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# **A Microphysics Guide to Cirrus – Part II: Climatologies of Clouds and Humidity from Observations**

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**Abstract.** This study presents airborne in situ and satellite remote sensing climatologies of cirrus clouds and humidity. The climatologies serve as a guide to the properties of cirrus clouds, with the new in situ database providing detailed insights into boreal mid-latitudes and the tropics, while the satellite-borne data set offers a global overview.

To this end, an extensive, quality checked data archive, the Cirrus Guide II in situ database, is created from airborne in situ measurements during 150 flights in 24 campaigns. The archive contains meteorological parameters, ice water content (IWC), ice crystal number concentration ( $N_{ice}$ ), ice crystal mean mass radius ( $R_{ice}$ ), relative humidity with respect to ice ( $RH_{ice}$ ) and water vapor mixing ratio ( $H_2O$ ) for each of the flights. Depending on the parameter, the database has extended by about a factor of 5-10 compared to earlier studies.

As one result of our investigations, we show that the medians of  $N_{ice}$ ,  $R_{ice}$  and  $RH_{ice}$  have distinct patterns in the IWC-T parameter space. Lookup tables of these variables as functions of IWC and T can be used to improve global model cirrus representation and remote sensing retrieval methods. Another outcome of our investigations is, that across all latitudes, the thicker liquid-origin cirrus predominate at lower altitudes, while at higher altitudes the thinner in situ-origin cirrus prevail. Further, exemplary investigations of the radiative characteristics of in situ-origin and liquid-origin cirrus show that the in situ-origin cirrus only slightly warm the atmosphere, while liquid-origin cirrus have a strong cooling effect.

An important step in completing the Cirrus Guide II is the provision of the global cirrus  $N_{ice}$  climatology, derived by means of the retrieval algorithm DARDAR-Nice from ten years of cirrus remote sensing observations from satellite. The in situ measurement database has been used to evaluate and adjust the satellite observations. We found that the global median  $N_{ice}$  from satellite observations is almost two times higher than the in situ median and increases slightly with decreasing temperature.  $N_{ice}$  medians of the most frequently occurring cirrus sorted by geographical regions are highest in the tropics, followed by austral/boreal mid-latitudes, Antarctica and the Arctic. Since the satellite climatologies enclose the entire spatial and temporal  $N_{ice}$  occurrence, we could deduce that half of the cirrus are located in the lowest, warmest (224-242 K) cirrus layer and contain a significant amount of liquid-origin cirrus.

A specific highlight of the study is the in situ observations of cirrus and humidity in the Asian monsoon anticyclone and the comparison to the surrounding tropics. In the convectively very active Asian monsoon, peak values of  $N_{ice}$  and IWC of 30 ppmv and 1000 ppmv are detected around the cold point tropopause (CPT). Above the CPT, ice particles that are convectively injected can locally add a significant amount of water available for exchange with the stratosphere. We found IWCs of up to 8 ppmv in the Asian monsoon in comparison to only 2 ppmv in the surrounding tropics. Also, the highest  $RH_{ice}$  (120-150%) inside of the clouds and in clear sky are observed around and above the CPT. We attribute this to the high  $H_2O$  mixing ratios (3 - 5 ppmv) observed in the Asian monsoon compared to 1.5 to 3 ppmv typically found in the tropics. The observed supersaturations above the

CPT suggest that the water transport into the stratosphere is 10-20% higher than expected in regions of weak convective activity and up to about 50% in the Asian monsoon.

## 40 1 Introduction

In Part 1 of the study (Krämer et al., 2016), a detailed guide to cirrus cloud formation and evolution is provided, compiled from extensive model simulations covering the broad range of atmospheric conditions and portrayed in the same way as field measurements in the Ice Water Content-Temperature (IWC-T) parameter space. The study was motivated by the continuing lack of understanding of the  
45 microphysical and radiative properties of cirrus clouds, which remains one of the greatest uncertainties in predicting the Earth's climate (IPCC, 2013). An important result is the classification of two types of cirrus clouds that differ in formation mechanisms and microphysical properties: relatively thin cirrus that form on-site below  $-38^{\circ}\text{C}$  (in situ-origin), and thicker cirrus originating from freezing in liquid clouds (liquid-origin cirrus) that are uplifted from warmer layers farther below.

50 Since then, a number of studies have been published that shed further light on the exploration of the high ice clouds. For example, some new studies, mostly based on aircraft or lidar observations, provide overviews and climatologies of cirrus cloud properties (Kienast-Sjögren et al., 2016; Petzold et al., 2017; Heymsfield et al., 2017a, b; Woods et al., 2018; Lawson et al., 2019) while others present a more specific view (Urbanek et al., 2017, 2018). Overviews of the properties of cirrus  
55 derived from global satellite remote sensing observations were also recently enhanced to include ice crystal number concentrations (Sourdeval et al., 2018; Gryspeerd et al., 2018; Mitchell et al., 2018). Several studies make use of the concept of in situ-origin and liquid-origin cirrus, e.g. Wernli et al. (2016), investigating the occurrence of in situ-origin and liquid-origin cirrus over the North Atlantic by analyzing ERA-interim data, Gasparini and Lohmann (2016) simulating, amongst other things,  
60 the global distribution of liquid-origin cirrus and Gasparini et al. (2018) presenting climatologies of in situ-origin and liquid-origin cirrus as seen by the CALIPSO satellite and the ECHAM-HAM global climate model. Wolf et al. (2018) studied the microphysical properties of Arctic in situ-origin and liquid-origin cirrus from balloon-borne observations, and Wolf et al. (2019) provide a cirrus parametrization demonstrating the dependence on the origin of the clouds.

65 The wealth of earlier (see e.g. references in Krämer et al., 2016) and new studies has provided insights into formation processes, life cycles and appearance of cirrus. Nevertheless, there are still gaps that need to be filled, on the one hand in the understanding of ice processes, and on the other in the representation of cirrus clouds in climate prediction. A way to accomplish this task requires large and high quality observational databases that can serve, for example, to evaluate global models  
70 or other data sets and be used to derive parameterizations for improved representation of different types of cirrus clouds in models (see e.g. Wolf et al., 2019). In addition, such databases allow detailed studies of special types of cirrus that are still poorly understood, e.g. cirrus in fast updrafts as

orographic cirrus or cirrus at the top of strong convection.

In this study, we approach these requirements as follows: we first compile a data archive of airborne in situ observations which is extended with respect to earlier versions (Schiller et al., 2008; Krämer et al., 2009; Luebke et al., 2013; Krämer et al., 2016) in terms of the size of the data set that contains all parameters needed for the desired studies, i.e. meteorological parameters, ice water content (IWC), number concentration of ice crystals ( $N_{ice}$ ), ice crystal mean mass radius ( $R_{ice}$ <sup>1</sup>), relative humidity with respect to ice ( $RH_{ice}$ ) and water vapor mixing ratio ( $H_2O$ ). However, though airborne in situ measurements best represent detailed microphysical properties of cirrus and their environment, they are always snapshots of specific situations that are also limited by the possibilities of the flight patterns and thus not suitable to derive spatial geographical or seasonal views of cirrus clouds. For this purpose, a globally complete dataset of remote sensing observations from satellite observations are the better option. Hence, as a next step of the study we use in situ climatologies to evaluate cirrus  $N_{ice}$  from satellite observations and, based on this, derive a global climatology of cirrus  $N_{ice}$ . From the portrayal of the two Cirrus Guide II data sets emerging from this study together with some more detailed analyses, we show that the combined evaluation of airborne in situ and satellite remote sensing observations enhances the insights into cirrus properties. The in situ observations are best suitable for the investigation of specific, smaller scale phenomena and for the evaluation of satellite observations or model simulations. Satellite-borne observations, on the other hand, allow a view of the larger scale and seasonal properties.

The article is structured as follows: The Cirrus Guide II in situ databases and the methods used are described in Section 2 and Appendix A. As an overview, in Section 3 we portray the in situ cirrus cloud and humidity database with respect to altitude for the latitudes covered by the observations. The usefulness of the data set is shown by discussing the characteristics and occurrences of in situ-origin and liquid-origin cirrus. Section 4 first presents cirrus and humidity climatologies of the extended Cirrus Guide II in situ data base in terms of temperature in comparison to the earlier studies mentioned above. Further, characteristic properties of mid-latitude and tropical climatologies are presented.

In Section 5 we show another example of a specific analysis extracted from the Cirrus Guide II in situ data base: the data set includes recent unique measurements in the tropical tropopause layer (TTL) region of the Asian monsoon anticyclone, where cirrus clouds and humidity are of special interest and observations are rare. The topic is briefly introduced before the special observations in the Asian monsoon are presented and compared with the conditions found in the surrounding tropical regions.

The last part of the study (Section 6) is the step to global climatology of cirrus  $N_{ice}$  from satellite remote sensing observations. For this purpose, the Cirrus Guide II in situ database is used to evaluate

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<sup>1</sup> mean mass radius  $R_{ice} = \left( \frac{3 \cdot IWC}{4\pi \rho \cdot N_{ice}} \right)^{1/3}$  with  $\rho = 0.92 \text{ g/cm}^3$



110 remote sensing cirrus observations. Based on this, a global  $N_{ice}$  climatology is derived and first analyses of the global and also regional  $N_{ice}$  are presented.

## 2 Databases

### 2.1 in situ data set

The observations presented here include the ice water content IWC, the ice crystal number concentration  $N_{ice}$ , the mean mass radius  $R_{ice}$  in-cloud as well as the clear sky  $RH_{ice}$  and clear sky water vapor volume mixing ratio  $H_2O$ . The complete in situ data set comprises 24 field campaigns: the 115 17 experiments shown in Part I of this study (Krämer et al., 2016, ; the campaigns were performed between 1999 and 2014 over Europe, Africa, Seychelles, Brazil, Australia, USA and Costa Rica), extended by the field campaigns SPARTICUS 2010 and START 2008 over Central US, LTU 2012-120 2018 over Kiruna, CONTRAST and ATTREX in 2014 and POSIDON 2016 over the tropical Pacific as well as StratoClim 2017 out of Nepal.

A map of flights during the various campaigns (extended map of Cirrus Guide: Part I) is shown in Figure 1. In the Appendix A, a summary of the field campaigns and deployed instrumentation is given in Table 4; Table 5 lists all campaigns and the measured parameters. Also, a discussion of new 125 data evaluation methods, data quality and data coverage is presented. An overview of each campaign is given in the Supplementary Material (SM in the following). Twenty campaigns are chosen to be included in the climatologies; four campaigns (marked in Table 5), where the data volume is very low (START 2008, LTU 2012-2018) or very massive (SPARTICUS in 2010, CONTRAST 2014), so that their contribution to frequency occurrences is either negligible or dominant, are shown in the 130 overview of measurements only (SM).

The climatologies are advanced in several aspects in comparison to the compilations of IWC by Schiller et al. (2008), Luebke et al. (2013) and Krämer et al. (2016) and  $N_{ice}$ ,  $R_{ice}$  and  $RH_{ice}$  by Krämer et al. (2009):

- The number of flights and total time in cirrus increased from 104 flights / 94 hours (Krämer 135 et al., 2016) to a total of 150 flights / 168 hours, i.e. the database disproportionally has extended by about a factor of 5-10 depending on the specific parameter.
- For IWC, a new data product has been developed that increases the observed data volume (Appendix A2.1).
- For  $N_{ice}$ , observations from advanced and extended instrumentation could be added to the 140 database; further, a new correction of the occurrence frequencies is applied (Appendix A2.2).
- The geographical spread of the observations has broadened, so that a portrayal of cirrus and humidity with respect to the geographical regions mid-latitude and tropics, and also latitude/altitude seems worthwhile.

- As in the earlier climatologies, all data underwent strict quality control.

## 145 2.2 Satellite data set

DARDAR-Nice provides observational-based estimates of  $N_{ice}$  obtained from CALIPSO and Cloud-Sat measurements (Sourdeval et al., 2018). This unique approach uses the sensitivity of lidar and radar measurements to small and large particles, respectively, to constrain two parameters of a particle size distribution (PSD) parametrization and  $N_{ice}$  is subsequently estimated by direct integration, from a minimum threshold size set in this study to  $5\text{ }\mu\text{m}$ . DARDAR-Nice uses the parametrization by (Delanoë et al., 2005), in which two normalization parameters (a slope parameter  $N_0^*$  and the volume-weighted diameter  $D_m$ ) are used to predict the shape of a PSD. Sourdeval et al. (2018) have demonstrated that the method of Delanoë et al. (2005) is theoretically capable of predicting  $N_{ice}$  from recent in situ campaigns, by comparing its prediction based on in situ  $N_0^*$  and  $D_m$  measurements to the actual in situ  $N_{ice}$  measurement. Good agreements were found, although it was noted that the inability of the modified gamma distribution to match the frequently bi-modal shape of the measured PSDs could lead to an overestimation of  $N_i$  in DARDAR-Nice. This typically occurs at temperature above  $-50^\circ\text{C}$  and is expected to be cloud-type dependent, but Sourdeval et al (2018) showed that  $N_i$  still was in reasonable agreement with the in situ (a factor of 2) down to  $T = -30^\circ\text{C}$ , which should cover the entire cirrus temperature ranges in this study.

This evaluation framework is here repeated on the basis of 5 in situ campaigns archived in the Cirrus Guide II in situ database: COALESC2011, ACRIDICON2014, ATTREX2014, MLCIR-RUS2014, STRATOCLIM2017. Based on the agreement between DARDAR-Nice and the in situ observations, a global  $N_{ice}$  climatology is derived from 10 years of satellite observations. Regional  $N_{ice}$  climatologies for the Arctic (90N - 67.7N), Northern mid-latitudes (67.7N-23.3N), Tropics (23.3N - 23.3S), Southern mid-latitudes (23.3S - 67.7S) and Antarctica (67.7S - 90S) are analysed in more detail. The results are presented in Section 6.

## 3 Vertical distribution of cirrus and humidity from in situ observations

As an introduction, atmospheric temperature profiles in the Arctic, at mid-latitudes and in the tropic are shown in Figure 2, (left panel, adopted from Schiller et al., 2008). Exemplarily, vertical profiles inside of cirrus clouds are shown for 28 flights, using blueish colors for Arctic, greenish for mid-latitude and reddish for tropical observations. It can be seen that in the tropics, with the strongest warming of the Earth's surface by the sun, the air is much warmer at higher altitudes than at mid-latitudes or in the Arctic. The coldest atmospheric temperatures are found at the points where the slopes of the temperature profile reverses: the cold point tropopause (CPT). This is the region where the transition from the troposphere to stratosphere occurs. Above the CPT, in the stratosphere, almost no cirrus clouds are observed because it is too dry for ice formation (Schiller et al., 2009). The

relations between temperature as well as altitude and the potential temperature  $\Theta$  are shown in Figure 2, middle and right panel for the different geographical regions.  $\Theta$  is often used in upper troposphere–lower stratosphere (UT/LS) research, since it allows a clear assignment of air parcels to the associated atmospheric layer, in contrast to the temperature, whose course reverses above the CPT. The range of the tropical tropopause layer (TTL) and the tropical CPT are marked by magenta lines (after Fueglistaler et al., 2009).

### 3.1 Latitude - altitude distributions

As a first application of the Cirrus Guide II data set the distribution of cirrus clouds and humidity is shown with respect to latitude and altitude in Figure 3. Plotted are IWC (color coded by volume mixing ratio),  $N_{ice}$  (color coded by concentration),  $R_{ice}$  (color coded by size), in-cloud as well as the clear sky humidity (color coded by  $RH_{ice}$ ) and  $H_2O$  (color coded by volume mixing ratio). The color codes range from yellow to blue with increasing amount of the respective parameter; note that the data points are plotted in the order of the colors from yellow to blue. The data were collected in the latitude range from about 70°North to around 20°South, i.e. the northern mid-latitudes and the tropics are covered by the observations. The altitude range is between about 5 to 20km. The times of data sampling are displayed in the respective panels; a more detailed description of the database is given in Section 4. The way the data is presented here as individual points was chosen because the entire range of measurements is visible. Although data overlap occurs in this type of display, it is possible to identify cirrus types and microphysical processes, especially based on extreme values. As additional overview information, we have created latitude-altitude intervals (500 m altitude, 0.5° latitude) and calculated the 25, 50 (median) and 75 % percentiles for all variables. These additional altitude-latitude climatologies are shown in the SM (Figures 1–3).

Cirrus clouds are found at lower altitudes at mid-latitudes and reach higher levels in the tropical region (Figure 2). Though this structure in the measurements is influenced by the maximum or minimum height the engaged aircraft can reach, it corresponds well with the CALIPSO latitudinal height distribution of cirrus clouds, which is largely caused by the decrease in tropopause height with increasing latitude (shown in the review article by Heymsfield et al., 2017a, original figure by Sassen et al. (2008)).

#### 3.1.1 in situ-origin and liquid-origin cirrus

Krämer et al. (2016), Luebke et al. (2016) and Wernli et al. (2016) describe two different cirrus types: (1) in situ-origin cirrus that form (by heterogeneous or homogeneous ice nucleation on ice nucleating particles, INP, or soluble solution aerosol particles, respectively) from water vapor directly as ice at  $T < 235K$ ,  $RH_{ice} > 100\%$  and  $RH_w < 100\%$ ; (2) liquid-origin cirrus that evolve (also heterogeneously or homogeneously) from freezing of liquid drops in clouds at  $T \gtrsim 235K$  and

$RH_w \sim 100\%$ <sup>2</sup>. In other words, in situ-origin cirrus are observed at the altitudes where they are formed, whereas liquid-origin cirrus are glaciated liquid clouds from further below which are lifted to the cirrus temperature region where liquid water no longer exists.

*Microphysical characteristics:* In the new in situ data set, containing advanced measurements and extended by several field campaigns in comparison to the earlier studies, some typical characteristics of the cirrus types and hints to ice nucleation mechanisms are visible. In the following, the cirrus types are briefly introduced, and, using Figures 3 and 4, the types and freezing mechanisms are discussed and summarized in Table 1.

IN SITU-ORIGIN cirrus can be divided in two sub-classes depending on the strength of the updraft: in *slow updrafts*<sup>3</sup>, in situ-origin cirrus form mostly heterogeneously and are rather optically thin with lower IWCs and few but large ice crystals. We like to note here that in an atmosphere free of INPs, cirrus clouds forming homogeneously would have similar characteristics, since also only a small number of ice crystals nucleate homogeneously in slow updrafts (see e.g. Spreitzer et al., 2017). Hence, in this regime, the freezing process is not relevant for determining the cirrus properties. In *fast updrafts*<sup>3</sup>, homogeneous freezing mostly occurs regardless of the presence of INPs, since fast updrafts cause  $RH_{ice}$  to reach the homogeneous freezing threshold even after heterogeneous freezing. The in situ-origin cirrus that emerge in these situations are optically thicker with higher IWCs and more but smaller ice crystals.

LIQUID-ORIGIN cirrus stem from lower altitudes, where more water is available, so they generally consist predominantly of thicker cirrus with higher IWC, together with more and larger ice crystals that are frozen heterogeneously at  $T > 235K$  in *slower updrafts*. In *fast updrafts*, liquid-origin cirrus with very high  $N_{ice}$  appear. The reason is that in such updrafts  $RH_w$  and  $RH_{ice}$  are both  $> 100\%$  and thus the Wegener-Bergeron-Findeisen process, where liquid drops evaporate and ice crystals remain at  $RH_w < 100\%$  and  $RH_{ice} > 100\%$ , is suppressed (Korolev, 2007). Thus, the numerous supercooled drops reach the altitude (temperature  $\sim 235K$ ) where they freeze homogeneously (Costa et al., 2017).

The meteorological situations where slow updraft in situ-origin cirrus frequently occur (see Krämer et al., 2016) are low- and high-pressure systems (frontal/synoptic cirrus). The warm conveyor belt in low pressure systems can also produce liquid-origin cirrus. Fast updraft in situ-origin and liquid-origin cirrus occur in gravity waves, often orographically induced, in jet streams and in convective systems and anvils.

As outlined in the Cirrus Guide I (Krämer et al., 2016), to a certain extent cirrus types can be identified by their typical characteristics. This applies to the initial stage, after which the clouds lose the signature of the formation process e.g. by sedimentation: as long as an updraft prevails (corre-

<sup>2</sup>For a more detailed description of the freezing mechanisms see Vali et al. (2015); Heymsfield et al. (2017a) and references therein.

<sup>3</sup>slow updrafts:  $\lesssim 10\text{ cm/s}$ ; fast updrafts:  $10\text{ cm/s} - 10\text{ m/s}$  (based on simulations of Kärcher and Lohmann, 2002; Krämer et al., 2016)

sponding  $RH_{ice} > 100\%$ ), smaller ice crystals in the size range  $\lesssim 20\mu\text{m}$  grow to larger sizes on a time scale of tens of minutes. The larger ice crystals sediment to lower altitudes, thus removing ice surface from the cloud volume which consequently reduces the depletion of  $H_2O_{gas}$  ( $\propto RH_{ice}$ ) by water vapor transport to the ice (for more detail see Spichtinger and Cziczo, 2010). The ice crystals that have fallen out of the layer deepen the cirrus extent to lower altitudes (fall streaks can extend the cirrus to several km below the nucleation level Jensen et al., 2012; Murphy, 2014), while at the same time, large ice crystals from above could sediment into the cloud volume. Altogether, the cirrus evolution is a dynamical process and the cirrus properties change in the course of a cirrus lifetime. At the final cirrus stage, i.e. when the temperature increases, the environment becomes subsaturated ( $RH_{ice} < 100\%$ ) and ice crystals evaporate, disappearing the faster the smaller they are (timescales of growth and evaporation of ice crystals are shown in Kübbeler et al., 2011, their Figure 12).

Cirrus types with the most striking features (high  $IWC / N_{ice}$ ) are the easiest to identify. They could be liquid-origin or in situ-origin cirrus in fast updrafts or also liquid-origin cirrus in slow updrafts. This will be shown on the basis of Figure 3 and Figure 4.

In Figure 4, the relation between  $N_{ice}$ ,  $R_{ice}$  and  $IWC$  is shown for  $\sim 87$  h of cirrus cloud observations. It can be nicely seen that in the cases where  $N_{ice}$  is high, the ice crystals are small – because the numerous ice crystals consumed all of the available vapor, thereby suppressing further growth – while low  $N_{ice}$  are related to large ice crystals for a given  $IWC$ . The  $IWC$  (in  $\text{mg}/\text{m}^3$ ) is indicated by the color code and result from the combination of both  $N_{ice}$  and  $R_{ice}$ . The thin black lines are isolines of  $IWC$  (in  $\text{ppmv}$ ) that appear in the order shown the legend. The scheme at the right side illustrates the partitioning of the clouds between liquid and in situ-origin: the thickest cirrus (blue points) are of liquid-origin, the thinnest (yellow points) of in situ-origin. As the thickness of the cirrus decreases, the fraction contributed by liquid-origin cirrus becomes smaller and smaller while an increasing fraction is due to in situ-origin cirrus.

Most of the highest  $IWC$ s ( $> 10 \text{ mg}/\text{m}^3$ , dark red and blue diamonds in Figure 3, top left panel) are of liquid-origin. They appear in the lower parts of the clouds, i.e. they are uplifted from farther below. A good example of this is the field campaign SPARTICUS in 2010, which is separately plotted in Figure 7 of the SM (right panel). About 23 hours of sampling in mostly liquid-origin clouds were performed over central USA. The clouds were observed in the temperature range 210–240 K, corresponding to altitudes between 5–10 km, i.e. they were rather low cirrus clouds. The  $IWC$ s are mostly high (red to blue colors) and, as shown by Muhlbauer et al. (2014), ice particles of greater than a thousand  $\mu\text{m}$  diameter were frequently encountered, indicative of liquid-origin cirrus (see Table 1). In the tropics (Figure 3), some blue points are also detected at high altitudes of about 17 km from measurements made above the Asian monsoon in strong convective updrafts (see also Section 5) and consist of many ice crystals ( $0.1\text{--}10 \text{ cm}^{-3}$ ) with medium  $R_{ice}$  (20–70  $\mu\text{m}$ ).

Generally, the  $IWC$  roughly shows a vertical structure of decreasing  $IWC$  with increasing altitude. This is caused on the one hand by the amount of available water that decreases with decreasing

285 temperature, but also because cirrus of liquid-origin predominate in lower layers, whereas cirrus  
with in situ-origin become more abundant at higher altitudes, i.e. colder temperatures. This is in  
accordance with the findings of Luebke et al. (2016); Wernli et al. (2016) and Wolf et al. (2018),  
where Luebke et al. (2016) and Wolf et al. (2018) experimentally investigated the two cloud types in  
mid-latitudes and in the Arctic, respectively, while Wernli et al. (2016) analyzed 12 years of ERA-  
290 interim data in the North Atlantic region (see also Section 6.2).

As IWC is an indication for the cirrus type,  $N_{ice}$  (Figure 3, middle left panel) provide a hint to the  
freezing mechanism, either heterogeneous or homogeneous: high ice crystal numbers  $N_{ice}$  ( $\gtrsim 0.5$   
 $\text{cm}^{-3}$ , dark red and blue diamonds, middle left panel of Figure 3) are an indicator for homogeneous  
ice formation in fast updrafts caused by waves or convection, both for in situ-origin as well as for  
295 liquid-origin cirrus. At mid-latitudes, they are found e.g. at the tops of mountain wave clouds (the  
ice nucleation zone), which were observed for example behind the Norwegian mountains at around  
 $62^\circ\text{North}$ . High  $N_{ice}$  values have also been observed in tropical deep convection around  $10^\circ\text{North}$ ,  
corresponding to measurements reported by Jensen et al. (2009).

Another source of mid-latitude high  $N_{ice}$  are young contrails. Note however, that these high  $N_{ice}$   
300 exist only for a short time. This is because high ice crystal numbers are associated with small ice  
crystal sizes (yellow diamonds in the bottom left panel, see also Figure 4) that grow/evaporate  
quickly. At lower altitudes,  $N_{ice}$  tends to be lower and the crystals are larger, because large crystals  
having lower concentrations sediment out from the cloud tops and, as mentioned above, liquid-origin  
cirrus with characteristic large ice crystals are also common in this altitude range. In the tropics, high  
305  $N_{ice}$  at high altitudes are induced by in situ homogeneous freezing in convection or gravity waves  
(see also Jensen et al., 2013a). At cloud bases, such high  $N_{ice}$  are most probably of liquid-origin,  
initiated by homogeneous freezing of supercooled drops.

A more detailed discussion of the microphysical properties of cirrus including frequencies of oc-  
currence of specific signatures which can not be seen from Figure 3, is given in Section 4.  
310

*Radiative characteristics:* A motivation to study cirrus clouds is to investigate the radiative prop-  
erties on the basis of the findings on their microphysical properties. In the Cirrus Guide I, Krämer  
et al. (2016) speculated that the physically and optically thinner in situ slow updraft cirrus cause a  
warming effect, while thicker fast updraft in situ-origin and, particularly, thick liquid-origin cirrus  
315 have the potential to cool. Here, we show a first estimate of the radiative forcing of typical in situ-  
origin slow and fast updraft as well as liquid-origin cirrus (Figure 5). To this end, radiative transfer  
calculations for idealized scenarios under specific conditions are realized, at noon at the equinox,  
which are briefly described in the caption of Figure 5.

In the left panel of Figure 5, the radiative forcing of the slow (light green) and fast (dark green)  
320 in situ-origin cirrus is displayed with respect to optical depth. This panel is expanded from the right  
panel, where the forcing of the liquid-origin cirrus is shown. Obviously, the net radiative forcing of

the in situ-origin cirrus is much smaller than that of liquid-origin and, moreover, change the sign from warming to cooling.

In more detail, the slow in situ-origin cirrus have only small optical depth ( $\tau$ ) between 0.001 - 0.05, resulting in a slight net warming effect of not larger than about 1.5 W/m<sup>2</sup>. The optical depth of fast in situ-origin cirrus is larger ( $\tau$ : 0.05 - 1), but most of them are also warming (2-10 W/m<sup>2</sup>). The thickest fast-updraft in situ-origin cirrus at the lowest altitudes change the sign of their net forcing, they switch to a slight cooling effect. The reason is the warmer temperature at lower altitude that reduces the warming effect of the longwave infrared radiation. The results of the radiative forcing calculations for the slow and fast updraft cirrus are in agreement with investigations from lidar observations reported by Kienast-Sjögren et al. (2016) and Campbell et al. (2016), who observed cirrus with optical depth up to 1 and 3, respectively, and found a decreasing warming effect with decreasing optical depth. Campbell et al. (2016) even reported a slight cooling effect at the warmest observed cirrus. The liquid-origin cirrus, however, mostly found in the warmest cirrus layers, have large optical depths ( $\tau$ : 1 - 12), which is larger than the range of cirrus optical depth reported in many studies (the maximum optical depth is often found to be 1-3, e.g. Sassen et al., 2008; Kienast-Sjögren et al., 2016; Campbell et al., 2016; Mitchell et al., 2018, ; note that this is likely because the lidar technique, often use to investigate cirrus cloud optical properties, has restrictions in the detection of thicker ice clouds). A consequence of the large optical thickness is a quite strong net cooling effect (- 15 to -250 W/m<sup>2</sup>) of liquid-origin cirrus. These values are of the same order of magnitude as reported from direct measurements inside of cirrus clouds (Wendisch et al., 2007; Joos, 2019).

Thus, from this first and very idealized simulations, we can conclude that in situ formed cirrus clouds are most likely warm the atmosphere, whereas liquid-origin ice clouds have the potential for strong cooling. Note here, that we only investigate local time 12 h, where the cooling is probably most pronounced. For lower sun position (i.e. larger zenith angle) the cooling is probably reduced and during night time, cirrus clouds can only warm the atmosphere (due to the thermal greenhouse effect). Thus, the net effect of cirrus clouds averaged over the whole daily cycle is not yet clear. Such investigations go beyond the scope of this study and are subject of future work.

### 3.1.2 Humidity

The distribution of in-cloud and clear sky RH<sub>ice</sub> as well as water vapor H<sub>2</sub>O with latitude and altitude is shown in the right column of Figure 3. It can nicely be seen how the amount of H<sub>2</sub>O decreases with altitude (bottom panel). The clear sky RH<sub>ice</sub> (middle panel) ranges from very dry conditions (< 70% green and orange diamonds) up to highly supersaturated regions (> 130%, dark red and blue diamonds), which mainly are found at high altitudes in the tropics. Such high supersaturations are also found inside of the tropical cirrus clouds (top panel). The behavior of RH<sub>ice</sub> will be further discussed in Sections 4 and 5.

## 4 In situ climatologies

With the term climatologies we refer here to statistical evaluations of the available variables with regard to temperature or potential temperature.

### 4.1 The IWC-T parameter space: median $N_{ice}$ , $R_{ice}$ , $RH_{ice}$

In the Cirrus Guide I (Krämer et al., 2016), observations and model simulations are portrayed in the Ice Water Content- Temperature (IWC-T) parameter space. One result of the simulations is that IWC and  $N_{ice}$  are correlated with each other. This relationship has been confirmed for some meteorological situations by examples from six individual field campaigns. The database of combined IWC- $N_{ice}$  measurements has grown considerably since then, and with it the covered temperature range of the observations. In the Cirrus Guide I, only few observations below 200 K were available.

With the extended Cirrus Guide II data set, we further investigate the appearance of  $N_{ice}$ , but also the corresponding mean mass size  $R_{ice}$  and the in-cloud  $RH_{ice}$  in the IWC-T parameter space. In Figure 6, medians of the respective parameters are presented in intervals (five iwc intervals per order of magnitude, 1K temperature intervals) covering the entire IWC-T parameter space. The variability of the parameters can be seen in Figures 1–3 of the SM, where the 25, 50 (median) and 75 % percentiles for the variables are shown.

#### 4.1.1 IWC-T- $N_{ice}$ and $R_{ice}$ relation

Observational evidence for the correlation between IWC and  $N_{ice}$  can nicely be seen in the upper panel of Figure 6. Almost symmetrical colored bands of  $N_{ice}$  can be seen across the entire IWC-T parameter space ( $N_{ice}$  concentration increases from yellow to blue). With decreasing temperature, the same  $N_{ice}$  numbers cause lower IWC values, which is caused by the likewise decreasing available water content of the air. This finding might be of importance for parametrizations used in global models or satellite retrieval algorithms, where IWC is often the only available parameter that characterizes cirrus clouds, but functions are used to assign  $N_{ice}$  to specific IWCs. This new analysis can be used (after some smoothing of the bands, which will be subject of a follow up study) as lookup table to derive  $N_{ice}$  from the information of temperature and IWC.

The ice crystal sizes also form colored bands in the IWC-T parameter space (Figure 6, middle panel;  $R_{ice}$  increases from yellow to blue, running diagonally through the IWC-T room with the size of the ice crystals increasing with increasing temperature (and thus decreasing amount of available water). This is because  $R_{ice}$  is calculated from the third root of the IWC divided by  $N_{ice}$ , as described in Section 1. However, the  $R_{ice}$  bands are somewhat less clear delineated from each other than the  $N_{ice}$  bands. They might be smoothed in the follow up study, so that  $R_{ice}$  can also be assigned to the IWC-T data points.



One could have expected to find distinct pattern for in situ-origin and liquid-origin cirrus in this type of analysis to quantify the characteristics of the cirrus types shown in Table 1. However, the differences are merged by the calculation of median values in the overlap regions of the types in the IWC-T parameter space. However, we believe that the more heterogeneous structure of the  $R_{ice}$  bands is caused by the different cirrus types, because the ice crystal size is their most pronounced difference. A longer term research goal is to derive the IWC-T- $N_{ice}$ - $R_{ice}$  relations separately for in situ-origin and liquid-origin cirrus.

#### 4.1.2 IWC-T- $RH_{ice}$ relation

In the bottom panel of Figure 6, median in-cloud  $RH_{ice}$  are shown in the IWC-T parameter space. Also for  $RH_{ice}$  pattern are visible:  $RH_{ice}$  decreases with decreasing IWC. Above the median IWC (black solid line),  $RH_{ice}$  is mostly between 90-110 % (orange data points), i.e. around saturation. This corresponds to the existence phase of the cirrus (between 110-100 / 100-90 %, the ice crystals slowly grow /evaporate). Below the median IWC, the median  $RH_{ice}$  decrease with decreasing IWC (green and yellow data points), reflecting the evaporation phase of the cirrus clouds. Interesting is the distribution of the higher supersaturations (red and blue points): at low temperatures, the clouds tend to higher supersaturations. This has been discussed by (Krämer et al., 2009) for thin cirrus (low IWC) and explained by the corresponding low  $N_{ice}$ , whose small ice surface cannot efficiently deplete the water vapor. However, the supersaturations are also seen at high IWC ( $\sim$  high  $N_{ice}$ ). The reason for that is the vertical velocity, which is -together with the ice surface and temperature- a driver of the  $RH_{ice}$  ( $RH_{ice} = H_2O/H_{2O_{sat,ice}}(T)$ ). The air inside of clouds is supersaturated in cases the depletion of  $H_2O$  on the available ice surface (decrease of  $RH_{ice}$ ) cannot compensate for the increase of  $RH_{ice}$  caused by the cooling of the air (decrease of  $H_{2O_{sat,ice}}(T)$  with decreasing temperature). High IWC, coinciding always with high  $N_{ice}$ , appear in high updrafts, in in situ-origin as well as in liquid-origin cirrus. These high updrafts allow that the supersaturations remain high despite the large ice surfaces available for deposition of excess water vapour. This was also reported by Petzold et al. (2017), who observed high  $RH_{ice}$  together with high  $N_{ice}$  in tropical convective cirrus on board of passenger aircraft (IAGOS).

## 4.2 Entire in situ climatologies

The Cirrus Guide II data set shown in Figure 3 is now displayed as frequencies of occurrence as a function of temperature (binned in 1 K intervals, Figure 7) and discussed in comparison to the earlier in situ climatologies presented by Schiller et al. (2008), Krämer et al. (2009) and Luebke et al. (2013) (Figure 6, SM). Further, as mentioned above, the new data set is large enough to split it in mid-latitude and tropical cirrus. The differing cirrus cloud properties are presented in Figures 8 and the respective clear sky and in-cloud  $RH_{ice}$  in Figures 9.

#### 425 4.2.1 Ice water content (IWC-T)

Figure 7 (top left panel) depicts the IWC. The black solid and dotted lines represent the median, minimum and maximum IWC of the core IWC band, that is the envelope of the most frequent IWC (>5% per IWC-T bin, Schiller et al., 2008).

The number of hours spent sampling in cirrus clouds increased from 27 hours in Schiller et al. (2008), 38 hours in Luebke et al. (2013) and 94 hours in (Krämer et al., 2016) to 168 hours in the new extended data set. Part of the additional data is due to the new IWC data product that is applied to all campaigns and combines the IWC from total water measurements as well as the IWC derived from cloud particle size distributions (see Appendix A2.1).

However, the median IWC and the core IWC band -decreasing with temperature as described by Schiller et al. (2008)- is still valid, showing that the IWC measurement techniques are robust and that the IWC is a stable parameter describing cirrus clouds. Note here that at temperatures  $\lesssim 200$  K data points underneath the lower dotted line are not unambiguously identified as clouds, while above about 200 K this threshold is 0.05 ppmv. For more detail see also Appendix A2.1.

#### 4.2.2 Ice crystal number ( $N_{ice}$ -T)

About 90 hours of  $N_{ice}$  observations are shown in Figure 7 (middle left panel <sup>4</sup>), which is an increase of about a factor of ten in comparison to the data set of Krämer et al. (2009), who compiled 8.5 hours (Figure 6, SM). For  $N_{ice}$ , the picture has greatly changed when comparing the old and the new data sets. This is on the one hand due to an extension of the lower detection limit of  $N_{ice}$  from  $4 \cdot 10^{-3}$  to  $10^{-4} \text{ cm}^{-3}$  (see Appendix A2.2), but also because the new data set represents a better mixture of different meteorological situations. For example, the higher  $N_{ice}$  at warmer temperatures in the Krämer et al. (2009) data set (Figure 6, SM) were caused by flights where lee wave cirrus behind the Norwegian mountains were probed (see also Figure 3, blue diamonds at around 60°North). Also, at temperatures colder than about 200 K,  $N_{ice}$  was most often very low. Further, the enhanced occurrence frequencies at the lowest concentrations seen in the earlier data set are corrected in the new data evaluation procedures (Figures 17 and Figure 6, SM, middle left panel).

90 hours of aircraft  $N_{ice}$  observations within of cirrus clouds is a tremendous amount when taking into account the necessary effort. However, this is still far from being representative for the distribution of  $N_{ice}$  in the atmosphere. We nevertheless calculated 10, 25, 50, 75 and 90% percentiles, which are shown as thin, dotted and solid lines in Figure 7 (middle left panel). Note that the 10 and 90% percentiles enclose the core region of  $N_{ice}$ , i.e. the envelope of the most frequent  $N_{ice}$  (>5% per  $N_{ice}$ -T bin). Fits through these percentiles and the median  $N_{ice}$  reveal no temperature dependence of  $N_{ice}$  (10%, median and 90%  $N_{ice}$ : 0.002, 0.03 and  $0.3 \text{ cm}^{-3}$ ). This is different to the slight decrease with

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<sup>4</sup>Note that for  $N_{ice}$  -and thus  $R_{ice}$ - the number of hours spent in clouds in Figure 7 is less than in Figure 3. For details see figure caption.

temperature of the minimum, middle and maximum  $N_{ice}$  lines shown by Krämer et al. (2009), which was, as discussed above, caused by two flights with high  $N_{ice}$  at comparatively warm temperatures.

It is a question why the  $N_{ice}$  do not increase with decreasing temperature, as would be expected because the homogeneous ice nucleation rate then increases. This has been investigated by Gryspeerdt et al. (2018) based on a 10 year global data set retrieved from satellite observations (DARDAR- $N_{ice}$ , Sourdeval et al., 2018, see also Section 6). Gryspeerdt et al. (2018) analyzed the  $N_{ice}$  ( $>5\mu m$ ) only at cloud tops and also those throughout the cirrus clouds. From the cloud top analysis, a clear increase of  $N_{ice}$  with temperature was obvious, while integrating throughout the cirrus this temperature dependence becomes much weaker, though it is still present. Gryspeerdt et al. (2018) propose that the missing temperature dependence in the in situ results could be due to a lack of in situ measurements near the cloud top, where the temperature dependence is strongest. Another reason could be that the higher  $N_{ice}$  are short-lived (see Section 3.1.1) and thus not easy to trace by aircraft.

A further consideration of  $N_{ice}$  frequencies of occurrence on a global as well as regional scales derived from satellite remote sensing will be presented in Section 6.

#### 4.2.3 Ice crystal mean mass radius ( $R_{ice}$ -T)

The ice crystal mean size is calculated as mean mass radius  $R_{ice}$  as shown in Footnote 1.  $R_{ice}$  is close to the common effective cloud particle radius  $R_{eff}$  ( $\sim$  ice Volume/Area), but can be calculated without knowing details of the ice particle size distribution (see also Krämer et al., 2016).

A total of 84 hours of observations are compiled in Figure 7 (bottom left panel). Overall, the mean mass  $R_{ice}$  ranges from about 1 to 100  $\mu m$ , while individual ice crystals in cirrus can reach sizes up to 1000  $\mu m$  or even larger. The  $R_{ice}$  core band (frequencies  $\gtrsim 5\%$  per  $R_{ice}$ -T bin) decreases slightly as the temperature decreases, which is caused by the decrease of the IWC core band, since the  $N_{ice}$  band is not dependent on temperature (see previous section).

The 10, 25, 50, 75 and 90% percentiles of the data set are plotted as thin, dotted and solid red lines. The black lines represent the minimum, middle and maximum  $R_{ice}$  shown by Krämer et al. (2009) (see Figure 6, SM). The range of  $R_{ice}$  has shifted slightly to larger sizes in the new data set, which is caused by the  $N_{ice}$  range extended towards lower concentrations that mainly consist of larger ice crystals.

Remarkable is the drop of the most frequent  $R_{ice}$  from larger to smaller ice crystals seen at around 215 K. This is probably the approximate temperature up to which liquid-origin clouds are detected (see Luebke et al., 2016; Sourdeval et al., 2018, and also Section 6.2), that are characterized by larger ice crystals than in situ-origin cirrus (Krämer et al., 2016). At higher temperatures where both liquid and in situ-origin cirrus prevail, higher IWCs - and thus also a larger  $R_{ice}$  - occur more often than at lower temperature where only the thinner in situ-origin clouds exist. This is especially true at mid-latitudes, in the tropics the liquid-origin cirrus reach also lower temperatures and thus no sudden drop of  $R_{ice}$  is observed (see Figure 8, left and right bottom panel). The bifurcated structure

of the most frequent  $R_{ice}$  which can be seen for temperatures  $\lesssim 190$  K is discussed in Section 4.3.1

495 (vii).

#### 4.2.4 Clear sky and in-cloud $RH_{ice}$ ( $RH_{ice}$ -T)

The new  $RH_{ice}$  clear sky and in-cloud data sets are displayed in the middle and top right panel of Figure 7. The respective earlier data sets of Krämer et al. (2009) are shown in Figure 6, SM. The overall picture of the  $RH_{ice}$  distributions has not changed substantially, though the amount of in-cloud  
500 data in the database has increased from 10 hours of measurements to about 96 hours, and the clear sky observation time from 16 to 186 hours. The only small difference is found in the in-cloud  $RH_{ice}$ , potentially caused by the larger amount of data: below about 200 K, where high supersaturations ( $>120\%$ ) occur more often and low subsaturations ( $<80\%$ ) less often. At higher temperatures, the peak of  $RH_{ice}$  frequencies at 100% is more pronounced.

505 For the new data set, in addition to the clear sky  $RH_{ice}$  the absolute water vapor volume mixing ratio  $H_2O$  in the bottom right panel of Figure 7 is plotted. To guide the eye, water vapor saturation wrt ice,  $H_2O_{sat,ice}$ , is drawn as black solid line. The decrease of  $H_2O$  with temperature is nicely seen in this panel and clear sky supersaturations appear in this portrayal as data points above  $H_2O_{sat,ice}$ .

### 510 4.3 Mid-latitudes and tropics

#### 4.3.1 Mid-latitude and tropical cirrus clouds

The data sets of mid-latitude/tropical cirrus consist of 67/101 hours of IWC, 29/61 hours of  $N_{ice}$  and 28/56 hours of  $R_{ice}$  measurements. This section describes some of the pronounced characteristics of mid-latitudes and tropical cirrus, which are displayed in Figure 8 (greenish/ reddish colors represent  
515 mid-latitude/tropical cirrus).

(i) *Temperature ranges*: Comparing mid-latitude and tropical cirrus, the first obvious difference -as expected when looking at the temperature profiles in Figure 2- is the temperature range. The observed mid-latitude cirrus rarely occur below 200 K, while tropical cirrus are detected down to  
520 temperatures of 182 K. The core IWC/ $N_{ice}$ / $R_{ice}$  range of both mid-latitude and tropical cirrus corresponds to the total climatology (see Figure 7).

(ii) *Mid-latitude WCBs and MCS*: At European mid-latitudes, the most frequent cirrus can be assigned to slow updrafts in frontal systems (WCBs: warm conveyor belts) containing both liquid-  
525 origin and in situ-origin cirrus. High IWCs stem mostly from liquid-origin WCB cirrus. Above the central USA, mesoscale convective systems (MCS) with faster updrafts are more frequent. The resulting liquid-origin cirrus are thicker than the European, i.e. the ice crystals are larger and the IWC is higher (see also Krämer et al., 2016).

(iii) *Contrails*: A striking feature in the cirrus observations are  $N_{ice}$  of up to several hundreds per  $cm^3$ , that are found at mid-latitudes in the temperature range of about 210-220 K, which corresponds to about 10 km altitude (see Figure 2), the typical cruising level of passenger aircraft. They can be attributed to young contrails, which were a topic of investigation during COALESC 2011 (Jones et al., 2012) and also ML-CIRRUS 2014 (Voigt et al., 2017). Higher mid-latitude  $N_{ice}$  at higher temperatures are most probably in situ-origin cirrus caused by stronger updrafts in e.g. mountain waves.

(iv) *Drop freezing*: High  $N_{ice}$  between ten and hundred per  $cm^3$  or even more are found above 235 K. Such high concentrations together with small  $R_{ice}$  are typical for supercooled liquid cloud drops that might be frozen by spontaneous homogeneous drop freezing (that occurs latest at around 235 K in the atmosphere) or not. In any case, these cloud particles are an indication for liquid-origin clouds caused in tropical or mid-latitude convective systems with fast updrafts.

(v) *Tropical convection*: In contrast, in tropical cirrus, and particularly for temperatures  $\gtrsim 220$  K, high IWCs and  $N_{ice}$  above the core range -corresponding to convective liquid-origin cirrus- become more frequent, while  $R_{ice}$  tends to be smaller. This is most likely caused by the fast convective vertical velocities that often let clouds in the mixed-phase temperature regime rise to the cirrus altitude range.

(vi) *Tropical deep convection*: Also remarkable is that in the tropics, massive convective liquid-origin cirrus carrying a high IWC -often accompanied by high  $N_{ice}$ - are detected down to very cold temperatures ( $< 200$  K), which corresponds to high altitudes up to 17 km. The thickest cirrus with very high IWC and  $N_{ice}$  can also be seen in Figure 3 (upper and middle left panel: distribution of cirrus with latitude; blue data points represent liquid-origin with a high IWC/ $N_{ice}$ ) at around  $25^\circ$  northern latitude and 16-18 km altitude. These exceptional thick and cold cirrus at high altitudes were observed in the Asian monsoon tropical tropopause layer (TTL). The observed many small  $N_{ice}$  are generated most likely by in situ homogeneous ice nucleation, triggered either by fast updrafts in gravity waves (see also Spichtinger and Krämer, 2013; Jensen et al., 2017) or deep convection. They often occur in the tops of to massive liquid-origin cirrus with very high IWC; a theoretical description of such clouds is given by Jensen and Ackerman (2006).

(vii) *TTL cirrus*: The discussion of cirrus clouds in the tropical tropopause layer is presented in Section 5.

#### 4.3.2 Mid-latitude and tropical humidity

The mid-latitude/tropical humidity data sets include 24/72 hours of in-cloud  $RH_{ice}$  and 51/132 hours of clear sky  $RH_{ice}/H_2O$  measurements, which are displayed in Figure 9 with the same color code as in Figure 8.

In clear sky at temperatures higher than about 200 K,  $RH_{ice}$  is most often below saturation and randomly distributed in both mid-latitudes and tropics (Figure 9, middle row). Clear sky supersatu-

rations occurs less frequently, simply because they only take place in those periods when moist air parcels are cooled towards the ice nucleation thresholds (heterogeneous or homogeneous), which are rare compared to drier conditions of the atmosphere.

Below about 200 K, i.e. in the TTL (see also Section 5), the clear sky  $RH_{ice}$  distribution looks very different. In this region,  $H_2O$  is low and its variability is only small (Figure 9, bottom row). We plotted lines of constant  $H_2O$  (1.5, 3 and 5 ppmv) in the clear sky  $RH_{ice}$  panels to illustrate that for constant  $H_2O$ ,  $RH_{ice}$  increases only due to the decrease in temperature, i.e.  $H_2O_{sat,ice}$ . Thus it can be seen that at mid-latitudes  $RH_{ice}$  mostly represent  $H_2O$  values around 3 ppmv and in the tropics between 1.5 and 3 ppmv. Since in the tropics much colder temperatures are reached, the respective  $RH_{ice}$  ranges from 10 up to about 150% or even more.

Clear sky  $RH_{ice}$  above the homogeneous freezing line are under discussion, because they would indicate that no supercooled liquid aerosol particles are present to initiate freezing, or that the homogeneous freezing is prevented, for example by organic material contained in the aerosol particles. More probably, these few data points are outliers; note also that the uncertainty of  $RH_{ice}$  rises from approximately 10% at warmer temperatures to about 20% at colder temperatures (Krämer et al., 2009).

Inside of cirrus, the peak of the  $RH_{ice}$  frequencies is mostly around the thermodynamical equilibrium value of 100% (saturation) at mid-latitudes as well as in the tropics (Figure 9, upper row). However, in the TTL, at the coldest prevailing temperatures ( $\lesssim 190$  K), supersaturation increasingly becomes the most common condition, which is discussed in more detail in Section 5. High supersaturations at low temperatures were also reported by (Krämer et al., 2009) and Jensen et al. (2013a), and the reason given for the existence of such high supersaturation is low  $N_{ice}$  concentrations, which were mostly present at low temperatures in these observations (Figure 6, SM). But, Jensen et al. (2013a) also showed that  $RH_{ice}$  rapidly drops to saturation in the presence of many ice crystals. As can be seen from Figure 8, middle right panel (and discussed in Section 4.2.2), in the new data set  $N_{ice}$  cover a broader concentration range in comparison to the earlier data (Figure 6, SM), while  $RH_{ice}$  is supersaturated in most cases. This is not straightforward to understand due to the complex relation between  $RH_{ice}$  and  $N_{ice}$ . We will investigate the TTL supersaturations in cirrus clouds in a follow-up study.

## 5 TTL in situ climatologies

### in- and outside of the Asian monsoon anticyclone

The tropical tropopause layer is the region above the upper level of main convective outflow, where the transition from the troposphere to stratosphere occurs. It is placed at temperatures  $<205$  K between  $\sim 150$  hPa/355 K potential temperature/14 km and 70 hPa/425 K/18.5 km (Fueglistaler et al., 2009, see also Figure 2). The coldest temperatures are found here at the point where the slope of

the temperature profile reverses (cold point tropopause, CPT). In the TTL, the prevailing dynamical conditions are very slow large-scale updrafts superimposed by a spectrum of high-frequency gravity waves (i.e. Spichtinger and Krämer, 2013; Dinh et al., 2015; Jensen et al., 2017; Podglajen et al., 2017). In addition, deep convection with fast updrafts that occasionally overshoot into the TTL.

Cirrus clouds and humidity in the TTL deserve a special consideration, because this region represents the main pathway by which water vapor enters the upper troposphere and lower stratosphere (UT/LS) where it is further distributed over long distances (e.g. Brewer, 1949; Rolf et al., 2018; Vogel et al., 2016; Ploeger et al., 2013). This is of importance, because water vapor is a greenhouse gas that has a significant impact on the climate, with greatest sensitivity in the tropical UT/LS (Solomon et al., 2010; Riese et al., 2012), but also in the LS at high latitudes where it is being transported from the tropics. Cirrus clouds are of particular relevance as regulators of the partitioning of H<sub>2</sub>O between gas and ice phase. Furthermore, they have a climate feedback themselves by influencing the Earth's radiation balance (e.g. Boucher et al., 2013). Thus, the simultaneous observation and analysis of both cirrus clouds and humidity (H<sub>2</sub>O) in this climatically crucial region are of special interest.

This section specifically reports cirrus clouds together with humidity (from the climatologies presented in Section 4), that was recently observed for the first time in the TTL in the Asian monsoon anticyclone (June to September) in comparison to observations in the surrounding tropical regions. As the Asian monsoon is characterized by strong convective activity, this comparison will show the difference to less active, calmer areas. Observations of Asian monsoon TTL cirrus clouds and humidity are of particular importance because, amongst other trace species, large amounts of H<sub>2</sub>O and also cloud particles are convectively transported upwards from far below, where the additional H<sub>2</sub>O in turn often cause cirrus formation (e.g. Ueyama et al., 2018, and references therein). Directly injected H<sub>2</sub>O or H<sub>2</sub>O from sublimated ice crystals can then be mixed up into the stratosphere. Thus, the Asian monsoon anticyclone represents a significant gateway for H<sub>2</sub>O between UT and LS (e.g. Fueglistaler et al., 2009; Ploeger et al., 2013) and currently it is under discussion to what extent cirrus cloud particles contribute to the amount of H<sub>2</sub>O entering the stratosphere (e.g. Ueyama et al., 2018, and references therein).

The airborne measurements in the Asian monsoon (see Figure 1 and Tables 4 and 5) were performed during July-August 2017 out of Khatmandu, Nepal, during a field campaign embedded in the StratoClim project (<http://www.stratoclim.org/>). An overview of the observations is given in Figures 10, 11 and 12, where the frequencies of IWC, N<sub>ice</sub>, R<sub>ice</sub> and in-cloud, clear sky RH<sub>ice</sub> as well as the clear sky H<sub>2</sub>O volume mixing ratio are shown in the temperature and also the potential temperature  $\Theta$  parameter space.<sup>5</sup> Most of the measurements during StratoClim are performed at temperatures  $\lesssim 205$  K, corresponding to potential temperatures  $\gtrsim 355$  K and altitudes about  $\gtrsim 14$  km, i.e. in the TTL (marked in the middle panels of the Figures).

<sup>5</sup> The additional  $\Theta$  portrayal provides a more detailed impression of the distribution of cirrus clouds and humidity around the CPT, i.e. at the transition between troposphere and stratosphere (see Figure 2).

The surrounding, in most (but not all) cases calmer tropical TTL regions are represented by observations during the campaigns shown in Figure 1 and listed in Tables 4 and 5. The majority of the data were sampled during ATTREX\_2014 and POSIDON\_2016. The TTL measurements in the temperature parameter space are shown in Figures 8 and 9; in these plots, the Asian monsoon observations are included, but since the measurements represent only a small part of all TTL observations, excluding the StratoClim campaign only slightly change the frequency distributions. Thus, the Figures are representative for the TTL outside of the Asian monsoon anticyclone, so we refrain from showing an additional Figure. The measurements in the  $\Theta$  parameter space (StratoClim excluded) are presented in the left panels of Figures 11 and 12.

## 5.1 TTL cirrus clouds

### 5.1.1 Temperature parameter space

In the tropics outside of the Asian monsoon (Figure 8, right column), cirrus IWC and  $N_{ice}$  range from very low to quite high values. We want to draw attention to a special feature of the most frequently occurring cirrus  $N_{ice}$  (middle right column). Two main branches of most frequent  $N_{ice}$  are found, one at very low ( $\sim 10^{-3} \text{ cm}^{-3}$ ) and the other at moderate ( $\sim 5 \cdot 10^{-2} \text{ cm}^{-3}$ )  $N_{ice}$ . These two branches are also reflected in  $R_{ice}$ , where the larger ice crystals are found together with lower concentrations and vice versa, as shown in Figure 4. More precisely, cirrus with  $N_{ice} \lesssim 1.5 \cdot 10^{-2} \text{ cm}^{-3}$  consist of ice particles larger than about  $20 \mu\text{m}$ <sup>6</sup>. That means that the ice crystal spectra of the low  $N_{ice}$  cirrus most likely represent aged clouds (in situ-origin or liquid-origin) where the smaller nucleation mode ice crystals are either grown to larger sizes in supersaturations or evaporated in case of subsaturated conditions (see also Section 3.1.1). The middle  $N_{ice}$  cirrus containing ice crystals smaller than  $20 \mu\text{m}$  are most likely young cirrus that have formed in situ, because these small crystals quickly (on a time scale of 10 - 20 minutes) grow to larger sizes. It is impossible to speculate if they have formed homogeneously, since both pathways might produce such  $N_{ice}$  in the slow updrafts prevailing in the TTL. The reason that the aged cirrus marked in Figure 8 (middle right column) become visible only in the TTL, though they certainly occur in all cirrus regions, is probably the calmer dynamic environment with -in comparison to lower altitudes- less frequently occurring temperature fluctuations. These fluctuations likely cause new ice nucleation events superimposed on the aged cirrus.

The clouds observed in the Asian monsoon include in situ formed cirrus as well as cirrus clouds from overshooting deep convection. In the much more convectively unstable Asian monsoon conditions, IWC and  $N_{ice}$  are most frequently above the median lines derived from the entire climatology (Figure 10, left column) and also high in comparison to the tropical climatology (Figure 8, right column). Here, the highest observed values (at temperatures  $< 205 \text{ K}$ ) are found with IWC mixing ratios

<sup>6</sup>It should be noted that concentrations  $\lesssim 0.1 \text{ cm}^{-3}$  of cloud particles  $\lesssim 20 \mu\text{m}$  are below the detection limit of cloud spectrometers, i.e. small ice crystals with such low concentrations could be present (see Appendix A2.2).



675 of up to 1000 ppmv and a maximum  $N_{ice}$  as high as  $30 \text{ cm}^{-3}$  (note that the ice crystal shattering was significantly minimized, see Section A2.2). Also, the ice crystals mean mass size,  $R_{ice}$ , is above the median, especially at very low temperatures, which means that large ice crystals are found around the cold point. These exceptional findings are recorded during flights in strong convection, where liquid-origin clouds from far below are detected in the upper part of the Asian monsoon anticyclone  
680 simultaneously with freshly homogeneously nucleated ice crystals. The observations were possible due to the pilot of the Geophysica aircraft, who dared to fly into the strong updrafts. Because of the dangerous nature of measurements under such conditions, the frequency of convective – and also orographic wave cirrus – is under-represented in the entire in situ climatology.

### 685 5.1.2 Potential temperature ( $\Theta$ ) parameter space

The distribution of Asian monsoon cirrus clouds in the  $\Theta$  parameter space is shown in the right panel of Figure 11. TTL average upper and lower boundaries and the CPT are marked in the middle panel of the Figure following the definition of Fueglistaler et al. (2009). In the left panel of Figure 11, the climatologies of the tropical observations without Asian monsoon are shown.

690 From Figure 11, upper row, it can be nicely seen how steeply the IWC increases during the transition from the TTL to the free troposphere. In the tropics outside of the Asian monsoon, the maximum  $\Theta$  where ice is detected is about 420 K ( $\sim 19 \text{ km}$ ). The mixing ratios of these highest cirrus are about 0.05 ppmv. At about 380 K (CPT,  $\sim 16\text{-}18 \text{ km}$ ), the range of most frequent IWCs broadens, ranging between about 0.005 and 1 ppmv. Some higher IWCs are also detected in the  
695 upper TTL, indicating that overshooting convection (liquid-origin cirrus) is also embedded in these measurements. Below about 380 K the most frequent IWC increases steadily up to values of around 10 ppmv at 355 K and 1000 ppmv at 340 K and farther below.

In the Asian monsoon, the maximum  $\Theta$  where ice is detected is about 415 K ( $\sim 0.5 \text{ ppmv}$ ) and at 400 K the IWC ranges between 0.05 - 0.1 ppmv, as outside of the Asian monsoon. But, at 380 K  
700 (CPT), the range of most frequent IWCs rises to 0.5 - 2 ppmv, then steadily increasing below about 380 K to values of around 50 - 500 ppmv at 355 K. Obviously, IWC near and below the CPT within the Asian monsoon anticyclone is enhanced by a factor of ten or more by injection of liquid-origin cirrus in overshooting events. The highest TTL IWCs (up to 1000 ppmv) are detected in the Asian monsoon up to 390 K.

705 These overshooting cirrus around and below the CPT are also seen in high ice crystal numbers  $N_{ice}$  ( $\gtrsim 0.5 \text{ cm}^{-3}$ ) when comparing the Asian monsoon with the other tropical regions (Figure 11 middle panels). Striking are again the high  $N_{ice}$  up to  $30 \text{ cm}^{-3}$  in the Asian monsoon, already discussed with respect to the temperature parameter space. Here it is visible that this burst of in situ homogeneous ice nucleation in a strong convective event (theoretically described by Jensen and Ackerman, 2006)  
710 took place below the CPT.

Above 390 K, at the transition to the stratosphere,  $N_{ice}$  is very low, mostly lower than  $0.1 \text{ cm}^{-3}$ . Recall that these low concentrations only contain particles  $> 20 \mu\text{m}$  (Footnote 6 and Appendix A2.2). This means that in the Asian monsoon (middle right panel of Figure 11) low concentrations of larger ice crystals (together with higher IWCs) are more often present at such altitudes in comparison to the surrounding tropics.

In the TTL outside of the Asian monsoon, the two branches of more frequent  $N_{ice}$  (and  $R_{ice}$ ) – discussed with respect to the temperature parameter space (Figure 8, right middle panel) – are also very clearly visible. The smaller/larger ice crystals with higher/lower concentrations were identified as young and aged cirrus clouds, which most probably have formed in situ. Farther below,  $N_{ice}$  further increases with decreasing  $\Theta$ /altitude and more and more small liquid-origin cloud particles (frozen or liquid) with higher concentrations appear.

## 5.2 TTL humidity

### 5.2.1 Temperature parameter space

Considering the in-cloud and clear sky  $RH_{ice}$  in the Asian monsoon (Figure 10, right column) in comparison to the entire tropical climatologies (Figure 9, right column), significant differences are visible. Below about 200 K, the most frequent in-cloud  $RH_{ice}$  are found in supersaturation in the Asian monsoon cirrus. In the entire climatology (Figure 9, right column) this occurs only below about 185 K. The same is seen in clear sky  $RH_{ice}$ : higher supersaturations occur frequently already at higher temperatures in the Asian monsoon comparison to the entire tropical climatology. These higher supersaturations reflect a higher amount of water vapor in the Asian monsoon TTL, visible through the dashed lines in the middle right panel of Figure 10. They show the increase of  $RH_{ice}$  caused by the decrease of  $H_2O_{sat,ice}$  during cooling of air at a constant  $H_2O$  mixing ratio. These lines correspond to  $H_2O$  between 3 and 5 ppmv in the Asian monsoon, while in the total tropical climatology the most frequent  $H_2O$  ranges only between 1.5 and 3 ppmv (Figure 9; compare also the bottom panels of the Figures, where  $H_2O$  is plotted). Due to the higher water vapor mixing ratios, supersaturation occurs already at higher temperatures. Implications of the observed supersaturations are further discussed in Section 5.3.

### 5.2.2 Potential temperature ( $\Theta$ ) parameter space

The high  $RH_{ice}$  in and outside of cirrus clouds in the Asian monsoon are also visible in the  $\Theta$  representation of the humidity (see Figure 12, top and middle right panels). In the tropical climatology outside of the Asian monsoon (top left panel of Figure 12),  $RH_{ice}$  most frequently center around saturation inside of the cirrus clouds. In the cloud free TTL outside of the Asian monsoon (middle left panel of Figure 12), a humidification of the layer between  $\sim 360$  and  $380$  K to  $RH_{ice}$  around 90% can be seen (for more detail see Schoeberl et al., 2019).

745 In the Asian monsoon, on the other hand, the in-cloud  $RH_{ice}$  in the TTL exceeds saturation (top right panel of Figure 12). Especially, between 380 and 400 K the most frequent  $RH_{ice}$  is around 130%. Also outside of clouds (middle right panel of Figure 12), around the CPT supersaturation is frequently detected and in general the humidification is higher than in the surrounding tropical regions. This is in agreement with Schoeberl et al. (2019), who reported high  $RH_{ice}$  coincident with  
750 the Himalaya Monsoon during summer and closely associated with convection. The higher amount of water vapor in the Asian monsoon in comparison to the other tropical regions can also be seen in the  $H_2O$  volume mixing ratios (bottom panels of Figure 12). In the tropics outside the Asian monsoon, the most frequent  $H_2O$  between 365-410 K is 1.5-4 ppmv, while in the Asian monsoon we found 3-8 ppmv, as also seen in the temperature parameter space. This finding is in accordance  
755 to other studies, but has been observed in situ from aircraft directly in the Asian monsoon for the first time. For example, analyzing the air mass histories of higher in situ  $H_2O$  observations at other locations (highest values of 8 ppmv), Schiller et al. (2009) found that those air masses has passed the Asian monsoon region. Also, Ueyama et al. (2018) (and references therein) reported 5-7 ppmv at 100 hPa from MLS observations and extensive model simulations over the Asian summer monsoon  
760 region.

### 5.3 $H_2O$ and IWC for transport to the stratosphere

The  $H_2O$  transport to the stratosphere is regulated by the coldest temperature an air parcel experiences during transition through the tropopause region. The water amount passing the tropopause is set by the freeze-drying process associated with this transition (Jensen and Pfister, 2004) and is  
765 discussed to be as low as  $H_2O$  saturation at the minimum temperature (e.g. Schiller et al., 2009).

However, Rollins et al. (2016) showed for the ATTREX 2014 observations, that the water vapor at the stratospheric entry point is higher by  $\sim 10\%$ , because the water vapor depletion by ice crystals becomes increasingly inefficient at temperatures below 200 K (note that this is a rough estimate of the excess water vapor at the stratospheric entry point based on the actual temperature, which could  
770 be somewhat different from the minimum temperature of the air parcel's back trajectory). This is of importance, since already small amounts of  $H_2O$  can influence the stratospheric radiation budget (Solomon et al., 2010; Riese et al., 2012).

High supersaturations at the coldest points of the TTL are also discussed in Section 5.2.1. The ATTREX 2014 and also POSIDON 2016 observations are included in Figure 9, right panels, where  
775 the most frequent in-cloud and clear sky  $RH_{ice}$  at these temperatures can be seen to increase with decreasing temperature as discussed by Rollins et al. (2016). But, in the Asian Monsoon measurements (Figure 10, right panels),  $RH_{ice}$  at the coldest temperatures is even higher above saturation than in the calmer tropics outside, the saturation is exceeded by about 20-50% below 195 K.

Therefore, we extend the conclusion of Rollins et al. (2016): taking saturation at the stratospheric  
780 entry of an air mass as set point for water vapor transport to the stratosphere, the transport is underes-

timated by  $\sim 10\%$  in regions of weak convective activity. In convective regions the underestimation increased to 20-50% in our observations.

As mentioned earlier, the question of how much  $H_2O$  from convectively injected ice crystals is transported from above the CPT further into the lower stratosphere is a subject of recent research. In the study of Ueyama et al. (2018), it is concluded that over the Asian monsoon at 100 hPa, convection is the dominant driver of the localized  $H_2O$  and that nearly all of the convective enhancements in  $H_2O$  is due to the effect of convective humidification, while convectively detrained ice crystals have negligible impact.

From our measurements in and above the Asian monsoon anticyclone, however, it is difficult to estimate to what extent cirrus cloud particles contribute to the amount of  $H_2O$  that might enter the stratosphere. Nevertheless, from Section 5.1 we know that the most frequent IWCs between about 365-400 K are 10 to 0.5 ppmv. Above 400 K, up to 415 K some overshoots with IWCs between 0.5 to 8 ppmv are detected (Figure 11, upper right panel). These IWCs are similar to the amount of gas phase  $H_2O$ , which is between 3 to 8 ppmv in this region above the CPT (see last Section). Such amounts of  $H_2O$  and IWC indicate that the air ‘masses originate’ from an altitude of about 15 km (Figure 3), which corresponds to the lower bound of the TTL (see Figure 2).

The question is if the ice particles found in the upper TTL will further grow and sediment out in supersaturated conditions (dehydration) or evaporate and add  $H_2O$  to the gas phase  $H_2O$  in subsaturation (hydration, see Jensen et al., 2007; Schoeberl et al., 2018). Inspecting the in-cloud  $RH_{ice}$  (see Figure 12, upper right panel) related to the respective IWCs, it can be seen that the air in the overshoots  $\gtrsim 400$  K with the fairly high IWCs (0.5 to 8 ppmv) is subsaturated, while at lower altitudes both super- and subsaturation occur. This means that convective overshoots in the Asian monsoon can locally contribute a significant amount of water that might be further transported into the stratosphere. Unfortunately, we can not say anything about the frequency of these events in this study.

Above the CPT in the surrounding tropics outside of the Asian monsoon, the in-cloud  $RH_{ice}$  at  $\gtrsim 400$  K are also subsaturated. Here, the IWCs range only between 0.02 and 2 ppmv, with some single data points up to about 2 ppmv. The related  $H_2O$  is 1.5 to 3 ppmv.

Comparing gas phase  $H_2O$  and IWC above the Asian monsoon CPT (both 3-8 ppmv) with the surrounding tropical regions (1.5-3/0.02-2 ppmv) might be an indication by in situ observations that, driven by overshooting convection, the Asian monsoon could be an important source for transport of  $H_2O$  to the stratosphere.

## 815 6 Global cirrus $N_{ice}$ climatology from satellite remote sensing

Though the new in situ cirrus climatologies presented in Section 4 represent a considerable data set from research aircraft from which substantial insights are gained, it is still a mixture of different meteorological situations encountered during various field campaigns that does not necessarily display a statistically representative overall picture of the distribution of cirrus cloud properties. For this purpose, long-term global satellite remote sensing observations would be the method of choice, though retrievals of cirrus microphysical properties also have their own difficulties and limitations. This is especially true for the ice crystal number  $N_{ice}$ , which is particularly challenging to estimate from satellite remote sensing. The new retrieval method for  $N_{ice}$  introduced in Section 2.2, DARDAR-Nice, has however demonstrated the ability to satisfactorily reproduce in situ ice concentrations (Sourdeval et al., 2018).

In this section, we show a global climatology of  $N_{ice}$  derived from DARDAR-Nice in the same presentation as the in situ  $N_{ice}$  climatology. Median  $N_{ice}$ , 25/75<sup>th</sup> and 10/90<sup>th</sup> percentiles are provided for a global data set. In addition, median  $N_{ice}$  are discussed for the Arctic region, the northern mid-latitudes, the tropics, the southern mid-latitudes and Antarctica.

### 6.1 Comparison of remote sensing and in situ $N_{ice}$

As a base for reliable  $N_{ice}$  climatologies from satellite remote sensing, in situ measurements of PSDs (ice particle size distributions) from five campaigns of the Cirrus Guide II are used for an evaluation of the DARDAR-Nice retrieval algorithm (see Section 2.2). This directly follows the evaluation of DARDAR-Nice presented in Sourdeval et al. (2018), a subset of campaigns from the Cirrus Guide II data set was used. This section therefore focuses on these five campaigns, which are nevertheless representative of a wide range of mid-latitude and tropical ice clouds (see Figure 1 and caption of Figure 13). Figure 13 (top panel) shows the  $N_{ice}$  frequencies of occurrence in 1K temperature bins of the in situ measurements, the corresponding frequencies obtained from DARDAR-Nice are shown in the bottom panel. The latter are derived on the basis of two input parameters which are extracted from the in situ PSDs of the five campaigns listed in the figure caption, rather than being constrained from lidar-radar measurements during usual retrievals. This approach allows to identify inherent incompatibilities between the satellite retrieval assumptions and the in situ measurements, by assuming that the in situ PSD parameters are perfectly constrained by the lidar-radar. Therefore, possible differences should only be attributed to other retrieval assumptions, such as the PSD shape. Sourdeval et al. (2018) showed that this approach is efficient for identifying algorithmic limitations while still being representative of actual satellite retrievals. Note that DARDAR-Nice provides concentrations of ice crystals  $> 5\mu m$ , as it has not been evaluated for smaller sizes, while  $N_{ice}$  from in situ observations includes sizes  $> 3\mu m$ ; however, the resulting difference in the ice concentrations is

negligible. Also, the 1-Hz in situ PSDs are here sampled into 10-s averages to simulate the 1.7-km horizontal resolution of the DARDAR-Nice retrievals (assuming a flight speed of about  $170 \text{ m s}^{-1}$ ).

The black solid and dotted lines of the in situ climatology indicate the 50, 25 and 75<sup>th</sup> percentiles, which agree well with those of the entire in situ climatology (see Figure 7, middle left panel). This demonstrates that the selected subset is statistically representative of the entire database (indeed, the entire  $N_{\text{ice}}$  climatology contains only one additional campaign, see Table 5). A satisfactory agreement between DARDAR-Nice and the in situ observations is also seen, demonstrating that DARDAR-Nice  $N_{\text{ice}}$  retrievals very well match the in situ database for known size distributions.

A detailed comparison of the DARDAR-Nice percentiles (black solid and dashed lines) with those of the in situ observations, however, yields a small offset by a factor of 1.73, which is visualized in Figure 13 by the average medians of DARDAR-Nice (straight black line,  $0.064 \text{ cm}^{-3}$ ) and in situ (straight blue line,  $0.037 \text{ cm}^{-3}$ ). One reason for this offset lies in the method of the retrieval. The two parameters,  $N_0^*$  and  $D_m$ , calculated here from the in situ PSDs (usually from lidar-radar observations) are feeded into a predefined four-parameter gamma-modified function to calculate ice particle size distributions (PSDs, see Section 2.2).  $N_{\text{ice}}$  is then calculated by summing up individual ice concentrations over a grid of size bins distributed over the gamma-shaped PSD for the entire range of observed ice crystal sizes (usually, a continuous integration of the PSD is performed). However, in the in situ measurements, there are often PSDs that do not contain ice particles  $< 20 \mu\text{m}$ , i.e. these size bins are empty. Such PSDs represent aged cirrus after the ice nucleation phase where the smaller ice crystals have grown to larger sizes (see Section 4.3.1). DARDAR-Nice, however, assumes a modified Gamma distribution including all size bins, which partly explains the described positive offset. This behavior was also found by Wolf et al. (2019) who parameterized in situ-origin and liquid-origin cirrus from balloon-borne measurements by Gamma functions. It is of note that when subtracting the ice concentrations of the 'empty bins' of in situ PSDs from the retrieved DARDAR-Nice in a direct intercomparison, the agreement of  $N_{\text{ice}}$  is improving (not shown here). This is an important finding, as gamma functions are often used to represent cirrus PSDs, both in remote sensing retrieval algorithms and in global models. On the other hand it has to be noted that an offset on the order of a factor of 1.73 is tolerable given the variability of  $N_{\text{ice}}$  (six orders of magnitude) and other possible error sources in the measurements, both in situ and remote sensing.

Other assumptions on the PSD shape by the satellite remote sensing method might also contribute to this bias. The PSD shape indeed is provided by 4 parameters, 2 of which are fixed and 2 are retrieved (see Section 2.2). Delanoë et al. (2014) showed that the two fixed PSD parameters defined by Delanoë et al. (2005) and used in DARDAR-Nice might lead to a too steep representation of the small ice mode (i.e. too high  $N_{\text{ice}}$ ) and should be updated in future algorithm versions. Also, the bi-modality of the PSD towards temperature where growth processes become important is not accounted for and usually leads to small positive  $N_{\text{ice}}$  biases (Sourdeval et al., 2018). The cause of these assumptions are difficult to account for, as they most likely depend on the cloud-type and on the

thermodynamical environment, but they should on a first order be reasonably captured by the 1.73 adjustment factor. Higher  $N_{ice}$  in DARDAR-Nice towards low temperatures are also not surprising, as Sourdeval et al. (2018) reported that the PSD parametrization used by DARDAR-Nice predicts higher concentrations of small particles ( $D_{ice} < 25 \mu m$ ) than the in situ comparative measurements. The overestimation increases with decreasing temperature, as the small ice particles dominate the PSDs more and more. This might be caused by a sharper representation of small ice concentrations in the PSD parametrization in comparison to the in situ small ice particle measurements below  $\sim 210$  K.

A further expected difference between the data sets arises from their detection limits. The lowest  $N_{ice}$  that can be detected by the in situ instruments is  $10^{-4} cm^{-3}$ , for the highest  $N_{ice}$  there is no limitation. Detection limits for DARDAR-Nice depend on lidar-radar sensitivity but are also influenced by the instrumental resolution, which may cause specific features to remain undetected. This effect, however, is represented in Fig. 13 by the 10-s sampling of 1-Hz in situ PSDs. Indeed, DARDAR-Nice misses the high  $N_{ice}$  in the temperature range 210-220 K, but these are young, line shaped contrails which are too small scale to be detected from satellite. Also, part of the thinnest cirrus are not represented by DARDAR-Nice. This will be further discussed in the next section describing the global  $N_{ice}$  distribution from DARDAR-Nice.

Overall, following the above-mentioned arguments, an overestimation within a factor of 2 in DARDAR-Nice by comparison to the in situ data set is expected, although there is still some uncertainty as to how this overestimation will propagate globally and over various ice cloud regimes. The ratio of 1.73 in  $N_{ice}$  between DARDAR-Nice and the in situ data set, found from Figure 13, should therefore be thought of as a minimum expected bias. Finally, the bias is expected to be stronger at low temperatures than at high temperatures, due to the increased likelihood of missing bins as well as the higher importance of the representation of the small ice particle mode in the PSDs. A correction that depends on temperature, and possibly IWC, might therefore be optimal but a simpler first-order correction of 1.73 for all T and IWC range should here be sufficient for the needs of this study and considering the multitude of processes that can lead to this bias. Future studies will be required to precisely understand such inherent differences between satellite and in situ dataset.

## 6.2 Global cirrus $N_{ice}$

The global frequency distribution of  $N_{ice}$  from ten years of satellite observations is shown in Figure 14. The data was collected twice a day, approximately at midday and at midnight (satellite equator-crossing time is 1.30 am/pm), from June 2006 to December 2016. Overall, the global satellite data set consists of nearly  $2 \times 10^{10}$   $N_{ice}$  retrievals. The color code represent frequencies of occurrence, the black contours the 25, 50 and 75<sup>th</sup> percentiles.

From the median (solid black line), a slight increase of  $N_{ice}$  with decreasing temperature is visible, which is somewhat different to the in situ median  $N_{ice}$  (solid blue line, from Figure 7, middle left

panel), where no temperature dependence is found. The DARDAR-Nice temperature dependence was already noted and discussed by Gryspeerdt et al. (2018). Another difference is that at  $T \gtrsim 210$  K, the maximum  $N_{\text{ice}}$  of DARDAR-Nice reaches up to  $100 \text{ cm}^{-3}$ , while the maximum in situ  $N_{\text{ice}}$  is only about  $10 \text{ cm}^{-3}$  (outside of young contrails).

We attribute the slightly increasing median  $N_{\text{ice}}$  with decreasing temperature to homogeneous ice nucleation events, because homogeneous ice nucleation rates increase with decreasing temperature, but their appearance in space and time is transient, as discussed in Section 4.2.2. Thus, such events are difficult to find by research aircraft. Hence, these events are likely underrepresented in the aircraft observations.

The thinnest cirrus with low  $N_{\text{ice}}$  are represented by DARDAR-Nice for temperatures  $\gtrsim 190$  K. At  $\lesssim 190$  K, however, a decreasing detectability of thin cirrus becomes apparent in the DARDAR-Nice climatology: in contrast to the in situ climatology, lesser or no thin cirrus are measured with decreasing temperature. The reason is that the colder the cirrus clouds, the smaller are the ice crystals and the lesser their IWC (Figure 10, top and bottom left panel). In the in situ  $N_{\text{ice}}$  climatology, in the TTL ( $\lesssim 205$  K), an increased occurrence of very thin cirrus with  $N_{\text{ice}}$  around  $0.001 \text{ cm}^{-3}$  is reported in Section 4.3.1 and explained as aged cirrus consisting of only larger ice crystals. Though this type of clouds is partly missing in the DARDAR-Nice climatology, the median  $N_{\text{ice}}$  decreases again at these temperatures. This might reflect the frequent presence of aged thin cirrus. Note, however, that the statistics at these temperatures is based on a lower number of observations than at higher temperatures (see Figure 15).

The median  $N_{\text{ice}}$  across all temperatures of the 10 years DARDAR-Nice climatology is about  $0.100 \text{ cm}^{-3}$  (Figure 14, plain cyan line). Adjusting this number to the offset factor of 1.73 between DARDAR-Nice and the in situ observations reported in the previous section yields an in situ average median of  $0.056 \text{ cm}^{-3}$  (dashed cyan line, see also Table 3). It is nevertheless important to note that such median is by construction computed assuming that all temperature bins have equal weight. When considering their actual relative occurrence as function of temperature (see Figure 15 and later discussion), the in situ adjusted global DARDAR-Nice median falls down to  $0.046 \text{ cm}^{-3}$ . This is still larger than the average median of  $0.03 \text{ cm}^{-3}$  reported from the  $N_{\text{ice}}$  in situ climatology (blue line; from Figure 7). Nevertheless, in the light of the offset between the data sets, the decreasing detectability of thin cirrus by DARDAR-Nice and other sources of error in both methods, the agreement between the DARDAR-Nice and the in situ  $N_{\text{ice}}$  climatology is good.

Altogether, both data sets have advantages and disadvantages: DARDAR-Nice has the advantage of the long, global time series where all atmospheric situations above the detection limit showing up at the times of observations are sampled. However, the thinnest cirrus at cold temperatures are not detected. Further, an offset is found between DARDAR-Nice and the in situ observations. On the other hand, the in situ  $N_{\text{ice}}$  climatology is based on a much smaller data set which is randomly



distributed between different atmospheric situations. But, nearly the complete range of possible  $N_{ice}$

960 can be detected by the in situ instruments.

For comparisons with other  $N_{ice}$  data sets or results from global models, the percentiles of the adjusted DARDAR-Nice as well as the average in situ  $N_{ice}$  are shown in Figure 14 and listed in Table ?? (note that an in situ data set slightly smaller than the data set presented here, but extended with regard to Krämer et al. (2009) is already used for this purpose by Bacer et al., 2018; Penner  
965 et al., 2018; Righi et al., 2019).

The DARDAR-Nice data set does not only provide the  $N_{ice}$  distribution with temperature as shown in Figure 14, but, as it contains the entire spatial and temporal  $N_{ice}$  information, also the actual appearance of cirrus clouds in the  $N_{ice}$ -T parameter space. This is shown in Figure 15, where the  
970 occurrences of DARDAR-Nice  $N_{ice}$  retrievals are indicated by the colored background. It can be noticed that, in regions of high frequency of occurrence (for  $T \gtrsim 210$  K), the agreement between DARDAR-Nice and the in situ data set is best.

From this portrayal it can be seen that the cirrus clouds do not spread evenly across all temperatures, but that about 50% of the cirrus appear at temperatures above about 225 K (see also Table 3,  
975 temperature range of global 'most frequent' median  $N_{ice}$ ). This clustering of cirrus at warmer temperatures is likely because in situ-origin and liquid-origin cirrus clouds are both found in this warmest and lowest cirrus layer, while at colder temperatures and higher altitudes only in situ-origin cirrus are present (see also Section 3.1 and Figure 3). This vertical structure of the cloud types is also reported from observations by Krämer et al. (2016); Luebke et al. (2016); Wolf et al. (2018), from global  
980 modeling by Gasparini and Lohmann (2016), from satellite and global modeling by Gasparini et al. (2018) and also from 12 years ERA-interim data analysis in the North Atlantic region by Wernli et al. (2016).

Gasparini et al. (2018) provide global relative frequencies of liquid-origin cirrus with respect to temperature: more than 50% of the cirrus clouds in the range between 235 K and 220 K are of liquid-  
985 origin, decreasing rapidly to about 20% between 200 K and 180 K. These findings from satellite observations and global modeling are reasonably consistent with those from trajectory analysis by Wernli et al. (2016), who reported the relative frequencies of liquid-origin cirrus with respect to pressure to decrease from about 55% of the cirrus at 500 hPa (roughly 240 K), to 50, 30, 5% at 400, 300, 200 hPa (roughly 230, 215, 200 K; see Figure 2).

990 The analysis presented here provides the information that globally, half of the cirrus are in the lowest, warmest cirrus layer between about 220 K and 240 K, where, as mentioned above, about half of the cirrus are of liquid-origin. This is of importance because of the differing radiative properties of in situ-origin and liquid-origin cirrus shown in Section 3.1.1 (see Figure 5): if the thick liquid-origin cirrus clouds occur with such a high frequency, their strong cooling effect might exceed the much  
995 smaller warming effect of the in situ-origin cirrus which could lead to a general cooling by cirrus.

Note that an analysis of  $N_{ice}$  in the IWC-T space, similarly to Figure 6, is shown in Figure 5 of the SM. This figure shows a good climatological agreement between the satellite product and the Cirrus Guide II data set, with very similar distribution of  $N_{ice}$  in the IWC-T space. Differences could be attributed to lack of statistics in Figure 6 (noisy patterns). However, one notable difference is the slope of the IWC-T relation, which appears much flatter in the satellite product than in the in situ data, as indicated by the density isolines.

### 6.3 Regional cirrus $N_{ice}$

Lastly, we provide an impression of the regional variations of  $N_{ice}$  (see Table 3). To this end, subsets of the DARDAR-Nice data for five regional latitude bands between  $90^\circ$ ,  $67.7^\circ$ ,  $23.3^\circ$ ,  $-23.3^\circ$ ,  $-67.7^\circ$ ,  $-90^\circ$ , corresponding to the Arctic, Mid-lat North, the Tropics, Mid-lat South and Antarctica, are considered, and three different  $N_{ice}$  medians are computed for each region and for the global data set. A first median (labeled “all Temp.”) corresponds to the median  $N_{ice}$  value across all temperatures and considering all temperature bins to be equiprobable, i.e. the same as used in Figure 14. A second (labeled “T-int weighted”) represents the median using the relative occurrence of each temperature interval as a weight. The last (“most frequent”) shows the most frequently appearing  $N_{ice}$ , considering those temperature intervals that contain 50% of the total  $N_{ice}$  occurrence; T-range denotes the respective minimum and maximum temperatures (note that in the in situ data set all three methods would yield to an identical median due to the flat temperature dependence).

Comparing the three median  $N_{ice}$ , it is obvious that in most cases the ‘T-int weighted’ and ‘most frequent’ medians are smaller than those for ‘all temperatures’. This behavior is found because the cirrus clouds appear often at warmer temperatures (see previous subsection) where  $N_{ice}$  is moderate, which can be seen particularly in the ‘most frequent’ medians, where the temperature range of the cirrus occurrence is given.

The global ‘most frequent’ median  $N_{ice}$  is  $0.031 \text{ cm}^{-3}$  (in comparison to  $0.056$  and  $0.046 \text{ cm}^{-3}$  for ‘all temperatures’ and ‘T-int weighted’) in the temperature range between 224-242 K (note that, to avoid representing mixed-phase clouds in the DARDAR-Nice analysis, 242 K is chosen as maximum temperature). The lowest  $N_{ice}$  are found in the Arctic (‘most frequent’ median  $0.016 \text{ cm}^{-3}$  between 230-242 K), which is most probably because the updrafts in the Arctic regions are generally lower than in other regions. In Antarctica, the ‘most frequent’ median is higher ( $0.029 \text{ cm}^{-3}$  between 224 and 242 K). This could be traced back to stronger orographic uplifts in austral winter. The boreal and austral mid-latitude cirrus clouds are similar to the global, with ‘most frequent’ medians of  $0.030$  and  $0.031 \text{ cm}^{-3}$  in the temperature range of 227-242 K. This points to moderate updrafts on average, though regionally orography and convection can cause higher updrafts and high  $N_{ice}$ , which is also shown by Sourdeval et al. (2018).

Tropical cirrus clouds are different from those in the other regions. The ‘most frequent’ cirrus have the highest median ( $0.074 \text{ cm}^{-3}$ ) and appear at much colder temperatures (197-221 K), i.e. higher

altitudes. This finding corresponds to the in situ observations presented in Section 3.1 (see Figure 3) and also the DARDAR-Nice analyses of the vertical distribution of  $N_{\text{ice}}$  shown by Sourdeval et al. (2018). The reason is the strong convection prevailing in tropical regions, lifting liquid-origin clouds up to high altitudes and cold temperatures while homogeneously nucleating a large number of in situ ice crystals in addition (see also Section 5).

For all regions, except the tropics, more than half of the cirrus clouds are found at temperatures warmer than about 225 K and contain a considerable amount of liquid-origin cirrus, as discussed in the previous subsection. The same is true in the tropics, but for colder temperatures.

## 7 Summary and Conclusions

The Cirrus Guide II aims to represent cirrus clouds and their environment from the perspective of in situ and satellite remote sensing observations. To this end, an in situ database is created, which is based on measurements with state-of the art instrumentation and extended in comparison to earlier studies (Schiller et al., 2008; Krämer et al., 2009; Luebke et al., 2013; Krämer et al., 2016). The in situ database consists now of about 168 hours of ice water content IWC, 90/84 hours of ice crystal number concentration  $N_{ice}$  and  $R_{ice}$ , 96/186 hours of in-cloud and clear sky  $RH_{ice}$  and, correspondingly, 186 hours of clear sky water vapor  $H_2O$ . The measurements span the altitude range between 5 and 20 km and cover the latitude band from 75 °North to 20 °South. The remote sensing database includes 10 years (2006-2016) of global recording of ice crystal number concentrations  $N_{ice}$ . The main results from the evaluation of the two data sets are summarized in the following.

### 7.1 Characteristics and distribution of in situ-origin and liquid-origin cirrus (Section 3)

From the extended Cirrus Guide II in situ data set, we generally *confirm the typical characteristics of in situ-origin and liquid-origin cirrus* introduced by Krämer et al. (2016) with some additions (see Table 1):

- in situ-origin as well as liquid-origin cirrus consist of two sub-classes determined by the updraft (1:slow updrafts - few large ice crystals form from heterogeneous freezing; 2:fast updrafts - many small ice crystals nucleate homogeneously).
- New in this concept is that also liquid-origin cirrus differ in the two updraft regimes. Often, the two subclasses occur in succession and form a bimodal size distribution.
- liquid-origin cirrus are thicker (higher IWC) than in situ-origin cirrus and are usually characterized by larger ice crystals.
- The differences between the cirrus types are most pronounced in the formation phase of the clouds and are blurred with increasing lifetime due to ice crystal growth and sedimentation or additional ice formation.

In addition, we present a *picture of the distribution of cirrus with respect to altitude and latitude*, including an impression of the vertical structure of liquid-origin and in situ-origin cirrus (Figure 3):

- Across all latitudes, the thicker liquid-origin cirrus predominate at lower altitudes, while at higher altitudes the thinner in situ-origin cirrus prevail. In between, the two cirrus types overlap. This finding is in agreement with Luebke et al. (2016) and Wernli et al. (2016) for mid-latitude and Wolf et al. (2018) for Arctic cirrus.

Finally, a *first estimate of the radiative characteristics of typical, specific and idealized in situ-origin and liquid-origin cirrus scenarios* is given (Figure 5):

- slow in situ-origin cirrus have a small optical depth ( $\tau$ : 0.001 - 0.05), resulting in a slight net warming effect of not larger than about 1.5 W/m<sup>2</sup>.
- the optical depth of fast in situ-origin cirrus is larger ( $\tau$ : 0.05 - 1), but most of them are also warming (2-10 W/m<sup>2</sup>). The thickest fast in situ-origin cirrus at the lowest altitudes can change the sign of their net forcing, they switch to a slight cooling effect.
- liquid-origin cirrus have large optical depths ( $\tau$ : 1 - 12), and consequently exhibit a quite strong net cooling effect (-15 to -250 W/m<sup>2</sup>).

## 7.2 Cirrus and humidity in the tropical tropopause layer (TTL) (Section 5)

The new in situ data set is extended by observations in the tropical TTL outside, but also, for the first time, inside of the Asian monsoon anticyclone. Therefore, we put special emphasis on the analysis of the TTL environment.

*TTL cirrus clouds* (Figure 11):

- Two types of most likely in situ formed cirrus are identified in slow large scale updrafts at low temperatures ( $T \lesssim 205$  K,  $\Theta \gtrsim 355$  K). The first is interpreted as young cirrus ( $N_{\text{ice}}$  around 0.1-1 cm<sup>-3</sup>  $\gtrsim 3$   $\mu\text{m}$  diameter), the second as aged cirrus ( $N_{\text{ice}}$  around 0.001 cm<sup>-3</sup>  $\gtrsim 20$   $\mu\text{m}$  diameter) where the smaller ice crystals have grown to larger sizes.
- The highest  $N_{\text{ice}}$  (up to 30 cm<sup>-3</sup> around the cold point tropopause, CPT) and IWC (up to 1000 ppmv around the CPT) are found in deep convective systems in the Asian monsoon. Such systems represent massive liquid-origin cirrus (very high IWC) superimposed by a fresh strong homogeneous in situ ice nucleation event (many small ice crystals  $\lesssim 20$   $\mu\text{m}$ ) in fast updrafts.

*TTL humidity* (Figure 12):

- In the Asian monsoon, in-cloud and clear sky  $\text{RH}_{\text{ice}}$  is higher (often supersaturated around and above the CPT) than in the surrounding tropics. This is caused by a higher amount of H<sub>2</sub>O in the Asian monsoon (most frequently 3 to 5 ppmv) in comparison to the tropics outside (1.5 to 3 ppmv).
- Taking saturation at the stratospheric entry of an air mass as set point for water vapor transport to the stratosphere, the transport is underestimated by  $\sim 10\%$  in regions of weak convective activity (see also Rollins et al., 2016). In convective regions, the underestimation increased to 20-50% in our observations in the Asian monsoon.
- Convectively injected ice over the Asian monsoon CPT ( $\Theta \gtrsim 400$  K,  $\text{RH}_{\text{ice}} \lesssim 100\%$ ) can locally contribute a significant amount of water (up to an IWC of 8 ppmv, in comparison to only 2 ppmv in the surrounding tropics) that might be evaporated and further transported into the stratosphere.

### 7.3 in situ and satellite climatologies (Sections 4 and 6)

1115 *Cirrus Guide II in situ cirrus and humidity climatologies:*

- Median  $N_{ice}$ ,  $R_{ice}$  and  $RH_{ice}$  are presented in the IWC-T parameter space (Figure 6).

Distinct patterns are found:  $N_{ice}$  form bands parallel to IWC, while  $R_{ice}$  are sorted diagonally in the IWC-T space. At tropical cold temperatures, supersaturations occur in thin as well as in thick cirrus.

- 1120
- The entire extended data set (Figure 7) is compared to the earlier studies (Figure 6, SM).

IWC: the median IWC and the core IWC band is the same in both data sets, showing that the in situ IWC measurement techniques are robust and that the IWC is a stable parameter describing cirrus clouds.

- 1125
- $RH_{ice}$ : the overall picture of the in-cloud and clear sky  $RH_{ice}$  distributions has also not changed, demonstrating that high altitude water vapor measurements that were under discussion earlier have improved and stabilized.

- 1130
- $N_{ice}$ : an extended view is presented for  $N_{ice}$ , which is due to the better lower  $N_{ice}$  detection limit and a better mixture of dynamical situation. The new  $N_{ice}$ -T percentiles are lower and show no distinct temperature dependence (average 10%, median and 90%  $N_{ice}$  percentiles: 0.002, 0.03 and  $0.3 \text{ cm}^{-3}$ ) in comparison to the earlier observations, that show a slight decrease of  $N_{ice}$  with temperature and an average median  $N_{ice}$  of about  $0.1 \text{ cm}^{-3}$ .

- The in situ data set is subdivided into mid-latitude and tropical climatologies (Figures 8 and 9) and typical cirrus and humidity characteristics of the respective regions are presented.

1135 *DARDAR-Nice satellite global cirrus  $N_{ice}$  climatology*  
(Figures 14 and 15):

- A global climatology of  $N_{ice}$  from 10 years (2006-2016) of satellite observations is provided that can be used for comparison with global models or other data sets.

- 1140
- The  $N_{ice}$  from satellite observations are validated by and adjusted to in situ measurements from a subset of five campaigns of the Cirrus Guide II.

- The global median  $N_{ice}$  from satellite observations is almost 2 times higher than the in situ median and increases slightly with decreasing temperature.

- $N_{ice}$  medians sorted by geographical regions are highest in the tropics, followed by austral/boreal mid-latitudes, Antarctica and the Arctic.

- 1145
- In the satellite climatologies of  $N_{ice}$ , half of the cirrus are located in the lowest, warmest cirrus layer and contain a significant amount of liquid-origin cirrus. Their global median  $N_{ice}$  is  $0.031 \text{ cm}^{-3}$ .

- Regarding the frequent appearance of liquid-origin cirrus together with strong cooling effect is a motivation to investigate their influence on the overall cirrus radiative feedback on climate in future studies.

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### **Acknowledgments**

This paper is dedicated to our colleague Cornelius Schiller, who passed away much too early in 2012. He was the initiator of the StratoClim project, including the tropical aircraft campaign. Through this campaign, he intended to complement his work on hydration and dehydration in the tropical tropopause layer, based on observations over Brazil, Australia and Africa (Schiller et al., 2009) with measurements over Asia. The campaign took place successfully out of Kathmandu, Nepal, in Summer 2017, after several years of planning and many hurdles that had to be overcome.

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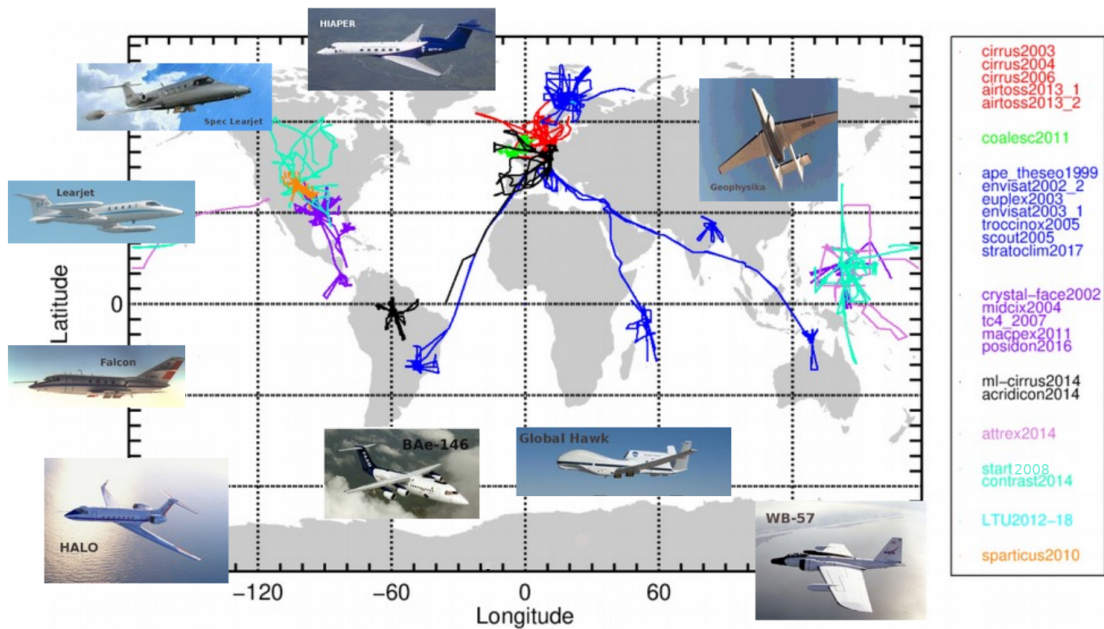
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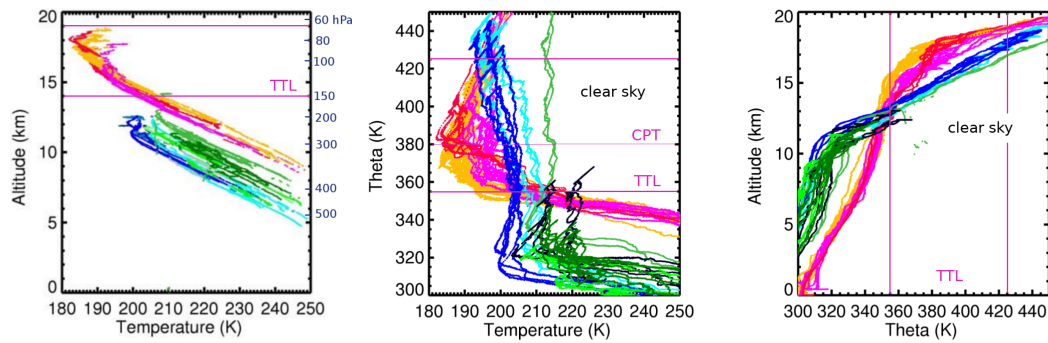
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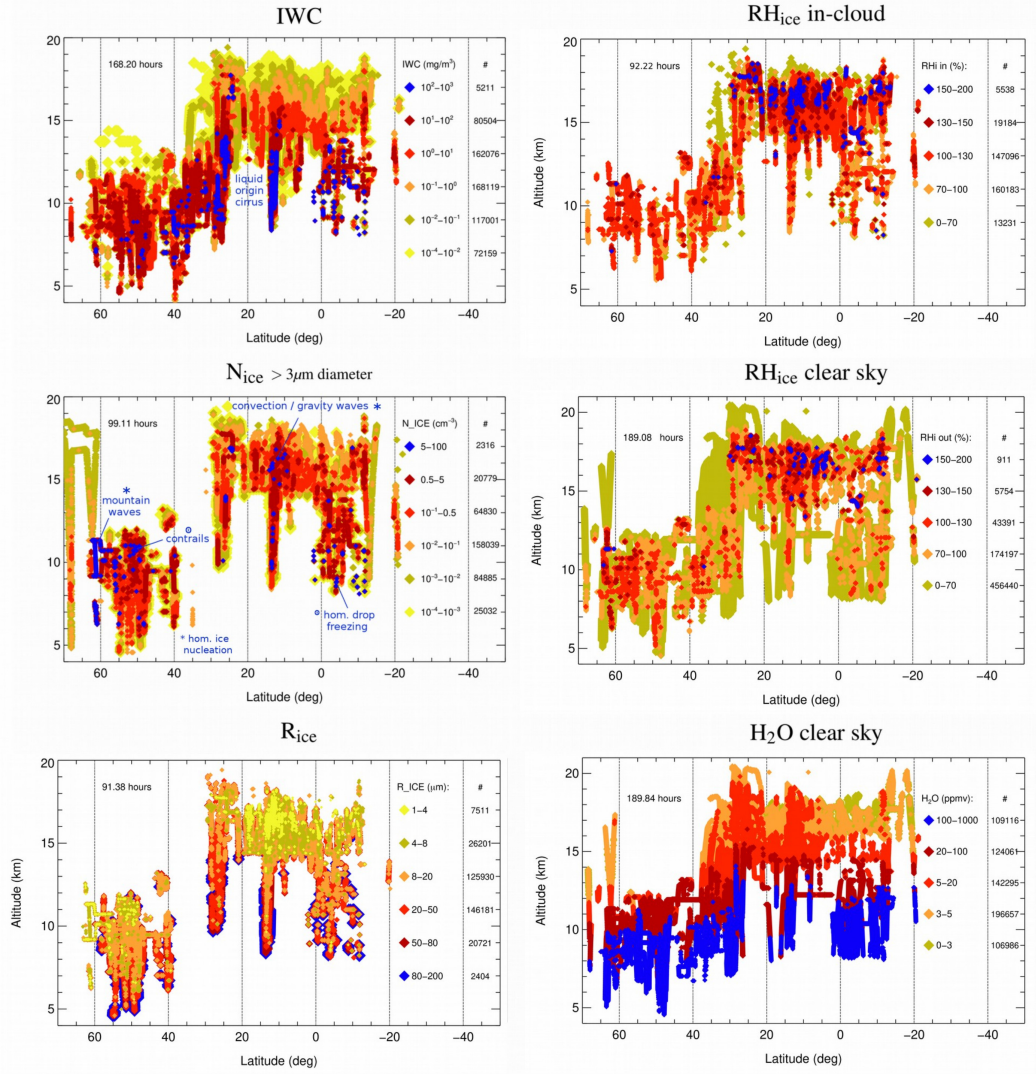


**Figure 1.** Aircraft flight paths during the 24 campaigns listed in Table 5. 185 flights, 192 h IWC measurements, 90 h  $N_{ice}$ , 84 h  $R_{ice}$ , 116/331 h  $RH_{ice}$  in/outside cirrus (campaign names: red – GfD Learjet, green – BAe-146, blue – Geophysika, purple – WB-57, black – HALO, light-purple – Global Hawk, cyan – GV HIAPER, orange – Spec Learjet). In comparison: Schiller et al. (2008) / Krämer et al. (2009) – 52 flights, 27 h IWC measurements, 8.5 h  $N_{ice}$ , 8.5 h  $R_{ice}$ , 10/16 h  $RH_{ice}$  in/outside cirrus.





**Figure 2. Left panel:** Temperature vs. altitude inside of cirrus clouds (adopted from Schiller et al., 2008), **middle panel:** Temperature vs. potential temperature  $\Theta$ , **right panel:** clear sky  $\Theta$  vs. altitude. Plotted are 28 flights from the data base, with blueish colors for Arctic, greenish colors for mid-latitude and reddish colors for tropical observations. Magenta lines: range of the TTL (tropical tropopause layer) and tropical CPT (cold point tropopause) (after Fueglistaler et al., 2009).



**Figure 3.** Cirrus cloud distribution with latitude and altitude of **Left column:** Ice water content (IWC, top panel), ice crystal number ( $N_{\text{ice}}$ , size  $> 3 \mu\text{m}$  diameter, middle panel) and mean mass radius ( $R_{\text{ice}}$ , calculated from  $\text{IWC}/N_{\text{ice}}$ , bottom panel). **Right column:** In-cloud and clear sky relative humidity wrt ice ( $\text{RH}_{\text{ice}}$ , top and middle panel), and clear sky water vapor volume mixing ratio ( $\text{H}_2\text{O}$ , bottom panel). The field campaigns are listed in Table 4, data evaluation methods and detection ranges of the parameters are described in Appendix A. The color codes range from yellow to blue with increasing amount of the respective parameter; for  $R_{\text{ice}}$ , the color code is reversed to indicate that high / low  $N_{\text{ice}}$  belong to small / large  $R_{\text{ice}}$ ; note that the data points are plotted in the order of the colors from yellow to blue; frequencies of occurrence of the parameters can be seen in Figure 7.

Typical characteristics of cirrus types in the initial stage				
ORIGIN	IWC	N <sub>ice</sub>	R <sub>ice</sub>	weather system
<i>slow updraft (heterogeneous ice nucl.)</i> <sup>mostly</sup>				
IN-SITU	low	few	large	frontals systems (WCBs)
LIQUID	high	more	larger	
<i>fast updraft (homogeneous ice nucl.)</i> <sup>mostly</sup>				
IN-SITU	high	many	small	gravity waves, convection
LIQUID	high	more	small & larger	

*slow updraft*:  $\lesssim 10$  cm/s; *fast updraft*:  $\gtrsim 10$  cm/s (Kärcher and Lohmann 2002, Krämer et al. 2016).

IWC high/low: above/below the IWC median (see Figure 7).

$N_{ice}$  few/more/many: below/in-between/above the 10 and 90%  $N_{ice}$  percentiles (see Figure 7).

$R_{ice}$  small/large/larger:

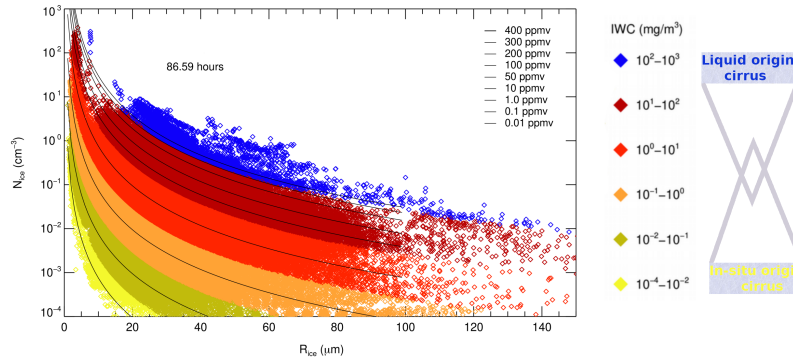
ice particles  $\lesssim 20\mu\text{m}$  dominate the PSD /

ice particles  $\gtrsim 20\mu\text{m}$  dominate the PSD, max. size several hundred  $\mu\text{m}$  diameter /

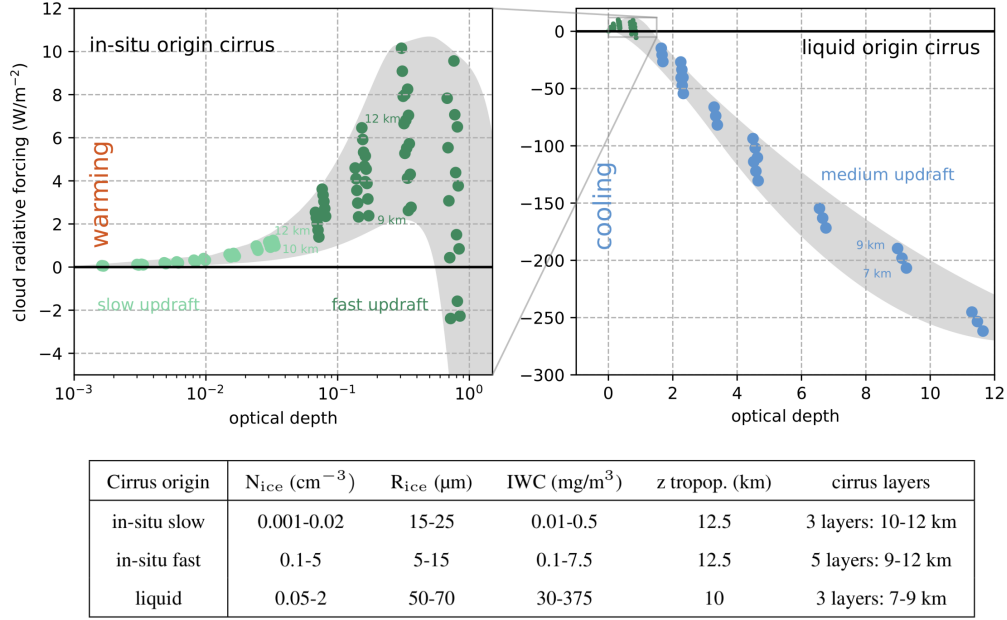
ice particles  $\gtrsim 20\mu\text{m}$  dominate the PSD, max. size up to thousand  $\mu\text{m}$  diameter,

PSD: particle size distribution.

**Table 1.** Typical characteristics of cirrus types in the initial stage.

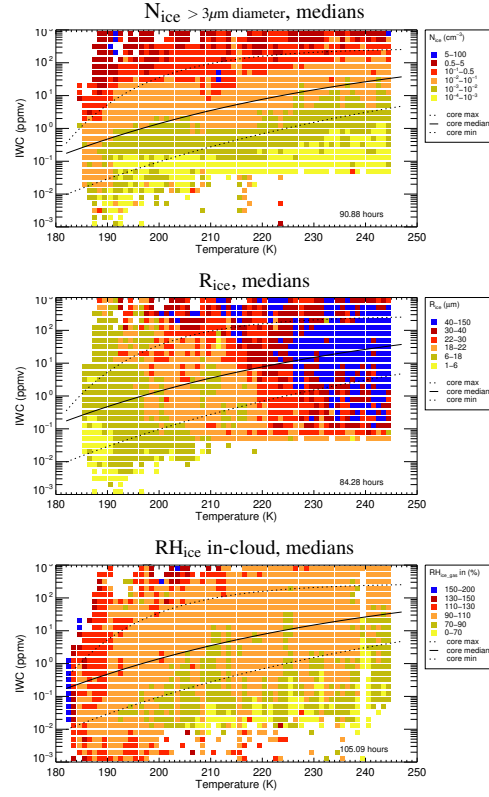


**Figure 4.** Relation between ice crystal concentration  $N_{ice}$  and mean mass radius  $R_{ice}$ , color coded by the ice water content IWC in  $\text{mg/m}^{-3}$ , from  $\sim 87$  h of cirrus cloud observations ( $R_{ice}$  is calculated by dividing  $\text{IWC}/N_{ice}$ ). The thin black lines are isolines of IWC in ppmv (in the order of the legend). The scheme at the right side illustrates the partitioning of the clouds between ‘liquid origin’ and ‘in situ origin’: the thickest cirrus (blue points) are of ‘liquid origin’, the thinnest (yellow points) of ‘in situ origin’. As the thickness decreases, the portion of ‘liquid origin’ cirrus becomes smaller and smaller while more and more ‘in situ origin’ cirrus appear.



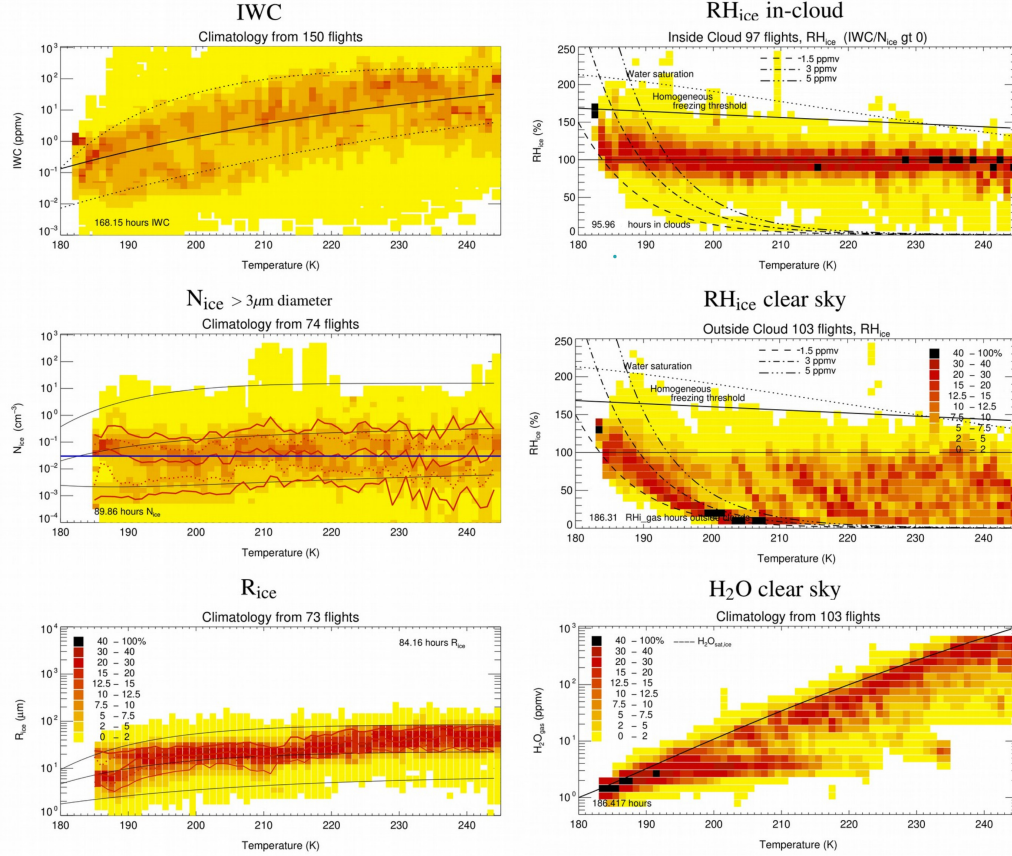
**Figure 5.** Simulated radiative forcing versus optical depth for exemplary ‘in situ origin’ slow and fast updraft (light and dark green dots) as well as ‘liquid origin’ (blue dots) cirrus; the idealized scenarios are summarized in the table.

Description of the idealized scenarios: a temperature profile is prescribed representative for midlatitude conditions, the surface temperature is set to 288 K (at  $p = 1000$  hPa,  $p$  exponentially decreases with height). The vertical profile of relative humidity over ice is also established, with a saturated layer of 1000 m thickness, constant subsaturation below the layer, and a strong decrease in humidity towards the stratosphere. A cirrus cloud with constant ice mass and number concentration is placed in the saturated layer. From the measurements (see Figure 4), typical values of  $N_{ice}$ ,  $R_{ice}$  and IWC are chosen for the three cirrus types (see table above). The vertical profiles are adjusted with respect to tropopause height ( $z$  tropop.) and placement of the cirrus layers in accordance with the cirrus types. For the radiative transfer calculations, the well-known two-stream radiative transfer model for ice particles by Fu and Liou (1993), with 6 bands in the solar and 12 bands in the thermal infrared regime, is used. The simulations are realized for a geographic latitude of  $\varphi = 50^\circ$ , solar surface albedo of 0.3, infrared surface emissivity of 1 and solar constant  $S = 1340$   $W/m^2$ ; we assume equinox conditions (e.g. end of March) at local time  $t = 12$  h (for these settings, we use the modified model of Joos et al., 2014). The net cloud radiative forcing (CRF) is calculated using the fluxes at the top of atmosphere in the short wave and long wave ranges in comparison with a clear sky case.

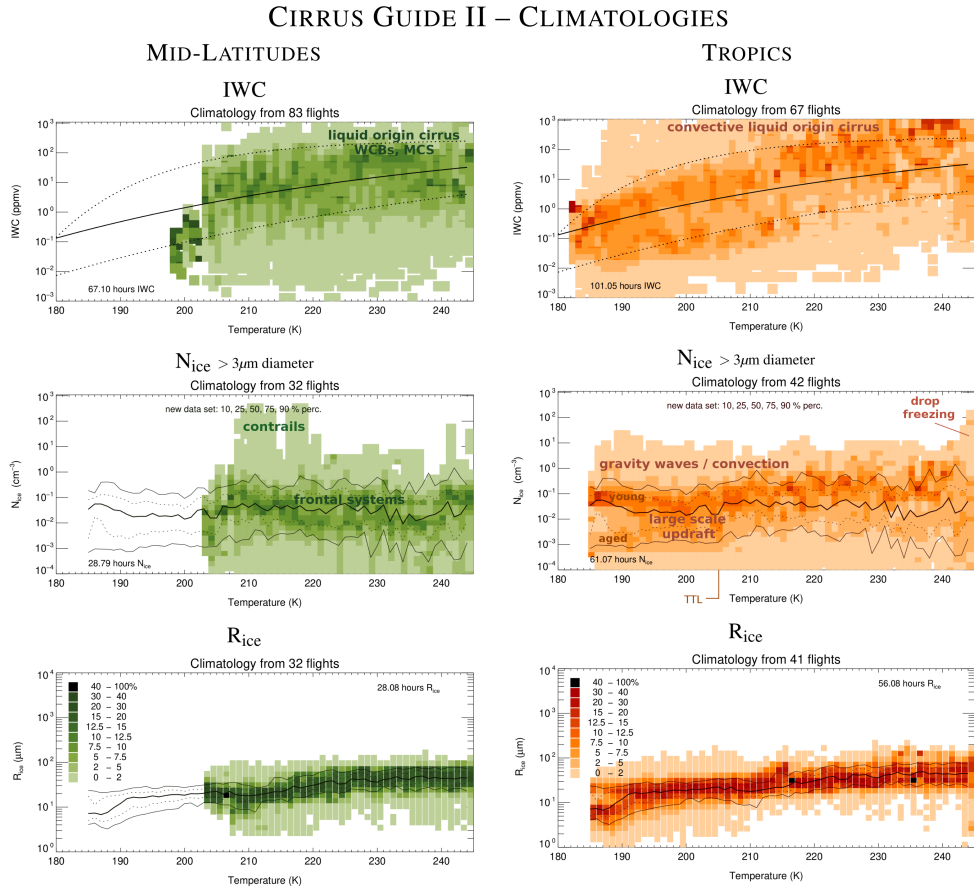


**Figure 6.** Median  $N_{ice}$ ,  $R_{ice}$  and  $RH_{ice}$  in intervals in the IWC-T parameter space (five iwc intervals per order of magnitude, 1K temperature intervals). The color codes of  $N_{ice}$  and  $RH_{ice}$  range from yellow to blue with increasing amount of the respective parameter; for  $R_{ice}$ , the color code is reversed to indicate that high / low  $N_{ice}$  belong to small / large  $R_{ice}$ . Black solid/dotted lines: median, min/max IWC of the core IWC band of Schiller et al. (2008). The variability of the parameters (25, 50 (median) and 75 % percentiles) is shown in Figures 1–3 of the supplementary material.

## CIRRUS GUIDE II – CLIMATOLOGIES

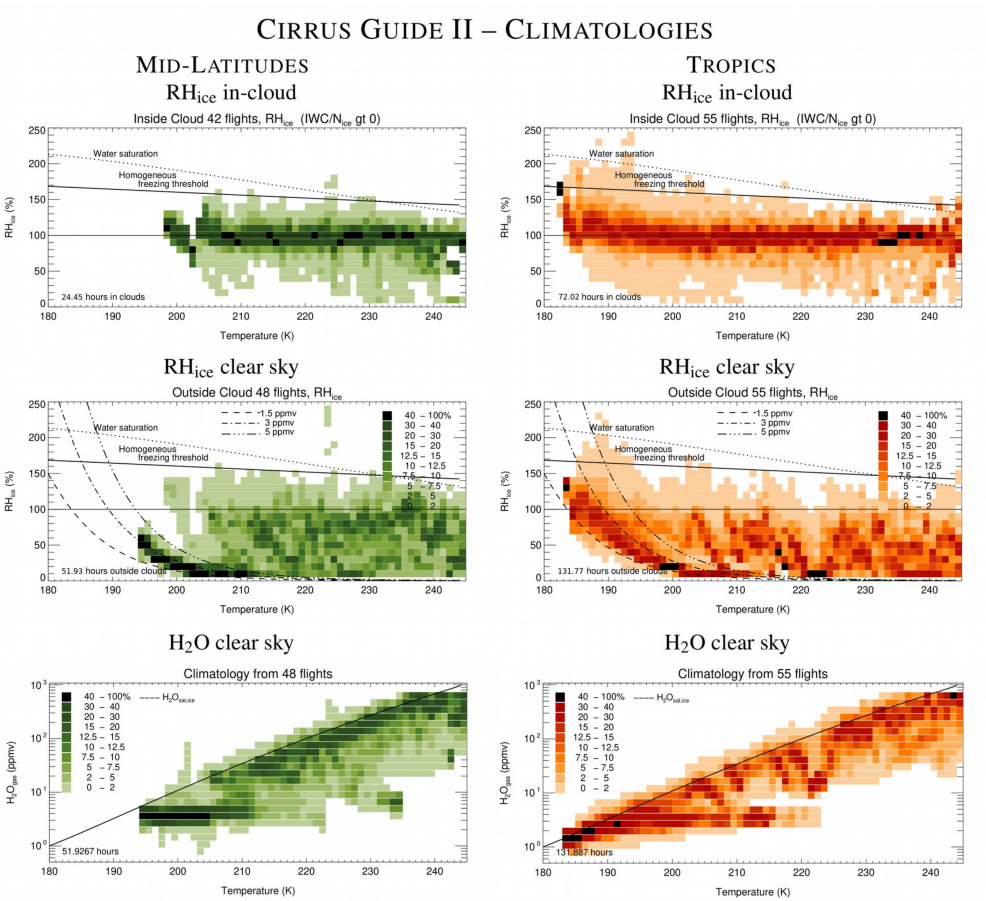


**Figure 7.** Frequencies of occurrence in dependence on temperature, binned in 1K intervals of: Ice Water Content (**IWC**; top left panel; black solid/dotted lines: median, min/max IWC of the core IWC band of Schiller et al. (2008)). Ice crystal number (**N<sub>ice</sub>**, size > 3 μm diameter, middle left panel; red lines: 10, 25, 50, 75, 90% **N<sub>ice</sub>** percentiles; blue line: fit through median **N<sub>ice</sub>** values (median **N<sub>ice</sub>**: 0.03 cm<sup>-3</sup>; 10, 25, 75, 90% percentiles: 0.002, 0.007, 0.102, 0.3 cm<sup>-3</sup>); black lines: minimum/middle/maximum **N<sub>ice</sub>** of Krämer et al. (2009)). Mass mean radius (**R<sub>ice</sub>**; calculated from **IWC**/**N<sub>ice</sub>**, bottom left panel; red lines: 10, 25, 50, 75, 90% **R<sub>ice</sub>** percentiles; black lines: minimum/middle/maximum **R<sub>ice</sub>** of Krämer et al. (2009)). In-cloud and clear sky relative humidity wrt ice (**RH<sub>ice</sub>**, top and middle right panel), and water vapor volume mixing ratio (**H<sub>2</sub>O**, bottom right panel). The field campaigns included in the data analysis are listed in Table 5. Data evaluation methods and detection ranges of the parameters are described in Appendix A. Note that for **N<sub>ice</sub>** -and thus **R<sub>ice</sub>**- the hours spent in clouds is less than in Figure 3. The reason is that for the calculation of data frequency distributions only measurements covering the same detection range are used (see also Appendix A2).



**Figure 8.** Same as Figure 7 (left column), but for mid-latitudes and tropics (WCBs: Warm Conveyor Belts; MCS: Mesoscale Convective Systems; TTL: Tropical Tropopause Layer). The field campaigns are listed in Table 5.

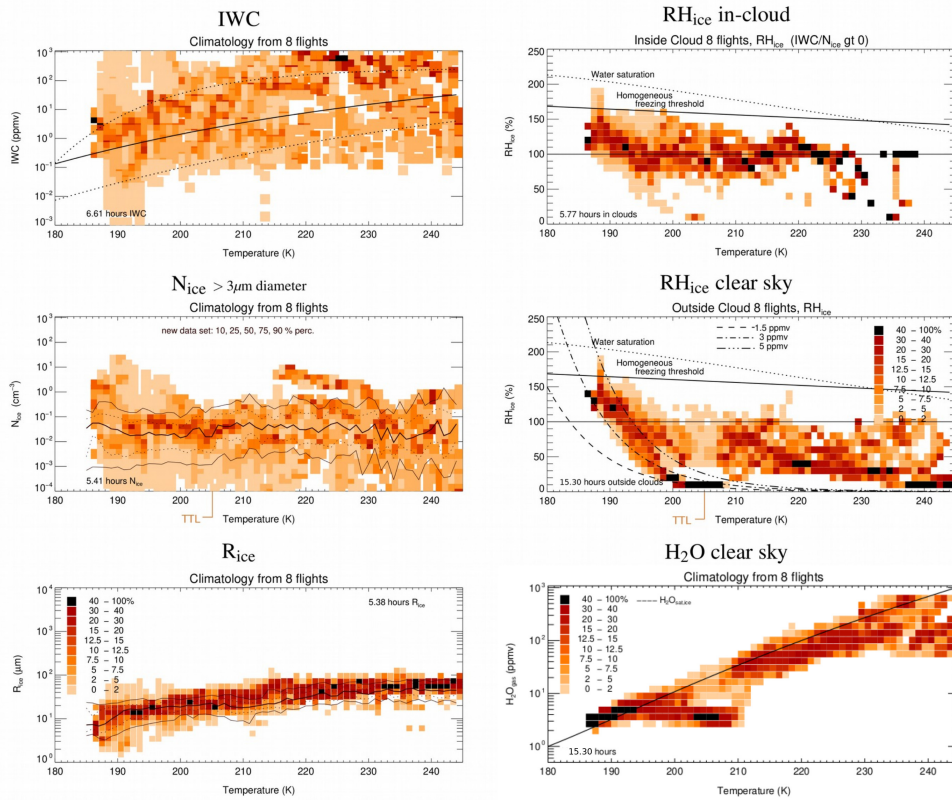




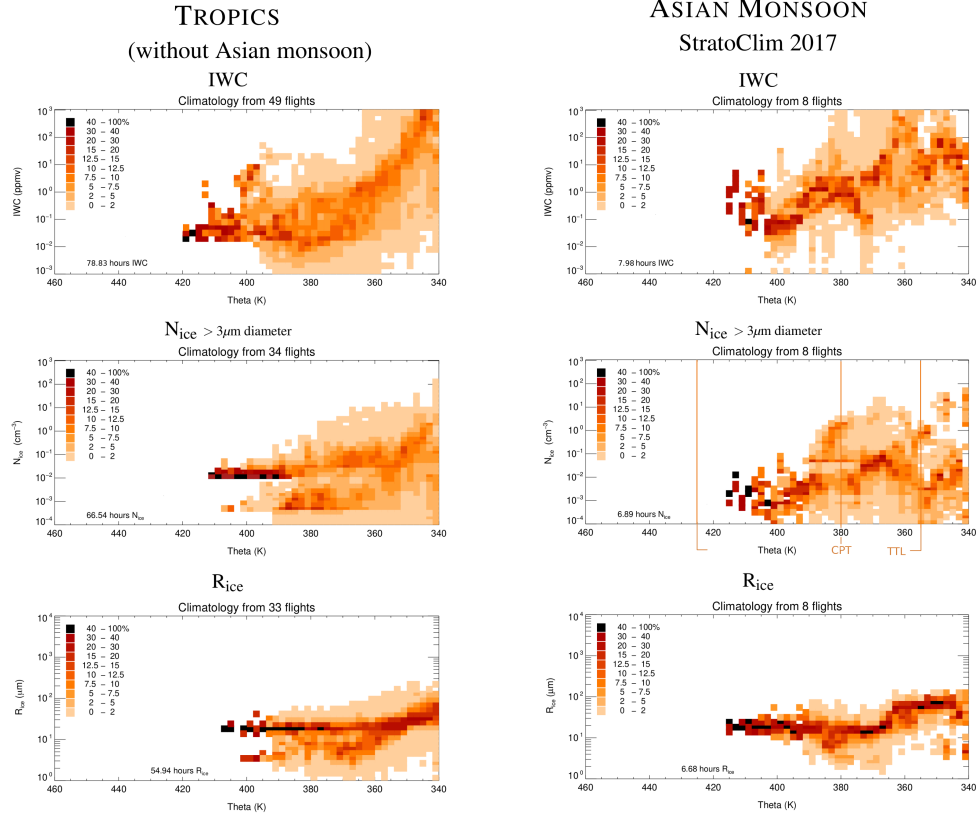
**Figure 9.** Same as Figure 7 (right column), but for mid-latitudes and tropics. The field campaigns are listed in Table 5.



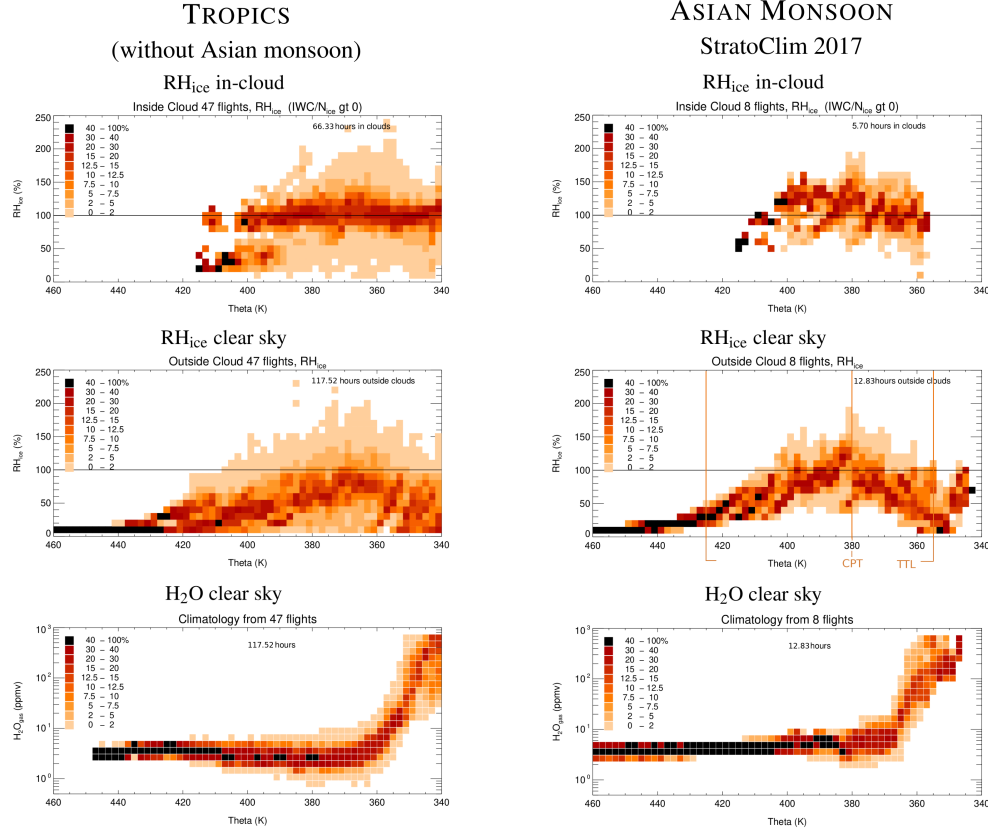
ASIAN MONSOON  
StratoClim 2017



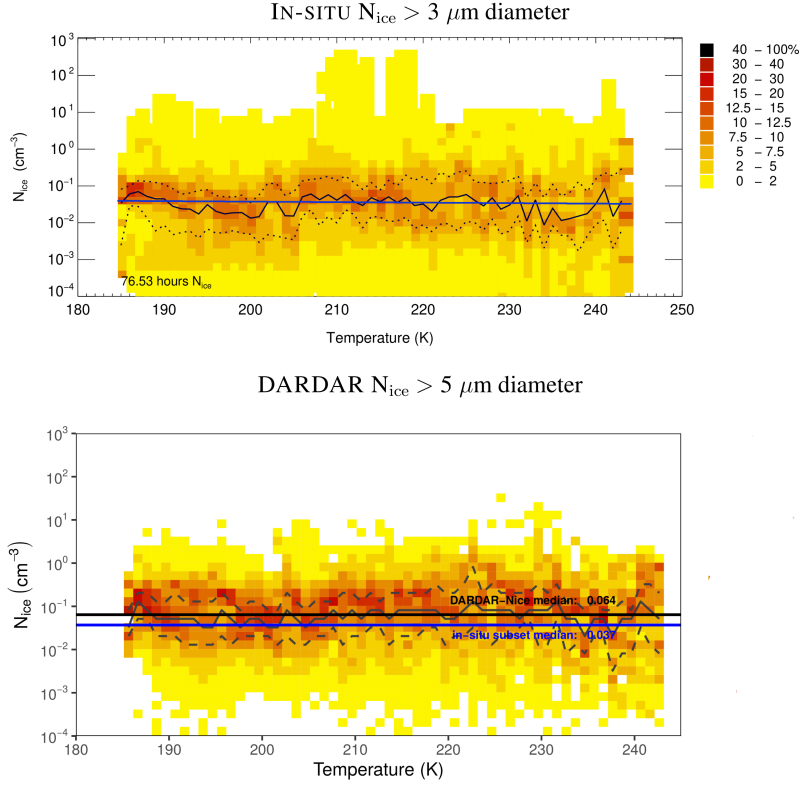
**Figure 10.** Same as Figure 7, but for the field campaign StratoClim 2017 above the Asian monsoon.



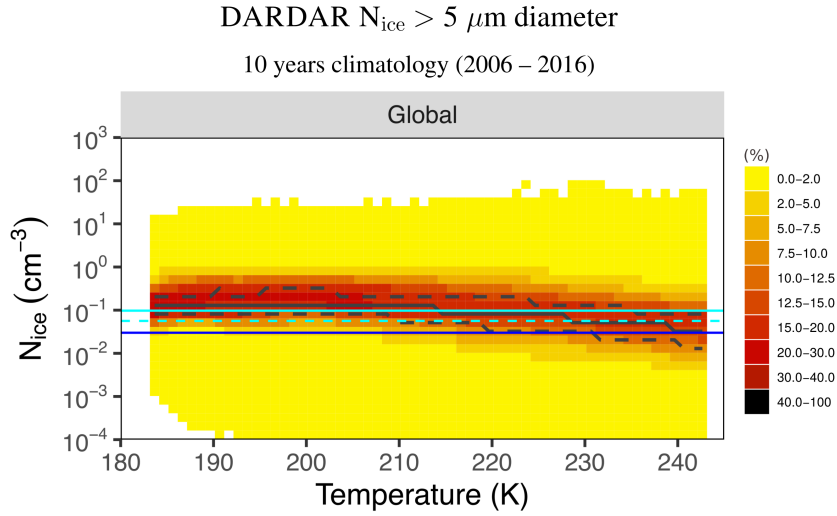
**Figure 11.** Climatology of cirrus clouds in dependence on potential temperature  $\Theta$  (left panels: tropics without Asian monsoon, right panels: Asian monsoon). In the middle panel, the range of the TTL (after Fueglistaler et al., 2009) and the cold point tropopause (CPT, derived from the observed temperature profiles) are marked; the corresponding altitude range is  $\sim 14$  to 20 km. Note that in the tropics  $355\text{--}330\text{ K} \approx 235\text{--}275\text{ K}$  ( $-38\text{--}0\text{ C}$ ) and that the TTL spans from about  $425\text{--}355\text{ K}$ . The field campaigns are listed in Table 5.



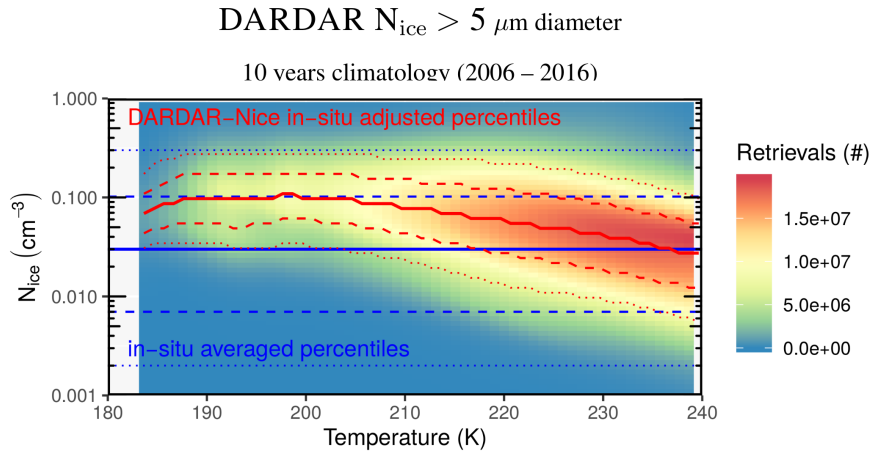
**Figure 12.** Climatology of humidity in dependence on potential temperature  $\Theta$  (left panels: tropics without Asian monsoon, right panels: Asian monsoon). In the middle panel, the range of the TTL (after Fueglistaler et al., 2009) and the cold point temperature (CPT, derived from the observed temperature profiles) are marked; the corresponding altitude range is  $\sim 14$  to 20 km. Note that in the tropics  $355\text{--}330\text{ K} \approx 235\text{--}275\text{ K}$  ( $-38\text{--}0\text{ C}$ ) and that the TTL spans from about  $425\text{--}355\text{ K}$ . The field campaigns are listed in Table 5.



**Figure 13.**  $N_{ice}$  – temperature climatologies from (top panel) in situ measurements during the field campaigns COALESC2011, ATTREX2014, ACRIDICON2014, ML-CIRRUS2014 and STRATOCLIM2017, (bottom panel) satellite remote sensing, applying the algorithm DARDAR-Nice  $f(N_0^*, D_m)$  to  $N_0^*$ ,  $D_m$  derived from the in situ observations. The black contours represent the 25, 50 and 75th percentiles. Straight black line: DARDAR-Nice average median  $N_{ice}$  ( $0.064 \text{ cm}^{-3}$ ), straight blue line: in situ average median  $N_{ice}$  ( $0.037 \text{ cm}^{-3}$ ). The excess of  $N_{ice}$  by a factor of 1.73 in DARDAR is caused by the retrieval method, for more information see text.



**Figure 14.**  $N_{ice}$  – temperature climatology from 10 years satellite remote sensing observations using DARDAR-Nice. The data was collected twice a day, at midday and at midnight. Thick black lines: 25, 50 and 75<sup>th</sup> percentiles; thin plain cyan line: DARDAR-Nice median  $N_{ice}$ ; thin dotted cyan line: DARDAR-Nice median  $N_{ice}$  adjusted to the offset factor of 1.73 between DARDAR-Nice and the in situ observations (see Figure 13); blue line: in situ average median  $N_{ice}$  (from Figure 7). Temperature intervals containing less than 10 retrievals were excluded (results non-sensitive to exact threshold).



**Figure 15.**  $N_{ice}$  – temperature climatology across the entire  $N_{ice}$ -T parameter space. Red lines: global DARDAR-Nice percentiles (adjusted to the in situ observations by the offset factor of 1.73, see Figure 13). The 10/90<sup>th</sup>, 25/75<sup>th</sup> and 50<sup>th</sup> percentiles are indicated by dotted, dashed and plain lines, respectively. Blue lines: in situ average percentiles (from Figure 7). The percentiles are recommended for intercomparison with other data sets (the corresponding numbers are listed in Table ??); regional median  $N_{ice}$  are shown in Table 3. The shaded background indicates the occurrence of retrievals, i.e. cirrus clouds. The cirrus appear most often at higher temperatures above about 225 K.

Temperature (K)	in-situ adj. DARDAR-Nice				
	$N_{ice} \text{ (cm}^{-3}\text{)}$				
	10% perc.	25% perc.	median	75% perc.	90 % perc.
180-190	0.034	0.051	0.087	0.149	0.243
190-200	0.033	0.055	0.100	0.173	0.274
200-210	0.028	0.051	0.091	0.163	0.265
210-220	0.017	0.034	0.068	0.133	0.239
220-230	0.011	0.022	0.049	0.102	0.195
230-240	0.007	0.015	0.033	0.068	0.130
Temperature all	In-situ				
	$N_{ice} \text{ (cm}^{-3}\text{)}$				
	10% perc.	25% perc.	median	75% perc.	90 % perc.
	0.002	0.007	0.03	0.102	0.3

**Table 2.** DARDAR-Nice and in situ observation  $N_{ice}$  percentiles (as portrayed in Figure 15) for intercomparison with global models or other data sets.

Region	in-situ adj. DARDAR-Nice			
	$N_{ice} \text{ (cm}^{-3}\text{)}$			
	medians			
	all Temp.	T-int weighted	most frequent	T-range
Arctic	0.036	0.023	0.016	230-242 K
Mid-lat North	0.057	0.040	0.030	227-242 K
Tropics	0.070	0.067	0.074	197-221 K
Mid-lat South	0.057	0.043	0.031	227-242 K
Antarctica	0.050	0.045	0.029	222-242 K
Global	0.056	0.046	0.031	224-242 K

**Table 3.** DARDAR-Nice median  $N_{ice}$  (as portrayed in Figure 15) for five regional latitude bands: Arctic:  $90^{\circ}$ – $67.7^{\circ}$ , Mid-lat North:  $67.7^{\circ}$ – $23.3^{\circ}$ , Tropics:  $23.3^{\circ}$ – $-23.3^{\circ}$ , Mid-lat South:  $-23.3^{\circ}$ – $-67.7^{\circ}$ , Antarctica:  $-67.7^{\circ}$ – $-90^{\circ}$  as well as global median  $N_{ice}$ . All Temp.: median without temperature binning; T-int weighted: median using the relative occurrence of each temperature interval as a weight; most frequent: median considering those temperature intervals that contain 50% of the total  $N_{ice}$  occurrence, T-range denotes the respective minimum and maximum temperatures.

## Appendix A: Methods

### A1 Field campaigns and instrumentation

The field campaigns, locations and deployed instruments are listed in Table 4, which is an extended version of the respective Table in the Cirrus Guide: Part I. For a brief description of the instruments already introduced there, we refer to Krämer et al. (2016). Other instruments used during START 2008, CONTRAST 2014, ATTREX 2014, POSIDON 2016 and LTU 2012-2018 are briefly introduced here.

ATTREX 2014 and POSIDON 2016: IWC and  $RH_{ice}$  are measured with the dual channel NOAA- $H_2O$  instrument, a dual channel TDL hygrometer which detects both  $H_2O_{tot}$  ( $= H_2O_{gas} + IWC$ ) and  $H_2O_{gas}$  by using a forward and a backward directed inlet. The detection limit of the IWC is 0.03 ppmv ( $\approx 0.003 \text{ mg/m}^3$ ). Cloud particle size distributions (PSDs) and thus  $N_{ice}$  are recorded by a FCDP and a 2D-S. The size range of the FCDP is 1–50  $\mu\text{m}$  diameter and the 2D-S counts cloud particles between 25–3005  $\mu\text{m}$ .

Start-08 and CONTRAST: IWC and  $N_{ice}$  are both derived from PSD measurements by a CDP and a 2D-C. The size range of the CDP is 2–50  $\mu\text{m}$  diameter and the 2D-C counts cloud particles between 60–1100  $\mu\text{m}$ .

LTU 2012-2018: IWC and  $N_{ice}$  are both derived from PSD measurements by the Balloon-borne Ice Cloud particle Imager (B-ICI), detecting ice particle sizes between 10 and 1200  $\mu\text{m}$  diameters. Note that the measurements are not from aircraft, but from balloons launched at Kiruna, Sweden.

### A2 Data evaluation methods

The data evaluation methods, detection ranges and data quality criteria of the measurements of IWC,  $N_{ice}$ ,  $R_{ice}$ ,  $RH_{ice}$  and  $H_2O$  are described in the following. The instruments measuring the respective parameters are listed in Table 4, the estimated uncertainties are mentioned in the Table caption. For the calculation of frequency distributions of the data, only those campaigns where the measured parameters cover the same detection range are used, since mixing of data with differing detection ranges influences the calculation of percentiles. Thus, the list of analyzed campaigns varies for each parameter, since not all parameters are available for the same detection ranges in all campaigns, or the data quality is considered as suspicious. In Table 5 the available parameters are listed for each campaign. The resulting data coverage of the parameters over the entire temperature range is shown in Figure 16.

### A2.1 Ice water content (IWC)

In the earlier studies of Schiller et al. (2008) and Luebke et al. (2013), the IWC was detected as difference between total water ( $H_2O_{tot}$ : water vapor + evaporated ice crystals) and gas phase water ( $H_2O_{gas}$ ); this product is named here  $IWC(H_2O)$ . Krämer et al. (2016) also determined the IWC by  
1510 integrating the ice particle size distributions (PSDs) measured by cloud spectrometers in cases where no total water measurements were available; we call this  $IWC(PSD)$ . A detailed description of both methods is given in Afchine et al. (2018). Also, Afchine et al. (2018) demonstrated good agreement between the two methods under reliable sampling conditions. Agreement between  $IWC(H_2O)$  and  $IWC(PSD)$  is also a criterion for the quality of the IWC data (see also Thornberry et al., 2017), as  
1515 well as for the measurements of  $H_2O$  and the PSDs (for the data quality of  $H_2O$  and  $N_{ice}$  see also the following subsections).

Unlike in the earlier studies, we here combine the two methods,  $IWC(H_2O)$  and  $IWC(PSD)$ . In case both measurements are available,  $IWC(H_2O)$  is used as first choice for each flight second. But if  $IWC(H_2O)$  is zero or missing,  $IWC(PSD)$  is used. This combination of IWCs can fill times of  
1520 measuring failures or signals beyond detection limits of one instrument and also compensates for some of the inhomogeneities in cirrus. If only one instrument is available, the respective IWC is taken.

As discussed by Schiller et al. (2008) and Afchine et al. (2018), the lower detection limit of  $IWC(H_2O)$  depends on the temperature and can not be clearly defined. Data points below the lower  
1525 dotted line drawn in the  $IWC$ - $T$  parameter space in Figure 16 (upper left panel) represent observations where the difference between both the  $H_2O_{tot}$  and  $H_2O_{gas}$  measurements is not significant to unambiguously identify a cloud. This is where the ratio  $H_2O_{enh}/H_2O_{gas} < 1.07$ ; note here that  $H_2O_{enh}$  is the originally measured total water quantity, which is enhanced due to the sampling characteristics of the system (for more detail see Schiller et al., 2008; Afchine et al., 2018). The lower  
1530 detection limit of  $IWC(PSD)$  is 0.05 ppmv ( $\approx 0.005 \text{ mg/m}^{-3}$ ) for NIXE-CAPS and 0.01 ppmv ( $\approx 0.001 \text{ mg/m}^{-3}$ ) for the combination of FCDP and 2D-C.

### A2.2 Ice crystal number ( $N_{ice}$ )

From the campaigns where cloud particle size distributions (PSDs) are measured, we chose those where cloud particles between 3 and  $\sim 1000 \mu\text{m}$  diameter are recorded when calculating  $N_{ice}$  (see  
1535 Table 5). We calculate  $N_{ice}$  as the sum of ice crystal concentrations over all size bins larger than  $3 \mu\text{m}$  on a 1 Hz time resolution, because this time span represents around 200 m flight distance - which might already not resolve cirrus inhomogeneities. Thus, when averaging  $N_{ice}$  over longer flight times, cloud free segments might influence the ice crystal concentrations.

Two instruments are needed to be deployed to cover the complete cirrus ice particle size range,  
1540 one for smaller and the other for larger cloud particles. The PSDs from NIXE-CAPS (CAS-Depol + CIPgs) and FCDP + 2D-S are merged between 20 and  $25 \mu\text{m}$  to avoid overlap of particle sizes,



those of CDP + 2D-C at 55  $\mu\text{m}$ . In earlier campaigns where only FSSP measurements (3–30  $\mu\text{m}$ ) are available, we calculate  $N_{\text{ice}}$  without the larger particles, since they contribute only a negligible part to  $N_{\text{ice}}$  (these campaigns are marked as  $y^*$  in Table 5). However, for the calculation of occurrence frequencies only the campaigns covering the whole size range are used.

*Concentrations and frequencies of occurrence of small ice crystals: the effect of limited sampling volumes*

$N_{\text{ice}}$  is given by the number of cloud particle events recorded in the cloud volume that is sampled. Thus, the lowest concentration that can be detected is when only 1 particle is recorded. For the particle imaging probes (CIPgs, 2D-S, 2D-C, used size range  $\sim 20\text{--}1000\ \mu\text{m}$ ) that results to  $\sim 10^{-4}\ \text{cm}^{-3}$ , while for the light scattering probes (CAS-Depol, FCDP, CDP, used size range  $\sim 3\text{--}20\ \mu\text{m}$ ) it reaches the higher value of  $\sim 0.015\ \text{cm}^{-3}$ . The difference in the recorded concentration range is caused by the differing sampling volumes of the two instrument types. As a consequence,  $N_{\text{ice}}$  concentrations  $\lesssim 0.015\ \text{cm}^{-3}$  contain only ice particles  $\gtrsim 20\ \mu\text{m}$ . Lower concentrations of smaller particles can not be detected with current particle measurement techniques (see also Krämer et al., 2016; Baumgardner et al., 2017).

Figure 17 (left panel) shows exemplarily  $N_{\text{ice}}$  in dependence on temperature, measured with NIXE-CAPS (NIXE-CAPS combines a CAS-Depol and a CIPgs) during the field campaign ML-Cirrus 2014, color coded by their frequencies of occurrence. CAS-DPOL samples  $47.5\text{cm}^{-3}@190\text{m/s}$  aircraft cruising speed in 1 second while CIPgs probes about  $1000\text{--}16000\text{cm}^{-3}$  in dependence on particle size in the same time interval and aircraft speed (Costa et al., 2017). Thus, one particle event in the CAS-DPOL corresponds to a higher concentration as in the CIPgs. The red line in the left panel of Figure 17 represents the lowest detectable  $N_{\text{ice}}$  of CAS-DPOL and the blue line the same for CIPg. A notable feature in the figure is that directly above the lowest detectable  $N_{\text{ice}}$  of CAS-DPOL (red line) the  $N_{\text{ice}}$  frequencies jump to a higher level than below. The reason for this peak is an effect caused by the small sampling volumes of these instruments, which is often called ‘bad statistics’. It concerns instruments whose lowest  $N_{\text{ice}}$  detection limit is above the naturally occurring particle concentrations. When clouds with concentrations smaller than the lowest detectable  $N_{\text{ice}}$  are probed, these concentrations appear in the sampling volume as ‘single particles events’ (in the size range  $\lesssim 20\ \mu\text{m}$ ) and are assigned to the minimum detectable concentration of the respective instrument, since the occurrence of one particle per time unit can not be related to the true larger air volume it belongs to. Therefore, the data sets of this study are cleared from the artificial concentration feature at the instrument’s detection limit by excluding  $N_{\text{ice}}$  caused by one single particle in the respective instruments.

The effect of this correction on the occurrence frequencies can be seen in Figure 17 (right panel), which shows the same data set as the left panel, but with ‘single particle events’ removed: the sharp edge at the lowest detectable  $N_{\text{ice}}$  of the CAS-DPOL has disappeared, instead more lower concen-

trations from the CIPg become visible, which were hidden by the artificial concentrations from the  
1580 CAS-DPOL ‘single particle events’ before.

‘Single particle events’ are less common if the measurements are averaged over larger time periods. The considered time interval can be adapted according to the required spatial resolution of the measurements. However, as outlined above, for this study we choose to show the lowest time interval (1 s) to achieve the highest possible spatial resolution.

#### 1585 *Ice crystal shattering*

Fragmentation of large ice crystals at the cloud probes housings distorted a correct recording of the ice crystal concentrations in earlier times. Nowadays, new inlets and also postprocessing algorithms based on the interarrival times of the crystals in the sampling volume (Korolev and Field, 2015)  
1590 have minimized this effect. Nevertheless, older data sets might be contaminated by small artifacts of shattered ice particles and needs to be marked in the data set and not used in the data analyses. Shattering can be recognized when looking at the  $N_{ice}$  frequencies of occurrence plotted versus the temperature, as can be seen in Figure 18, where an example of a data set including shattered ice crystals is shown. Clearly, high frequencies of occurrence appear at  $N_{ice}$  concentrations between 10-  
1595  $100\text{ cm}^{-3}$  for all temperatures. This ice particle ‘mode’ is not present in data sets not influenced by shattered ice crystals, as can be seen when comparing Figures 18 and 17.

### **A2.3 Water vapor and Relative Humidity wrt ice ( $H_2O$ and $RH_{ice}$ )**

Water vapor measurements where, as ice crystal concentrations, under discussion in the last decade  
1600 (Peter et al., 2006). After some efforts to improve the quality of the measurements (e. g. Fahey et al., 2014), the water vapor instruments today provide high-precision data (see Rollins et al., 2014; Meyer et al., 2015; Thornberry et al., 2015; Buchholz et al., 2013; Kaufmann et al., 2018).

Nevertheless, we check the  $H_2O$  data quality for each campaign, including the recent ones, best by comparison of several  $H_2O$  instruments as done by e.g. Kaufmann et al. (2018). In case this is  
1605 not possible, the frequencies of occurrence of in-cloud  $RH_{ice}$  are used to evaluate the quality of the measurements: inside of cirrus clouds, at temperatures  $\gtrsim 200\text{ K}$ , the frequencies should center around saturation, as can be seen in Figure 7 (upper right panel). Figure 19 gives an example of a data set with a bias in the in-cloud  $RH_{ice}$ . We should note that such a distortion might also be caused by a shift in the temperature measurements, which we, however, have not yet observed. In any case,  
1610 such data sets are not used for the data analysis.

Aircraft	Locations	IWC	RH <sub>ice</sub>	N <sub>ice</sub>
Geophysica	Seychelles <sup>1</sup> , Europe <sup>2</sup> , Brazil <sup>3</sup> , Australia <sup>4</sup> , Africa <sup>5</sup>	FISH <sup>a</sup>	FLASH <sup>c</sup>	FSSP-100 <sup>i</sup>
Geophysica	Nepal <sup>14</sup>	FISH <sup>a</sup>	FLASH <sup>c</sup>	NIXE-CAPS <sup>m</sup>
Learjet	Europe <sup>6</sup>	FISH <sup>a</sup>	OJSTER <sup>d</sup> , SEAL <sup>e</sup>	FSSP-300 <sup>j</sup>
BAe-146	UK <sup>9</sup>	NIXE-CAPS <sup>m</sup>	–	NIXE-CAPS <sup>m</sup>
HALO	Europe <sup>10</sup> , Brazil <sup>11</sup>	NIXE-CAPS <sup>m</sup>	SHARC <sup>h</sup>	NIXE-CAPS <sup>m</sup>
Balloon	Europe <sup>15</sup>	B-ICI <sup>n</sup>	–	B-ICI <sup>n</sup>
WB-57	USA <sup>7,16</sup> , Costa Rica <sup>8</sup>	CLH <sup>b</sup>	HWV <sup>f</sup> , JLH <sup>g</sup>	CAPS <sup>k</sup> , 2D-S <sup>l</sup>
WB-57	Guam <sup>13</sup>	NOAA-H <sub>2</sub> O <sup>o</sup>	NOAA-H <sub>2</sub> O <sup>o</sup>	FCDP <sup>p</sup> , 2D-S <sup>l</sup>
Global Hawk	Guam <sup>12</sup>	NOAA-H <sub>2</sub> O <sup>o</sup>	NOAA-H <sub>2</sub> O <sup>o</sup>	FCDP <sup>p</sup> , 2D-S <sup>l</sup>
GV HIAPER	USA <sup>17</sup> , Guam <sup>19</sup>	CDP <sup>q</sup> , 2D-C <sup>r</sup>	VCSEL <sup>s</sup>	CDP <sup>q</sup> , 2D-C <sup>r</sup>
Learjet	USA <sup>18</sup>	2D-S <sup>l</sup>	–	FSSP <sup>i</sup> , 2D-S <sup>l</sup>

1: APE-THESEO 1999

2: ENVISAT 2002, EUPLEX 2003, ENVISAT 2003

3: TROCCINOX 2005

4: SCOUT-O<sub>3</sub> 2005

5: AMMA 2006

6: CIRrus 2003, CIRrus 2004, CIRrus 2006,

AIRTOSS-ICE 2013

7: MidCix 2004, MACPEX 2011

8: TC-4 2007

9: COALESC 2011

10: ML-CIRrus 2014

11: ACRIDICON-CHUVA 2014

12: ATTREX 2014

13: POSIDON 2016

14: StratoClim 2017

15: LTU 2012-2015

16: CRYSTAL-FACE 2002

17: START 2008

18: SPARTICUS 2010

19: CONTRAST 2014

a: Lyman- $\alpha$  fluorescence hygrometer (Schiller et al., 2008; Krämer et al., 2009; Meyer et al., 2015)

b: Tunable diode laser hygrometer (Luebke et al., 2013)

c: Lyman- $\alpha$  fluorescence hygrometer (Sitnikov et al., 2007)

d: Tunable diode laser hygrometer (Krämer et al., 2009)

e: Tunable diode laser hygrometer (Buchholz et al., 2013)

f: Lyman- $\alpha$  fluorescence hygrometer (Rollins et al., 2014)

g: Tunable diode laser hygrometer (May, 1998)

h: Tunable diode laser hygrometer (Meyer et al., 2015)

i: Light scattering cloud probe (Baumgardner et al., 2017)

j: Light scattering cloud probe (Baumgardner et al., 2017)

k: Light scattering and optical imaging cloud probe (Baumgardner et al., 2001)

l: Optical imaging cloud probe (Lawson et al., 2006)

m: Light scattering and optical imaging cloud probe (Meyer, 2012; Luebke et al., 2016)

n: Cloud particle Imager (Wolf et al., 2018)

o: Tunable diode laser hygrometer (Thornberry et al., 2017)

p: Light scattering cloud probe (McFarquhar et al., 2007)

q: Light scattering cloud probe (Baumgardner et al., 2017)

r: Optical imaging cloud probe (Baumgardner et al., 2017)

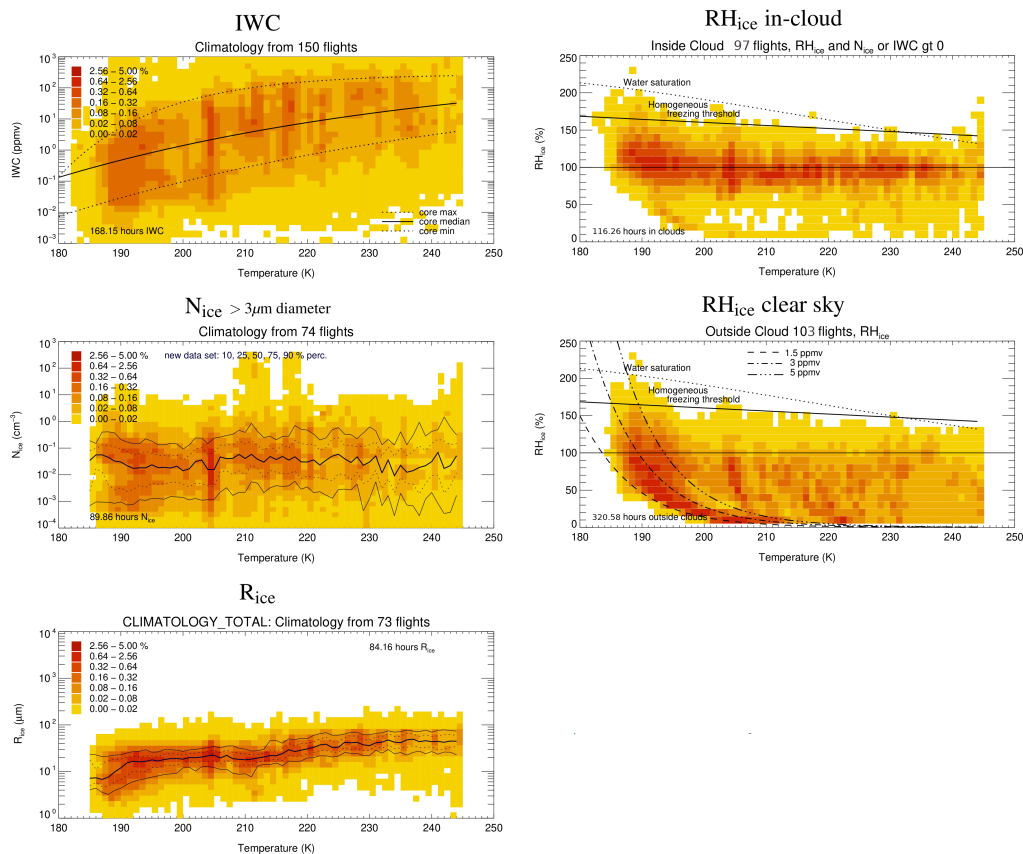
s: Tunable diode laser hygrometer (Zondlo et al., 2010)

The campaigns under 1-6 are described by Schiller et al. (2008), Krämer et al. (2009) and Finger et al. (2015), 7-8 by Luebke et al. (2013); Jensen et al. (2013), 9 Jones et al. (2012), 10 Voigt et al. (2017), 11 Wendisch et al. (2016), 12 Jensen et al. (2017), 13 <https://espo.nasa.gov/posidon>, 14 Stroh et al. ACP, in prep., 15 Wolf et al. (2018), 16 <https://espo.nasa.gov/crystallface>, 17 Pan et al. (2010), 18 Muhlbauer et al. (2014), 19 Pan et al. (2017).

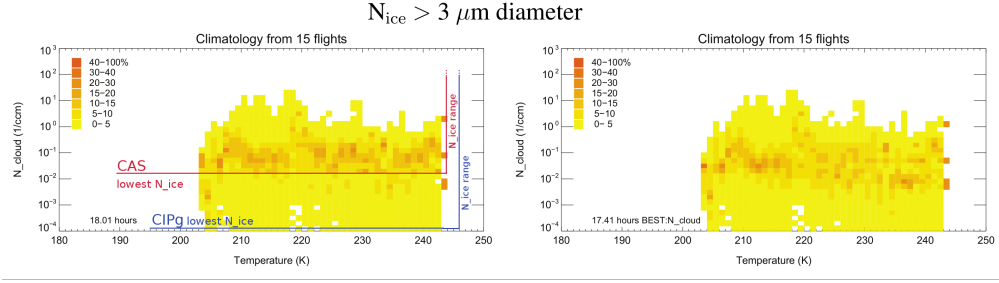
**Table 4.** Overview of campaigns and instruments. IWC: ice water content (uncertainties  $\sim 20\%$ ), RH<sub>ice</sub>: relative humidity over ice (uncertainties  $\sim 10\text{-}15\%$ ), N<sub>ice</sub>: ice crystal number concentration (uncertainties  $\sim 10\text{-}100\%$ ); the sampling rate of all aircraft instruments is 1 Hz.

Campaign	data sets <sup>†</sup>	IWC	N <sub>ice</sub>	RH <sub>ice</sub> /H <sub>2</sub> O
<b>Arctic</b>				
EUPLEX_2003	SKL	y	y*	y
ENVISAT_2003_1	SKL	y	y*	y
ENVISAT_2003_2	SKL	y	y*	y
<i>LTU_2012-18</i>		y	y <sup>spe</sup>	–
<b>Mid-latitude</b>				
ENVISAT_2002_2	SK	y	y*	y
CIRRUS_2003	SKL	y	y*	y
CIRRUS_2004	SKL	y	y*	y
CIRRUS_2006	SKL	y	y*	y
<i>START_2008</i>		y	y <sup>spe</sup>	y
MIDCIX_2004	Lk	y	y>15	
–				
<i>SPARTICUS_2010</i>		y	y>15	–
MACPEX_2011	k	y	y>15	y
COALESC_2011	k	y	y <sup>spe</sup>	–
AIRTOSS_2013	k	y	y>15	y
ML-CIRRUS_2014	k	y	y <sup>spe</sup>	y
<b>Tropics</b>				
APE_THESEO_1999	SKl	y	y*	y
CRYSTAL-FACE_2002	L	y	–	–
TROCCINOX_2005	SKL	y	y*	y
SCOUT_2005	SKL	y	y*	y
TC-4_2007	L	y	y>15	–
<i>CONTRAST_2014</i>		y	y <sup>spe</sup>	–
ACRIDICON_2014	k	y	y <sup>spe</sup>	y
ATTREX_2014		y	y <sup>spe</sup>	y
POSIDON_2016		y	y <sup>spe</sup>	y
STRATOCLIM_2017		y	y <sup>spe</sup>	y

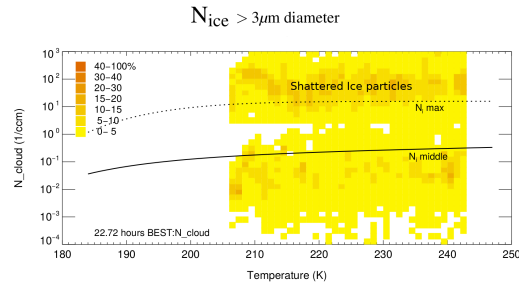
**Table 5.** Cirrus Guide II data base for the IWC, N<sub>ice</sub> and RH<sub>ice</sub> climatologies. y: quality checked measurements; –: no/questionable measurements; an overview of each campaign is given in the [Supplementary Material](#); y<sup>spe</sup>: N<sub>ice</sub> is corrected for single particle events (spe, see Section A2.2); y\*: N<sub>ice</sub> from size limited detection range (3 - 30 µm → contains  $\gtrsim 90\%$  of the ice particles), not spe corrected; y>15: N<sub>ice</sub> from size limited detection range (> 15 µm → underestimating the full ice particle concentration), not included in the data analysis presented here; the campaigns written in *italics* are not contained in the occurrence frequencies. <sup>†</sup> S K L k: campaigns contained in the data sets of Schiller et al. (2008); Krämer et al. (2009); Luebke et al. (2013); Krämer et al. (2016) (for S&K see Figure 5 of the Supplementary Material)



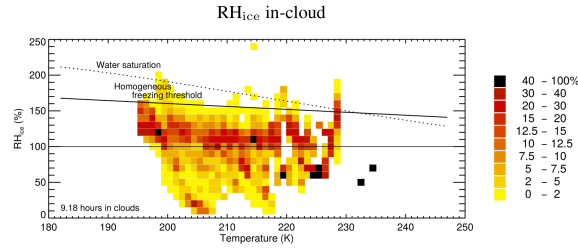
**Figure 16.** Data coverage of the Cirrus Guide II in situ data base: IWC, N<sub>ice</sub>, R<sub>ice</sub>, in-cloud and clear sky RH<sub>ice</sub> frequencies of occurrence in dependence on temperature.



**Figure 17.**  $N_{ice}$  frequencies of occurrence for ML-Cirrus 2014; left: original data set, right: corrected for 'single particle events'. A further example of an uncorrected data set can be seen in Figure 5 of the Supplementary Material, (middle left panel). For more details see text.



**Figure 18.** Example of  $N_{ice}$  measurements biased by shattering of large ice particles, visible in the frequencies of occurrence: high frequencies appear at  $N_{ice}$  concentrations between  $10\text{--}100 \text{ cm}^{-3}$  for all temperatures, which are not present in undisturbed measurements (see Figure 17). The black lines denote the middle and maximum  $N_{ice}$  lines from Krämer et al. (2009).



**Figure 19.** Example of biased  $H_2O$  measurements, visible in the in-cloud  $RH_{ice}$  frequencies: inside of cirrus clouds, at temperatures  $\gtrsim 200 \text{ K}$ , the frequencies should center around saturation (see Figure 7 upper right panel).