### The origin of midlatitude ice clouds and the resulting influence on their microphysical properties

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#### **Responses to Referees**

We would like to thank both referees for providing constructive comments to aid us in improving our manuscript. We have carefully considered each comment and trust that you will find our responses and revisions to be appropriate and sufficient. The comments are listed here in their entirety followed by our responses in a different font.

#### **Responses to Referee #1**

#### **General Comments:**

1. (a) First, I think that they should considerably improve the comparison of their results and methodology with that presented in previous manuscripts. Currently, they mainly compare their approach to approaches that have previously been used within their group, both in terms of comparing the calculated microphysical properties and in terms of comparing their classification mechanism against the homogeneous/heterogeneous classification mechanisms that their own group previously used. But, there are copious ice crystal data sets that have been collected in other field projects and other methodologies for classifying cirrus used by other groups that also merit comparison. For example, there are extensive papers by Heymsfield where ice cloud properties are classified according to origin (convective or stratiform) or papers where cirrus properties are presented for one specific classification of cirrus. How do the properties observed between the in-situ and liquid origin clouds compare against some of the microphysical properties that were measured in these previous experiments. Further, recent observations collected in ice clouds during the Small Particles in Cirrus (SPARTICUS) campaign have been analyzed by Muhlbauer et al. (2014) and Jackson et al. (2015), with different schemes used to classify cirrus according to their origin. How do the classification systems presented in this study compare to the classification systems that were used in those previous studies? Is there any reason why their classification system is advantageous? If so, it should be explicitly stated what these reasons are, or at the very least, the advantages and disadvantages of their new scheme discussed. I think some discussion about the relative merits of all the different classification systems is sorely needed. Some more detailed comparison with prior observations would be useful to place their study within the appropriate context.

**Response:** At the beginning of Section 2, we state that other schemes are often used, typically classifying cirrus into synoptic and convective groups. However, we see that such comparisons with external data and analysis are missing from the current

manuscript, and thank the referee for this comment. To more clearly acknowledge that this analysis component frequently appears in the literature, we have revised the text starting on page 4, line 23 (of the revised manuscript) to include citations of previous work and more thoroughly describe how our classification scheme differs from what is used elsewhere (see also text starting on page 7, line 27 of the revised manuscript).

In regard to comparisons with the microphysical properties of other studies, a brief description of an evaluation and comparison to the SPARTICUS dataset (as presented by Muhlbauer et al., 2014) to our own larger climatology has been added starting on page 14, line 17. This has already been carried out more extensively in Krämer et al. (2016, ACP), where the classification scheme and ML-CIRRUS dataset from NIXE-CAPS are first introduced. Thus, we only provide a summary of those findings. Briefly, the overall comparison of the observed properties is good, but it is important to note the types of cirrus that are represented in each dataset. Notably, there is a difference between what we would consider to be low/high IWC and what Muhlbauer et al. call low/high IWC. This stems from the difference in IWC ranges that were sampled by each group (Krämer et al.:  $0.001 \text{ mg/m}^3 - 300 \text{ mg/m}^3$ ; Muhlbauer et al.: 1  $-400 \text{ mg/m}^3$ ). The IWCs in Krämer et al. extend to lower values because observations of in situ origin cirrus in slow updrafts are contained in their dataset. Also, though it is not shown here or in Krämer et al. (2016, ACP), we have also found this to be the case if we compare our results to datasets from analyses from Heymsfield et al. for example.

2. Second, when discussing the in-situ and liquid origins of cirrus clouds, I think it would be very advantageous to discuss which of the different previously hypothesized ice nucleation mechanisms apply to each case. For example, wouldn't homogeneous nucleation be only associated with liquid origin clouds because such liquid origin particles would freeze while being lifted to cirrus temperatures. Some of the heterogeneous mechanisms that involve a transient occurrence of liquid could also be associated with liquid clouds? I'm not sure how the in-situ clouds could be forming from homogeneous nucleation? I think some more discussion in relation to nucleation

#### mechanisms could clarify some of the confusion I experienced here.

**Response:** It is very important to understand that whether heterogeneous or homogeneous ice nucleation takes place is independent of the cirrus origin type. Both processes are possible for liquid origin and in situ origin cirrus. The process that occurs depends on the dynamics within the cirrus that we sampled (i.e. cooling rates, water vapor concentrations, etc.). It is already stated in the text (see Section 2) how homogeneous and heterogeneous ice nucleation works for in-situ origin cirrus (taking place within the cirrus regime, T < 235K), and how heterogeneous and homogeneous drop freezing work for a liquid origin cirrus cloud (taking place within the mixedphase regime, where both liquid and ice are present). For a more clear distinction between these processes throughout the paper, we now refer to drop freezing in the case of liquid origin cirrus and nucleation in the case of in situ origin cirrus.

3. The third major critique of the current manuscript I have is that some of the comments are overly speculative and not fully justified according to the data that are presented in the manuscript. For example, there are a lot of comments about the role of secondary ice nucleation and secondary homogeneous ice nucleation in the later parts of the manuscript. There needs to be better justification of these comments. The manuscript should restrict itself to statements that can be definitely shown, rather than saying certain observations are likely indicators of the operation of some process.

**Response:** We respectfully disagree with this assessment of the work that we have presented. The mechanisms that we suggest as being responsible for the appearance of various features in our data are based on more than speculation. Many of the arguments that we make are established by the modeling work described in Krämer et al. (2016, ACP), where the authors have used various cirrus formation scenarios to replicate the full range of cirrus IWCs, ice crystal concentrations, etc. that are observed in nature. The observations used for their work also include the ML-CIRRUS campaign. Additionally, the cirrus origins used in our manuscript are first presented by Krämer et al. and discussed within the framework of their modeling

study. Therefore, we feel that we already have the foundation in place to make good arguments for our suggestions. However, in order to make this clearer we have added text to reference the work in Krämer et al., which this paper is a companion paper to. Furthermore, in regard to the identification of homogeneous ice nucleation events within our data, we can support our arguments with information derived from Barahona and Nenes (2009, ACP) for example, which shows that homogeneous ice nucleation can be identified where  $N_{ice}$  is greater than 0.5 cm<sup>-3</sup>.

Also, in case there is any confusion, when we refer to secondary ice nucleation, we mean cases where a second, or subsequent, homogeneous ice nucleation event takes places when ice crystals are already present. The text has been revised to refer to these events as "subsequent homogeneous ice nucleation events" in order to make this more clear.

4. Fourth, with regards to the microphysics probes it would seem that the combination of the CIP and CAS is not sufficient for detecting any larger particles if they are present. Particle reconstruction techniques would only work up to a certain extent to give concentrations of particles that are larger than the widths of the photodiode array. How were larger particles handled? What did the mass distribution functions look like? Does the absence of direct measurements of large particles affect the derivation of the microphysical quantities? In addition, some estimates of the uncertainties associated with the derived products would be beneficial.

**Response:** There are limitations to the size range that the CAS and CIP detect, but we think that it is appropriate for use in cirrus clouds, where the largest particles are often found within the CIP size range (Lawson et al., 2008; McFarquhar and Heymsfield, 1997; DeReus et al., 2008). Also, particle reconstruction was not used in the processing of the CIP data for this analysis, therefore particles larger than or partially outside of the diode array are not included in our analysis.

In the figure below, we present the mass distributions for the whole ML-CIRRUS campaign, divided into 5 K temperature bins. It is clear that the bulk and peak of the mass that has been sampled sits well within the range of the NIXE-CAPS instrument.

However, we realize that it is possible that we have missed larger particles, and therefore the IWC and modal mass diameter could be underestimated in some cases. We think that this would be more likely to occur in the case of liquid origin cirrus when the particles have originated in a mixed-phase cloud. Large particles that are outside the range of our instrumentation are not often found in in situ origin cirrus. However, an underestimation would not change the main message of the paper, as this would mean that the IWCs and modal mass diameters are even larger than we are reporting now.



Normalized masses per size bin (3-937.5 um) and T interval

Concerning the uncertainties of the measurements, we have added some information from Meyer (2012). The uncertainties associated with the particle concentration for the NIXE-CAPS sum up to a total of  $\pm 20\%$ .

5. The authors apply the mass-diameter relationship of Mitchell et al. (2010) to the results of their study. But, past studies have shown that there is a lot of variation in the mass- diameter relationships depending on the meteorological situation and the location of the measurements. How can the authors be confident that this relationship derived for tropical clouds apply to data collected in a different geographical location? How much of au uncertainty might be induced by the use of this relationship? I am also having trouble reconciling this statement with the statement on line 17 on page 34252 that the mass in each size bin is calculated using a

simplified assumption that all crystals are spheres. Assuming all particles is spheres is a huge error, so I can't understand why the mass-diameter relationships are not being used here.

**Response:** The new, extensive analysis from Erfani and Mitchell (2015, ACPD) provides observation based m-D relationships and shows that the relationship is nearly independent of cirrus type. We have compared the modified Mitchell et al. (2010) relationship that we use (and chose because this study had shown that the m-D relationship does not depend on cirrus type) to that from Erfani and Mitchell (2015) and find a good agreement (see figure below). Thus, we feel confident that the mass-dimensional calculation that we have chosen is appropriate for use with our data. This has been summarized and added to the text, page 11, lines 7 - 10.



Furthermore, we compared the PSD derived IWC with IWC derived from total water (TW) and gas phase water measurements. Unfortunately, the total water based IWC has a large uncertainty caused by the position of the sampling inlet, which resulted in an oversampling of ice crystals that was dependent on their size and the shape of the PSD. However, the ratio of median IWC values (PSD\_IWC/IWC\_TW) is 1. This is an additional argument in favor of the m-D relationship that we have chosen to use.

In regard to the use of this relationship for determining the mass in each bin, we agree that this is a more accurate method and have produced new results based on its use (see Figs. 8, 9, and 12). While this is a necessary change to make, it should be noted that the recalculation and subsequent results do not have an impact on the message of this study.

6. The authors state that they are able to use the CLaMS-ice model to classify the flight segments by origin type. I would have expected to have seen more information about the validation of the model if the model is playing such a critical role in the classification procedures being used in the paper.

**Response:** The validation and the detailed microphysical comparison between the model and in situ data has not yet been finalized and will be published in an additional manuscript. Nevertheless, we can offer a preview (but only in the responses) and say that the CLaMS-Ice simulations compare better to the observed cloud occurrence than ECMWF-only data. The advantage of the model setup in CLaMS-Ice is the Lagrangian approach combined with microphysical model along the trajectories, which results in better representations of cirrus clouds compared to the underlying meteorological ECMWF data. The amount of measured liquid origin cirrus clouds reproduced by the model is 73.2% compared to 68.7% using ECMWF data only. For in-situ origin cirrus clouds, we found an even better agreement of 84.9% (CLaMS-Ice) compared to 66.1% (ECMWF). Therefore, we are very confident that we can reproduce observed cirrus clouds from the ML-CIRRUS campaign with CLaMS-Ice and can use CLaMS-Ice to classify their origin.

7. The authors state on page 34259 that cirrus typically have small ice at the top, larger ice crystals at the bottom, and that the smallest crystals are found where nucleation is occurring. But, isn't this a contradiction to statement that for liquid origin cloud that nucleation occurs when liquid particles are ascending to some temperature? Also with regards to in-situ clouds it is stated that they form by heterogeneous and homogeneous ice nucleation whereby an air parcel rises and cools to a point at which a freezing threshold is crossed and ice crystals can form and continue to grow as conditions allow. I think the dynamical activity of cirrus and the role of sedimentation also have to be considered in order to understand the observed structure of cirrus.

**Response:** This is not a contradiction. A typical cirrus cloud sets up this way. Of course, dynamics and sedimentation play a role and the text has been revised to comment on that. The initial text represents a simplified view of the microphysical processes of cirrus, which in any case represent the basic structure of cirrus clouds. This is seen e.g. in the study from Spichtinger and Gierens (2009). Also, one of the points of showing the PSD is to demonstrate that the liquid origin cirrus have a structure different from in-situ origin cirrus, where the small particles and large particles are not sorted into the same kind of structure. The structure found in liquid origin cirrus more closely resembles that found in a mixed-phase cloud. The liquid particles are ascending and will have some kind of sorting structure depending on when we sample them.

#### **Specific Comments:**

1. Page 34250, line 7: Did the probe have anti-shatter tips?

**Response**: Yes, the tips on the CIP probe are designed to mitigate shattering.

2. Page 34250, line 14: Given the lack of sphericity of ice particles, it would appear that *Mie theory would not apply for sizing particles because this only refers to spheres.* 

**Response:** As described in Meyer (2012), the sizing is detected by applying T-Matrix calculations to the CAS probe. The differences of the sizes of spheres in comparison to particles with aspect ratios of 0.75 or even 0.5 are so small that we decided to keep the size bins derived for spheres. The figure shown below is Fig. 3.5 from Meyer (2012), and demonstrates this point.



Figure 3.5.: CAS size bins as determined by the different theories for scattering on spherical (black) and aspherical (red and blue) cloud particles (compare Figures 3.2 and 3.4). The dots indicate the middle of each bin and the vertical black lines define the bin boundaries.

3. Page 34252, line 5: Were these particles truly spherical, or was simply there not enough photodiodes shadowed to resolve their shape?

**Response:** The particles measured at this size (< 10  $\mu$ m) are detected by the CAS-DPOL probe, and therefore do not rely on the photodiodes of the CIP-Grayscale or shadow imaging techniques. As stated in the original manuscript on page 34250 line 15 – 18, the CAS-DPOL includes a detector for measuring the parallel and perpendicularly polarized components of the light scattered by the particles. This information allows us to separate spherical from aspherical particles. To answer the question, yes, the particles are spherical at that size.

4. Page 34252, Eq (2): Note that equations are not unit specific. The equation should work regardless of what units are used for different variables and will be reflected in the final units. Remove conversion terms in equation and statement that certain parameters must be in certain units.

**Response:** For convenience, we provided the units, though we are aware that the equation is not unit specific. Thus, we prefer to keep the manuscript as is.

5. Page 34253, line 10: Are any ensembles run to give any indication about the uncertainty of these trajectories. This would seem to be important given the role of the model in determining the origin of the cirrus.

**Response:** Please see the answer to General Comment 6.

6. Page 34257, line 2: Most papers present IWC in units of g/m3. I would recommend converting to these units for more easy comparison with past studies.

**Response:** The analyses from our group have typically presented IWC in ppmv (since it is independent of temperature and pressure), thus for our own consistency, we will continue to do so with this analysis. However, we agree with the referee that  $g/m^3$  is very useful for comparison with other studies. Thus, we have included the new Fig. 5, which presents the IWC for the whole ML-CIRRUS campaign in both ppmv and  $g/m^3$  to satisfy this request.

7. Page 34259: There are many other studies that have measured PSDs in addition to the few that are referenced here. I would recommend also comparing results with many of the prior other studies of cirrus PSDs to better understand the context of these new observations from CIRRUS.

**Response:** The classification of PSDs in this study by cirrus origin type is something new and has not been done before. In addition, many older measurements are affected by shattering. Thus, it is difficult to make a comparison. However, we have added text (starting on page 21, line 3 of the revised version) that compares our PSDs to those from previously published studies, including Jackson et al. (2015), Jensen et al. (2013), and Lawson et al. (2006). We agree with the referee that this is necessary and useful for framing the methods and results of our study.

#### **Response to Referee #2**

#### **General Comments:**

1. Overall terminology: The authors state in the abstract that "It has recently been proposed that there are two types of cirrus clouds – in situ and liquid origin". For decades, cirrus clouds have been separated into "in situ" and "convectively generated" categories. The authors need to state much earlier how their categories differ from the traditionally used categories. Currently, this difference isn't stated until the beginning of the second section.

**Response:** We thank the referee for pointing this out, and have revised this sentence in the abstract to read: "Recently, two types of cirrus differing by formation mechanism and microphysical properties have been classified – in situ and liquid origin cirrus." Additionally, in response to General Comment 1 from Referee #1, we have revised the text starting on page 4, line 23, to more clearly frame our classification scheme in comparison with the other schemes used more commonly in the literature.

2. Throughout the manuscript, the authors use ppmv as the unit of measure for IWC. The microphysics community generally uses grams per cubic meter. As this is a microphysics oriented publication, I'd suggest that the authors use grams per cubic meter rather than ppmv when referring to IWC values.

Response: Please see answer to Specific Comment (6) from Referee #1.

#### **Specific Comments:**

1. 34245 line 2: Re-word this sentence without "despite". Using the word 'despite' suggests that there has been controversy in this.

**Response:** We have rephrased this sentence to read, "Though difficulties and uncertainties associated with measuring and parameterizing cirrus cloud properties and the complex processes involved exist, ...".

2. Line 10: Is the 2002 Boudala paper still the most up to date on how ice crystals are represented in models?

**Response:** The aim of this sentence was to briefly highlight the intricacies of representing cirrus clouds in GCMs. In an effort to more clearly convey that message using a more recent publication, the revised manuscript now reads: "Furthermore, recent studies, such as that from Joos et al. (2014), and references therein, highlight the intricacies of representing cirrus clouds accurately in simulations and reveal that this issue leads to questions in regard to the radiative role of cirrus clouds in the present and future climate." (see page 3, lines 12 - 15).

3. 34246 line 8: The large particle mode is between 40 and 100 um? This seems small when you state in the abstract that median sizes are around 200 um with large particles up to 750 um.

**Response:** We thank the referee for pointing this typo out to us! The text should read that the modes are separated by a level area in the distribution between 40 and 100 µm. This has been corrected in the text.

4. 34248 line 1-2: This suggests that "ice only" cirrus clouds cannot form "in situ" at temperatures warmer than 235K.

**Response:** We see how this suggestion is misleading. For the purposes of our analysis, we did not include in situ origin cirrus at T > 235 K. As indicated, it is possible to get mixed phase clouds at those warmer temperatures. However, we have removed the temperature restriction suggested at this point, and instead limit it to the description of the parameters of our analysis.

5. Line 24: The authors state that "aggregation and riming are not typically seen in cirrus environments". If cirrus environments include cirrus clouds which were formed in convective systems, then aggregation and riming do exist and are very important factors in particle development.

**Response:** The cirrus environments sampled in ML-CIRRUS do not include convective systems. However, we thank the referee for pointing out that these are still important processes in cirrus clouds. Thus, we have revised the text at this point to include that aggregation and riming are important for liquid origin cirrus clouds, but mainly at higher vertical velocities (i.e. in strong convection).

6. 34251: What is the resolution of the CIP (10 microns or 15). Do you do out of focus corrections for small sizes? How much would the results differ if you used CAS PSDs out to 50 um rather than only to 20 um? Some would consider the use of the CIP for 20 micron particles to be unsatisfactory as this is where there is larger uncertainty due to depth of field and focus issues. Is Ntot calculated for all particles larger than 3 um?

**Response:** The resolution of the CIP-Grayscale is 15  $\mu$ m. At this time, we do not sort out of focus particles out of the data. However, we have looked into this further and found that no significant differences appeared when corrections were made since the cases with out of focus particles were quite rare. Furthermore, as the referee suggests, the CIP is not perfectly suited for measuring particles in the smaller sizes  $(20 - 45 \ \mu\text{m})$  due to depth of field issues. However, we chose to use the CIP instead of the CAS for this particle size range due to the issues associated with bad counting statistics from the CAS. In comparisons between the CIP and CAS, we found cases where the CAS was reporting no or too few ice crystals in this range during a cloud pass. The CIP was able to successfully detect those crystals. Using one instrument instead of the other could result in differences of up to  $\pm 20\%$  in the ice crystal concentrations in this size range. However, we feel that it would not be likely to have a significant effect overall, and would not change the message of the paper. Most of the reported ice crystal concentrations are totals, which should not be affected by this at all. Also, Ntot is calculated for all particles larger than 3  $\mu$ m (up to 937  $\mu$ m).

As a concluding remark, we would like to say that the uncertainties in measuring cloud particle concentrations in the size range of  $20 - 100 \mu m$  are a general issue for standard cloud instrumentation. This is not something that is specific to the NIXE-CAPS. At this point, we are at the limits of current measurement techniques.

7. Section 3.1.2, the authors derive IWC from the NIXE-CAPS measurements. There are numerous publications available where mass dimensional relationships are derived from "in situ" and "convective" cirrus yet the authors use one mass dimensional relationship which was derived from the TC4 project, which was for convective cirrus. There can be large differences in particle mass when comparing in situ vs convective mass dimensional relationships. The authors need to justify the use of a "convective" mass dimensional relationship for their non convective cases.

**Response:** Please see answer to General Comment (5) from Referee #1.

8. You state on 34250 line 25 that you calculate "area equivalent diameter" for CIP. Is the Mitchell et al (2010) relationship for area equivalent size? How does the Mitchell

et al (2010) mass dimensional relationship compare to Heymsfield et al (JAS 61, 982-1003) where they used PSDs and CVI data in convective and in situ cirrus separately? What is the maximum particle size considered? Cirrus, especially anvil cirrus, can have particles up to 1 cm and larger.

**Response:** The Mitchell et al. (2010) relationship is used in their study with maximum dimension. For our analysis, we have used this relationship with both maximum dimension and area equivalent sizing and found that area equivalent sizing yields more appropriate IWC values. We compared the PSD-derived IWC (from both maximum dimension and area equivalent sizing) with IWC derived from total water (TW) and gas phase water measurements. Unfortunately, the total water based IWC has a large uncertainty caused by the position of the sampling inlet, which resulted in an over- or undersampling of ice crystals that was dependent on their size and the shape of the PSD. However, the ratio of median IWC values (PSD\_IWC/IWC\_TW) is 1 when using area equivalent size. When max dimension was used, the ratio was 3. Also, the IWC values appeared unrealistically high when using maximum dimension (higher than what has been observed in North American campaigns in situations with much higher vertical velocities, such as in an MCS) given the fact that no convective systems were sampled during ML-CIRRUS.

We can only speculate that the reason we can create a working modified relationship is that the m-D relationship of Mitchell et al. is derived from data obtained with other instruments that have more pixels than the CIP-Grayscale. Thus, we believe a modification of this relationship to fit the PSD\_IWC/IWC\_TW ratio is appropriate.

In regards to a comparison between Mitchell et al. (2010) and Heymsfield et al. (2004), we draw your attention to the response to General Comment (5). The results of the Erfani and Mitchell (2015) study demonstrate that there is little influence from cirrus type on m-D relationships, which is different from what Heymsfield et al. (2004) suggest.

The largest particles we observed were 937  $\mu$ m in size, which is already noted on page 34249 line 23 of the original manuscript. Ice crystals of D > 1000  $\mu$ m might be

found in all situations. However, these do not seem to occur very frequently when looking at the largest sizes found in our study as well as other studies. Please see General Comment (4) from Referee #1.

9. Section 3.1.4: What is the reason that you chose to not use particle mass for each size bin from the mass dimensional relationship and instead assume that the ice crystals were spheres when calculating the modal mass diameter?

**Response:** We thank the referee for this comment and have reevaluated this decision. The revised version of the paper uses  $D_{ice, mode}$  and the mass dimensional relationship for calculating the mass in each size bin. We find that this is the more accurate method. However, the results do not greatly differ from the original calculations. See Figs. 8, 9, and 12 for revisions.

10. Section 3.2: ML-CIRRUS was a field campaign looking at cirrus clouds over Europe where convective systems aren't as common as over North America. It would be interesting for the authors to conduct a cirrus origin study for North American field campaigns.

**Response:** We agree with this comment completely! The comparison to MidCiX in Section 5 was a start in this direction.

11. 34256, first paragraph: Were these small ice cases near the tropopause? Tropopause cirrus in many regions contain only small ice crystals. Especially in the tropics where aviation is less likely. Aviation is likely important here, but it isn't the only source of high concentrations of small particles.

**Response:** It is very clear from the time series data in particular that these cases are contrails. As indicated, they show up at 210 K and appear in the time series as very narrow bands of very high ice crystal concentrations, at values that would be uncharacteristic for the cirrus that was sampled over Europe. Also, they often appeared embedded within a cirrus cloud and were distinctly different from the surrounding cirrus. In addition, N<sub>ice</sub> is very high in these observations, > 5 - 20 cm<sup>-3</sup>. Natural cirrus with high N<sub>ice</sub> (and small sizes) form mostly in fast updrafts, but the

 $N_{ice}$  values are > 0.5 - 5 cm<sup>-3</sup> in our observations.

12. 34260 line 2-3: The lack of evidence of homogeneous freezing could be due to the lack of observations of strong convection given that strong convection is less frequent in Europe.

**Response:** We also think that this is likely to be the case and have revised the text at this point to direct the reader to this possibility. In fact, during the HALO ACRIDICON campaign in tropical convection, we were able to observe homogeneous drop freezing.

13. 34260 line 2-3 then lines7-17: On lines 2-3 you state that no evidence of homogeneous drop freezing was found, then on lines 7-17 you describe a "strong homogeneous freezing event". "strong events" such as you describe could be more common in North American mid latitude cirrus.

**Response:** We agree with the referee that "strong events" are more likely to be observed in North America where convection is more prevalent. In regard to the lack of evidence of homogeneous drop freezing and the homogeneous ice nucleation event that was observed, it seems that it was not clear that we are discussing two different processes. Homogeneous drop freezing refers to the freezing of liquid water drops that appear initially within a mixed-phase cloud. Homogeneous ice nucleation refers to homogeneously nucleated particles at temperatures below -38 °C. As stated above, we have observed in data from the tropics, for example, evidence of liquid drops that froze into small, round crystals. This results in a high ice crystal concentration, which was not observed during ML-CIRRUS. To make it more clear for readers that we are discussing two different things, we have revised the text starting on page 6, line 21, where we first introduce these concepts. The description of homogeneous ice nucleation includes that these are frozen aerosol particles in solution. The description of homogeneous drop freezing now reads, "Homogeneous drop freezing, which is different than the homogeneous ice nucleation discussed in the previous paragraph,...". Additionally, we have revised the text throughout the manuscript to specifically say "homogeneous drop freezing" and "homogeneous ice nucleation" to

make it more clear which process we are referring to.

14. Section 6: The authors should clearly state that the results shown in Figure 12 are for the ML-CIRRUS project and may not be representative of all mid-latitudes, ie. North America and Asia where more convection is common.

**Response:** We thank the referee for bringing attention to this, and have revised the text in the conclusions to reflect this important point (starting on page 26, line 8). Also, on page 34262, line 25 of the original manuscript, it was already stated that these results only pertain to ML-CIRRUS.

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# The origin of midlatitude ice clouds and the resulting influence on their microphysical properties

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# Discussion Paper

Discussion Paper

#### Abstract

5

10

The radiative role of ice clouds in the atmosphere is known to be important, but uncertainties remain concerning the magnitude and net effects. However, through measurements of the microphysical properties of cirrus clouds, we can better characterize them, which can ultimately allow for their radiative properties to be more accurately ascertained. It has recently been proposed that there are Recently, two types of cirrus clouds – differing by formation mechanism and microphysical properties have been classified – in situ and liquid origin cirrus. In this study, we present observational evidence to show that two distinct types of cirrus do exist. Airborne, in situ measurements of cloud ice water content (IWC), ice crystal concentration ( $N_{ice}$ ), and ice crystal size from the 2014 ML-CIRRUS campaign provide cloud samples that have been divided according to their origin type. The key features that set liquid origin cirrus apart from the in situ origin cirrus are a higher frequency of high IWC (> 100 ppmv), higher  $N_{ice}$  values, and larger ice crystals. A vertical distribution of  $N_{ice}$  shows that the in situ origin cirrus clouds exhibit a median value of around 0.1 cm<sup>-3</sup>,

<sup>15</sup> while the liquid origin concentrations are slightly, but notably higher. The median sizes of the crystals contributing the most mass are less than 200 µm for in situ origin cirrus, with some of the largest crystals reaching 550 µm in size. The liquid origin cirrus, on the other hand, were observed to have median diameters greater than 200 µm, and crystals that were up to 750 µm. An examination of these characteristics in relation to each other and their relationship to temperature provides strong evidence that these differences arise from the dynamics and conditions in which the ice crystals formed. Additionally, the existence of these two groups in cirrus cloud populations may explain why a bimodal distribution in the IWC-temperature relationship has been observed. We hypothesize that the low IWC mode

### is the result of in situ origin cirrus and the high IWC mode is the result of liquid origin cirrus.

#### 25 **1** Introduction

Discussion Paper

Despite the Though difficulties and uncertainties associated with measuring and parameterizing cirrus cloud properties and the complex processes involved exist, the fact that cirrus clouds are a key component in the Earth's radiative budget is well established. Numerous studies have demonstrated the intricate details involved in putting together a complete and accurate portrayal of the radiative properties of cirrus clouds. For example, analyses have reported on the sensitivity to ice crystal sizes, shapes, and concentrations, cloud top height, optical depth, etc. and how these factors change within and between regions of the globe (e.g. Stephens et al., 1990; Jensen et al., 1994; Heymsfield and McFarguhar, 1996; Zhang et al., 1999). However, as Boudala et al. (2002) point out, models still

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- typically use parameters like fixed ice crystal sizes or other values that are inappropriate for global application of radiative calculations in Global Climate Models (GCMs). Thus, it is not surprising that questions still remain Furthermore, recent studies, such as that from Joos et al. (2014), and references therein, highlight the intricacies of representing cirrus clouds accurately in simulations and reveal that this issue leads to questions in regard to the magnitude of the radiative role of cirrus clouds in the present and future climate.
- In situ observations and subsequent analyses of cirrus microphysical properties such as ice water content (IWC), ice crystal concentration ( $N_{ice}$ ), and ice crystal size contribute to the construction of a more accurate characterization of cirrus clouds by providing values that are the basis for creating and validating parameterizations developed for GCMs. These three properties are found to vary naturally over several orders of magnitude (Luebke et al., 2013; Schiller et al., 2008; Krämer et al., 2009; Lawson et al., 2010; Heymsfield et al., 2013), therefore it is more reasonable and useful to explore them in the context of their relationship to other environmental variables (e.g. temperature). This in turn allows us to infer other information such as the mechanism of ice crystal formation and growth and go on to develop classifications of cirrus clouds based on these relationships.

An analysis of a large database of cirrus data from Luebke et al. (2013) showed that there is a bimodal frequency distribution of IWC as a function of temperature. They hypothesized that the two modes are representative of the two formation pathways of cirrus ice crystals, homogeneous and heterogeneous <u>ice</u> nucleation. Both modes

sion Paper

are observed over the complete cirrus temperature range, and the peak values of the modes increase with temperature. Furthermore, the low and high IWC modes correspond to respective N<sub>ice</sub>. A bimodality is also commonly observed in ice crystal size distributions. Studies such as Muhlbauer et al. (2014) While that study points to differences in nucleation pathways as being the key to understanding these bimodalities, studies like Muhlbauer et al. (2014) suggest that differences in larger scale dynamics are the important

factor. Their study reported that two populations of ice crystals were observed in particle size distributions (PSDs) from the Small Particles in Cirrus (SPARTICUS) campaign. They found a narrow small-particle mode and a broader large-particle mode (separated

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- by a level area in the distribution, usually between about 40 and 100 µm). However, this 10 bimodality was not consistently evident. Further, they found that subtropical and anvil cirrus types were more likely to display a bimodal PSD, while ridge-crest and frontal cirrus PSDs were more typically monomodal. ? also observed bimodal size distributions in cirrus in their ground-based remote sensing data from the Atmospheric Radiation Measurement
- (ARM) site near Lamont, Oklahoma. It was orginally suspected that the small ice crystal 15 mode seen in the airborne measurements was simply an artifact resulting from shattering of large crystals on in situ instrumentation, but they found that this was not the case. While there were inconsistencies between their  $N_{ice}$  measurements and those from airborne instrumentation, they did find that a bimodality was frequently observed in warm cirrus clouds (T > 243) but was rare at temperatures below 223 An analysis of other microphysical 20
- properties also demonstrated strong ties to the large-scale dynamics of the environment in which they were observed.

A scan of the literature surrounding cirrus clouds shows that classification schemes based on large-scale dynamics or meteorology are commonly used (e.g. Sassen , 2002; Heymsfield et al., 2002; Lynch et al., 2002; Muhlbauer et al., 2014; Jackson 25 Typically, the cirrus clouds are classified as "synoptic" or "convective", or they are classified based on more specific meteorology. However, Krämer et al. (2016) has recently proposed new definitions for a cirrus classification scheme based on the origin of the ice crystals - in situ and liquid origin cirrus clouds. The details of this scheme are further discussed

in Sect. 2. Voigt et al. (2016) followed this scheme in their study, however, in situ origin cirrus is classified as "non-LWC-origin" (LWC: liquid water content) and liquid origin cirrus is "LWC-origin".

While continuing our efforts to better understand and characterize the microphysical

- <sup>5</sup> properties of ice clouds with the concept of two modes in mind, one of our lingering questions has been in regard to the high IWC values that have been observed as a function of temperature (see Luebke et al., 2013). In the recent work of Briefly, in Krämer et al. (2016), various cirrus production and development scenarios are discussed. These scenarios are explored through extensive and detailed modeling work from a microphysics box
- <sup>10</sup> model, MAID (Model for Aerosol and Ice Dynamics), and compared to in situ observations from several airborne campaigns. However, the frequently observed high IWC values in combination with high  $N_{ice}$  are not represented in the model simulations, thus indicating that "classic" cirrus microphysics does not lead to such conditions. One feature that is not included in the MAID model is the possibility for preexisting ice. Preexisting ice means that
- <sup>15</sup> the ice crystals are formed in the mixed phase mixed-phase regime at warmer temperatures (T > 235 K), but are eventually incorporated into a cirrus cloud where they contribute to the overall microphysics. This pathway could lead to a cirrus cloud that contains many, large ice crystals and thus the high IWC values, particularly if the ice crystals first developed in an environment that allows them to grow larger.

The analysis presented here follows from Krämer et al. (2016) by using observational evidence to further explore and explain the two distinct types of cirrus proposed – in situ and liquid origin cirrus clouds. Krämer et al. (2016) used model results and a more broad campaign-case method to introduce this concept. The following study seeks to demonstrate the existence of these two cirrus cloud types by delving more deeply into how the micro-

<sup>25</sup> physical properties differ from one type to the other. Specifically, we focus on IWC, N<sub>ice</sub>, and ice crystal size. This is especially important for fully understanding cirrus clouds and how they should be properly represented in modeling scenarios as changes in microphysical properties will affect the radiative properties of cirrus clouds, both locally and globally.

# Discussion Paper

#### 2 Cirrus cloud origins

Cirrus analyses often categorize naturally occurring, non-aviation induced cirrus clouds into two groups based on the meteorology associated with their development- synoptic cirrus

- and convective or anvil cirrus. However, the recent study from Krämer et al. (2016) introduced an updated classification of these two types, which instead refers to their origin – in situ and liquid. This classification is based on (i) the formation mechanism of the cloud particles (directly as ice or frozen liquid droplets), and is therefore tied to a temperature threshold of -38°C, below which liquid water drops do not exist, and, (ii) the vertical
- velocity, which determines the thickness of the cirrus. By default, the a meteorological classification is also embedded within this classificationscheme, but with some modification. For example, parts of the cirrus clouds that develop as a result of warm conveyor belt systems as well as convective/anvil cirrus are classified as liquid origin cirrus. The reasons for doing so are explained in the following paragraphs. This is discussed at the end of this section.
- <sup>15</sup> Cirrus clouds whose ice crystals have formed and grown within an ice cloud only environment (T < 235) are referred to as in situ origin cirrus clouds. This definition is also used by Krämer et al. (2016). These clouds form via heterogeneous and homogeneous ice nucleation whereby an air parcel rises and cools to a point at which a freezing threshold is crossed (i.e. the supersaturation with respect to ice needed to initiate nucleation) is crossed,
- and ice crystals can form and continue to grow as conditions allow. The freezing threshold is determined with respect to ice nuclei in the case of heterogeneous freezing ice nucleation or with respect to the homogeneous freezing threshold. This process is simply illustrated supercooled solution particles in the case of homogeneous ice nucleation. Homogeneous ice nucleation refers to the process by which supercooled particles in solution freeze. This
- development process is illustrated simply in the schematic shown in Fig. 1, left. In situ cirrus clouds may also be observed at temperatures greater than 235 in the form of fall streaks, i.e. where large ice crystals have sedimented to lower altitudes/higher temperatures. However, this phenomena was not observed in the dataset used for this analysis.

Cirrus clouds whose ice crystals originally formed as liquid drops lower in the atmosphere (T > 235 K), which subsequently froze while being lifted into the cirrus temperature region of the atmosphere, are referred to as liquid origin cirrus clouds (Fig. 1, right). This difference is important because liquid and mixed phase mixed-phase clouds develop and are controlled by different microphysical processes, such as the mechanism described by the Köhler equation, than those found in ice-only atmospheric environments. These warmer 5 clouds exist in a regime that supplies a greater amount of water vapor for cloud particle formation and growth. Futhermore Furthermore, the population of effective cloud condensation nuclei (CCN) can result in clouds with many liquid cloud particles. Heterogeneous drop freezing will be triggered in those particles containing an insoluble ice nucleus. Homogeneous drop freezingis, which is something different from the homogeneous ice nucleation 10 of aerosol particles in solution discussed in the previous paragraph, is also possible but will only occur at -38 °C if supercooled liquid water droplets still remain. These conditions also allow for other growth mechanisms, such as aggregation and riming, that are not typically always seen in the cirrus environment. Aggregation and riming can be important processes in liquid origin cirrus clouds, but mainly at higher vertical velocities (i.e. in strong convection). 15 As shown in Fig. 1, we suggest that if the vertical motion is strong enough, any existing ice crystals or liquid droplets can also be lifted into the cirrus environment. Any ice crystals or

- frozen liquid drops observed within this space would then be identified as a cirrus cloud, regardless of their origin. Additionally, liquid origin cirrus clouds can be connected to in situ origin cirrus clouds. If the conditions allow for it (i.e. if the supersaturation reaches the homogeneous freezing ice nucleation threshold), further ice nucleation events producing small ice crystals may take place in addition to the existing, large liquid origin cirrus crystals. This cirrus origin The liquid origin cirrus type is where convective cirrus is classified. The uplift mechanisms associated with warm Warm conveyor belt cirrus and, in some cases, lee wave induced cirrus class make these alouds are class applied applied to far inclusion in the third.
- <sup>25</sup> induced cirrus, also make these clouds are also good candidates for inclusion in the this category because they can involve a lifting of clouds to T < -38 °C.

Though this classification is based on the ice crystal origin, it can also be compared to categorization based on meteorology. For example, as noted in the

Discussion Paper

introduction, Muhlbauer et al. (2014) classify observations from the SPARTICUS campaign into groups such as ridge-crest, frontal, subtropical jet stream, and anvil cirrus. Krämer et al. (2016) explain that ridge-crest cirrus is comparable to in situ cirrus in a fast updraft case, while frontal, subtropical jet stream, and anvil cirrus fit into the liquid origin category –and represent both slow and fast updraft cases. As discussed more thoroughly therein, this is further supported by observational similarities and differences between SPARTICUS and the campaigns (including ML-CIRRUS) used by Krämer et al. for their analysis.

#### 3 ML-CIRRUS 2014

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The primary dataset used for this study comes from the recent ML-CIRRUS campaign, which took place in the spring of 2014. The campaign was based out of Oberpfaffenhofen, Germany using the HALO aircraft and comprises 16 flights in total covering various locations over the European continent (?)(Voigt et al., 2016). Only 13 flights are used in the analysis presented here. The remaining three flights have been excluded from our analysis because they were aimed at sampling pure contrail/aviation induced cirrus or liquid clouds.

#### 3.1 Measurement of ice crystal properties: NIXE-CAPS particle spectrometer

#### 3.1.1 Instrument description

The instrument used in this study is a version of the Cloud Aerosol and Precipitation Spectrometer (CAPS) that was developed in 2001 to measure the properties of cloud and aerosol
 particles (Baumgardner et al., 2001). The CAPS is a combination probe that integrates two techniques for measuring the particle size distribution (PSD): the PSD of particles 0.6 to 50 µm in diameter is measured with the Cloud and Aerosol Spectrometer (CAS) using light scattered from individual particles that pass through a focused laser beam. For measurements of particles 15–93015–937 µm in diameter, the Cloud Imaging Probe (CIP), which
 utilizes the the optical array probe (OAP) technique is used. The new version of CAPS op-

utilizes the the optical array probe (OAP) technique, is used. The new version of CAPS, op-

erated by Forschungszentrum Jülich, is called NIXE-CAPS (Novel Ice eXperimEnt-CAPS) and is described in more detail by Meyer (2012).

The improved features of the new instrument are briefly described here. Both the CAS and CIP are now equipped with the "particle-by-particle" option, meaning each particle is recorded with its own time stamp. This option makes a particle interarrival time analysis, and therefore the removal of most shattered ice crystal fragments, possible (Field et al., 2006). Additionally, the CIP has been upgraded to imaging at a higher resolution with three gray-scale levels (CIP-Grayscale), which improves the discrimination of out of focus particles.

The CAS has undergone several modifications as well. Firstly, the inlet tube, which orig inally had a stepped, slight expansion, has been replaced by a completely straight tube to ensure that the velocity in the inlet equals the aircraft speed so that the particles are sampled nearly isokinetically. Secondly, the entry of the CAS inlet tube has been sharpened to a knife edge to minimize the area susceptible to shattering of ice particles. Lastly, a new detector was implemented that allows the separation of spherical from non-spherical (as pherical) shapes (CAS-DPOL). Briefly, it measures the intensity of the parallel and perpendicularly polarized components of the scattered light caused by single atmospheric particles (see Baumgardner et al., 2014, for more details).

In addition to the instrument improvements, a data processing library (NIXE-Lib) was developed for fast and precise simultaneous data analysis of the NIXE-CAPS measurements, which has been described in Meyer (2012). A flowchart of the NIXE-Lib is shown in Fig. 2, where all subsequent standard data processing procedures are displayed, including time synchronization of the measurements, velocity correction, corrections of particle counts, particle sizing ( $D_p$ : optical equivalent diameter for CAS-DPOL, area equivalent diameter for CIP-Grayscale), interarrival time analysis, and finally, calculation of the particle concentra-

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tions (d*N*: particle concentration per size bin,  $N_{tot}$ : total ambient particle concentration; the true air speed (TAS) is used for the calculations), and the PSDs (d*N*/dlog*D*<sub>p</sub>) for CAS-DPOL and CIP-Grayscale.

The sphericity classification is performed for the size range 3–50 µm by using the polarization channel of the CAS-DPOL (to be discussed further in an upcoming analysis) and for

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sizes 70–240  $\mu$ m from CIP-Grayscale measurements using the habit identification algorithm of Korolev and Sussman (2000).

As a last step, the PSDs of CAS-DPOL and CIP-Grayscale are merged into a single PSD covering the range of 0.6 to  $930937 \,\mu$ m. Henceforth, the size bins up to 20  $\mu$ m are taken

- <sup>5</sup> from the CAS-DPOL and those larger than 20  $\mu$ m from the CIP-Grayscale. This threshold is used since the CIP-Grayscale has a larger sampling volume than the CAS-DPOL, thus providing better particle sampling statistics. Particles larger than 3  $\mu$ m in diameter are classified as cloud, while the smaller particles are considered aerosols. Thus, for this analysis, particles in the size range 3 937  $\mu$ m are used. Also, as noted in Meyer (2012),
- the uncertainties associated with the particle concentration for the NIXE-CAPS sum up to a total of  $\pm$  20 %.

#### 3.1.2 IWC from NIXE-CAPS measurements

During ML-CIRRUS 2014, the IWC is was derived from the PSD information from NIXE-CAPS by integrating the particle mass in each size bin. The mass-dimension relation that
 we used for the different sizes is based on Mitchell et al. (2010) since it was developed using a good agreement between aircraft measurements (during the Tropical Composition, Cloud and Climate Coupling mission, TC4). Namely, this IWC derivation comes from PSD measurements using another type of optical array probe, 2D-S (with interarrival time correction to remove shattered particles), and simultaneous measurements with a CVI (Counterflow Virtual Impactor). The Mitchell et al. (2010) relationship is

$$m = a \cdot D^b$$

where m is ice particle mass in mg and

 $a = 0.082740, \quad b = 2.814 \quad \text{for} \quad D < 240 \,\mu\text{m}$  $a = 0.001902, \quad b = 1.802 \quad \text{for} \quad D > 240 \,\mu\text{m}$  As shown in Fig. 3, we modified the relationship for ice crystals with  $D < 240 \,\mu\text{m}$  so that

for	$D < 10\mu m$	crystals are spheres	
for	$D = 10-240\mu\text{m}$	<i>a</i> = 0.058,	b = 2.7
for	$D > 240\mu m$	a = 0.001902,	b = 1.802

<sup>5</sup> This modification is derived from an inspection of the sphericity of the ice crystals (see previous section), which shows that there are many spherical ice particles present during the campaign, especially at the smaller sizes. Also, the confidence in using such a relationship has recently been discussed in the new, extensive analysis from Erfani and Mitchell (2015) where they provide observation based m-D relationships and demonstrate that the relationship is nearly independent of cirrus type.

#### 3.1.3 $N_{ice}$ and $R_{ice}$ from NIXE-CAPS measurements

 $\mathit{N}_{ice}$  and mass mean radius ( $\mathit{R}_{ice}$ ) observations for this analysis also come from the NIXE-CAPS.  $\mathit{R}_{ice}$  in  $\mu m$  is calculated with

$$R_{\rm ice} = 1e^4 \cdot \left(\frac{1 \cdot e^{-6} \mathsf{IWC}}{N_{\rm ice}} \cdot \frac{3}{4\pi\rho}\right)^{1/3} \tag{2}$$

where IWC is in mg m<sup>-3</sup>,  $N_{\text{ice}}$  is in cm<sup>-3</sup>, and  $\rho$  is 0.92 g cm<sup>-3</sup>. Note that  $R_{\text{ice}}$  is only discussed in Sect. 4.2 and is used for consistency in a discussion that includes a figure taken directly from Krämer et al. (2016). Elsewhere in the paper, ice crystal sizes are referred to in diameter.

#### 3.1.4 Modal mass diameter

<sup>20</sup> The primary ice crystal size variable used in this analysis is modal mass diameter ( $D_{ice,mode}D_{ice,mode}$ ). This variable is calculated by considering the observed ice crystal size distribution for each time step. The mass in each size bin is calculated by making a simplified assumption that all crystals are spheres. using the modified Mitchell et al. (2010) relationship discussed in Sect. 3.1.2. Then, the bin size where the maximum amount of mass is located is determined to be the modal mass size. It is worth considering this variable in addition to the traditionally used size variables, such as  $R_{ice}$ , because we are interested in visualizing large particles and determining whether those particles are in fact related to very high IWC values.

#### 3.2 Origin classification

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In order to categorize each ML-CIRRUS flight, or flight segment when appropriate, by origin type, information from the CLaMS-Ice model was used. A detailed description of the modelcan be found in ?, including a validation study and comparison between model and

- in situ data, will take place in an additional analysis, but is briefly discussed here. The Chemical Lagrangian Model of the Stratosphere (CLaMS; McKenna et al., 2002; Konopka et al., 2007) performs a back trajectory analysis using location information from the aircraft along the flightpath (time, location) and ECMWF operational analysis data. The trajectories are performed over a time frame specified by the operator. Next, the CLaMS-Ice model is run
- <sup>15</sup> in the forward direction and uses the two-moment box-model developed by Spichtinger and Gierens (2009a) to simulate cirrus cloud development. This modeling scheme only considers those trajectories that end at T < 235 K. If a part of the trajectory existed at T > 235 K before crossing into the colder cirrus environment, then it is possible for the forward model to be initialized with preexisting ice from mixed-phase clouds, if present. Whether or not preexisting ice exists is determined by the IWC values found in the ECMWF data.

The resulting simulated clouds show a clear difference between the two origins. An example of each origin type is shown in Fig. 4. The flights from 7 and 11 April were chosen to represent in situ and liquid origin clouds, respectively. The figure illustrates the location of the aircraft in terms of the distance flown and pressure, and is marked with a solid black line to form a flight track. The simulated clouds are depicted in a curtain format using the IWC values calculated by CLaMS-Ice at each point along the flight track. Grey areas appear for T < 235T > 235 K. The liquid origin cirrus clouds (top) are found at lower altitudes (higher pressures) and exhibit a very high IWC (on the order of 100 ppmv) consistently throughout

the base of the cloud. They are easily identified identified by eye due to the bright orange colors. On the other hand, the in situ origin clouds (bottom) are found at higher altitudes. The simulations show a more cellular appearance to the cloud structure and IWC values that are lower than their liquid origin counterparts. These clouds are also observed on top or to the sides of the liquid origin cirrus, which is also illustrated by the 11 April flight in Fig. 4.

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We were able to use this information along each of the flight tracks to determine whether the flight or individual flight segments represent in situ or liquid origin cirrus. Flights and flight segments were then divided accordingly. Temperature criteria were also applied to the classification. For the in situ origin cases, only cirrus sampled at T < 235 K are considered. Clouds warmer than this temperature are likely to be influenced by mixed-phase cloud microphysics. Thus, for the liquid origin cases, the temperature range is extended to capture that influence and ice-only clouds at T < 250 K are considered. Clouds above that temperature threshold are likely to be mixed-phase (containing both ice and liquid) and were not used in this analysis. Additionally, Nice information from NIXE-CAPS was considered to aid us in determining in-cloud flight segments and for visualizing characteristics of 15 the clouds that were sampled.

The classification scheme was also validated using a different method based only on the trajectory information from the CLaMS model and without the visual cues like those shown in Fig. 4. A trajectory is classified as liquid origin if: (1) it contains ice at the beginning of the trajectory that does not dissipate before reaching the flightpath, (2) if the first valid 20 temperature of the trajectory is warmer than 235 K, and (3) if the flightpath at the time of observation is at a higher altitude than the 500 hPa level. The trajectories classified as in situ must satisfy one of the following criteria: the trajectory does not contain ice water at the beginning, or if it does, it must first appear at a temperature colder than 235 K or must evaporate before the trajectory reaches the flightpath if it began at a temperature warmer 25 than 235 K. Good agreement was observed between the classification used in this analysis and the trajectory-based scheme. This demonstrates the robustness of our classification.

Seven flights were found to contain in situ origin cirrus only and two flights contain liquid origin cirrus only. Four flights contain a combination of both origin categories and have therefore been divided into respective segments. This information is listed in Table 1.

#### 5 4 Microphysical properties of in situ and liquid origin cirrus

#### 4.1 IWC differences

As stated in the introduction, our work until this point has focused primarily on the relationship between IWC and temperature. Thus, we begin our exploration of our first impressions of the ML-CIRRUS dataset are also based on the observations of this relationship that

- were collected during this campaign. This is shown in Fig. 5 (in both ppmv and g/m<sup>3</sup>) and includes 15 flights from ML-CIRRUS (the excluded flight does not contain data from NIXE-CAPS). The most frequently observed IWC values (darker colors in Fig. 5) as a function of temperature are generally found along the "core median" fit line, which was calculated based on the larger climatological dataset found in Schiller et al. (2008). Also
- <sup>15</sup> notable are the high IWC values ( > 100 ppmv, or approximately > 0.05 g/m<sup>3</sup>) that were observed.

In comparison to another midlatitude dataset, such as SPARTICUS, the most frequent values from ML-CIRRUS appear low. The range of IWC values found in SPARTICUS are between 0.001–0.4 g/m<sup>3</sup> (Muhlbauer et al., 2014), while the values from ML-CIRRUS are found in a larger range between 10<sup>-5</sup>–0.2 g/m<sup>3</sup>. As a result, the definition of what is considered "low" and "high" IWC is different between our study and others. However, when the meteorology that was encountered during each campaign is considered, the reasons for differing IWC ranges is explained. The ML-CIRRUS dataset does not contain the higher IWC values associated with anvil cirrus, while the SPARTICUS dataset does not include observations of in situ cirrus in slow updrafts, which contributes the low IWC values ( <

 $0.001 \text{ g/m}^3$ ). This is more thoroughly discussed in Krämer et al. (2016).

To explore the differences between these two cirrus types by looking at this particular relationship, as shown (in situ and liquid origin), we also begin with IWC as a function of temperature. As seen in Fig. 6. It, it is already possible to see from this figure that our hypothesis concerning the difference in IWC magnitude between the two origins can be demonstrated. Not only are the higher IWC values sorted into the liquid origin cirrus category, but the distribution is also different. As illustrated by the distribution relative to the median line(in solid black), the most frequent IWC values (darker colors) found in liquid origin cirrus are higher than those observed in the in situ origin cirrus clouds. The next

sections take a more detailed look at how the microphysics of the two cirrus types differ, the mechanisms that can potentially explain those differences, and <u>underscoring underscore</u> that two distinct cirrus types do indeed exist.

#### 4.2 IWC, $N_{ice}$ , and $R_{ice}$

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While IWC, N<sub>ice</sub>, and R<sub>ice</sub> are often investigated individually, this analysis considers all three variables together, as shown in the two top panels of Fig. 7a and b. This representation was first used in Krämer et al. (2016)and the top, left panel of , and Fig. 7a comes directly from
their article. The plots show N<sub>ice</sub> as a function of R<sub>ice</sub> with the colors representing IWC. The black lines in the plot also denote IWC, but represent a value that is calculated using Eq. (2). Comparing the plots side by side, one of the most obvious differences is once again (as in Fig. 6) that the highest IWC values are found in the liquid origin cirrus. Also, the high IWCs occur in combination with higher N<sub>ice</sub> values than in the in situ origin cirrus, which
a key indicator of liquid origin cirrus. Additionally, the R<sub>ice</sub> values observed in the liquid origin cirrus cases begin to taper off above approximately 75 µm in radius.

Another feature that should be noted is the high  $N_{ice}$  values at small  $R_{ice}$  and low IWC values that are exhibited in the in situ origin panel. These data are likely to be the result of aviation induced cirrus (see also Krämer et al., 2016). Although they are also ice clouds, aviation induced cirrus clouds, or contrails, develop in different environmental conditions than naturally occurring cirrus clouds and display different microphysical properties as a result. This includes lower IWC values, high  $N_{\rm ice}$ , and quite small ice crystals between about 10–20  $\mu$ m in diameter. For that reason, it is more appropriate to consider and analyze this type of cirrus separately. In the study presented here, aviation induced cirrus were not filtered out due to the complexities of doing so, particularly since contrails are often embedded within naturally occurring cirrus. However, within the NIXE-CAPS ice crystal data

set, there are some instances in which we can observe a strong contrail signal occurring during flight legs around 210 K, which is the average temperature at the cruising altitude for commercial aircraft in the midlatitudes. Thus, we have an indication of which flights are more representative of aviation induced cirrus as well as how the microphysical properties of those segments appear. Any strong features resembling those found in aviation induced cirrus should be explored with some amount of caution as they may be the result of contrail samples.

From the plots in the top panels Fig. 7a and b alone it is difficult to say anything about the frequency with which these observations have occurred. For this purpose, we can look at the bottom two panels of Fig. 7c and d. The same information from the top panels Fig. 7a and b is presented regarding  $N_{\rm e}$  and  $R_{\rm e}$  occurred to compare the frequency of Fig. 7b and b is presented regarding  $N_{\rm e}$  and  $R_{\rm e}$  occurred.

<sup>15</sup> 7a and b is presented regarding N<sub>ice</sub> and R<sub>ice</sub>, except the colors represent the frequency of observation instead of IWC. However, the IWC information is not completely lost as the IWC lines provide a rough indication of the expected IWC. Here, the differences between these two cirrus types become more clear. Not only are the upper bounds of IWC and R<sub>ice</sub> reaching higher and larger values, respectively, in the liquid origin case, but the overall distribution is shifted to higher IWC, N<sub>ice</sub>, and R<sub>ice</sub> values in terms of where the highest frequency observations are occurring. For example, the most frequently observed IWC for in situ origin cirrus are 1–10 ppmv, while the most common liquid origin cirrus IWCs lie between 10 and 100 ppmv. Also, for the same N<sub>ice</sub> value, R<sub>ice</sub> values are shifted to larger sizes in the liquid origin cirrus relative to the values in the in situ origin cirrus.

#### <sup>25</sup> 4.3 IWC, $N_{\text{ice}}$ , and $\underline{\mathcal{D}_{\text{ice,mode}}}$

Another way of looking at the size of the particles is by considering the  $\frac{D_{\text{ice,mode}}}{D_{\text{ice,mode}}}$  instead of  $R_{\text{ice}}$ . Figure 8 shows the same IWC and  $N_{\text{ice}}$  as Fig. 7, but now with  $\frac{D_{\text{ice,mode}}}{D_{\text{ice,mode}}}$ 

 $D_{\text{ice.mode}}$  as the size parameter. An advantage to looking at the sizes of the crystals contributing the most mass is that the differences between the cirrus types become more clear. For example, the fact that there are more high IWC values in the case of liquid origin cirrus than in the in situ origin cirrus becomes more obvious given the abundance of the orange

- and red colors. Also, we can see that *D*<sub>ice,mode</sub> *D*<sub>ice,mode</sub> values reach approximately 550 μm in the in situ origin cirrus, but extend out to approximately 750 μm for the liquid origin cirrus. This provides a visual link between the high IWCs and large crystals. Furthermore, a relationship between *N*<sub>ice</sub> and the range of *D*<sub>ice,mode</sub> *D*<sub>ice,mode</sub> values appears in the liquid origin cirrus. As *N*<sub>ice</sub> increases, the upper bound of *D*<sub>ice,mode</sub> *D*<sub>ice,mode</sub> also increases. For
   example, at 0.01 cm<sup>-3</sup>, the largest *D*<sub>ice,mode</sub> *D*<sub>ice,mode</sub> values are around 500 μm while they
- are up to 750  $\mu$ m for concentrations of 0.5 cm<sup>-3</sup>. The relationship between size and concentration, as well as possible explanations for the PSDs in each origin type, are discussed in more detail in Sect. 4.4.2.

### 4.4 Ice crystal properties: vertical and temperature profiles of concentration and size

We have already shown that there is a variability of IWC as a function of temperature and that there are differences in this variability and the magnitude of the IWC values between origin types. Also, we have determined that there are differences in the concentrations and sizes of the ice crystals. In the following sections, we examine the ice crystals in a profile format in order to better examine these differences, as well as look for information concerning the mechanisms involved.

#### 4.4.1 Vertical profiles

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Figure 9 illustrates the vertical profiles of  $N_{ice}$  and  $D_{ice, mode}$  for in situ origin cirrus and liquid origin cirrus. Median values of each variable were calculated for 500 m altitude intervals along with the lower and upper quartiles (horizontal lines). Starting with the top panelsIn Fig. 9a and b, it is clear that the  $N_{ice}$  values in the liquid origin cirrus type are

Discussion Paper

Discussion Paper

Discussion Paper

larger than those in the in situ origin cirrus type by up to nearly an order of magnitude depending on the altitude. The ranges between the lower (LQ) and upper quartiles (UQ) also reveal another difference. This range is larger for in situ origin cirrus than for most of the liquid origin cirrus. The median values in the liquid origin are consistently greater than the midlatitude modal value of  $0.1 \text{ cm}^{-3}$ , which we use here as a guideline, whereas the in situ origin values are distributed around the modal value.

The bottom panels Figure 9c and d also demonstrate a clear difference between these origin types in terms of their  $\mathcal{D}_{\text{ice,mode}}\mathcal{D}_{\text{ice,mode}}$ . Nearly all of the median values in the in situ origin type are less than 200 µm, while the opposite is true in the liquid origin case. The range between the LQ and UQ is mostly narrower in the in situ origin cirrus compared to the

liquid origin cirrus. As for trends in  $\frac{D_{\text{ice,mode}}}{D_{\text{ice,mode}}}$  as a function of altitude, it is demonstrated there is not a clear trend for in situ origin cirrus, but  $\frac{D_{\text{ice,mode}}}{D_{\text{ice,mode}}}$  is decreasing with increasing altitude in liquid origin cirrus, which is likely a result of sedimentation.

Another piece of information that becomes clear at this point is that while there is an overlap region in regard to altitude, the liquid origin cirrus can be found at the lower end of expected cirrus altitudes, while the in situ origin cirrus are found at higher altitudes. This result is not surprising considering our hypothesized development mechanism and the indications from the CLaMS-Ice model. It makes sense that the liquid origin cirrus have strong ties to lower regions in the atmosphere.

#### 20 4.4.2 PSDs as a function of temperature

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Further inferences about the formation and evolution of the clouds in each origin type can be made based on how the overall population of ice crystals is behaving as a function of temperature. Figure 10 shows a comparison between the PSDs in 5 K temperature bins observed in liquid origin and in situ origin cirrus. For both origin cases, the general trend is that as the temperature increases, the number of small crystals decreases while the number of larger ice crystals increases, which is consistent with reports from other studies such as Boudala et al. (2002). Cirrus clouds are typically structured with small ice crystals at the top and large ice crystals at the bottom. The smallest crystals are found where nucleation is occurring. Larger crystals develop mostly through diffusional growth by water vapor and then fall to lower cloud layers and warmer temperatures as they grow. Despite the Of course, dynamics and processes like sedimentation are also important for determining the structure of a cirrus cloud (e.g. Spichtinger and Gierens, 2009b). Nevertheless, despite the fact that PSDs from both origins fit this simplified description, clear differences remain.

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The most obvious difference between the overall PSDs, is that the concentrations of both small and large crystals are greater overall in the liquid origin cirrus clouds (right panel, Fig. 10). This is consistent with the observations that have been discussed in regard to the previous figures. The other clear difference is that the PSD range in the liquid origin cirrus reaches higher ice crystal diameters ( $D_p$ ). Excluding the PSD at 210 K, which contains a smaller number of data points, the upper limit of the  $D_p$  range in the liquid origin cirrus clouds goes from 400–1000 µm as the temperature increases while the in situ origin clouds reach only 300–700 µm.

- If we consider the origin of the ice crystals, the reasons for the differences between the PSDs become more clear. For example, though the liquid origin cirrus PSDs are structured similarly to the in situ origin PSDs, they are in fact also consistent with what is observed in ice crystal PSDs from glaciated mixed phase clouds (to be demonstrated in an upcoming analysis from ?)mixed-phase clouds (to be demonstrated in an upcoming analysis). As explained in Sect. 2, the ice particles in glaciated mixed phase mixed-phase clouds stem from heterogeneous drop freezing.
- <sup>20</sup> glaciated mixed phase mixed phase clouds stem from heterogeneous drop freezing. Thus, the higher overall concentrations of cloud particles is indicative of the abundance of active ice nuclei (IN) lower in the atmosphere where the crystals first formed (see Krämer et al., 2016, for more detailed discussion). In the observations used here, we have not found evidence that homogeneous drop freezing also contributed to the devel-
- opment of the liquid origin PSDs. This, which would have resulted in even higher overall concentrations. The lack of this feature in our data is likely due to the fact that the strong convection necessary for producing such events is not typically found over Europe.

However, the result of secondary ice nucleation a subsequent homogeneous ice nucleation event (a second nucleation event after heterogeneous nucleation has already

<u>taken place</u>) can be observed. The liquid origin PSDs at 215 and 220 K both show an increased concentration of small particles around  $20 \,\mu m$ . This feature can be traced back to a strong homogeneous freezing ice nucleation event that was sampled during the flight on

- <sup>5</sup> 29 March. Figure 11 shows a time series of the PSDs observed by the NIXE-CAPS during this flight. Additional information concerning temperature and pressure as well as RH (with respect to water and ice) from the BAHAMAS and SHARC instruments, respectively, is also presented. Two passes into the homogeneous freezing-ice nucleation event were made between 16:50–17:10, one at 215 K followed by another at 220 K. High RH<sub>ice</sub> up to 150 % and
- $N_{ice}$  as high as  $5 \text{ cm}^{-3}$  were observed during the event, which are both a good indication of homogeneous freezingice nucleation (e.g. Barahona and Nenes, 2009). As evidenced by the yellows and oranges, there was an increase in the concentration of small particles at these points, which is consistent with the increased concentrations in the PSD in Fig. 10.
- It is also possible that secondary subsequent homogeneous ice nucleation contributed to the in situ origin cirrus, but such strong, visible indications are not observed in the PSDs from ML-CIRRUS. The high concentrations of the smallest crystals seen at 210 K in the in situ origin cirrus (left panel, Fig. 10) are attributable to aviation induced cirrus, not homogeneous freezingice nucleation. Overall, the lower concentrations of ice crystals in the in situ origin cirrus relative to the liquid origin cirrus are indicative that the number of available IN is lower, thus minimizing the impact of heterogenous freezing. Futhermoremight be lower
- (see Krämer et al., 2016, for more detailed discussion). Furthermore, in cases of homogeneous freezingice nucleation,  $N_{ice}$  is unlikely to be enhanced to the same degree as what what observed during the 29 March flight.

The difference in sizes between the largest crystals observed in each origin type is likely to be the result of the more desirable growth conditions found in the mixed-phase regime (i.e. more water vapor). Also, it is possible for the ice crystals to continue growing after arriving in the cirrus regime. When the air parcel is lifted already containing containing many large crystals, they will continue to grow by diffusion, if the concentration is low and the air is supersaturated, or by aggregation when the concentration is high. In comparison, in situ origin cirrus development essentially starts from the beginning. Cirrus clouds with a liquid origin have a PSD to begin with and build upon.

The classification of PSDs in this study by cirrus origin type is something that has not been done before. In addition, many older measurements are influenced by shattering artifacts. Furthermore, differences in instrumentation, data processing/analysis techniques, and the conditions in which observations were made also exist between datasets, thus, making it difficult to draw a good comparison between the PSDs presented in Fig. 10 and previous studies.

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Overall, the ice crystal concentrations in the PSDs from ML-CIRRUS are low throughout

- the sampled temperature range relative to PSDs from other midlatitude observations made by e.g. Jackson et al. (2015), Jensen et al. (2013), and Lawson et al. (2006). In the case of the Jensen et al. (2013) measurements, two PSDs that are provided in the analysis come from observations of convective outflow (typical for that dataset). In comparison to both the case of in situ and liquid origin cirrus, the concentrations from the convective case are
- higher, which is expected given that this is not a dynamic situation that was observed during ML-CIRRUS.

The observations reported in Jackson et al. (2015) concerning the SPARTICUS campaign (also found in Muhlbauer et al. (2014)) result in a similar findings, but is perhaps a more appropriate comparison since their observations have been classified as either "synoptic" or "convective". The in situ origin cirrus concentrations in our study are within

- <sup>20</sup> "synoptic" or "convective". The in situ origin cirrus concentrations in our study are within the range of the synoptic concentrations from SPARTICUS, but still consistently below the median values for all temperatures. The comparison between the liquid origin and convective cirrus shows better agreement between 219–233 K, but is again lower for the warmer temperatures. These differences could be attributed to, (i) differences in the way
- that the data was categorized, or, (ii), differences in the observed dynamics as noted earlier. The difference in categorization could mean, for example, that clouds we would classify as liquid origin (e.g. lee wave, warm conveyor belt), which have the associated high IWC and high N<sub>ice</sub>, are being classified in Jackson et al. (2015) as synoptic cirrus. Another consistent

and notable feature from the SPARTICUS data in comparison to the PSDs shown here, are the high concentrations of large particles, which was also not seen in ML-CIRRUS.

A third comparison to a dataset from Lawson et al. (2006) demonstrates an overall better comparison in regard to ice crystal concentrations than the previous two. In this case, the cirrus observations only come from synoptically generated cirrus, but could also be

- orographically enhanced. It should be noted that the very high concentrations of small particles in the Lawson et al. (2006) PSDs are suggested to be the result of shattering, and are therefore not considered in the comparison here. In the three temperature ranges (210–223 K, 224–233 K, and 234–243 K), the median concentration values in the lowest temperature range in Lawson et al. agree well with the in situ origin PSD from Fig. 10,
- while the middle and highest temperature range compare better to the liquid origin PSD. Considering the vertical distributions of in situ and liquid origin cirrus shown in Fig. 9 and that it is possible for "synoptic" to include liquid origin cirrus, this result is not surprising. In general, the comparisons that we have made demonstrate how using a formation-based classification versus the more traditional meteorology-based ones can result in differences
   expressed in the PSDs.

#### 5 Comparisons to MidCiX

Despite having a clear picture of the properties associated with the two cirrus origin types, there are questions concerning whether they are also found in other locations and regions, i.e. how cirrus produced by other meteorological situations (e.g. anvil outflow cirrus) fit in
to this classification scheme, and if the frequency with which they occur is similar. In an effort to begin exploring this idea, we have compared the results from ML-CIRRUS to the data from the Midlatitude Cirrus eXperiment (MidCiX), which took place in the spring of 2004 and was based out of Houston, Texas. Figure 12 shows the relationships between IWC, N<sub>ice</sub>, and *D*<sub>ice,mode</sub> in the same format as Fig. 8 for each campaign. The top panel shows the observations from the ML-CIRRUS campaign without any division between in situ and liquid origin cirrus. The bottom panel shows data from the MidCiX campaign.

For this campaign, the IWC values were measured by the Closed-path Laser Hygrometer (CLH) from the University of Colorado (Davis et al., 2007), while the  $N_{ice}$  and  $\mathcal{D}_{ice,mode}$  (CLH) from the University of Colorado (Davis et al., 2007), while the  $N_{ice}$  and  $\mathcal{D}_{ice,mode}$  and  $\mathcal{D}_{ice,mode}$  values come from a different CAPS instrument, but also covering a similar size range as NIXE-CAPS. It is interesting to compare these two campaigns because they are representative of different dynamics. The MidCiX campaign took place in the springtime when the large scale dynamics in the US are shifting from the winter frontal systems to the summer convective systems. As a result, this dataset is representative of cirrus stemming from jet streams, convection, and closed low pressure systems.

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It can be seen in Fig. 12 that there is a difference in IWC,  $N_{ice}$ , and  $\mathcal{D}_{ice,mode}$ ,  $\mathcal{D}_{ice,mode}$ , values. The MidCiX IWC content values are much larger overall and appear at larger  $\mathcal{D}_{ice,mode}$ ,  $\mathcal{D}_{ice,mode}$  than in ML-CIRRUS. Also, these large IWC values are observed at both low and high  $N_{ice}$ . From this comparison, we hypothesize, that conditions with more prevalent convection will lead to more liquid origin cirrus with higher IWC values. However, the very high  $N_{ice}$  values reported by the CAPS could be an overestimation caused by ice crystal shattering. This data set has not been corrected by an interarrival-time-based algorithm for such features. Instead, the concentrations of the particles in the overlapping ranges of the CAS and CIP probes incorporated into the CAPS have been adjusted to each other (see Krämer et al., 2016, for more details). However, an overestimation of  $N_{ice}$  does not change the im-

portant message conveyed by this comparison in regard to the high IWC and large  $\mathcal{D}_{\text{ice,mode}}$   $\mathcal{D}_{\text{ice,mode}}$  values, the appearance of which should be mostly unaffected by shattering.

<sup>20</sup> Unfortunately, due to the important small scale features in these dynamic systems, the CLaMs-Ice model was unable to accurately portray each MidCiX flight, and therefore, we do not currently have the same information with respect to where the appropriate divisions between in situ origin and liquid origin cirrus cases should be. Although we cannot demonstrate it in the current analysis, we suspect that in MidCiX, and other campaigns sampling from similar dynamics, the liquid origin cirrus clouds are more prevalent relative to the in situ origin cirrus clouds than what is observed in the ML-CIRRUS dataset. Further analysis and additional data, which can be found in **?**an upcoming analysis, are necessary to answer this critical question.

# Discussion Paper

Discussion Paper

#### 6 Distribution of in situ and liquid origin cirrus

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The differences between the cirrus cloud origins that have been described here offer new insights into how cirrus can be classified. To demonstrate that two groups do in fact exist within

- one campaign dataset, Fig. 13 shows the IWC-temperature relationship from ML-CIRRUS. Similar to Fig. 6, the data are presented in 5 K temperature bins and provide information on the frequency with which each variable occurs within a given temperature bin. Furthermore, the percentage by which each point is more representative of in situ origin cirrus (greens) or liquid origin cirrus (blues) is also shown here. The most frequent observations at low IWC
- <sup>10</sup> are at low temperatures and are predominantly in situ origin cirrus while the most frequent observations at warmer temperatures are predominantly liquid origin cirrus and exhibit high IWC values. There is an overlap region in the mid-range temperatures where in situ origin cirrus becomes less prevalent and liquid origin cirrus becomes increasingly dominant, but there is still a distribution around the median fit line of the IWC-T relationship. It can be argued that at T > 235 K the data will show 100 % liquid origin because we have selected for it in the data processing, but this is not true for T < 235 K.

The emergence of two distinct groups of cirrus clouds is reminiscent of the bimodal IWC distribution from Luebke et al. (2013) mentioned in the introduction, particularly since one group is more representative of low IWC, while the other is more representative high IWC. Thus, after completing this analysis, we now hypothesize that the two modes are the result of the presence of the two origin types. However, the heterogeneous and homogeneous freezing-ice nucleation mechanisms are still highly influential in driving the microphysical development of a cirrus cloud and are will be discussed further in ?future work.

Finally, classifying the data in this way could be more accurate for representing cirrus clouds in the climate system because it includes the potential for also classifying the clouds according to their radiative role. The distribution shown in Fig. 13 appears very similar to what is shown in Fig. 11 in Krämer et al. (2016). Further analysis is planned to evaluate this as well.

# Discussion Paper

Discussion Paper

#### 7 Conclusions

The analysis presented here has expanded upon Luebke et al. (2013) and Krämer et al. (2016) by showing that cirrus clouds can be divided into two groups according to the origin

- of their ice particles. Here, we have used airborne, in situ observations of IWC,  $N_{ice}$ , and ice crystal size from the 2014 ML-CIRRUS campaign to demonstrate clear differences between the microphysical properties of each origin type. Notably, we demonstrate that observations of high IWC and  $N_{ice}$  values in combination with large crystals are found in the liquid origin cirrus type. The highest frequency IWC values for in situ origin cirrus were observed to be
- <sup>10</sup> between 1–10 ppmv, while they were 10–100 ppmv in the liquid origin cirrus. The  $N_{\text{ice}}$  values appear to be similar between the origin types, but median values demonstrate that there is a difference. Using the modal  $N_{\text{ice}}$  value for midlatitude cirrus (0.1 cm<sup>-3</sup>) as a guideline, it was found that median values of  $N_{\text{ice}}$  for in situ cirrus are distributed around this value, but liquid origin cirrus clouds are above it. Similar to IWC, ice crystal size (both  $R_{\text{ice}}$  and
- <sup>15</sup>  $\mathcal{D}_{\text{ice,mode}} \mathcal{D}_{\text{ice,mode}}$  proved to also show distinct differences dependent on origin.  $\mathcal{D}_{\text{ice,mode}} \mathcal{D}_{\text{ice,mode}}$  in the in situ origin clouds had median values that were mostly less than 200 µm with the largest particles reaching sizes of 550 µm. Ice crystals in the liquid origin cirrus had median  $\mathcal{D}_{\text{ice,mode}} \mathcal{D}_{\text{ice,mode}}$  values that were larger than the 200 µm guideline and even larger crystals of nearly 750 µm.
- PSDs in 5 K temperature bins allowed a more in depth look at the details of the cloud structures based on the different populations of ice crystals and how they change with temperature. Once again, it was clear that differences exist between the concentrations and sizes of the particles. In particular, as noted throughout this analysis, the liquid origin cirrus could be characterized by higher concentrations of particles and a size range that is patientable breader and exert interval.
- noticeably broader and containing larger crystals. From this information combined with the existing knowledge concerning the details of cloud development in the cirrus environment versus lower in the atmosphere (mixed-phase regime), we could speculate on infer the mechanisms and conditions that contributed to create the PSD for each origin type. This indicates that the origin of the ice crystal matters and the influence of that origin can be

observed. Moreover, an example was given demonstrating how the PSD for a liquid origin cirrus cloud can continue to be built upon through <u>secondary subsequent</u> homogeneous ice nucleation after arriving in the cirrus regime.

- One of the uncertainties still existing within the work that is presented here is what the ratio of in situ to liquid origin clouds is on a local or even global scale. A The concept that the two different formation-based cirrus types have different microphysical properties has been demonstrated based on the observations from the midlatitude field campaign ML-CIRRUS. However, this campaign may not be representative of the midlatitudes as a whole. The cloud observations mostly took place in moderate updrafts, typical for the region over Europe that
- 10 was probed during the campaign. A comparison between the results from ML-CIRRUS and MidCiX provides evidence to suggest that convective conditions will result in higher IWCs, which gives an indication of how different dynamics in different locations can different dynamics will influence the relative frequency of occurrence of in situ versus liquid origin cirrus. Additional data and analysis are necessary to carry this conclusion further Faster
- <sup>15</sup> updrafts (e.g. convection) will result in higher IWCs and a larger influence from liquid origin cirrus, as demonstrated by the MidCiX dataset. One of the uncertainties still existing within the work that is presented here is what the ratio of in situ to liquid origin clouds is on a local or even global scale. Thus, it would be informative to also analyze additional data from locations such as North America and Asia, where the dynamics are known to be more
   <sup>20</sup> convective than what is typically observed over Europe.

The existence of these two cirrus groups also leads us to examine how we define a cirrus cloud. The major identifier of a cirrus cloud is that it is composed solely of ice. Other measurable properties may be assigned to different cloud samples to tell us more about the position, thickness, etc. of the cloud. However, as Lynch et al. (2002) suggest, subclassifications of cirrus based on their ice content would be useful. Information concerning the origin of an ice crystal and how that influences the microphyscial properties of a cirrus cloud is something that moves our understanding of cirrus in a direction that begins to provide a more clear representation of the radiative role of cirrus clouds. As stated by the 2013 IPCC report (Boucher et al., 2013), there remains a very large uncertainty in the role of ice clouds in the atmosphere. Simply put, it is unclear whether ice clouds have a warming or cooling effect on the atmosphere. Krämer et al. (2016) suggest that in situ origin cirrus clouds may have the tendency toward a cooling effect, while the thicker liquid origin clouds may tend toward warming. Future work is planned to address this topic. While these clouds will be called "cirrus" in any case, the study presented here demonstrates that a categorization scheme based on the two origins is more appropriate for describing the variety of

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**Table 1.** ML-CIRRUS flight dates and respective origin categorization. Classification as "combination" means that both in situ and liquid origin cirrus were observed. Some days contain more than one flight.

Date	Origin Category
19 Mar	In situ
21 Mar	In situ
22 Mar (1)	Liquid
22 Mar (2)	Liquid
26 Mar	In situ
27 Mar	Combination
29 Mar	Combination
1 Apr	In situ
3 Apr	In situ
7 Apr	In situ
11 Apr (1)	Combination
11 Apr (2)	Combination
13 Apr	In situ



**Figure 1.** Schematic of the basic mechanism surrounding in situ origin cirrus (left) and liquid origin cirrus (right). Each scenario illustrates the movement of air and/or cloud particles from their origin to a cirrus cloud. Left panel (in situ origin): the "freezing threshold" indicates where heterogeneous and/or homogeneous freezing ice nucleation takes place and cirrus development begins. Right panel (liquid origin): the cloud particles first form in the mixed-phase region of the atmosphere and become ice through heterogeneous or homogeneous drop freezing. After crossing the 235 K threshold, liquid water no longer exists, which indicates the boundary of the cirrus region of the atmosphere.



**Figure 2.** Flowchart of the NIXE-CAPS data processing library, NIXE-Lib. The data first undergoes time synchronization and velocty correction. It continues into various corrections of particle counts and sizing. The final steps produce a particle concentration for CAS-DPOL and CIP-Grayscale, respectively. SODA: a software program developed at the National Center for Atmospheric Research (NCAR) in Boulder, Colorado, USA. This program is embedded in the NIXE-Lib. See Meyer (2012) for more details.



**Figure 3.** *m*–*D* relationship for spheres (black) and cirrus cloud particles (blue), as in Mitchell et al. (2010), and the modified relationship for this analysis (turquoise).

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**Figure 4.** Examples of CLaMS-Ice simulations from ML-CIRRUS showing a liquid origin cloud sample (top) from the 11 April flight and an in situ origin sample (bottom) from the 7 April flight. The flightpath is illustrated by the black line and represents the pressure at which the aircraft was flying (*y* axis) and the distance since take-off (*x* axis). The colors in each plot represent the simulated IWC (orange: high IWC, blue: low IWC). Grey areas indicate T > 235 K and do not contain simulated clouds.



**Figure 5.** The frequency of IWC observations as a function of temperature for 15 flights from the ML-CIRRUS campaign. IWC is plotted in 1 K temperature bins and is show in ppmv (top) and  $g/m^3$  (bottom). The core max, median, and core min lines (black) are from Schiller et al. (2008).

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**Figure 6.** The frequency of IWC observations as a function of temperature. IWC is plotted in 1 K temperature bins for in situ origin (top) and liquid origin (bottom) data. The core max, median, and core min lines (black) are from Schiller et al. (2008).



**Figure 7.**  $N_{\text{ice}}$  as a function of  $R_{\text{ice}}$  sorted by observed IWC. The solid black lines in all panels represent IWC levels as calculated by Eq. (2). Top panels(a) and (b): The colors indicate the IWC (in ppmv) that were observed for each observed  $N_{\text{ice}}-R_{\text{ice}}$  combination. Bottom panels(c) and (d): The colors indicate the frequency of observation for each  $N_{\text{ice}}-R_{\text{ice}}$  combination. The cutoff at small  $R_{\text{ice}}$  and  $N_{\text{ice}} < 0.03 \text{ cm}^{-3}$  represents the lower  $N_{\text{ice}}$  detection limit of the CAS-DPOL when it is operated at 1 Hz. (a) is also shown in Krämer et al. (2016).

39



Figure 8. Same as the top panels of Fig. 7(a) and (b) but with  $D_{ice,mode}$  instead of  $R_{ice}$ .

40



**Figure 9.** Vertical profiles of median values of  $N_{ice}$  (topa, b) and  $\underline{\mathcal{D}_{ice,mode}}$  (bottomc, d) for in situ origin (lefta, c) and liquid origin (rightb, d) cirrus. The horizontal bars represent the range from the lower quartile to the upper quartile. The black, vertical line at  $0.1 \text{ cm}^{-3}$  in the top two panels a and b represents the modal  $N_{ice}$  observed in midlatitude cirrus. The red, vertical line at  $200 \,\mu\text{m}$  in the bottom two panels c and d was arbitrarily chosen as a reference for comparing the  $\underline{\mathcal{D}_{ice,mode}}$   $\underline{\mathcal{D}_{ice,mode}}$  values.



**Figure 10.** Particle size distributions of in situ origin cirrus (left) and liquid origin cirrus (right) for 5 K temperature bins. The temperatures listed in the key are the middle of the temperature bin.  $D_p$ : optical equivalent diameter for CAS-DPOL, area equivalent diameter for CIP-Grayscale ( $D_p > 20 \,\mu$ m).



**Figure 11.** Time series example from flight on 29 March demonstrating the observation of a homogeneous freezing ice nucleation event in a liquid origin cirrus cloud. The top panel of the figure shows the atmospheric data for the flight – time (red), pressure (green),  $RH_{ice}$  (turquoise), and  $RH_{water}$  (blue). The bottom panel shows the PSD observed by the NIXE-CAPS (diameter is on the *y* axis, time is on the *x* axis). The colors indicate the concentration of particles ( $dN/d\log D_R$  in cm<sup>-3</sup>).



**Figure 12.** Same as Fig. 8 but for all 13 ML-CIRRUS flights (both in situ and liquid origin; top) and MidCiX (bottom). The blank spaces between sizes are due to the merged bins for MidCiX.



**Figure 13.** Probability distribution of IWC as a function of temperature for ML-CIRRUS. The size of the points represent the frequency of occurrence of each value within a 5 K temperature bin, similar to the data shown in Fig. 6. The colors represent the percentage by which each point is more representative of in situ origin cirrus (greens) or liquid origin cirrus (blues). The maximum, core max, median, and core min lines (black) are from Schiller et al. (2008).