# Answers to the editor comments on acp-2020-40

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

by Krämer et al.

Many thanks for your further revisions and for submitting the revised version. I am happy to accept your paper for publication in ACP, however, I recommend that you do some further technical corrections in order to facilitate the typesetting process.

Also many thanks for the smooth and quick review process of the long manuscript and also your helpful comments.

1) I found a few language issues in the new paragraph:

L922: "are not considered IN this study"

Done

L923: "to quantify IN/FROM lidar/radar products"

Done

L925: "The color code represent" --> represents

Done

2) Of course the general language level is fine, but there are several places where a native speaker should be able to make things even more elegant. Just an example, in L35 "inside of clouds": why not just "in clouds"? You have several native speaker co-authors - would one of them be ready to do some language corrections? My impression is that this would be worth the effort, given the outstanding dataset and science you present in your paper.

One of the native speaking co-authors has revised the manuscript for language and also made some more small changes and technical corrections. I hope the paper is now in a shape that it can be published.

3) Please carefully check your list of references, for instance for Dinh et al., there is a 2016 version in ACP (instead of the 2015 ACPD); some journal names are written with capital letters (anyway, always use abbreviations); some journal names are just given as "JGR" (please correct); and some references are incomplete (e.g., Righi et al.).

Thanks for pointing this out, I have checked the list and corrected a number of references.

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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 ( $N_{\rm ice}$ ), ice crystal mean mass radius ( $R_{\rm ice}$ ), relative humidity with respect to ice ( $RH_{\rm ice}$ ) and water vapor mixing ratio ( $H_2O$ ) for each of the flights. Depending on the parameter, the database has been extended by about a factor of 5-10 compared to earlier studies.

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As one result of our investigations investigation, we show that the medians of  $N_{\rm ice}$ ,  $R_{\rm ice}$  and  $RH_{\rm ice}$  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 , investigation 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 examination 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  $N_{\rm ice}$  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 improve the satellite observations. We found that the global median  $N_{\rm ice}$  from satellite observations is almost two times higher than the in situ median and increases slightly with decreasing temperature.  $N_{\rm ice}$  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  $N_{\rm ice}$  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  $N_{\rm ice}$  and IWC of 30 ppmv cm<sup>-3</sup> 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  $RH_{\rm ice}$  (120-150%) inside of clouds and in clear sky are observed around and above the CPT. We attribute this to the high  $H_2O$  mixing ratios (typically 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 CPTsuggest that the water transport into the stratosphere is Above the CPT, supersaturations of 10-20% higher than expected are observed in regions of weak convective activity and up to about 50% in the Asian monsoon. This implies that the water available for transport into the stratosphere might be higher than the expected saturation value.

#### 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 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 in situ below -38°C (in situ-origin), and thicker cirrus originating from freezing in liquid clouds (liquid-origin cirrus) that are uplifted from warmer layers farther below.

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 derived from global satellite remote sensing observations were also recently enhanced to include ice crystal number concentrations (Sourdeval et al., 2018; Gryspeerdt 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, 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.

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 models. A way to accomplish this task Accomplishing these tasks requires large and high quality observational databases that can serve, for example, to evaluate global models 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<sub>ice</sub>), ice crystal mean mass radius (R<sub>ice</sub><sup>1</sup>), relative humidity with respect to ice (RH<sub>ice</sub>) and water vapor mixing ratio (H<sub>2</sub>O). However, though Although 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 Nice from satellite observations and, based on this, derive a global climatology of cirrus Nice. 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 situorigin 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.

<sup>&</sup>lt;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 g/cm<sup>3</sup>

The last part of the study (Section 6) is the step to global climatology of cirrus  $N_{\rm ice}$  from satellite remote sensing observations. For this purpose, the Cirrus Guide II in situ database is used to evaluate remote sensing cirrus observations. Based on this, a global  $N_{\rm ice}$  climatology is derived and first analyses of the global and also regional  $N_{\rm ice}$  are presented.

#### 2 Databases

#### 115 2.1 in situ data set

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The observations presented here include the ice water content IWC, the ice crystal number concentration N<sub>ice</sub>, the mean mass radius R<sub>ice</sub> in-cloud as well as the clear sky RH<sub>ice</sub> and clear sky water vapor volume mixing ratio H<sub>2</sub>O. The complete in situ data set comprises 24 field campaigns: the 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 inclusion of the field campaigns SPARTICUS 2010 and START 2008 over Central US, LTU 2012-2018 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 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 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  $N_{\rm ice}$ ,  $R_{\rm ice}$  and  $RH_{\rm ice}$  by Krämer et al. (2009):

- The number of flights and total time in cirrus increased from 104 flights / 94 hours (Krämer et al., 2016) to a total of 150 flights / 168 hours, i.e. the database disporportionally has extended by about a factor of 5-10 depending on the specific parameter.
- For IWC, a new data product analysis method has been developed that increases the observed
   data volume (Appendix A2.1).
  - For N<sub>ice</sub>, observations from advanced and extended instrumentation eould be has been added to
    the 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 tropiestropical geographical regions, and also with respect to latitude/altitude seems worthwhile.
- As in the earlier climatologies, all data underwent strict quality control.

#### 2.2 Satellite data set

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DARDAR-Nice provides observational-based estimates of  $N_{\rm ice}$  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  $N_{\rm ice}$  is subsequently estimated by direct integration of the size distribution from a minimum threshold sizeset in this studyto. For this study, we use the threshold diameter of 5 µm. 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  $D_{\rm m}$ ) 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  $N_{\rm ice}$  from recent in situ campaigns, by comparing its prediction based on in situ  $N_0^*$  and  $D_{\rm m}$  measurements to the actual in situ  $N_{\rm ice}$  measurements. Good agreements were agreement was 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  $N_{\rm ice}$  in DARDAR-Nice.

This This problem typically occurs at temperature above -50°C and is expected to be cloud-type dependent. Nevertheless, Sourdeval et al. (2018) showed that the satellite  $N_{ice}$  remains in reasonable agreement with the in situ (within a factor of 2) from T = -30°C down to -90°C, which should cover the entire cirrus temperature ranges in this study.

This evaluation framework of DARDAR-Nice is here repeated on the basis of 5 in situ campaigns archived in the Cirrus Guide II in situ database: COALESC2011, ACRIDICON2014, ATTREX2014, MLCIRRUS2014, STRATOCLIM2017. Based on the agreement between DARDAR-Nice and the in situ observations, a global N<sub>ice</sub> climatology is derived from 10 years of satellite observations. Regional N<sub>ice</sub> 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 tropics are shown in Figure 2, (left panel, adopted from Schiller et al., 2008). Exemplarily, vertical 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 upper tropospheric 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)(Smith et al., 2001; 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),  $N_{\rm ice}$  (color coded by concentration),  $R_{\rm ice}$  (color coded by size), in-cloud as well as the clear sky humidity (color coded by  $RH_{\rm ice}$ ) 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  $70^{\circ}N$  orth to around  $20^{\circ}S$  outh, i.e. the northern mid-latitudes and the tropics are covered by the observations. The altitude range is between about 5 to  $\frac{20km}{20}$  km. 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,  $0.5^{\circ}$  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)) (Sassen et al., 2008).

# 210 3.1.1 in situ-origin and liquid-origin cirrus

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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 $_{\rm ice}>100$ % and RH $_{\rm w}<100$ %; (2) liquid-origin cirrus that evolve (also heterogeneously or homogeneously) from freezing of liquid drops in clouds at T  $\gtrsim$  235K and RH $_{\rm w}\sim100$ %  $^2$ . 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 . The cirrus types and freezing mechanisms are discussed and summarized in Table 1.

IN SITU-ORIGIN cirrus can be devided in two sub-classes depending on the strength of the updraft: in *slow updrafts* <sup>3</sup> , in situ-origin cirrus form mostly heterogeneously (Krämer et al., 2016) and are rather opitcally 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 may have similar characteristics, since also to heterogeneously-formed cirrus, since 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 may not be 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<sub>ice</sub> 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 abundant 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 in *slower updrafts* together with larger ice crystals that are frozen heterogeneously at T > 235K in slower updrafts. In fast updrafts, liquid-origin cirrus with very high appear. The reason is that in such updrafts and are both > 100235 % and thus K. Small supercooled liquid cloud drops, which are much more numerous than heterogeneously formed N<sub>ice</sub>, does not reach the cirrus cloud altitude (temperature  $\leq$  235K) in the slow updrafts, the clouds completely glaciate before because of the Wegener-Bergeron-Findeisen process, where liquid drops evaporate and ice crystals remain grow at RH<sub>w</sub>  $\leq$  100% below

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

and  $RH_{ice} \rightarrow$  above 100 %, is suppressed (Korolev, 2007). Thus, the numerous supercooled drops. In *fast updrafts*, liquid-origin cirrus with very many small  $N_{ice}$  appear. The reason is that here the supercooled liquid cloud drops can reach the altitude (temperature  $\sim 235K$ ) where they freeze homogeneously (Costa et al., 2017). This is because the high updrafts keep both  $RH_w$  and  $RH_{ice}$  above 100 % and thus the Wegener-Bergeron-Findeisen process does not take place (Korolev, 2007).

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 (WCB) of low pressure systems can also produce slow updraft liquid-origin cirrus. Fast updraft in situ-origin and liquid-origin cirrus occur in gravity waves, often orographically induced, in jet streams, mesoscale 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 (corresponding to  $RH_{ice} > 100\%$ ), smaller ice crystals in the size range  $\lesssim 20\mu m$  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 deposition on 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 sublimate, with small crystals disappearing faster than larger ones (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 and/or  $N_{\rm ice}$ ) 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  $N_{\rm ice}$ ,  $R_{\rm ice}$  and IWC is shown for  $\sim\!87$  h of cirrus cloud observations. It can be nicely seen that in the cases where  $N_{\rm ice}$  is high, the ice crystals are small – because the numerous ice crystals consumed all of the available vapor, thereby suppressing further growth – while low  $N_{\rm ice}$  are related to large ice crystals for a given IWC. The IWC (in mg/m³) is indicated by the color code and result from the combination of both  $N_{\rm ice}$  and  $R_{\rm ice}$ . The thin black lines are isolines of IWC (in ppmv) that appear in the order shown the legend. The scheme at the right side illustrates symbols to the right of the figure indicate 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.

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Most of the highest IWCs (> 10 mg/m³, 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 was 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 IWCs are mostly high (red to blue colors) and, as shown by Muhlbauer et al. (2014), ice particles of greater than a thousand μm 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 (0.1-10 cm<sup>-3</sup>) with medium R<sub>ice</sub> (20-70 μm).

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 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-interim data in the North Atlantic region (see also Section 6.2).

As-While IWC is an indication for the cirrus type,  $N_{\rm ice}$  (Figure 3, middle left panel) provide a hint to the freezing mechanism, either heterogeneous or homogeneous: high ice crystal numbers  $N_{\rm ice}$  ( $\gtrsim 0.5~{\rm cm}^{-3}$ , 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 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°North. High  $N_{\rm ice}$  values have also been observed in tropical deep convection around  $10^{\circ}$ North, corresponding to measurements reported by Jensen et al. (2009).

Another source of mid-latitude high  $N_{\rm ice}$  are young contrails. Note however, that these high  $N_{\rm ice}$  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,  $N_{\rm ice}$  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  $N_{\rm ice}$  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  $N_{\rm ice}$  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 occurrence of specific signatures which can not be seen from Figure 3, is given in Section 4.

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Radiative characteristics: A motivation to study cirrus clouds is to investigate the radiative properties 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 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 conducted, with solar zenith angles corresponding to 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) 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; i.e., they switch to a slight cooling effect. The reason for the sign change 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  $\tau$  determination 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 these first very idealized simulations, we can conclude that in situ formed cirrus clouds are most likely to 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

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The distribution of in-cloud and clear sky  $RH_{ice}$  as well as water vapor  $H_2O$  with latitude and altitude is shown in the right column of Figure 3. It can nicely be seen how the amount of  $H_2O$  decreases with altitude (bottom panel). The clear sky  $RH_{ice}$  (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 occasionally found inside of the tropical cirrus clouds (top panel). The behavior of  $RH_{ice}$  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<sub>ice</sub>, R<sub>ice</sub>, RH<sub>ice</sub>

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<sub>ice</sub> 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<sub>ice</sub> 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 statistics of  $N_{\rm ice}$ , but also the corresponding mean mass size  $R_{\rm ice}$  and the in-cloud  $RH_{\rm ice}$  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- $N_{\rm ice}$ and $R_{\rm ice}$ relation

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Observational evidence for the correlation between IWC and  $N_{\rm ice}$  can nicely be seen in the upper panel of Figure 6. Almost symmetrical colored bands of  $N_{\rm ice}$  can be seen across the entire IWC-T parameter space ( $N_{\rm ice}$  concentration increases from yellow to blue). With decreasing temperature, the same  $N_{\rm ice}$  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  $N_{\rm ice}$  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  $N_{\rm ice}$  from the information of temperture and IWC.

The ice crystal sizes also form colored bands in the IWC-T parameter space (Figure 6, middle panel;  $R_{\rm ice}$  inreases 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  $R_{\rm ice}$  is calculated from the third root of the IWC divided by  $N_{\rm ice}$ , as described in Section 1. However, the  $R_{\rm ice}$  bands are somewhat less elear clearly delineated from each other than the  $N_{\rm ice}$  bands. They might be smoothed in the follow up study, so that  $R_{\rm ice}$  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 Nevertheless, we believe that the more heterogeneous structure of the R<sub>ice</sub> 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<sub>ice</sub>-R<sub>ice</sub> relations seperately for in situ-origin and liquid-origin cirrus.

#### 4.1.2 IWC-T-RH<sub>ice</sub> relation

In the bottom panel of Figure 6, median in-cloud  $RH_{\rm ice}$  are shown in the IWC-T parameter space. Also for  $RH_{\rm ice}$  patterns are visible:  $RH_{\rm ice}$  decreases with decreasing IWC. Above the median IWC (black solid line),  $RH_{\rm ice}$  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  $RH_{\rm ice}$  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  $N_{\rm ice}$ , whose small ice surface cannot efficiently deplete the water vapor. However, the supersaturations are also seen at high IWC ( $\sim$  high  $N_{\rm ice}$ ). 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

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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  $RH_{\rm lice}$  in Figures 9.

#### **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  $\lesssim 200~\rm K$  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<sub>ice</sub>-T)

About 90 hours of N<sub>ice</sub> 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

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

455 (Figure 6, SM). For N<sub>ice</sub>, the picture has greatly changed when comparing the old and the new data sets. This change is on the one hand due to an extension of the lower detection limit of N<sub>ice</sub> from  $4 \cdot 10^{-3}$  to  $10^{-4}$  cm<sup>-3</sup> (see Appendix A2.2), but also because the new data set represents a better mixture of different meteorological situations. For example, the higher N<sub>ice</sub> 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<sub>ice</sub> 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  $N_{\rm ice}$  observations within 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  $N_{\rm ice}$  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  $N_{\rm ice}$ , i.e. the envelope of the most frequent  $N_{\rm ice}$  (>5% per  $N_{\rm ice}$ -T bin). Fits through these percentiles and the median  $N_{\rm ice}$  reveal no temperature dependence of  $N_{\rm ice}$  (10%, median and 90%  $N_{\rm ice}$ : 0.002, 0.03 and 0.3 cm<sup>-3</sup>). This is different to the slight decrease with temperature of the minimum, middle and maximum  $N_{\rm ice}$  lines shown by Krämer et al. (2009), which was, as discussed above, caused by two flights with high  $N_{\rm ice}$  at comparatively warm temperatures.

It is a an open question why the N<sub>ice</sub> do not increase with decreasing temperature, as would be expected because the homogeneous ice nucleation rate then increases from theoretical homogeneous freezing calculations (see e.g. Kärcher and Lohmann, 2002). This has been investigated by Gryspeerdt et al. (2018) based on a 10 year global data set retrieved from satellite observations (DARDAR-N<sub>ice</sub>, Sourdeval et al., 2018, see also Section 6). Gryspeerdt et al. (2018) analyzed the N<sub>ice</sub> (>5µm) only at cloud tops and also those throughout the cirrus clouds. From the cloud top analysis, a clear increase of N<sub>ice</sub> with decreasing 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<sub>ice</sub> are short-lived (see Section 3.1.1) and thus not easy to trace by aircraft.

A further consideration of  $N_{\rm ice}$  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 (Rice-T)

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The ice crystal mean size is calculated as mean mass radius  $R_{\rm ice}$  as shown in Footnote 1.  $R_{\rm ice}$  is close to the common effective cloud particle radius  $R_{\rm eff}$  ( $\sim$  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  $R_{\rm ice}$  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  $R_{\rm ice}$  core band (frequencies  $\gtrsim 5\%$  per  $R_{\rm ice}$ -T bin) decreases slightly as the temperature decreases, which is caused by the decrease of the IWC core band, since the  $N_{\rm ice}$  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  $R_{\rm ice}$  shown by Krämer et al. (2009) (see Figure 6, SM). The range of  $R_{\rm ice}$  has shifted slightly to larger sizes in the new data set, which is caused by the  $N_{\rm ice}$  range extended towards lower concentrations that mainly consist of larger ice crystals.

Remarkable is the drop of the most frequent  $R_{\rm ice}$  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  $R_{\rm ice}$  - occur more often than at lower temperature where only the thinner in situ-origin clouds exist. This is especially true at mid-latitudes, in . In the tropics the liquid-origin cirrus reach also lower temperatures and thus no sudden drop of  $R_{\rm ice}$  is observed (see Figure 8, left and right bottom panel). The bifurcated structure of the most frequent  $R_{\rm ice}$  which can be seen for temperatures  $\lesssim 190$  K is discussed in Section 4.3.1 (vii).

# 510 4.2.4 Clear sky and in-cloud $RH_{\rm ice} \, (RH_{\rm ice}\text{-}T)$

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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 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.

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  $H_2O_{sat,ice}$ .

### 4.3 Mid-latitudes and tropics

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# 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  $N_{\rm ice}$  and 28/56 hours of  $R_{\rm ice}$  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 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 temperatures of 182 K. The core IWC/N<sub>ice</sub>/R<sub>ice</sub>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-origin and in situ-origin cirrus. High IWCs stem mostly from liquid-origin WCB cirrus. Above the central USA, small and 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 is N<sub>ice</sub> of up to several hundreds per cm<sup>3</sup>, 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<sub>ice</sub> at higher temperatures are most probably in situ-origin cirrus caused by stronger updrafts in e.g. mountain waves.
  - (iv) *Drop freezing:* High  $N_{\rm ice}$  values between ten and one hundred per cm<sup>3</sup> or even more are found above 235 K. Such high concentrations together with small  $R_{\rm ice}$  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 by 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  $N_{\rm ice}$  above the core range -corresponding to convective liquid-origin cirrus- become more frequent, while  $R_{\rm ice}$  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 liquidorigin cirrus carrying a high IWC -often accompanied by high N<sub>ice</sub>- 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<sub>ice</sub> 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<sub>ice</sub>) at around 25°
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 are
generated most likely numerous observed small ice crystals are most likely generated by in situ homogeneous ice nucleation, triggered either by fast updrafts in gravity waves (see also Spichtinger and Krämer, 2013; 1)
(see also Spichtinger and Krämer, 2013; 1)
(see also Spichtinger and Krämer, 2013; 2)
(see also Spichtinger and Krämer, 2013; 3)
(see also Spichtinger and Krämer, 2013; 4)
(see also Spichtinger and Krämer, 2013; 5)
(see also Spichtinger and Krämer, 2013; 5)
(see also Spichtinger and Krämer, 2013; 5)

(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

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The mid-latitude/tropical humidity data sets include 24/72 hours of in-cloud RH<sub>ice</sub> and 51/132 hours 580 of clear sky RH<sub>ice</sub>/H<sub>2</sub>O 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<sub>ice</sub> is most often below saturation and randomly distributed in both mid-latitudes and tropics (Figure 9, middle row). Clear sky supersaturations 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<sub>ice</sub> distribution looks very different. In this region, H<sub>2</sub>O is low and its variability is only small (Figure 9, bottom row). We plotted lines of constant H<sub>2</sub>O (1.5, 3 and 5 ppmv) in the clear sky RH<sub>ice</sub> panels to illustrate that for constant H<sub>2</sub>O, RH<sub>ice</sub> increases only due to the decrease in temperature, i.e. H<sub>2</sub>O<sub>sat,ice</sub>. Thus it can be seen that at mid-latitudes RH<sub>ice</sub> mostly represent H<sub>2</sub>O 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<sub>ice</sub> ranges from 10 up to about 150% or even more (see also Jensen et al., 2017b; Krämer et al., 2009, , who already presented parts of the data show

Clear sky  $RH_{\rm ice}$  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  $RH_{\rm ice}$  rises from approximately 10% at warmer temperatures to about 20% at colder temperatures (Krämer et al.,

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Inside of cirrus, the peak of the RH<sub>ice</sub> frequencies frequency distribution is mostly around the thermodynamical equilibrium value of 100% (saturation) at mid-latitudes as well as in the tropics (Figure 9, upper row; see also Jensen et al. (2017b); Krämer et al. (2009)). 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<sub>ice</sub> concentrations, which were mostly present at low temperatures in these observations (Figure 6, SM). But, Jensen et al. (2013a) also showed that RH<sub>ice</sub> 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<sub>ice</sub> cover a broader concentration range in comparison to the earlier data (Figure 6, SM), while RH<sub>ice</sub> is supersaturated in most cases. This is not straightforward to understand due to the complex relation between RH<sub>ice</sub> and N<sub>ice</sub>. We will investigate the TTL supersaturations in cirrus clouds in a follow-up study.

The tropical tropopause layer is the region above the upper level of main convective outflow, where

# 5 TTL in situ climatologies

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

the transition from the troposphere to stratosphere occurs. It is placed at temperatures <205 K between ~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; ?; ?; Podglajen et al., 2017)(i.e. Spichtinger and Krämer, 2013; Dinh et al., 2016; Jense In addition, deep convection with fast updrafts that occasionally overshoot occasionally overshoots into the TTL.

sents 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., 2016; Ploeger 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,

Cirrus clouds and humidity in the TTL deserve a special consideration, because this region repre-

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 (note here that the ATTREX and POSIDON campaigns were in Western Pacific highly convective regions; however, the flights from these campaigns generally did not sample fresh convective outflow). 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 as part of 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,  $N_{\rm ice}$ ,  $R_{\rm ice}$  and in-cloud, clear sky  $RH_{\rm ice}$  as well as the clear sky  $H_2O$  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).

The surrounding, in most (but not all) cases typically 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 changes the frequency distributions. Thus, the Figures are representative for the TTL outside of the Asian monsoon anticyclone, so and 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.

<sup>&</sup>lt;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).

#### 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  $N_{\rm ice}$  range from very low to quite high values. We want to draw attention to a special feature of the most frequently occurring cirrus  $N_{\rm ice}$  (middle right column). Two main branches of most frequent  $N_{\rm ice}$ are found, one at very low ( $\sim 10^{-3}~\text{cm}^{-3}$ ) and the other at moderate ( $\sim 5 \cdot 10^{-2}~\text{cm}^{-3}$ )  $N_{\rm ice}$ . These two branches are also reflected in Rice, where the larger ice crystals are found together with lower concentrations and vice versa, as shown in Figure 4. More precisely, cirrus with  $N_{\rm ice} \lesssim 1.5 \cdot 10^{-2}$  $\text{cm}^{-3}$  consist of ice particles larger than about 20  $\mu\text{m}^{\,6}$  . That means that the ice crystal spectra of the low  $N_{\rm ice}$  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-higher N<sub>ice</sub> cirrus containing ice crystals smaller than 20 µm 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 homo- or heterogeneously, since both pathways might produce such  $N_{
m ice}$  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<sub>ice</sub> 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 K) are found with IWC mixing ratios of up to 1000 ppmv and a maximum N<sub>ice</sub> as high as 30 cm<sup>-3</sup> (note that the ice crystal shattering was significantly minimized, see Section A2.2). Such high values have also been observed before during the TC-4 mission 2007 in tropical anvils (Jensen et al., 2009) and even higher in lee wave cirrus during Cirrus 2006 (Krämer et al., 2009). Also, the ice crystals mean mass size, R<sub>ice</sub>, 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 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

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

705 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.

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

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The distribution of Asian monsoon cirrus clouds in the  $\Theta$  parameter space is shown in the right panel panels of Figure 11. TTL average upper and lower boundaries and the average 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 excluding the Asian monsoon measurements are shown.

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 km). The mixing ratios of these highest cirrus are about 0.05 ppmv. At about 380 K (CPT,  $\sim$  16-18 km), the range of most frequent IWCs broadens, ranging between about 0.005 and 1 ppmv. Some higher IWCs are also detected in the 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 ppmv), and at 400 K the IWC ranges between 0.05 - 0.1 ppmv, as outside of the Asian monsoon. But, at 380 K (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.

These overshooting cirrus around and below the CPT are also seen in high ice crystal numbers  $N_{\rm ice}$  ( $\gtrsim 0.5~{\rm cm}^{-3}$ ) when comparing the Asian monsoon with the other tropical regions (Figure 11 middle panels). Striking are again the high  $N_{\rm ice}$  up to  $30~{\rm cm}^{-3}$  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) took place below the CPT.

Above 390 K, at the transition to the stratosphere,  $N_{\rm ice}$  is very low, mostly lower than  $0.1~{\rm cm}^{-3}$ . Recall that these low concentrations only contain particles  $> 20~\mu m$  (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 wit 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  $N_{\rm ice}$  (and  $R_{\rm ice}$ ) – 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,  $N_{\rm ice}$  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

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#### **5.2.1** Temperature parameter space

Considering the in-cloud and clear sky RH<sub>ice</sub> 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<sub>ice</sub> are found in supersaturation supersaturated air 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<sub>ice</sub>: higher supersaturations occur frequently already at higher temperatures in the Asian monsoon comparison to the entire tropical climatology. These higher supersaturations are likely to be influenced by the stronger dynamics in the Asian mosoon, but additionally 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<sub>ice</sub> caused by the decrease of H<sub>2</sub>O<sub>sat,ice</sub> during cooling of air at a constant H<sub>2</sub>O mixing ratio. These lines correspond to H<sub>2</sub>O between 3 and 5 ppmv in the Asian monsoon, while in the total tropical climatology the most frequent H<sub>2</sub>O ranges only between 1.5 and 3 ppmv (Figure 9; compare also the bottom panels of the Figures, where H<sub>2</sub>O 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  $RH_{\rm ice}$  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),  $RH_{\rm ice}$  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  $RH_{\rm ice}$  around 90% can be seen (for more detail see Schoeberl et al., 2019).

In the Asian monsoon, on the other hand, the in-cloud RH<sub>ice</sub> in the TTL exceeds saturation (top right panel of Figure 12). Especially, beween 380 and 400 K the most frequent RH<sub>ice</sub> 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<sub>ice</sub> coincident with the Himalaya Monsoon during summer and closely associated with convection. The higher amount of water vapor RH<sub>ice</sub> in the Asian monsoon in comparison to the other tropical regions, influenced on

the one hand by the stronger dynamics as mentioned in the previous Section, can also be seen in the H<sub>2</sub>O volume mixing ratios (bottom panels of Figure 12). In the tropics outside the Asian monsoon, the most frequent H<sub>2</sub>O 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 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<sub>2</sub>O 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 region.

#### 5.3 H<sub>2</sub>O and IWC for transport to the stratosphere

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The H<sub>2</sub>O 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 discussed to be as low as H<sub>2</sub>O saturation at the minimum temperature (e.g. Schiller et al., 2009).

However, e.g. 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 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 the most frequent in-cloud and clear sky RH<sub>ice</sub> 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<sub>ice</sub> 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 hypothesize that the conclusion of Rollins et al. (2016) might be extended: 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. In convective regions the underestimation increased to 20-50% in our observations. Note however, that this hypothesize is based on a limited set of measurements and needs to be confirmed for the climatological conditions that prevail in the Asian monsoon TTL.

As mentioned earlier, the question of how much H<sub>2</sub>O 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<sub>2</sub>O and that nearly all of the convective enhancements in H<sub>2</sub>O 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<sub>2</sub>O that might enter the stratosphere. Nevertheless, from Section 5.1 we know that the most frequent IWCs between about 365-400 K are range from about 10 down 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<sub>2</sub>O, which is between 3 to 8 ppmv in this region above the CPT (see last Section). Such amounts of H<sub>2</sub>O and IWC indicate that the air 'masses originate' masses originate from an altitude of about 15 km (Figure 3), which corresponds approximately to the lower bound boundary of the TTL (see Figure 2).

The question is if remains whether 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 incloud  $RH_{ice}$  (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<sub>2</sub>O 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 by an important source for transport of H<sub>2</sub>O to the stratosphere.

# $\label{eq:optimize} 6 \quad \text{Global cirrus $N_{\rm ice}$ climatology} \\ \text{from satellite remote sensing}$

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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  $N_{\rm ice}$ , which is particularly challenging to estimate from satellite remote sensing. The new retrieval method for  $N_{\rm ice}$  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  $N_{\rm ice}$  derived from DARDAR-Nice in the same presentation as the in situ  $N_{\rm ice}$  climatology. Median  $N_{\rm ice}$ , 25/75<sup>th</sup> and 10/90<sup>th</sup> percentiles are provided for a global data set. In addition, median  $N_{\rm ice}$  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 $N_{\rm ice}$

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As a base for reliable N<sub>ice</sub> 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), in which 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<sub>ice</sub> 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  $N_{\rm ice}$  from in situ observations includes sizes > 3µm; 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 m s<sup>-1</sup>). The black solid and dotted lines of the in situ climatology indicate the 50, 25 and 75<sup>th</sup> percentiles,

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<sub>ice</sub> 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<sub>ice</sub> retrievals very well match the in situ database for known size distibutions.

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 cm<sup>-3</sup>) and in situ (straight blue line, 0.037 cm<sup>-3</sup>). 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 fed into a predefined four-parameter gamma-modified function to calculate ice particle size distributions (PSDs, see Section 2.2). N<sub>ice</sub> 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 µm, 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<sub>ice</sub> is improving improved (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  $N_{\rm ice}$  (six orders of magnitude) and other possible error sources in the measurements, both in situ and remote sensing.

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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<sub>ice</sub>) 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<sub>ice</sub> 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 to first order be reasonably captured by the 1.73 adjustment factor. Higher N<sub>ice</sub> 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 ( $D_{\rm ice} < 25\,\mu m$ ) 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  $N_{\rm ice}$  that can be detected by the in situ instruments is  $10^{-4} {\rm cm}^{-3}$ , for the highest  $N_{\rm ice}$  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  $N_{\rm ice}$  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  $N_{\rm ice}$  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<sub>ice</sub> 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 empty size 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  $N_{\rm ice}$  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  $\frac{1.30}{1.30}$  am/pm), from June 2006 to December 2016. It should still be noted that satellite retrievals are not continuously available throughout this period due to instrumental limitations and night-time retrievals are only available until 2011. This does not impact the conclusions presented here as diurnal cycles in  $N_{\rm ice}$  are not considered in this study. Such variations are also difficult to quantify from lidar/radar products, although a diurnal signature has been investigated in the DARDAR-Nice  $N_{\rm ice}$  data for tropical anvils (Wall et al., 2020). Overall, the global satellite data set consists of nearly  $2 \times 10^{10}$   $N_{\rm ice}$  retrievals. The color code represent represents 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  $N_{\rm ice}$  with decreasing temperature is visible, which is somewhat different to the in situ median  $N_{\rm ice}$  (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  $N_{\rm ice}$  of DARDAR-Nice reaches up to  $100 \, {\rm cm}^{-3}$ , while the maximum in situ  $N_{\rm ice}$  is only about  $10 \, {\rm cm}^{-3}$  (outside of young contrails).

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We attribute the The slightly increasing median N<sub>ice</sub> with decreasing temperature might be attributed 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 Such events are difficult to find by research aircraft. Hence, these events are likely and could be underrepresented in the aircraft observations.

The thinnest cirrus with low  $N_{\rm ice}$  are represented by DARDAR-Nice for temperatures  $\gtrsim 190~\rm K$ . At  $\lesssim 190~\rm K$ , 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  $N_{\rm ice}$  climatology, in the TTL ( $\lesssim 205~\rm K$ ), an increased occurrence of very thin cirrus with  $N_{\rm ice}$  around 0.001 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  $N_{\rm ice}$  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<sub>ice</sub> across all temperatures of the 10 years DARDAR-Nice climatology is about  $0.100\,\mathrm{cm^{-3}}$  (Figure 14, plain-solid 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  $0.056\,\mathrm{cm^{-3}}$  (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  $0.046\,\mathrm{cm^{-3}}$ . This is still larger than the average median of  $0.03\,\mathrm{cm^{-3}}$  reported from the N<sub>ice</sub> 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<sub>ice</sub> 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  $N_{\rm ice}$  climatology is based on a much smaller data set which is randomly distributed between different atmospheric situations. But, nearly the complete range of possible  $N_{\rm ice}$  can be detected by the in situ instruments.

For comparisons with other  $N_{\rm ice}$  data sets or results from global models, the percentiles of the adjusted DARDAR-Nice as well as the average in situ  $N_{\rm ice}$  are shown in Figure 14 and listed in Table 2

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(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 alred (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 alred (2009)

The DARDAR-Nice data set does not only provide not only provides the  $N_{\rm ice}$  distribution with temperature as shown in Figure 14, but, as it also contains the entire spatial and temporal  $N_{\rm ice}$  information, also as well as the actual appearance of cirrus clouds in the  $N_{\rm ice}$ -T parameter space. This is shown in Figure 15, where the occurrences of DARDAR-Nice  $N_{\rm ice}$  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, temperature range of global 'most frequent' median  $N_{\rm ice}$ ). This clustering of cirrus at warmer temperatures, also reported by Yang et al. (2010) from CALIPSO cloud climatologies, 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 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-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).

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 smaller warming effect of the in situ-origin cirrus which could lead to a general cooling by cirrus.

Note that an analysis of  $N_{\rm ice}$  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  $N_{\rm ice}$  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.

#### 1035 6.3 Regional cirrus N<sub>ice</sub>

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Lastly, we provide an impression of the regional variations of  $N_{\rm ice}$  (see Table 3). To this end, subsets of the DARDAR-Nice data for five regional latitude bands between 90°, 67.7°, 23.3°, -23.3°, -67.7°, -90°, corresponding to the Arctic, Mid-lat North, the Tropics, Mid-lat South and Antarctica, are considered, and three different  $N_{\rm ice}$  medians are computed for each region and for the global data set. A first median (labeled "all Temp.") corresponds to the median  $N_{\rm ice}$  value across all temperatures and considering all temperature bins to be equiprobable equally probable, 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  $N_{\rm ice}$ , considering those temperature intervals that contain 50% of the total  $N_{\rm ice}$  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  $N_{\rm ice}$ , 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  $N_{\rm ice}$  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  $N_{\rm ice}$  is  $0.031~{\rm cm^{-3}}$  (in comparison to 0.056 and  $0.046~{\rm cm^{-3}}$  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 DADAR-Nice analysis, 242 K is chosen as maximum temperature). The lowest  $N_{\rm ice}$  are found in the Arctic ('most frequent' median  $0.016~{\rm cm^{-3}}$  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 ( $0.029~{\rm cm^{-3}}$  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  $0.030~{\rm and}~0.031~{\rm cm^{-3}}$  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  $N_{\rm ice}$ , 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  $(0.074~cm^{-3})$  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  $N_{\rm ice}$  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, while also 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

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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 and mass-mean radius N<sub>ice</sub> and R<sub>ice</sub>, 96/186 hours of in-cloud and clear sky RH<sub>ice</sub> and, correspondingly, 186 hours of clear sky water vapor (H<sub>2</sub>O). The measurements span the altitude range between 5 and 20 km and cover the latitude band from 75-oNorth to 20-oNorth. The remote sensing database includes 10 years (2006-2016) of global recording of-ice crystal number concentrations N<sub>ice</sub>measurements. The main results form 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* 1090 *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 up-draft (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):

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- − Two types of most likely in situ formed cirrus are identified in slow large scale updrafts at low temperatures (T  $\lesssim$  205 K, Θ  $\gtrsim$  355 K). The first is interpreted as young cirrus (N<sub>ice</sub> around 0.1-1 cm<sup>-3</sup>  $\gtrsim$  3 μm diameter), the second as aged cirrus (N<sub>ice</sub> around 0.001 cm<sup>-3</sup>  $\gtrsim$  20 μm diameter) where the smaller ice crystals have grown to larger sizes.
  - The highest  $N_{\rm ice}$  (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  $\lesssim 20~\mu m$ ) in fast updrafts.

# TTL humidity (Figure 12):

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- In the Asian monsoon, in-cloud and clear sky RH<sub>ice</sub> is higher (often supersaturated around and above the CPT) than in the surrounding tropics. This is caused by the stronger dynamics and 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, RH<sub>ice</sub>  $\lesssim 100\%$ ) 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)

Cirrus Guide II in situ cirrus and humidity climatologies:

- $\mbox{ Median $N_{\rm ice}$, $R_{\rm ice}$ and $RH_{\rm ice}$ are presented in the IWC-T parameter space (Figure 6).} \\ Distinct patterns are found: $N_{\rm ice}$ form bands parallel to IWC, while $R_{\rm ice}$ are sorted diagonally in the IWC-T space. At tropical cold temperatures, supersaturations occur in thin as well as in thick cirrus.}$ 
  - 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.

RH<sub>ice</sub>: the overall picture of the in-cloud and clear sky RH<sub>ice</sub> distributions has also not changed, demonstrating that high altitude water vapor measurements that were under discussion earlier have improved and stabilized.

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

DARDAR-Nice satellite global cirrus N<sub>ice</sub> climatology

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A global climatology of N<sub>ice</sub> from 10 years (2006-2016) of satellite observations is provided
that can be used for comparison with global models or other data sets.

The  $N_{\rm ice}$  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<sub>ice</sub> from satellite observations is almost 2 times higher than the in situ median and increases slightly with decreasing temperature.
  - N<sub>ice</sub> medians sorted by geographical regions are highest in the tropics, followed by austral/boreal mid-latitudes, Antarctica and the Arctic.
  - In the satellite climatologies of  $N_{\rm ice}$ , half of the cirrus are located in the lowest, warmest cirrus layer and contain a significant amount of liquid-origin cirrus. Their global median  $N_{\rm ice}$  is 0.031 cm<sup>-3</sup>.
    - 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.

#### 1185 Acknowledgments

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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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