with ~~large~~ high N ($> 500 \text{ L}^{-1}$) seen near the top of the geometrically thin clouds ($T_{base} - T_{top} < 15^\circ \text{C}$) ~~clouds~~, when $T_c - T_{top}$ is smaller than about 5°C . In contrast, the N dependence on T_c is very weak (consistent with Figs. ~~7a, b, c+10a-e~~). As seen in Figs. ~~103~~ and S11, the $T_c - T_{top}$ (and $T_{base} - T_{top}$) difference tends to be larger in the tropics than at mid- and high latitudes. ~~The strong N dependence on $T_c - T_{top}$ appears to support the lower updraft cirrus cloud simulations of Spichtinger and Gierens (2009a, b) and the in situ measurements of Diao et al. (2015), although other possible explanations for this dependence cannot be ruled out.~~ The strong dependence of N on $T_c - T_{top}$ (Fig. 130) appears weakest at high latitudes and at mid-latitudes during winter over land, with N being relatively high there. ~~If N near cloud top is due to hom as predicted by Spichtinger and Gierens (2009a, b), then hom appears more active near cloud top. Other attributes of Fig. 14 have been discussed in relation to Fig. 12.~~

5.4 Effective diameter

The dependence of median D_e on the representative temperature T_c is shown in Fig. 45-11 for each latitude zone (tropics, mid- and high latitudes), with profiles for summer and winter for each zone (based on both hemispheres), over oceans and land. The analysis is using the SPARTICUS unmodified $N(D)_1$ (top) and the TC4 $N(D)_1 = 0$ (bottom) formulations. The TC4 curve fits yield a larger range of D_e values than the SPARTICUS ones. At least 100 samples contributed to each data-point, and data from both years (2008 and 2013) were combined to generate these profiles. Consistent with the reported β_{eff} -altitude relationships (Fig. 844), the D_e -temperature relationships show that D_e for a given temperature and season is generally largest in the tropics, intermediate at mid-latitudes and smallest in the high latitudes. The profiles exhibit a considerable latitudinal and seasonal dependence, with latitudinal differences up to $\sim 40 \mu\text{m}$ for a given temperature. Seasonal differences may be up to about $20 \mu\text{m}$ for a given temperature at mid-latitude over land. Combined with the latitudinal and seasonal dependence of selected cirrus cloud frequency of occurrence (e.g. Table 4), these D_e differences are likely to produce substantial variations in cirrus cloud net radiative forcing relative to a constant D_e profile assumption.

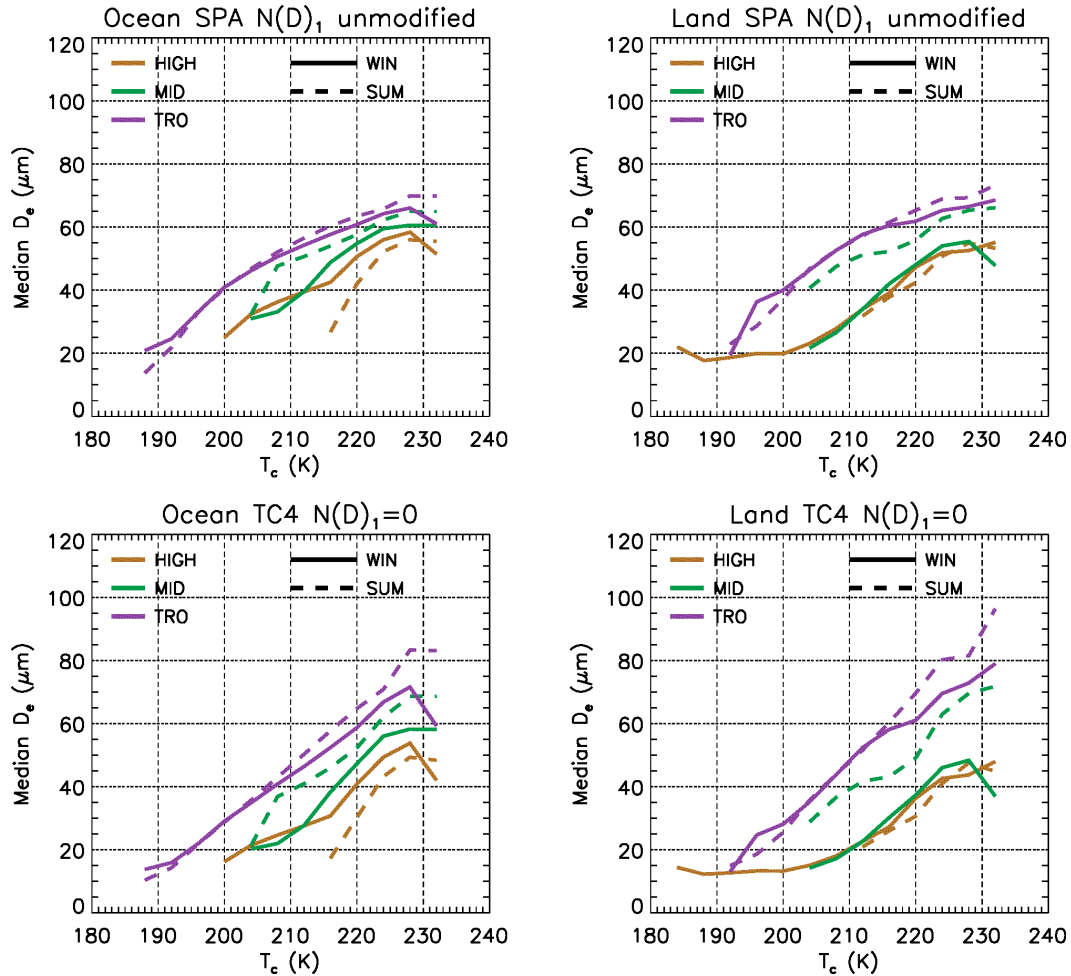


Figure 115. Temperature dependence of the retrieved median effective diameter D_e (μm) in winter (solid) and summer (dashed) during 2008 and 2013, based on the SPARTICUS $N(D)_1$ unmodified (upper panels) and the TC4 $N(D)_1 = 0$ (lower panels) formulations. Latitude zones are denoted by colors: purple: 0° – 30° (TRO); green: 30° – 60° (MID), brown: 60° – 82° (HIGH).

Following the same reasoning as previously for N , Fig. S12+6 shows the dependence of median D_e (using the SPARTICUS $N(D)_1 = 0$ unmodified assumption) on T_c and $T_c - T_{\text{top}}$, for the same latitude ranges and seasons as in Figs. 103 and 115. Unlike N (Fig. 103), median D_e depends strongly on T_c with a somewhat weaker dependence on $T_c - T_{\text{top}}$. For instance, in the tropics, small median D_e ($< 50 \mu\text{m}$) are found at any T_c colder than about 205 K, but only near cloud top when T_c is warmer than 205 K. At $T_c = 220$ K, median D_e increases with $T_c - T_{\text{top}}$ from less than $50 \mu\text{m}$ near cloud top up to the upper retrieval limit around $80 \mu\text{m}$ at $T_c - T_{\text{top}} = 35$ K. This weaker dependence on $T_c - T_{\text{top}}$ relative to N is expected since D_e is proportional to $\text{IWC}/A_{\text{PSD}}$. For a gamma function PSD and assuming power law relationships for ice particle area and mass, D_e is proportional to $\lambda^{\sigma-\beta}$ where λ = PSD slope parameter and σ and β are power law exponents for area and mass, respectively. The difference $\sigma - \beta$ is slightly more than -1, showing that D_e is strongly related to the PSD mean size (inversely proportional to λ). The derivation also shows that D_e is not dependent on N . The decrease in D_e with decreasing T_c is likely due to a decrease in IWC with decreasing T_c .

6 Discussion and perspectives

6.1 Global and seasonal distribution of N

In the tropics, most cirrus are anvil cirrus and there is little difference in N between land and ocean, nor is there much seasonal dependence. A seasonal variation is seen in the NH at mid-latitude, more over land than over oceans, with larger N (and smaller D_e) during the winter than during the summer season (see Fig. 9 & S9+2a). This behavior is less evident over land in the Southern Hemisphere (SH) where mid-latitude land mass is relatively small, but over the southern oceans where ice nuclei (IN) concentrations are relatively low (Vergara-Temprado, 2018), N is larger (and D_e smaller) during winter. At high latitudes, N is relatively high and D_e relatively low during both seasons. These observations appear generally consistent with the modeling results of Storelvmo and Herger (2014), where the Community Atmosphere Model version 5 (CAM5) was used to predict the global distribution of mineral dust at 200 hPa by season and the fraction of ice crystals produced by ~~heterogeneous and homogeneous ice nucleation (henceforth het and hom)~~. Dust concentrations in both hemispheres were minimal at high latitudes, especially during DJF. If dust is the main source of ~~ice nuclei~~ IN (e.g. Cziczo et al., 2013) and more ice crystals are produced through hom when dust concentrations are relatively low (e.g. Haag et al., 2003), then the above observations appear consistent with the predicted latitude dependence of dust concentrations. That is, lower ~~ice nuclei~~ IN concentrations may result in higher RH_i sufficient for hom to occur since the limited numbers of ice crystals produced via het may not exhibit sufficient surface area to draw down the RH_i and prevent hom from occurring.

While hom does not always result in relatively high N (e.g. Spichtinger and Krämer, 2013), hom can produce much higher N than het within sustained appreciable updrafts and is generally associated with higher N (e.g. Barahona and Nenes, 2008). Other observational studies indicating that hom is often important in determining the microphysics of cirrus clouds are Mitchell et al. (2016), Zhao et al. (2018), Sourdeval et al. (2018a) and Gryspeerdt et al. (2018).

5 Atmospheric dynamics may also help to explain the results in Fig. 9+2a. The tropical troposphere is well mixed due to deep convection, whereas in the mid-latitudes during winter, deep mixing is more limited. This reduced mixing should reduce the transport of ~~INice nuclei~~ to cirrus cloud levels (relative to the tropics). Snow cover during winter may also limit dust transport. Indeed, N is considerably higher during winter over land in the NH mid-latitudes, and a similar but somewhat weaker seasonal relationship exists over oceans in the NH and SH mid-latitudes.

10 In general, we find higher N over mountainous regions, and mountain-induced waves may be responsible for higher N in the ~~NH~~ mid-latitudes ~~over North America~~, especially during the winter season (having higher winds). The Andes Mountains in South America provide abrupt orography for mountain-induced waves that provide high updrafts favoring hom (Jiang et al., 2002; Hoffmann et al., 2016). This abrupt orography combined with minimal mineral dust concentrations in this region, especially during DJF (Storelvmo and Herger, 2014), may explain the
 15 higher N over the Andes. ~~However, N over the Andes was not higher in the tropics (Mitchell et al., 2016not), perhaps due to higher IN concentrations in the tropics, lower sustained winds and the OD range sampled.~~ Lower dust concentrations (Vergara-Temprado, 2018) may also be responsible for the higher N over the southern oceans.

6.2 Dependence of N on distance from cloud top

The temperature dependence of N on the cloud representative temperature T_c , as shown in Figs. ~~7a, b, c, 40a-b-c-d-e~~,
 20 is an important relationship for understanding the physics of cirrus clouds. However, the in-cloud dependence of N on height below cloud top may also be important, ~~as shown in Fig. 13.~~ The strong N dependence on $T_c - T_{top}$ shown in Fig. 10 appears to support the lower updraft cirrus cloud simulations of Spichtinger and Gierens (2009a, b) and the in situ measurements of Diao et al. (2015). It could be enhanced by the fact that $T_c - T_{top}$ is somewhat correlated with geometrical thickness. In geometrically thin cirrus, there is less dilution of the mean N due to ice sedimentation
 25 (Jensen et al., 2013a). Also, in geometrically thicker cirrus, sedimenting ice particles from above tend to quench hom in the mid-to-lower cloud by decreasing the RH_i , preventing the threshold RH_i needed for hom. This can decrease N considerably (Spichtinger and Geirens, 2009a, b). This effect would tend to produce higher N in geometrically thin cirrus relative to the thicker cirrus. Moreover, aggregation has less time to decrease N in thin cirrus.

30 ~~It was also shown with the SPARTICUS case study (Fig. 8a) that β_{eff} can be sensitive to the $T_c - T_{top}$ temperature difference. The explicit synoptic cirrus cloud model of Spichtinger and Gierens (2009a, b) that describes the impact~~

of aerosols on different types of nucleation mechanism indicates that the $T_e - T_{top}$ difference can be an important metric for understanding physical processes. Using a constant vertical velocity of 0.05 m s^{-1} in a modeled cirrostratus cloud, they show that the portion of an ice supersaturated region (ISSR) reaching the hom RHi threshold initiates cloud formation via hom, and though new ice crystal growth reduces the RHi, the RHi is not reduced much at this level since the rapid growth allows the ice crystals to fall more rapidly to lower levels. In this way a continuous supply of new ice crystals is maintained via hom near cloud top. As the ice crystals descend to lower levels and grow, they prevent the RHi from reaching the hom threshold and N decreases substantially (since N near cloud top is spread vertically over the whole depth of cloud). In this way ice sedimentation plays a crucial role in the development and evolution of the cirrus cloud microphysical structure. These modeling results are also supported by the in situ measurements reported in Diao et al. (2015), which show the altitude dependence of the four evolution phases for synoptic cirrus clouds, which are ice nucleation, early ice crystal growth, later growth and sedimentation/ sublimation. The ice nucleation phase generally occurs near the tropopause and cloud top while the other phases (e.g. growth of ice crystals) occur primarily below the ice nucleation layer. The above findings are also broadly consistent with those of Jensen et al. (2012; 2013a) that show N depends strongly on the ice sedimentation and entrainment/dilution processes.

6.3 Representativeness of these results

As mentioned, the cirrus clouds sampled in this study are optically thick ($0.3 < OD < 3.0$) and contribute a relatively small fraction of total cirrus cloud coverage according to some cirrus cloud climatologies (e.g. Kienast-Sjögren et al., 2016; Goldfarb et al., 2001; Immler and Schrems, 2002). Thus it is possible that the cirrus cloud properties retrieved here are not representative of cirrus clouds in general. However, these are ground-based lidar studies conducted at point locations whereas the global cirrus cloud climatology in Hong and Liu (2015) is based on the CALIPSO and CloudSat satellites. In that satellite study, where the ice cloud OD ranged from < 0.03 to > 20 , the frequency of occurrence for cirrus having $0.3 < OD < 3.0$ is much larger than these ground-based climatologies indicate, and this OD category appears to have the highest frequency of occurrence (see Fig. 7 of Hong and Liu, 2015). Moreover, in Kienast-Sjögren et al. (2016) where cirrus having $OD > 0.3$ comprised a very small fraction of the cirrus sampled, cirrus having $OD > 0.3$ still accounted for about half of the cloud net radiative forcing for cirrus cloud overcast conditions. If the Hong and Liu study is more realistic, then cirrus having $0.3 < OD < 3.0$ should strongly dominate the overall cirrus cloud net radiative forcing.

6.4 Comparisons with other studies

Figure 127 shows the geographical distribution of median N based on the years 2008 and 2013, with N retrieved using the SPARTICUS unmodified (left) and TC4 $N(D)_1 = 0$ formulations (right), which correspond to the upper and lower N bounds, respectively. Figure 172 is for layers of representative temperature, T_e , between 218 and 228 K, so that these results can be directly compared with the retrieved N values in Gryspeerd et al. (2018). When

averaging over all temperature levels sampled (not shown), our results are still similar to those reported in Fig. 12. The geographical distributions of the number of samples and of median $T_c - T_{top}$ corresponding to Fig. 12 are shown in Fig. S13 under Supplementary Materials. Median $T_c - T_{top}$ is up to 20 °C and is smaller at mid- to high latitudes than in the tropics. While the color legend in Fig. 12 changes color in increments of 50 L^{-1} , the 1st color bin is from zero to 100 L^{-1} and the last color bin is from 500 L^{-1} to infinity. When comparing the two plots, regions corresponding to the highest N values may not change much due to this legend convention.

Results in Fig. 12 can be compared with the retrieved N values near cloud top at 223 K shown in Fig. 1 of Gryspeerd et al. (2018). The geographical distributions are qualitatively similar to those reported in Fig. 1 of Gryspeerd et al. (2018), including higher N over mountainous regions and over the southern oceans, with minimal N over the tropics. Sourdeval et al. (2018a) have developed a new satellite retrieval for N regarding ice clouds, based on the CloudSat radar and the CALIPSO lidar (built upon the so-called DARDAR retrieval and referred to as the DARDAR-LIM scheme; LIM standing for Leipzig Institute for Meteorology). This retrieval uses the operational DARDAR retrieval products of IWC and N_0^* (a PSD normalization factor that is a function of the IWC and D_m , where D_m is the ice particle mean volume diameter, defined as the PSD moment ratio M_4/M_3), and also the “universal” normalized PSD described in Delanoë et al. (2005), to estimate N. Although their retrieval scheme is based on different physics than our scheme, these

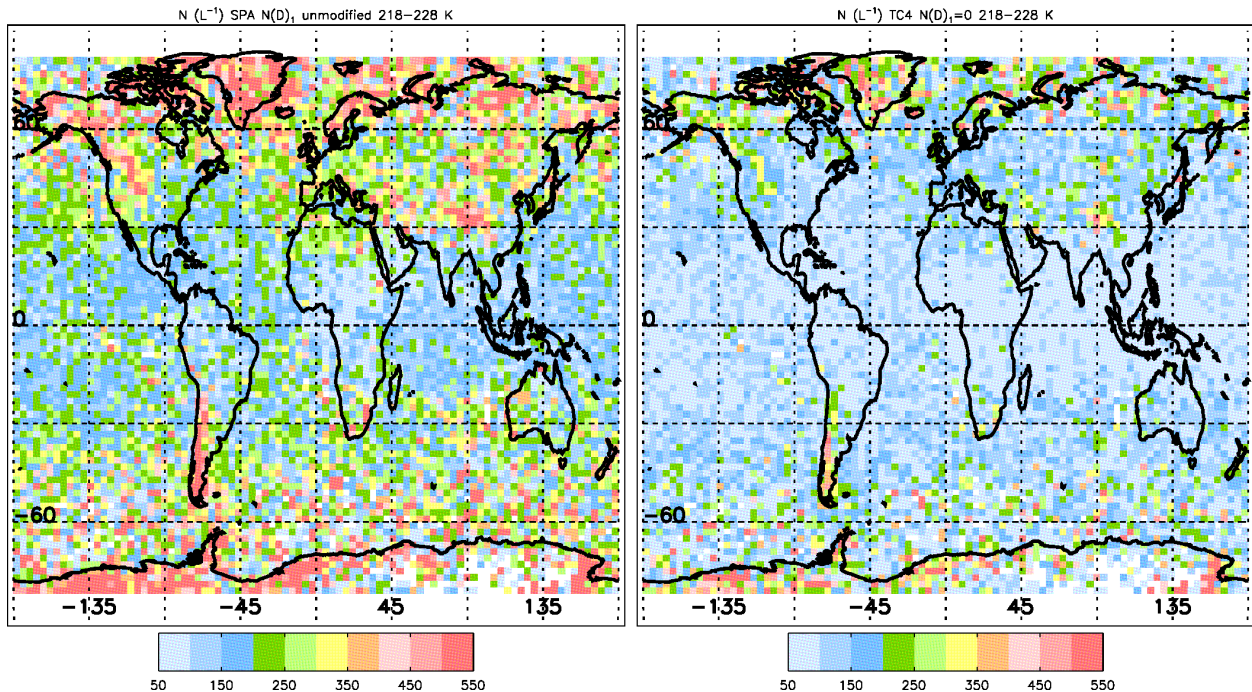


Figure 127: Geographical distribution of median ice particle number concentration N (L^{-1}) during 2008 and 2013 where the layer representative temperature, T_c , is between 218 and 228 K. The retrievals are using two formulations: SPARTICUS $N(D)_1$ unmodified (left) and TC4 $N(D)_1 = 0$ (right).

These schemes yield similar results. However, there are important differences. Since our scheme is based on in situ cirrus cloud PSDs, especially the smallest ice crystal sizes, it naturally suffers from uncertainties endemic to the PSD probes and uncertainties in how PSD shape may vary between regions (i.e. field campaigns). When β_{eff} is greater than about 1.06, N retrievals vary by about a factor of 2 (Fig. 56), but N uncertainties are greater at smaller β_{eff} where N/IWC and D_e lose sensitivity to β_{eff} . This limits our ability to retrieve at relatively low N. On the other hand, the DARDAR-LIM scheme estimates N using the normalized universal PSD described in Delanoë et al. (2005). This normalized PSD has the form $N(D_{\text{eq}}) = N_0 D_{\text{eq}}^\alpha \exp(-k D_{\text{eq}}^\beta)$, where N_0 and k are constrained by the radar and lidar measurements, α and β are fixed constants, and D_{eq} is the melted equivalent diameter of an ice particle. Whereas β_{eff} determines the PSD fraction of small ($D < 50 \mu\text{m}$) ice crystals in our scheme, this fraction is determined by α and k in the DARDAR-LIM scheme. Moreover, α depends on environmental conditions (Herzogh and Hobbs, 1985), with lower α values (which promote higher N) associated with higher updrafts and ice crystal production rates (such as might be found in cirrus clouds over mountainous terrain). Furthermore, radar measurements are not sensitive to small particles (Deng et al., 2010, 2013), in contrast to the lidar and to the IIR, and DARDAR-LIM retrievals using the lidar-only mode rely strongly on a priori relationships to retrieve N. In a general sense, in the case of clouds detected only by the radar or only by the lidar, the DARDAR algorithm does not benefit from the synergism between these two measurements, making the a priori relationships essential to obtain closure.

The highest N values reported in Sourdeval et al. (2018a) and Gryspeerd et al. (2018) are for ice particles larger than $5 \mu\text{m}$. These values would be higher by a factor of about 1.7 if all ice particles larger than $1 \mu\text{m}$ were considered (Sourdeval et al., 2018b). This is important to note since the CALIPSO IIR retrieval has no cut-off size, and it is very sensitive to ice crystals in the 1 to $5 \mu\text{m}$ range (Fig. 1). Keeping this in mind, the CALIPSO N retrievals are comparable in value to the DARDAR-LIM N retrievals.

~~It is evident that N varies by about a factor of 4 in Fig. 1 of Gryspeerd et al. (2018) and by about a factor of 7 in Fig. 17 of this study. The greater N range of this study might be partly due to the use of β_{eff} without the constraint of a parameterized PSD.~~

~~The DARDAR-LIM retrieval also shows that N has a strong temperature dependence, with N increasing with decreasing temperature T. This dependence was not observed in the field campaigns analyzed in Krämer et al. (2009), nor is it predicted by our retrieval for $T > 200 \text{ K}$ (Fig. 10e).~~

~~The dependence of N on distance from cloud top was also evaluated in Fig. 5 of Gryspeerd et al. (2018). A decrease in N with increasing distance from cloud top was evident in that study for cloud top distance more than $\sim 0.5 \text{ km}$ (based on ice particle sizes $> 5 \mu\text{m}$), although the extent of decrease appears less than often observed in Fig. 104. A comparison is complicated by the different scales used (linear vs. log) and different data treatments.~~

The regional variation in median N might be greater than indicated by Fig. 172. Regarding TC4 and Fig. 68b, the $N(D)_1 = 0$ assumption yielded the best agreement between in situ and retrieved β_{eff} , whereas a similar comparison with SPARTICUS data (Fig. 68a) may arguably favor the unmodified $N(D)_1$ assumption (since PSD sampled at $T < -60^\circ\text{C}$ were from a single cirrus event). Thus it is possible that the TC4 $N(D)_1 = 0$ formulation of our retrieval scheme is most representative for tropical cirrus and that the SPARTICUS $N(D)_1$ unmodified version is most appropriate for mid-to-high latitudes. This would double the N range estimated from the color-bar legend in Fig. 127, with N varying by a factor of about 14 (note that $N \ll 50 \text{ L}^{-1}$ is within the dynamic retrieval range of the TC4 $N(D)_1 = 0$ scheme, which is evident from Fig. 56 and Eq. 8).

Ice cloud CALIPSO-CloudSat retrievals of D_e are reported in Fig. 12 of Hong and Liu (2015) against temperature in terms of season and latitude zone for ODs ranging from < 0.03 to > 20 . Keeping in mind the different cloud sampling (in particular OD range and our cloud base temperature being colder than 235 K), comparing their seasonal D_e changes with those in this study is not straight-forward. Nevertheless, mean D_e values are comparable, in particular for the TC4 $N(D)_1 = 0$ assumption at temperatures colder than 235 K (the upper limit in this study). Their D_e in the tropics at 190 K is about $40 \mu\text{m}$ compared to $20 \mu\text{m}$ in this study. At 220 K, their D_e around $70 \mu\text{m}$ is in fair agreement with our D_e found between 60 and $70 \mu\text{m}$. Heymsfield et al. (2014, their Fig. 11) report small D_e around $20 \mu\text{m}$ at temperature colder than -72°C , in agreement with this study, but they find a steeper increase of D_e with temperature. Detailed comparisons with other work, which are beyond the scope of this paper, should account not only for temperature, but also for distance from cloud top.

6.5 A possible link between high latitude cirrus and mid-latitude weather

The retrieval results in Table 4 indicate that at high latitudes there tends to be the greatest cirrus cloud coverage during winter in the Arctic and during spring (SON) in the Antarctic (where relatively high N and small D_e occur throughout the year in both regions, as shown in Figs. 942, 115 and 127). While this study only considers a subset of cirrus clouds and two years of retrievals, our findings on the seasonal dependence of cirrus cloud coverage are consistent with other satellite cirrus cloud studies that consider a broader range of conditions over longer periods (Nazaryan et al., 2008; Hong and Liu, 2015). Independent of the macro- and microphysical cirrus cloud attributes found in this study, at high latitudes when the sun is near or below the horizon, cirrus clouds ~~there are important seasonal changes to the cirrus cloud shortwave (SW) and longwave (LW) radiative forcing due to a changing solar zenith angle. The SW and LW components almost cancel during summer, but during winter, the LW component strongly prevails,~~ producing a strong net warming at the top of atmosphere (TOA) and at the surface (Hong and Liu, 2015; Storelvmo et al., 2014). This indicates that the strongest net radiative forcing by cirrus clouds on Arctic (Antarctic) climate occurs during winter (spring). ~~This seasonal cycle of the solar zenith angle e~~ Combined with the unique macro- and microphysical properties of Arctic cirrus during winter reported here, this suggests that

wintertime Arctic cirrus may have a significant warming effect on Arctic climate. A satellite remote sensing study of ice clouds ($T < 0^{\circ}\text{C}$) by Hong and Liu (2015) found that at high latitudes, ice cloud net radiative forcing at the TOA and at the surface during the cold season is $> 2 \text{ W m}^{-2}$ for a cirrus cloud OD of 1.5. Since the most severe effects of global warming occur at high latitudes, it is critical to understand the factors controlling the macro- and microphysical properties of high latitude cirrus clouds.

A potential link to mid-latitude winter weather is the possible impact of the winter Arctic cirrus on the meridional (north-south) temperature gradient between the Arctic and mid-latitudes. The cirrus-induced winter warming described above will occur throughout the troposphere (Chen et al., 2000; Hong and Liu, 2015), and will thus act to reduce this temperature gradient in the upper troposphere (UT). While it is not clear how this would impact weather, some type of impact is likely if the warming is significant, and several possible scenarios are described in Cohen et al. (2014) and Barnes and Screen (2015). While many papers have been published recently regarding potential effects of Arctic Amplification (henceforth AA; the observation that the mean Arctic temperature rise due to greenhouse gases is at least a factor of two greater relative to the adjacent mid-latitudes) on mid-latitude weather, it is important to note that the AA related to sea ice loss and associated increases in sea surface temperature ~~increases~~ primarily affects temperatures between the surface and 700 hPa (Screen et al., 2012), while heating due to winter cirrus would strongly affect the UT. A theoretical link between AA and the jet-stream is found in the thermal wind balance, which states that a reduced meridional temperature gradient tends to produce a reduced vertical gradient in the zonal-wind field, depending on other factors like changes in surface winds, storm tracks and the tropopause height (Barnes and Screen, 2015). Thus, AA could lead to a weaker jet-stream having more amplified Rossby waves and associated extreme weather events as hypothesized by Francis and Vavrus (2012; 2015), but it is currently not clear whether such a phenomenon is occurring or will be occurring (Barnes and Screen, 2015).

As described in Barnes and Screen (2015), GCM simulations from the fifth Coupled Model Intercomparison Project (CMIP5) show that while the lower troposphere during Arctic winter is projected to warm substantially by 2100, this is not happening in the Arctic UT where little warming is projected. Moreover, in the tropics the models predict the strongest warming in 2100 occurs in the UT. These effects decrease the meridional temperature gradient at low levels and increase the temperature gradient in the UT. These low- and high-level gradients have competing effects on the jet-stream, with a decreasing low-level gradient acting to weaken the jet-stream and shift it towards the equator, while an increasing UT gradient acts to strengthen the jet-stream and shift it poleward (Barnes and Screen, 2015). An interesting question to ask here is whether the CMIP5 GCMs adequately describe the changes in winter Arctic cirrus that satellite remote sensing studies observe. If they do not, and the winter heating from Arctic cirrus clouds is underestimated in the models, then the meridional UT temperature gradient may be overestimated during winter. ~~If this were the case, then increasing Arctic cirrus coverage during winter in the models would tend to weaken the simulated jet stream and shift it further towards the equator.~~ Future GCM research should determine

~~whether predicted cirrus cloud coverage and microphysics is consistent with the results from satellite studies such as this one, and~~ strive for consistency with Arctic cloud observations from satellites~~these remote observations. Then it could be~~ to determined whether the UT heating from the winter Arctic cirrus could be a significant factor affecting the simulated NH mid-latitude circulation.

- 5 A related question is whether wintertime Arctic cirrus are increasing, causing a change in jet-stream behavior. Poleward transport of heat and moisture is a fundamental attribute of the Earth's climate system, and the atmosphere of a warming climate can hold more water vapor (Dufour et al., 2016). While mid-latitude frontal systems are a primary component of this poleward transport, extreme transport events that can be described as water vapor intrusions (WVI) may account for 28% of the total moisture transport into the Arctic (Woods et al., 2013). During
- 10 WVIs, low level winds from below the Arctic Circle (66 °N) penetrate deep into the Arctic mostly during winter (Johansson et al., 2017). These WVI destabilize the Arctic boundary layer, and can increase both low- and high-cloud coverage over the Arctic, with cirrus clouds increasing mostly in winter by 15-30%. Both Screen et al. (2012) and Francis and Vavrus (2015) found evidence of increased remote energy transport into the Arctic, especially after 2000. This occurred mostly during the fall through enhanced planetary wave amplitudes (Francis and Vavrus,
- 15 2015). This transport may have contributed to the observed increase of Arctic cirrus clouds during winter (Table 4). These remote effects may enhance Arctic winter cirrus and their associated heating rates, which may affect jet-stream dynamics.

7 Summary and conclusions

- A new satellite remote sensing method was developed to retrieve the ice particle number concentration N within
- 20 cirrus clouds, along with effective diameter D_e , extinction coefficient and ice water content (IWC)~~and path (IWP), and visible optical depth (OD)~~. This was made possible by exploiting the fact that most of the ~~ratio of effective difference in retrieved absorption optical depth (τ_{abs})~~ cloud emissivity difference between the split-window channels at ~~4-12.05 and 12.06 μm (β_{eff})~~ is due to wave resonance absorption, a process sensitive to the smallest ice crystals that dominate N (Mitchell et al., 2010). Due to this process, a tight relationship between
- 25 N/IWC and β_{eff} $[\tau_{\text{abs}}(12 \mu\text{m})/\tau_{\text{abs}}(10.6 \mu\text{m})]$ was obtained using PSD aircraft measurements. This relationship, and a similar tight relationship between D_e and β_{eff} , are the unique aspects of this retrieval and make it self-consistent through the shared dependence on β_{eff} . Perhaps the most unique aspect of this retrieval method is its sensitivity to small ice crystals via β_{eff} . The sensitivity ranges ($N/\text{IWC} > \sim 10^7 \text{ g}^{-1}$ & $D_e < 90\text{-}110 \mu\text{m}$) are usually compatible with cirrus clouds ($T < -38 \text{ }^\circ\text{C}$) since PSDs tend to be narrower at these temperatures, containing relatively small ice
- 30 particles.

The N retrieval uses β_{eff} derived from IIR-CALIOP measurements. Although the retrieval is restricted to single-layer cirrus cloud ODs between about 0.3 and 3.0 (which excludes most TTL cirrus), this OD range is likely to be the

most radiatively significant range due to the lower cirrus cloud frequency of occurrence at ~~higher~~ ODs ≥ 3 and a much lower cirrus cloud mean emissivity at ~~the lower~~ ODs ≤ 0.3 (Hong and Liu, 2015). In other words, for the sampled single layer clouds, the cirrus clouds that the IIR senses best in the window channels will also have the most influence on the Earth's longwave radiation budget. Four formulations of the retrieval were used, based on either the SPARTICUS or TC4 field campaigns, with the smallest size-bin of the 2D-S probe either assumed valid ($N(D)_1$ unmodified) or by assuming $N(D)_1=0$. The SPARTICUS unmodified $N(D)_1$ assumption gives the highest N values while the TC4 $N(D)_1 = 0$ assumption yielded the lowest N values. The N predicted from these two formulations differed by a factor of two (see Fig. 5), thus defining a possible systematic uncertainty in N. The random relative uncertainty, $\Delta N/N$, associated to a N retrieval is most of the time < 0.5 for small crystals ($D_e < 35$ -
50 μm), but increases up to more than 2 at the sensitivity limit.

A two-year global and seasonal analysis of these CALIPSO observations reveals that N depends on the latitude zone, season and surface type (land vs. ocean). In the relatively pristine high latitudes, N was relatively high and D_e was relatively low, suggesting that homogeneous ice nucleation may be an important process at these latitudes. In the tropics, N was lowest and D_e was largest on average, relative to the high- and mid-latitudes, with little seasonal dependence. There was considerable seasonal dependence regarding median N and D_e in the mid-latitudes, with N being higher and D_e being smaller during winter (for a given temperature), especially over land in the NH and over ocean in the SH.

The objective of this paper was not to determine absolute magnitudes for the retrieved quantities, but rather to show how they vary in terms of temperature, cloud thickness, latitude, season and topography using any of the four formulations of this retrieval. For a given formulation, these relative differences were similar and were not sensitive to the retrieval formulation used. When the sample selection criteria were relaxed to accept samples having cloud OD > 0.1 (instead of OD > 0.3), median N was reduced by a factor of 1.5 on average while $\Delta N/N$ was more than doubled.

The N retrieval described herein is an advancement of the work described in Mitchell et al. (2016), which supports many of the findings in Gryspeerdt et al. (2018), including the dependence of homogeneous ice nucleation in cirrus clouds on topography and latitude. Consistent with other satellite studies on ice clouds, the optically thicker cirrus clouds we studied exhibited a ~~strong~~ seasonal dependence in the Arctic in regards to frequency of occurrence, with cirrus during winter at least twice as~~at least twice as~~ likely to occur relative to other seasons. This might ~~possibly~~ have a significant effect on jet stream dynamics.

Future cirrus cloud field campaigns designed to sample small ice crystals in regions not representative of the TC4 and SPARTICUS domains (such as at high latitudes or over mid-latitude oceans) may further improve upon this

retrieval framework. It may also be beneficial if more information relevant to remote sensing synergism is taken during in situ observations.

Appendix: Retrieval uncertainty analysis

We begin this analysis with our retrieval equation for the ice particle number concentration, N:

$$N = \frac{\rho_i}{3} \times \frac{\left[\frac{2}{Q_{abs,eff}(12\mu m)} \right] \cdot \tau_{abs}(12.05\mu m)}{\Delta z_{eq}} \times D_e \times \left(\frac{N}{IWC} \right) \quad (A1)$$

with $\rho_i = 0.917 \times 10^6 \text{ g m}^{-3}$. The quantities N/IWC , D_e , and $2/Q_{abs,eff}(12 \mu m)$ are retrieved from β_{eff} using the regression curves given in Table 1 for the four formulations. By writing $x = \beta_{eff}$, they can be expressed as

$$\left(\frac{N}{IWC} \right) (g^{-1}) = 10^9 (a2.x^2 + a1x + a0) \quad (A2)$$

$$D_e (\mu m) = (b2.x^2 + b1.x + b0)^{-1} \quad (A3)$$

$$2/Q_{abs,eff}(12 \mu m) = c2.x^2 + c1.x + c0 \quad (A4)$$

Equation. (A1) can be re-written as:

$$N(L^{-1}) = f(x) \times \alpha_{abs}(km^{-1}) \quad (A5)$$

with

$$f(x) = 10^{-6} \left(\frac{2}{Q_{abs,eff}(12\mu m)} \right) \cdot \left(\frac{N}{IWC} \right) (g^{-1}) \times D_e (\mu m) \times \frac{1}{3.27} = \frac{a2.x^2 + a1x + a0}{b2.x^2 + b1x + b0} \cdot \frac{c2.x^2 + c1x + c0}{1} \times \frac{10^3}{3.27} \quad (A6)$$

and

$$\alpha_{abs} = \frac{\tau_{abs}(12.05\mu m)}{\Delta z_{eq}} \quad (A7)$$

Assuming a negligible error in ΔZ_{eq} , and writing $\tau_{abs}(12.05 \mu m)$ as τ_{12} and $\tau_{abs}(10.6 \mu m)$ as τ_{10} for more clarity, so that $x = \tau_{12}/\tau_{10}$, the derivative of N can be written:

$$\frac{dN}{N} = \frac{1}{f} \frac{\partial f}{\partial x} \cdot x \cdot \left(\frac{d\tau_{12}}{\tau_{12}} - \frac{d\tau_{10}}{\tau_{10}} \right) + \frac{d\tau_{12}}{\tau_{12}} \quad (\text{A8})$$

In Eq. (A8), the derivative of $x = \beta_{\text{eff}}$ is:

$$dx = d\beta_{\text{eff}} = x \cdot \left(\frac{d\tau_{12}}{\tau_{12}} - \frac{d\tau_{10}}{\tau_{10}} \right) \quad (\text{A9})$$

Errors in τ_{12} and in τ_{10} are computed by propagating errors in i) the measured brightness temperatures T_m , ii) the background brightness temperatures T_{BG} , and iii) the blackbody brightness temperatures T_{BB} (Garnier et al., 2015).

The uncertainties in T_{m10} at 10.6 μm and in T_{m12} at 12.05 μm are random errors set to 0.3 K according to the IIR performance assessment established by the Centre National d'Etudes Spatiales (CNES) assuming no systematic bias in the calibration. They are statistically independent.

Because the same cloud temperature is used to compute τ_{12} and τ_{10} , the uncertainty ΔT_{BB} is the same at 10.6 and at 12.05 μm . A random error of $\pm 2\text{K}$ is estimated to include errors in the atmospheric model.

After correcting for systematic biases based on differences between observations and computations (BTDoc) in cloud-free conditions, the random error ΔT_{BG} in T_{BG} is set from the standard deviation of the resulting distributions of BTDoc. [As illustrated in Fig. S1a in Supplementary Materials](#), ~~Over ocean~~, nighttime and daytime standard deviations at 12.05 μm are similar [over ocean](#), and found smaller than over land, where the deviations tend to be larger during daytime than at night. For simplicity, ΔT_{BG} at 12.05 μm is set to $\pm 1\text{K}$ over ocean, and to $\pm 3\text{K}$ over land for both night and day. Standard distributions of BTDoc(10.6 μm) - BTDoc(12.05 μm) ([Fig. S1b](#)) indicate whether the errors in T_{BG} at 10.6 and 12.05 μm are canceling out or not, after accounting for the contribution from the observations, which is estimated to $\sqrt{2} \times 0.3 = 0.45\text{K}$. Standard deviations of [BTDoc(10.6 μm) - BTDoc(12.05 μm)] are found smaller than 0.5 K over ocean and over land during nighttime, which indicates that the errors ΔT_{BG} in T_{BG} at 12.05 μm and at 10.6 μm can be considered identical. They are found locally up to 1 K during daytime over land, which could reflect a variability of the 10.6-12.05 difference in surface emissivity, but also the presence of residual clouds.

Finally, the relative uncertainty $\Delta N/N$ is written as:

$$\left(\frac{\Delta N}{N} \right)^2 = \left[\frac{1}{f} \frac{\partial f}{\partial x} \cdot x \cdot \left(\frac{\partial \tau_{12}}{\tau_{12} \cdot \partial T_{BG}} - \frac{\partial \tau_{10}}{\tau_{10} \cdot \partial T_{BG}} \right) + \frac{\partial \tau_{12}}{\tau_{12} \cdot \partial T_{BG}} \right]^2 \cdot \Delta T_{BG}^2 + \left[\frac{1}{f} \frac{\partial f}{\partial x} \cdot x \cdot \left(\frac{\partial \tau_{12}}{\tau_{12} \cdot \partial T_{BB}} - \frac{\partial \tau_{10}}{\tau_{10} \cdot \partial T_{BB}} \right) + \frac{\partial \tau_{12}}{\tau_{12} \cdot \partial T_{BB}} \right]^2 \cdot \Delta T_{BB}^2$$

$$+ \left[\left(\frac{1}{f} \frac{\partial f}{\partial x} \cdot x + 1 \right) \cdot \frac{\partial \tau_{12}}{\tau_{12} \cdot \partial T_{m12}} \right]^2 \cdot \Delta T_{m12}^2 + \left[\left(\frac{1}{f} \frac{\partial f}{\partial x} \cdot x \right) \cdot \frac{\partial \tau_{10}}{\tau_{10} \cdot \partial T_{m10}} \right]^2 \cdot \Delta T_{m10}^2 \quad (\text{A10})$$

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Supplementary material for “CALIPSO (IIR-CALIOP) Retrievals of Cirrus Cloud Ice Particle Concentrations”, by David L. Mitchell, Anne Garnier, Jacques Pelon and Ehsan Erfani

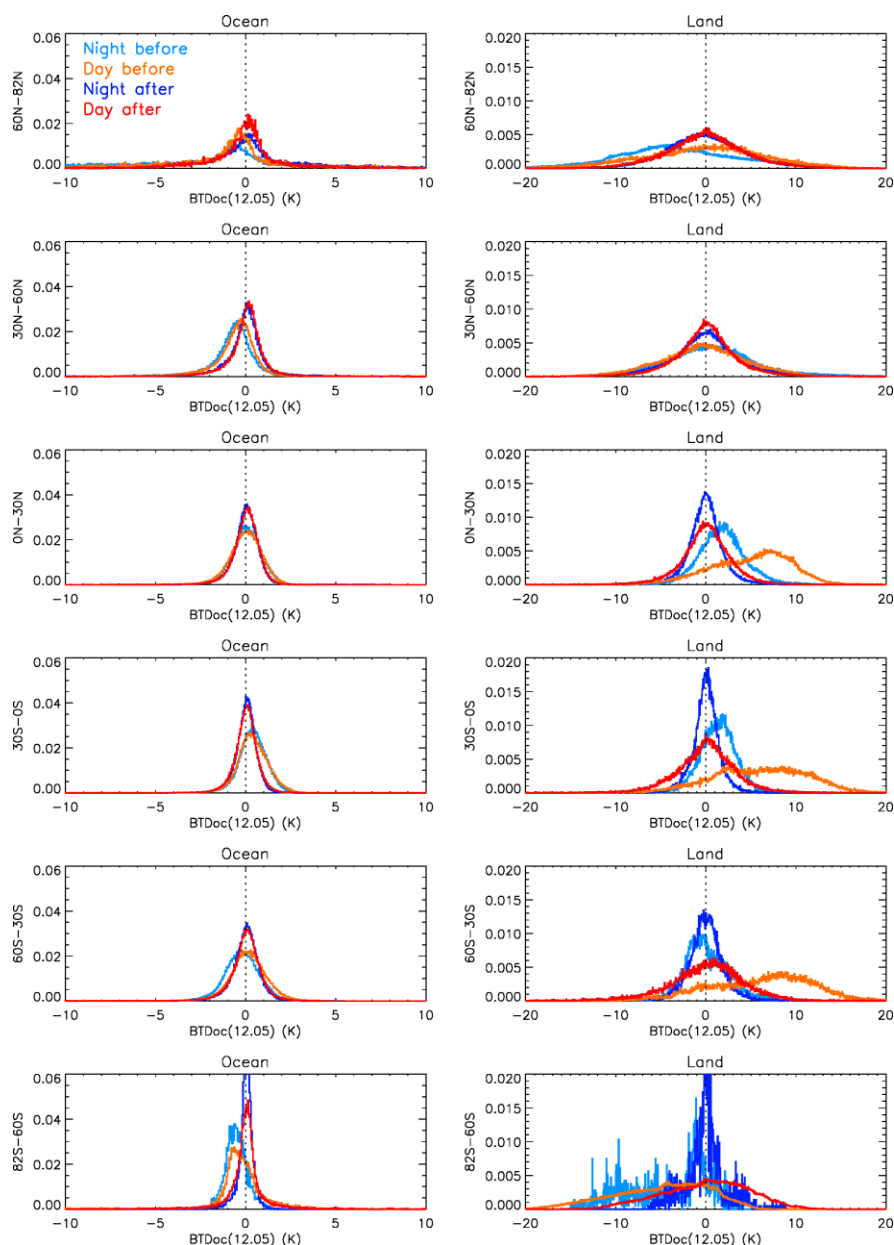


Figure S1a: Probability density functions of the differences between clear sky observations and computations of brightness temperature (BTDoc) at 12.05 μm , before (night: light blue; day: orange) and after (night: navy blue; day: red) correction are shown below for six latitude bands over ocean (left) and over land (right) in January 2008. Note the different scales over ocean and over land.

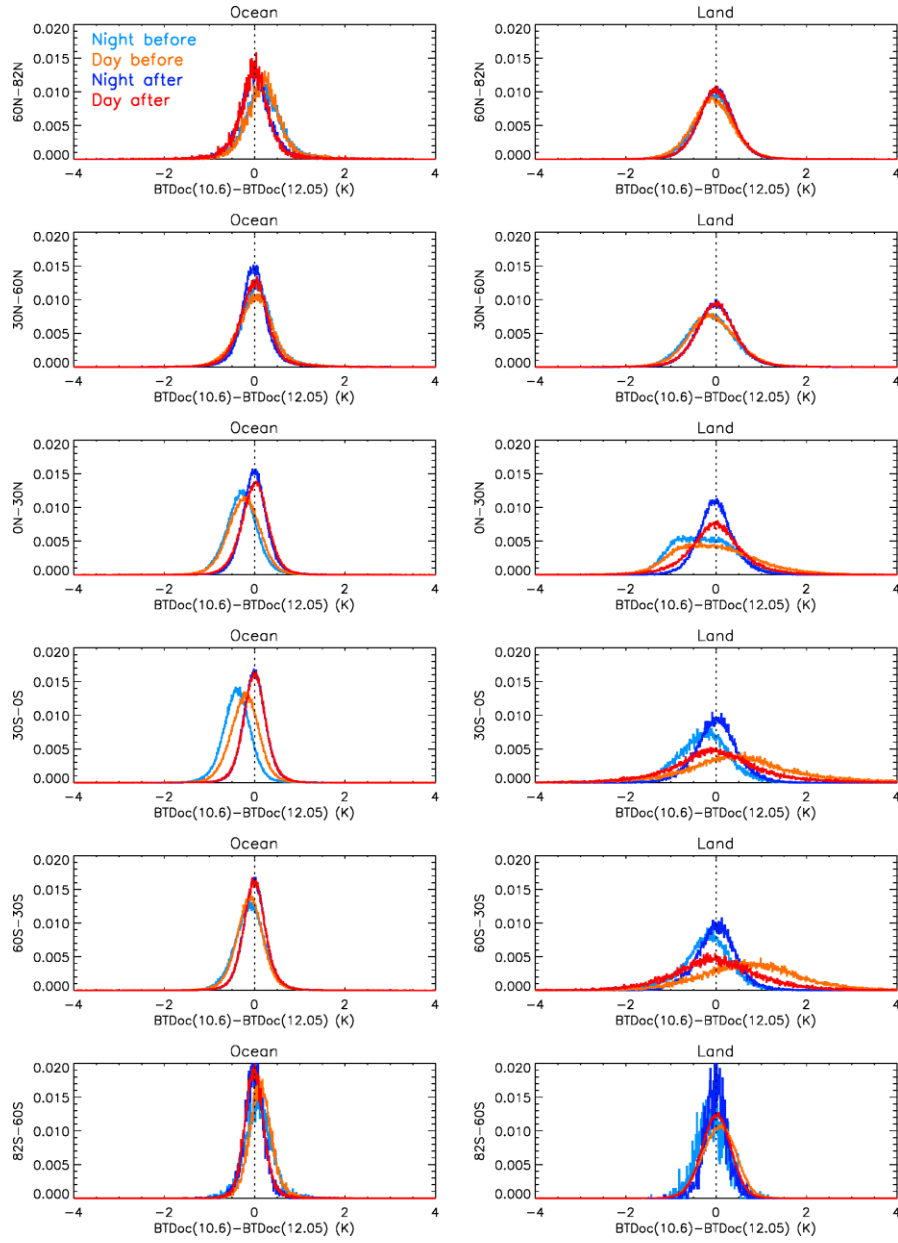


Figure S1b: Same as Fig. S1a, but for the inter-channel difference between observations and computations, $\text{BTDoc}(10.6 \mu\text{m}) - \text{BTDoc}(12.05 \mu\text{m})$.

Impact of the smallest size bin in PSD measurements

Jensen et al. (2013a) argues that the PSD first bin $N(D)_1$ as measured by the 2D-S probe is anomalously high since it tends to be considerably higher than the adjacent size-bins [$N(D)_2$ for example] and that these small ice crystals should rapidly grow or sublimate to larger or smaller sizes ($> 15 \mu\text{m}$ or $< 5 \mu\text{m}$) due to the relative humidity with respect to ice, RH_i , being significantly different than ice saturation ($RH_i = 100\%$). Therefore, $N(D)_1 > N(D)_2$ would imply frequent ice nucleation events to sustain these higher $N(D)_1$ values, which appears unlikely. This argument provided an additional incentive to formulate this retrieval assuming $N(D)_1=0$.

However, there are also physical reasons that argue in favor of assuming that $N(D)_1$ is a valid measurement. For example, if strong competition for water vapor due to a relatively high small ice crystal concentration (e.g. due to a homogeneous ice nucleation event) rapidly reduces the RH_i to $\sim 100\%$, then this relatively high concentration may last for time periods comparable to the lifetime of the cirrus cloud. High ice crystal concentrations (~ 300 to $10,000 \text{ L}^{-1}$) associated with $RH_i \sim 100\%$ were documented by aircraft measurements in the tropical tropopause layer (TTL), existing in layers ranging from meters to 0.4 km in depth (Jensen et al., 2013b). These layers were embedded within a deeper cirrus cloud having N typically less than 20 L^{-1} (where RH_i was higher). Evidence that RH_i near 100% is common in cirrus clouds is shown in Figs. 6 and 7 of Krämer et al. (2009), where for the relationships most representative of cirrus clouds, the relaxation time τ for RH_i to develop a quasi-steady state (i.e. dynamical equilibrium denoted RH_{qsi}) is on the order of 5-10 minutes. RH_{qsi} is where $d(RH_i)/dt \approx 0$, where the rate of vapor uptake by ice approximately balances the rate of supersaturation development. Between the time of initial in-cloud supersaturation (corresponding to cloud formation) and τ , the cloud updraft w tends to be higher than the w occurring after RH_{qsi} is attained. Since cirrus cloud lifetimes tend to be considerably longer than 5-10 minutes, w is relatively low with $RH_i \sim 100\%$ for time $t > \tau$. This may explain the relatively high frequencies of occurrence of RH_i near 100% in Fig. 7 of Krämer et al. (2009). It can also be argued that cirrus clouds are formed by atmospheric wave activity, and that RH_i near 100% results from averaging transient wave-induced fluctuations of RH_i . However, Fig. 1 in Krämer et al. (2009) shows water vapor concentrations being fairly constant with time over periods of 30 to 50 minutes, while also showing evidence of wave-induced fluctuations in RH_i during another period.

Comparisons between the Fast Cloud Droplet Probe (FCDP) and the 2D-S probe during ATTREX when sampling tropical tropopause layer (TTL) cirrus clouds (Woods et al., 2018, Fig. 3) show that the smallest 2D-S size bin often, but not always, measured a lower $N(D)_1$ than did the FCDP. These clouds were not in close proximity to deep convection and were sustained over relatively long periods.

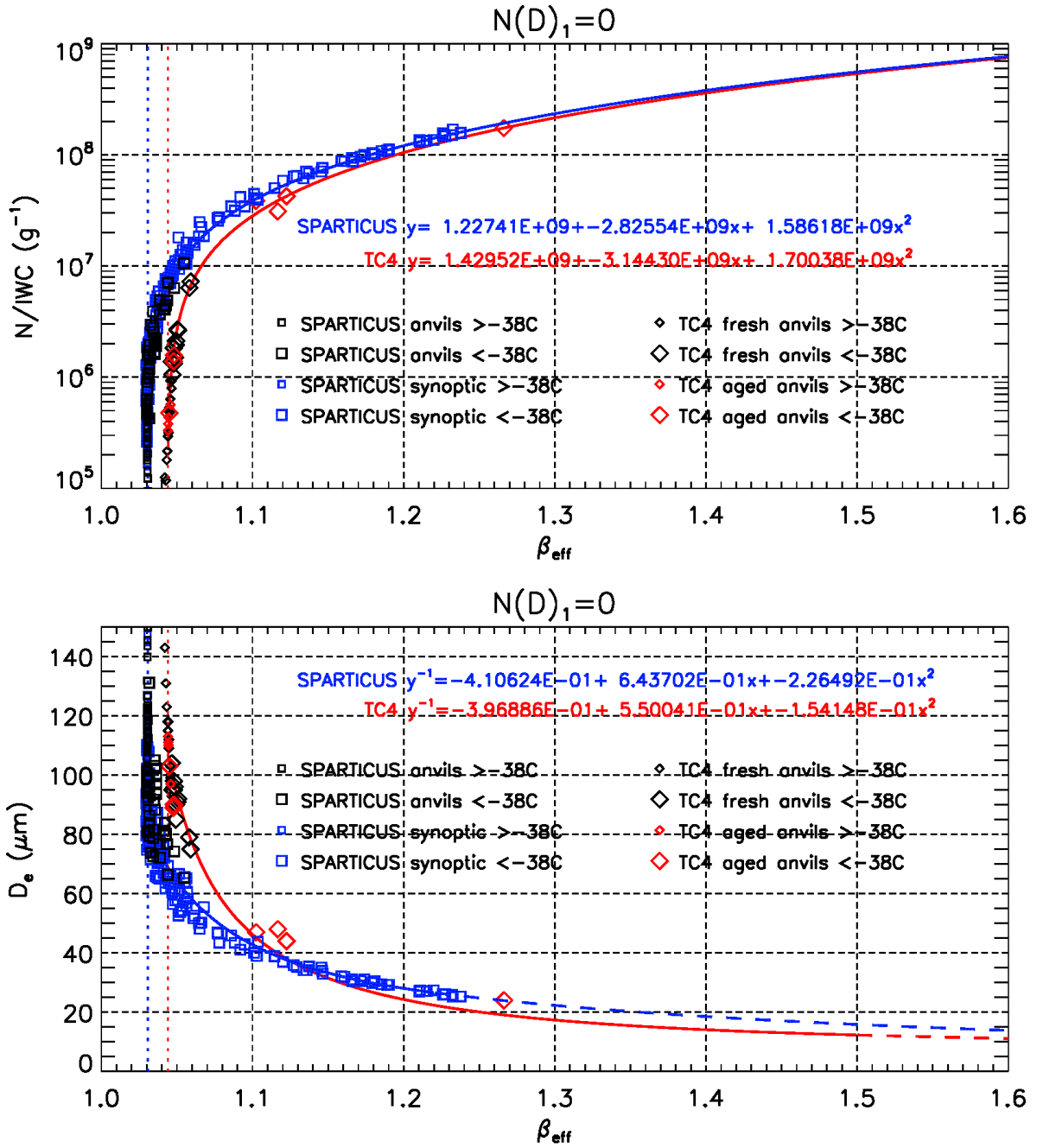


Figure S2: Same as Fig. 2 (top) and Fig. 3 (bottom) in the main paper, but in this case the first size-bin of the PSD is not included (i.e. $N(D)_1=0$). The dashed lines in the lower panels are where the curve-fit equations giving $1/D_e$ in μm^{-1} are extrapolated (see Table 1 in main paper).

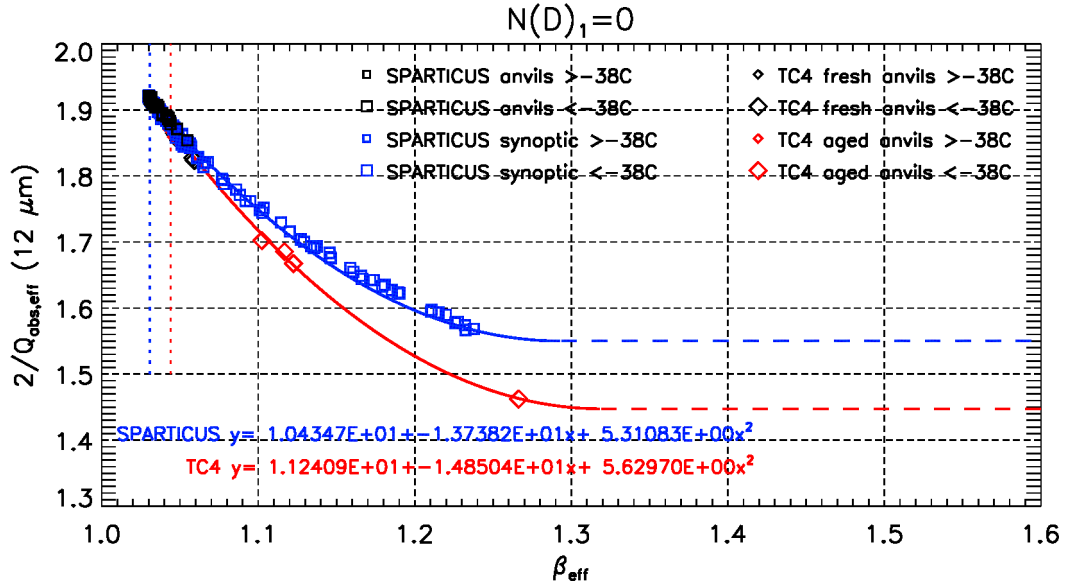


Figure S3: The β_{eff} dependence of the term that converts the absorption optical depth τ_{abs} into visible optical depth in Eq. 7, based on PSDs from SPARTICUS synoptic cirrus (blue squares) and anvils (black squares), and TC4 aged (red diamonds) and fresh (black diamonds) anvils, where the first size-bin is included (top, $N(D)_1$ unmodified), or not included (bottom, $N(D)_1=0$). The larger (smaller) symbols denote PSDs measured at a temperature colder (warmer) than -38°C . The curve-fit equations are for SPARTICUS synoptic cirrus (blue) and for TC4 aged and fresh anvils (red). The dashed lines are where the curve-fit equations are extrapolated (see Table 1 in main paper).

Relationship between β_{eff} , α_{ext} , IWC, and N

As seen from Eq. (6), (7) and (8), β_{eff} and α_{ext} are the two key parameters retrieved from the CALIPSO IIR to derive N/IWC, IWC, and finally N. The interrelationship between β_{eff} , α_{ext} , IWC, and N is illustrated in Fig. S4 (top row) for the SPARTICUS relationships using the unmodified $N(D)_1$ assumption, which also shows the range encountered for these properties in the selected cloud population. The red dashed lines are where $N = 100 \text{ L}^{-1}$, 500 L^{-1} and 1000 L^{-1} . The pink dashed lines are where $\text{IWC} = 0.5 \text{ mg g}^{-3}$, 5 mg g^{-3} , or 30 mg g^{-3} . The horizontal red dotted lines for $\beta_{\text{eff}} < 1.031$ (or $D_e > 83 \mu\text{m}$) indicate where the retrieval is not sensitive to N/IWC. For $\beta_{\text{eff}} < 1.031$, N/IWC is set to its limiting (minimum) value so that N is a priori overestimated in these conditions, but typically smaller than 100 L^{-1} . For $\beta_{\text{eff}} < 1.031$, D_e is set to $83 \mu\text{m}$, as denoted by the horizontal pink lines, and IWC is a priori underestimated for these conditions. For our data selection, α_{ext} is mostly between 0.05 km^{-1} and 5 km^{-1} . Large values of N ($> 500 \text{ L}^{-1}$) result from larger values of β_{eff} (yielding smaller D_e and much larger N/IWC) and sufficiently large values of α_{ext} so that IWC is sufficiently large for these small values of D_e . Low values of N ($< 100 \text{ L}^{-1}$) can be retrieved for small values of β_{eff} , yet larger than the low limit of 1.031, only if α_{ext} is sufficiently small.

This same analysis was repeated in Fig. S5, except the sample selection criteria for minimum OD was changed from 0.3 to 0.1. This increased the sample population considerably. The larger dispersion in β_{eff} and in particular the

larger portion of samples with β_{eff} much smaller than 1 (Fig. S5, top row) are due to large uncertainties at OD between 0.1 and 0.3, which also explain the larger portion of samples with $\Delta N/N > 1$ (Fig. S5, bottom row). More samples now correspond to lower values of α_{ext} (down to 0.016 km^{-1}), IWC, and N.

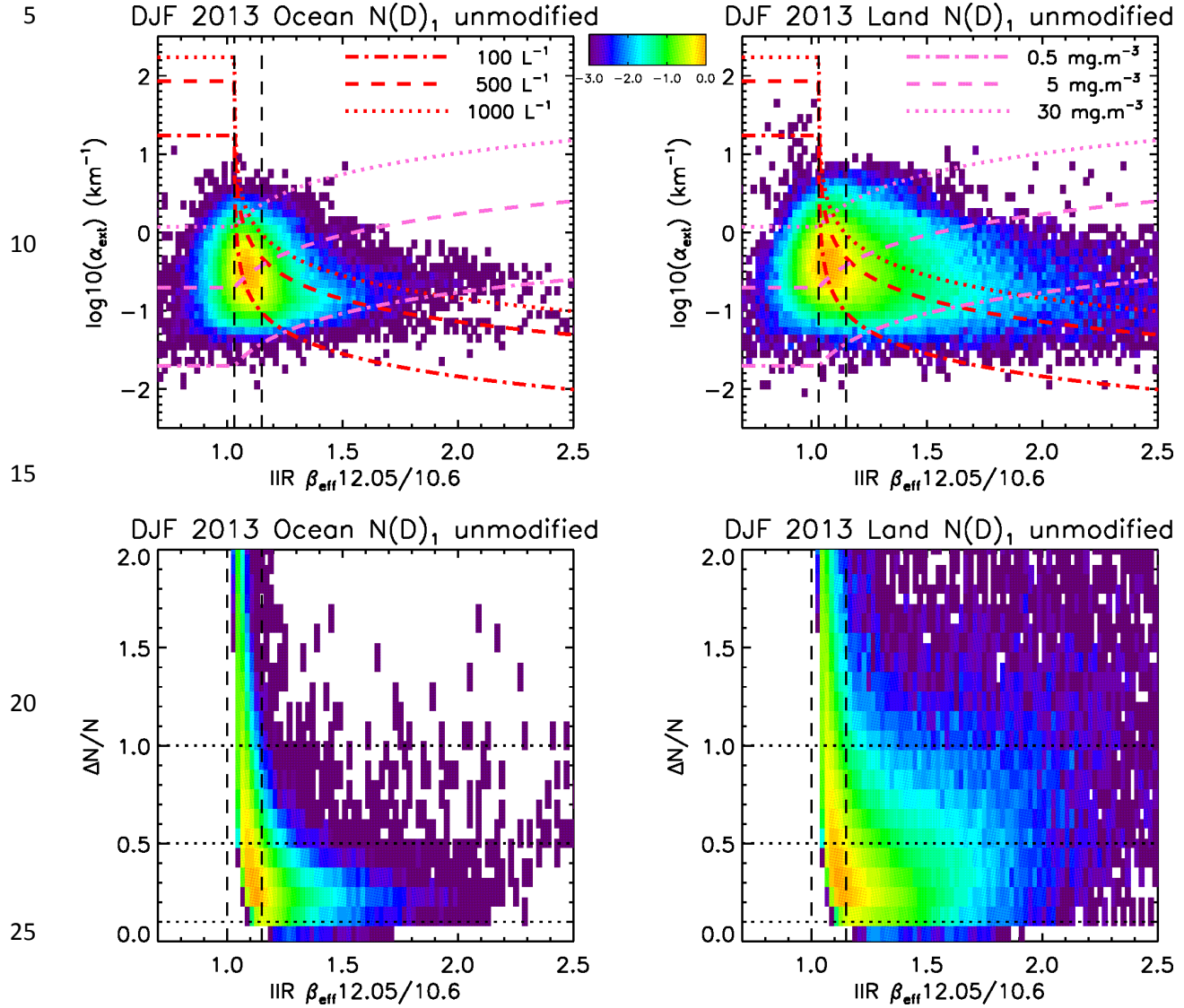


Figure S4: Top: The interrelationship between β_{eff} (X-axis), layer extinction coefficient α_{ext} (km^{-1}) (Y-axis, log10 scale), ice water content IWC, and ice particle number concentration N for the SPARTICUS $N(D)_1$ unmodified assumption. The red dashed lines are where N is equal to 100, 500, or 1000 L^{-1} . The pink dashed lines are where IWC is equal to 0.5, 5, or 30 mg.m^{-3} . Bottom: 2D-distribution of β_{eff} (X-axis) and relative uncertainty estimate $\Delta N/N$. The color bar gives the log of number of samples normalized to the maximum value. Relative uncertainty tends to be considerably smaller at larger β_{eff} values. Left: ocean; right: land; all latitudes; based on December 2013, January and February 2014.

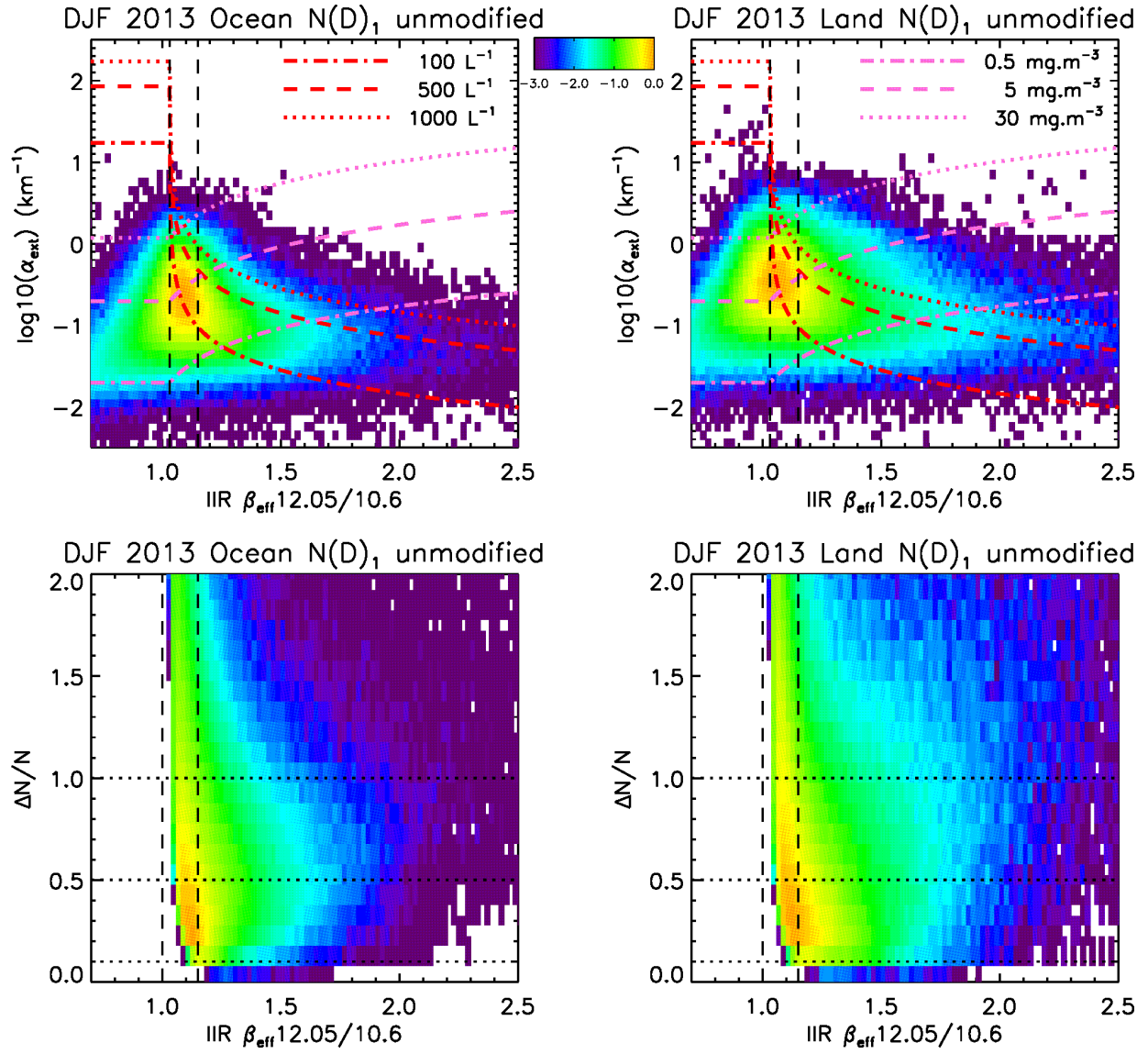


Figure S5: Same as Fig. S4 but sample selection criteria was changed to accept samples having $\text{OD} > 0.1$ approximately. Note the larger portion of samples having $\Delta N/N > 1$.

Comparison of N/IWC with the Krämer cirrus dataset

Although the cirrus cloud measurements in Krämer et al. (2009) occurred over both land and ocean, no distinction was made in this regard. But since CALIPSO IIR β_{eff} uncertainties are greater over land, Fig. S6 separates in situ and satellite retrievals of N/IWC and β_{eff} over ocean (top) and land (bottom). CALIPSO values are averaged over all seasons for 2013 and over the latitude range roughly corresponding to the field measurements (70°N to 25°S). Temperature intervals are 4°C.

Shown in the left panels is N/IWC vs. T_c . The N/IWC curve fits describing the in situ measurements of Krämer et al. (2009) are shown by the grey curves, and correspond to the maximum, minimum and middle (i.e. mid-point) value of a cloud property as a function of temperature. They are compared with corresponding retrieved median values, based on our four formulations: SPARTICUS unmodified $N(D)_1$ (solid navy blue), SPARTICUS $N(D)_1 = 0$ (solid light blue), TC4 unmodified $N(D)_1$ (solid red), and TC4 $N(D)_1 = 0$ (solid orange), all derived from IIR β_{eff} shown in the right panels (black curves). The dashed curves give the 25th and 75th percentile retrieval values. Using our four formulations, in situ N/IWC is converted into four in situ β_{eff} plotted in the right panels for comparison with IIR β_{eff} in black. Comparing both N/IWC and β_{eff} allows visualizing the non-linear relationship between N/IWC and β_{eff} .

Note that the Krämer et al. (2009) data used in Fig. S6 contain several non-zero bins between 5 and 15 microns (i.e. the 1st size-bin of the 2DS probe). Thus, the in situ PSD do not conform with the $N(D)_1 = 0$ assumption. However, as shown in Fig. S6 (left panels), the retrieved N/IWC is weakly sensitive to the $N(D)_1$ assumption. Given the above ambiguities and uncertainties, the agreement between the median retrieved and in situ N/IWC is noticeable, especially for both SPARTICUS relationships over land. Both CALIPSO IIR β_{eff} and in situ β_{eff} are smaller than about 1.25 for temperatures greater than 203 K (-70 °C), in agreement with CALIPSO IIR β_{eff} retrieved during SPARTICUS (Fig. 6a in main paper) and during TC4 (Fig. 6b in main paper).

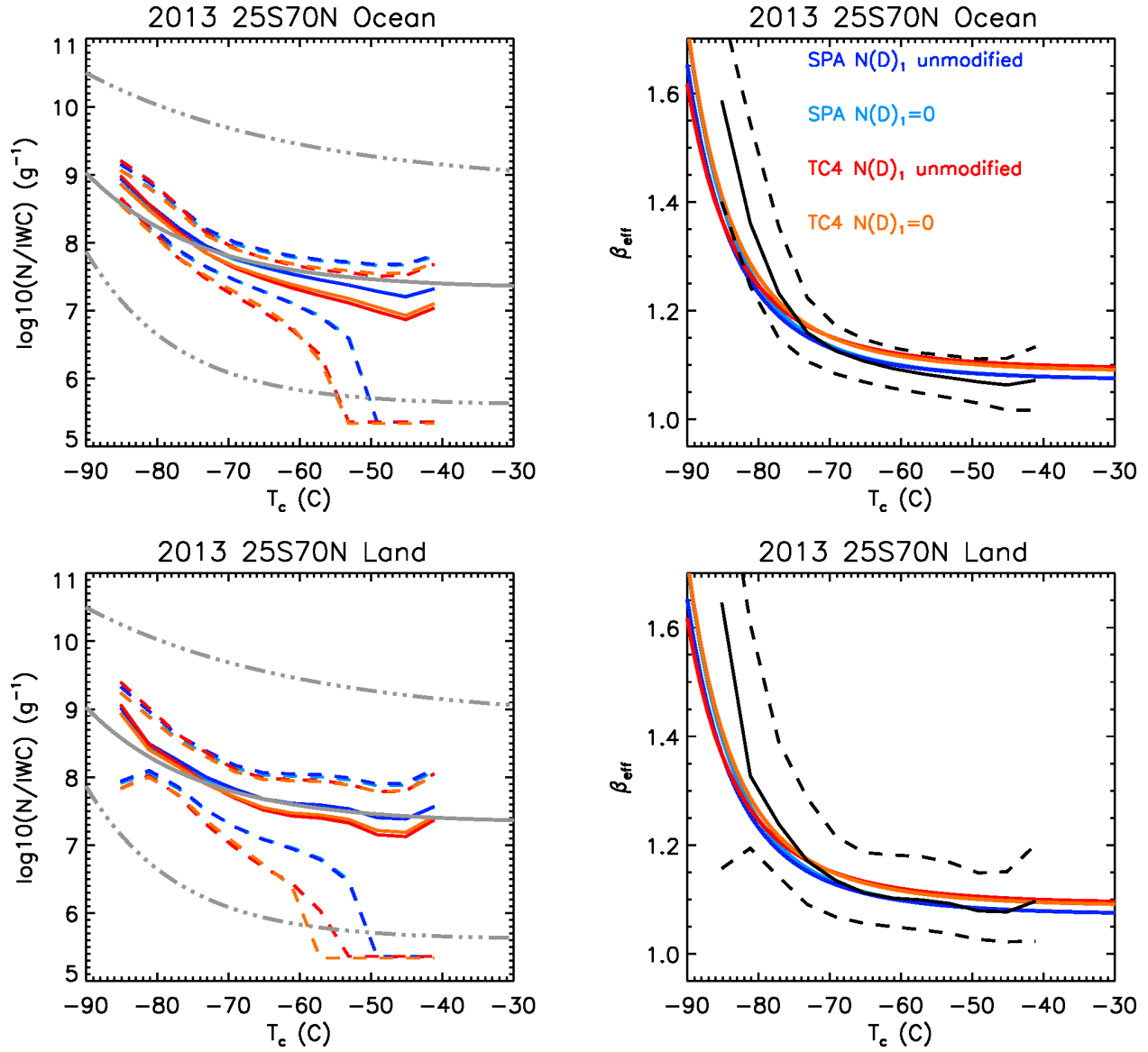
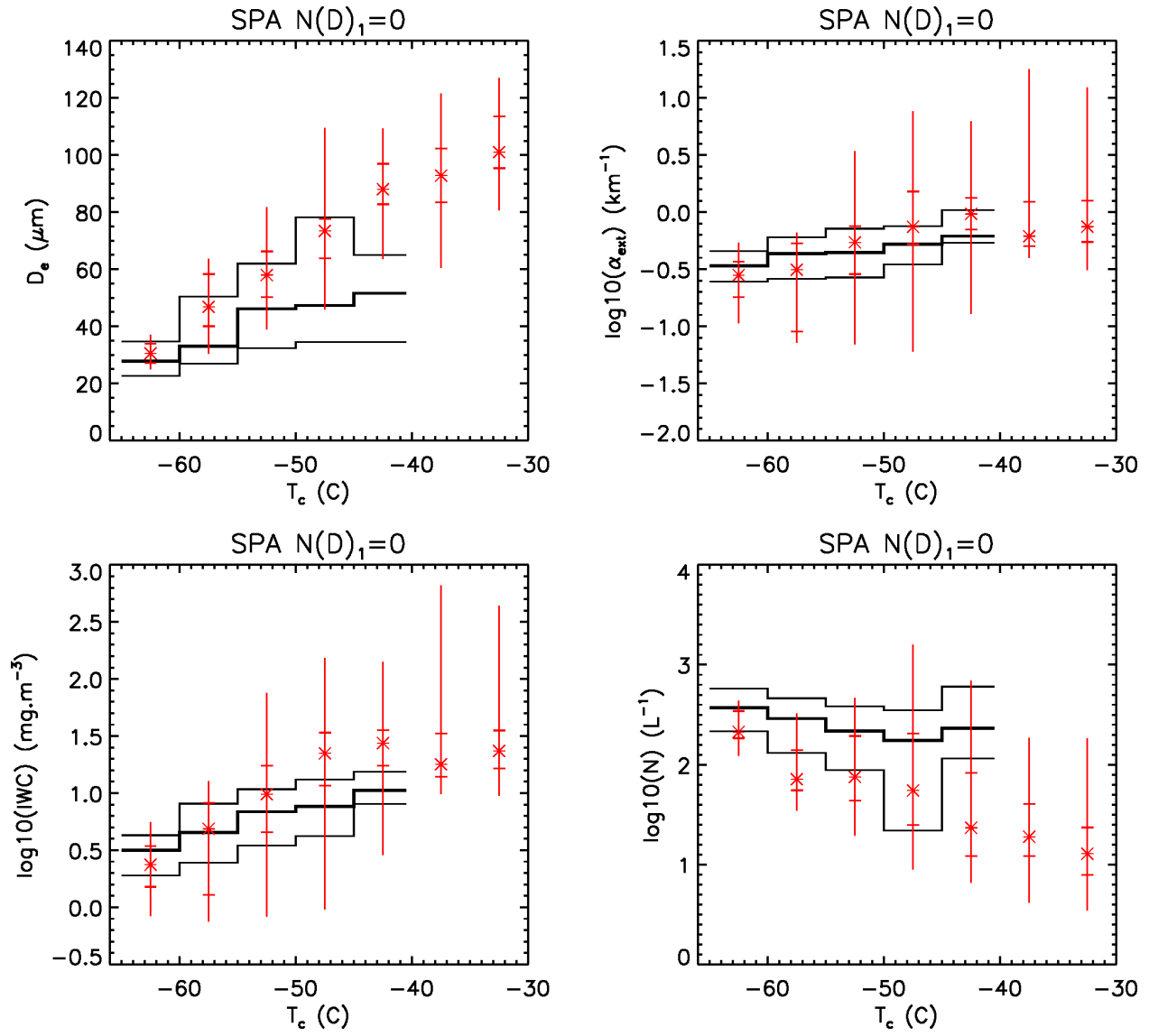


Fig. S6. Left: Comparisons of the median CALIPSO IIR N/IWC (g^{-1}) for the four formulations (colored) with in situ measurements from Krämer et al. (2009) shown by the grey curves; top and bottom being minimum and maximum values and middle grey solid curve being the middle value. Colored solid curves are median values while dashed curves indicate the 25th and 75th percentile values. Right: Comparisons of CALIPSO IIR β_{eff} shown by the black curves (solid curve gives the median value while dashed curves indicate the 25th and 75th percentile values) with the four (colored) in situ β_{eff} inferred from in situ N/IWC (from Krämer et al. (2009)) using the four formulations. Corresponding percentiles are not shown. The navy and light blue curves correspond to the SPARTICUS formulations for the unmodified $N(D)_1$ assumption and the $N(D)_1 = 0$ assumption, respectively. The red and orange curves are using the TC4 formulations for the $N(D)_1$ unmodified and $N(D)_1 = 0$ assumptions, respectively. The CALIPSO IIR retrievals are from 2013 and are for the approximate latitude range (25 °S to 70 °N) of the in situ data, over oceans (top) and over land (bottom).



5 Figure S7. Same as Fig. 7 in the main paper (comparing CALIPSO retrievals with SPARTICUS data), except using the $N(D)_1 = 0$ assumption.

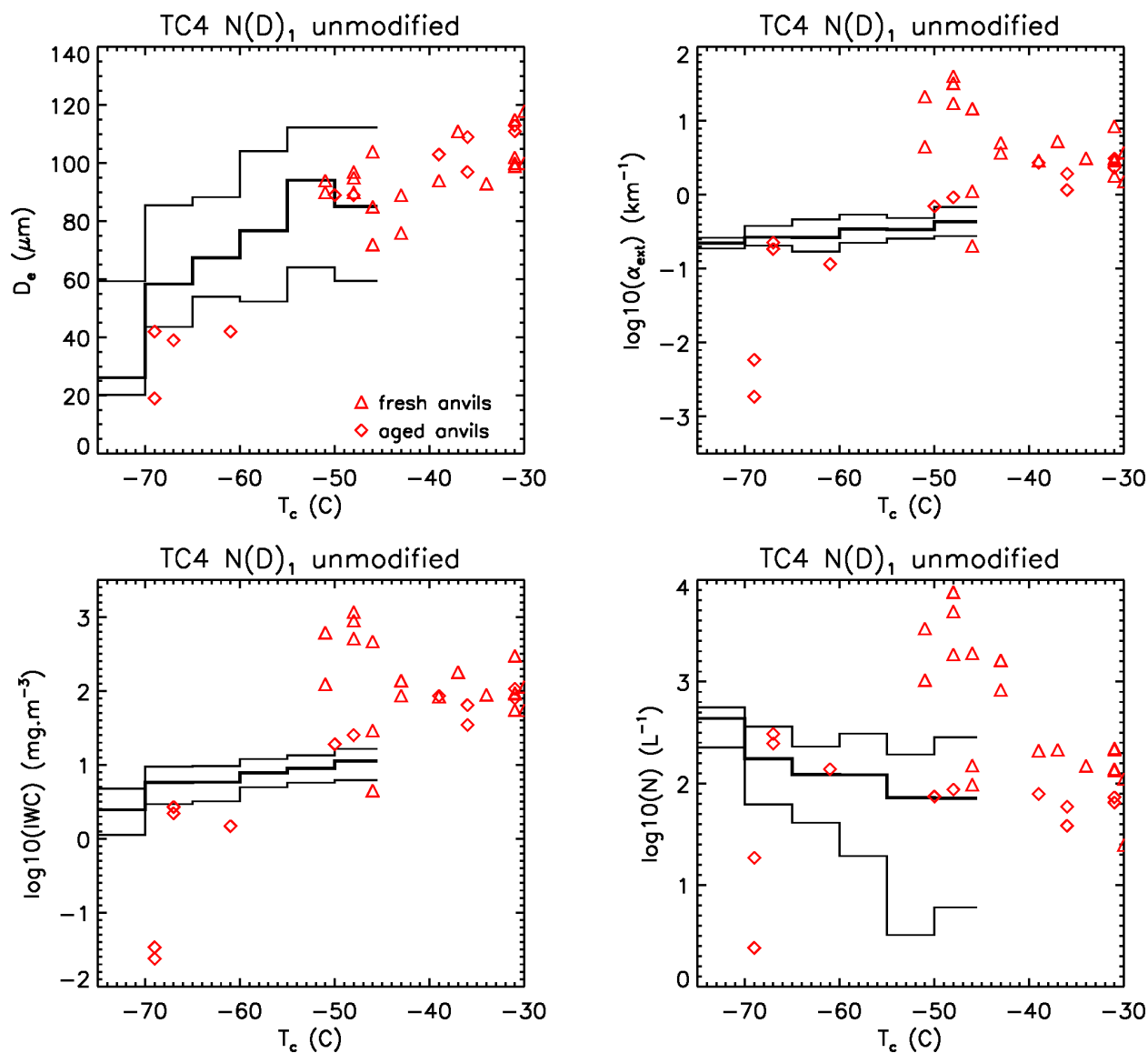


Figure S8. Temperature dependence of effective diameter D_e (μm), extinction coefficient a_{ext} (km^{-1}), ice water content IWC (mg.m^{-3}) and ice particle number concentration N (L^{-1}) during TC4 based on unmodified $N(D)_1$. In situ TC4 measurements in red are for fresh (triangles) and aged (diamonds) anvils. Corresponding CALIPSO IIR retrieved median properties using the TC4 relationships are given by the thick black histograms, with thin black histograms giving the 25th and 75th percentile values.

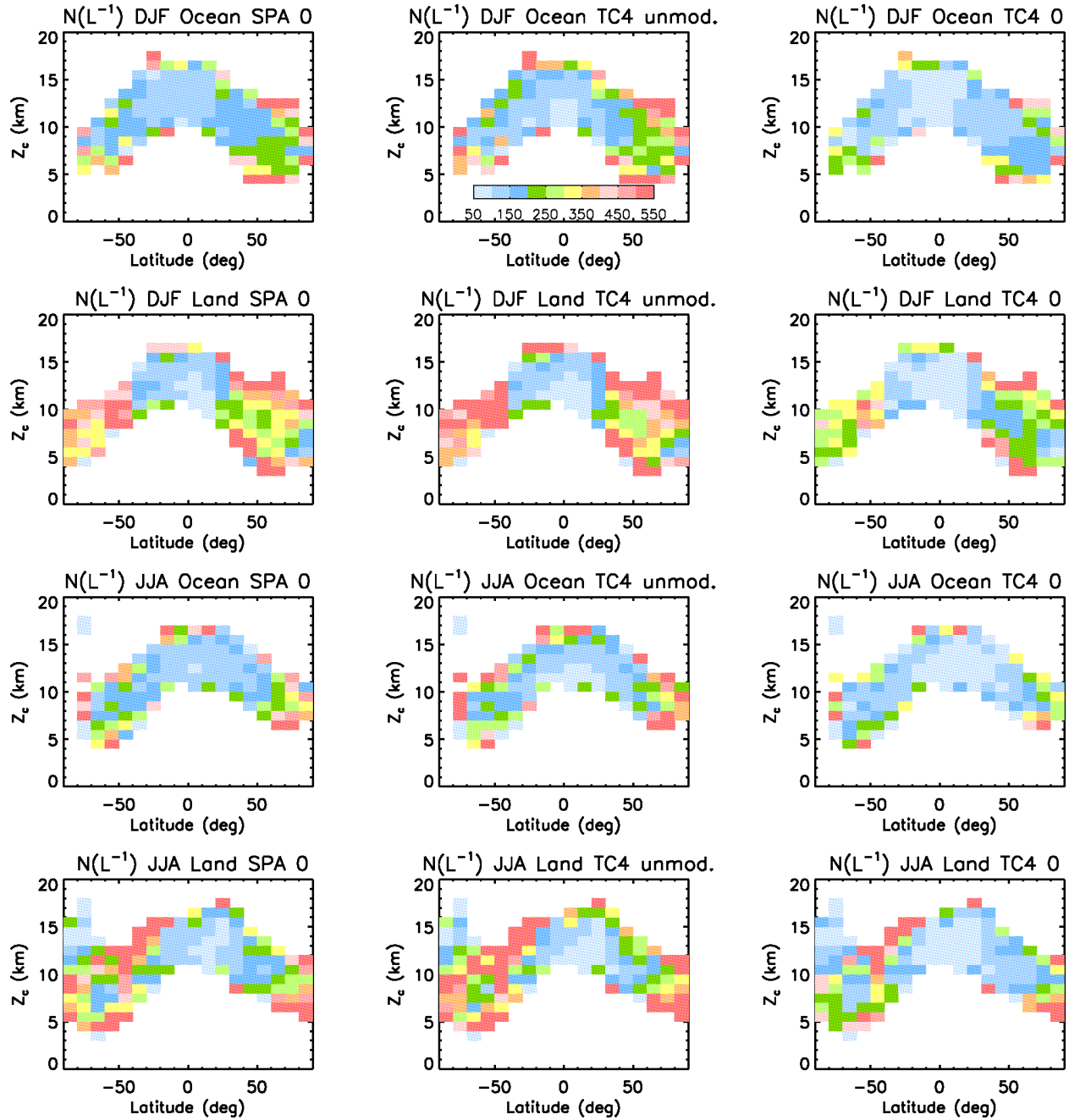


Figure S9: Median ice particle number concentration N (L^{-1}) vs. latitude and representative cloud altitude, Z_c , during 2008 and 2013 using three formulations: SPARTICUS $N(D)_1=0$ (left), TC4 $N(D)_1$ unmodified (center), and TC4 $N(D)_1=0$ (right). Panels from top to bottom are for DJF over oceans, DJF over land, JJA over oceans, and JJA over land.

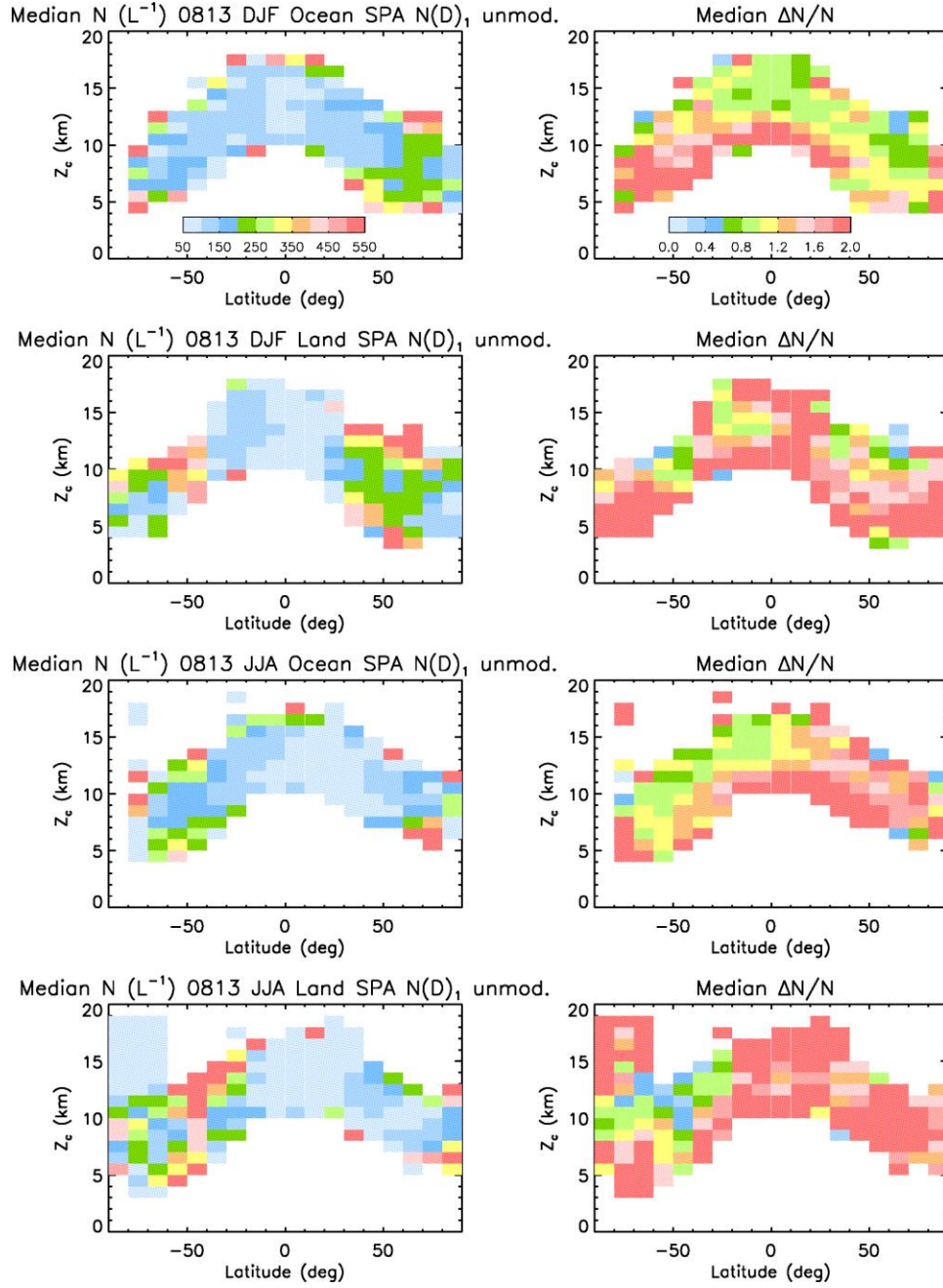


Fig. S10. Same as Fig. 9 in the main paper, but by relaxing the OD threshold to $OD > 0.1$.

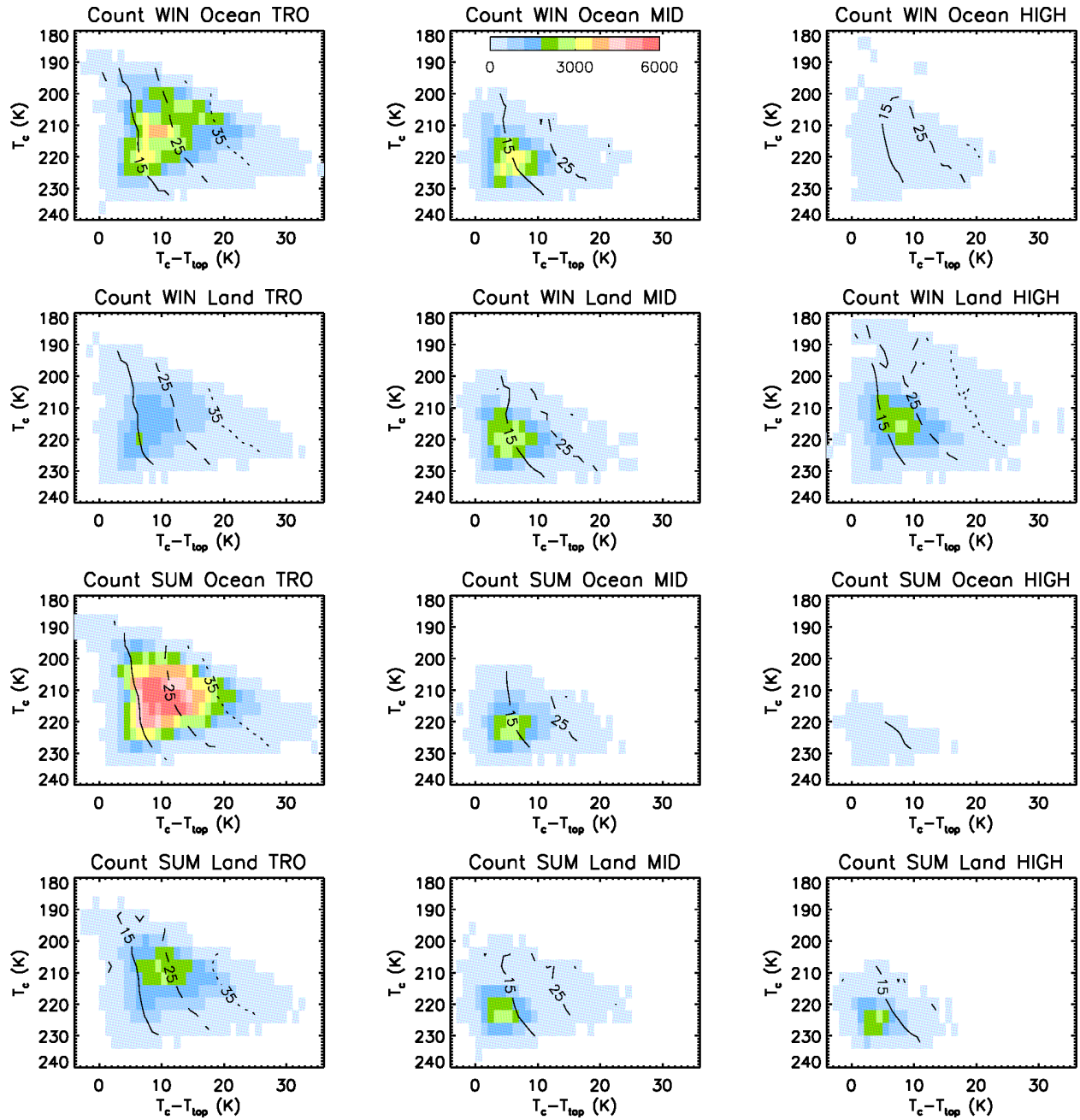


Figure S11. Samples count vs. the representative cloud temperature T_c and $T_c - T_{top}$ at 0° - 30° (TRO, left), 30° - 60° (MID, center), and 60° - 82° (HIGH, right) during 2008 and 2013. Overplotted are isolines of $T_{base} - T_{top}$ (solid: 15 K, dashed: 25 K; dotted: 35 K). Panels from top to bottom are for winter over oceans, winter over land, summer over oceans, and summer over land.

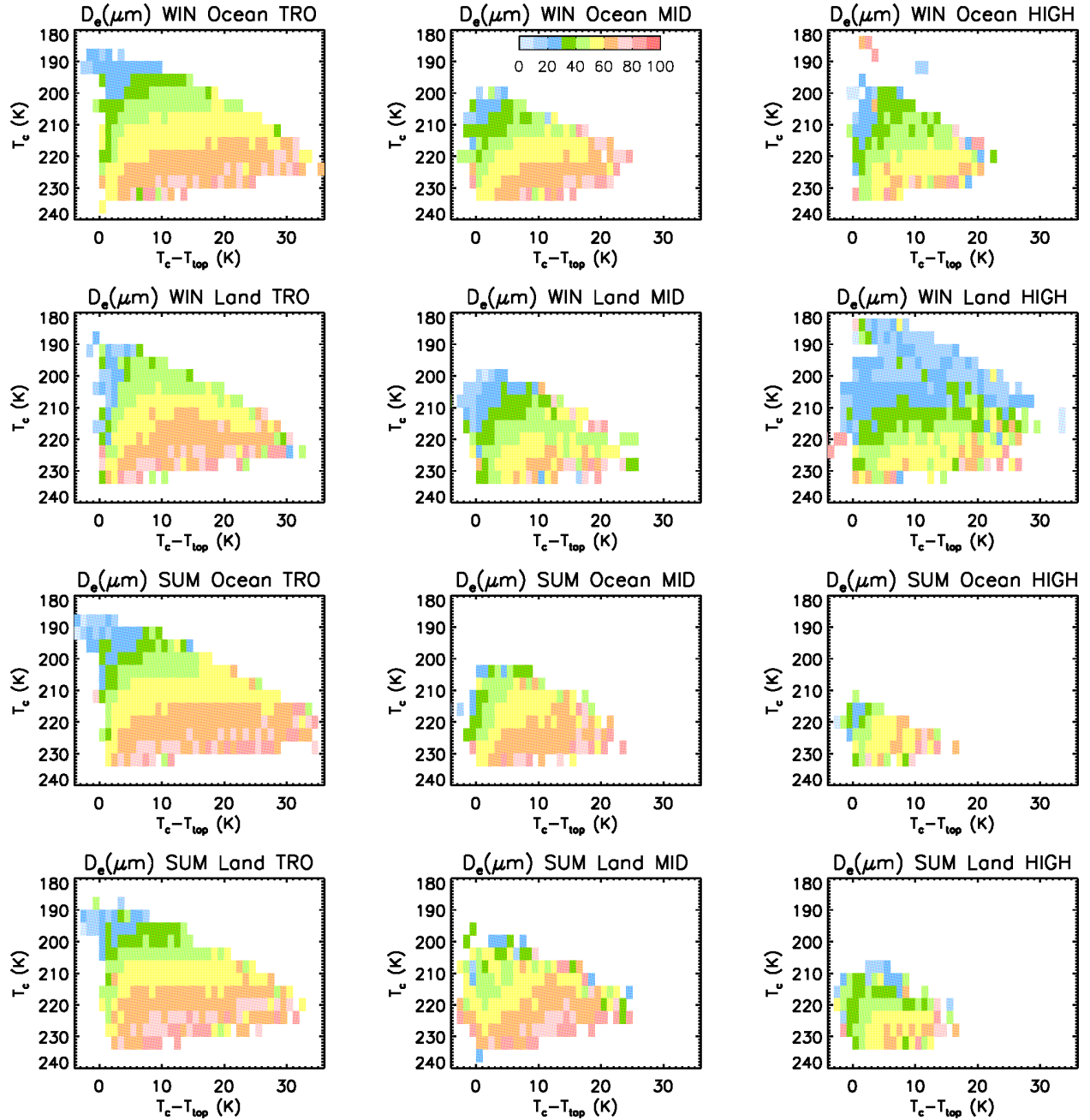


Figure S12: Median retrieved D_e (μm) using the SPARTICUS $N(D)_1$ unmodified formulation vs. the representative cloud temperature T_c and $T_c - T_{\text{top}}$ at 0° - 30° (TRO, left), 30° - 60° (MID, center), and 60° - 82° (HIGH, right) during 2008 and 2013. Panels from top to bottom are for winter over oceans, winter over land, summer over oceans, and summer over land.

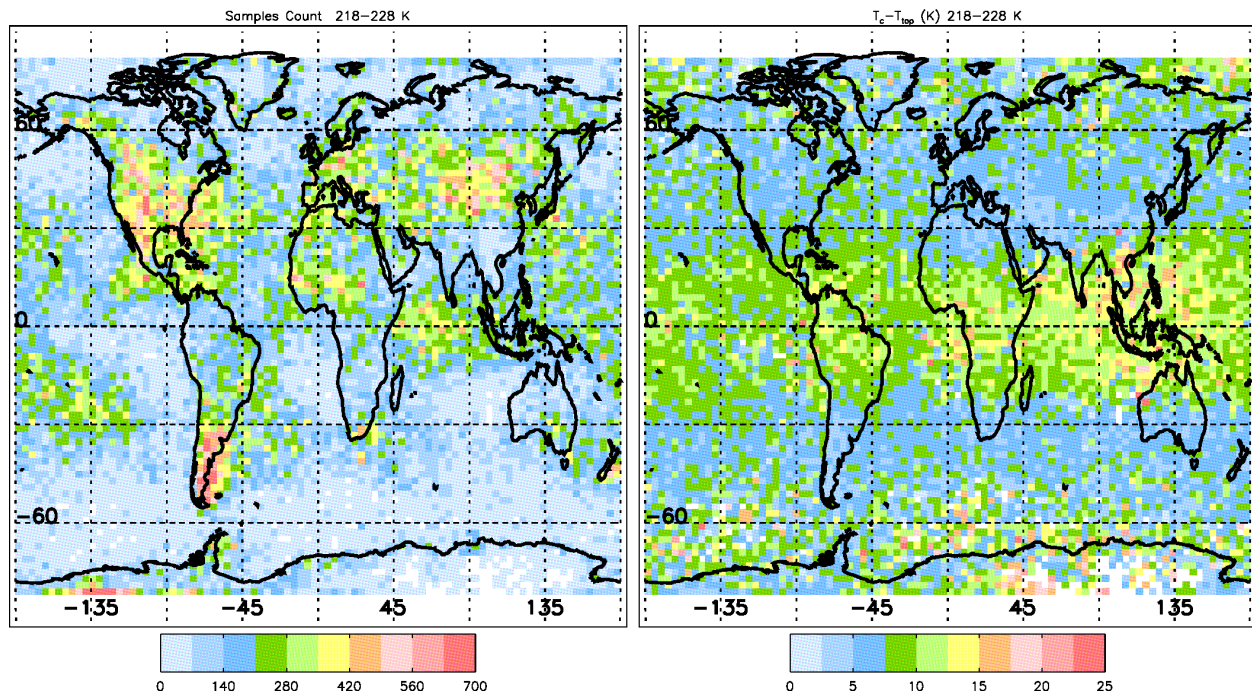


Figure S13: Geographical distribution of (left) number of samples and (right) median $T_c - T_{top}$ values during 2008 and 2013 where the cloud layer representative temperature, T_c , is between 218 and 228 K.