Author's final response

Review RC3 by Anonymous Referee #2

We thank Referee #2 for his suggestions to improve the quality of our work. In the following we will answer to his specific comments.

General Comment from Referee

In this study a classification scheme for stages of cirrus life-cycle is presented. The scheme is based on LIDAR data in combination with meteorological data (temperature and pressure) from ECMWF. In a case study of orographic cirrus clouds as measured during the ML-CIRRUS campaign the scheme is applied and the results are interpreted.
Generally this is an interesting and important contribution to ice cloud research; thus, this study is an appropriate contribution for AMT. However, there are some issues, which must be clarified before this manuscript can be accepted for publication. Therefore, I recommend major revisions of the manuscript. In the following I will explain my concerns in details.

Comment 1 from Referee (Major comment)

Classification scheme and interpretation of results
The general aim of the scheme is not really clear to me. I recommend that the authors give a bit more information about the aim and the possible use of the scheme.
In general, I agree with the discrimination between regions of potential ice nucleation, moderate supersaturation and subsaturation, since this reflects the different thermodynamic states of the system. However, the role of the class HET is not clear to me and seems to cause severe problems:

(a) Since heterogeneous ice nucleation is not well understood, and ice nucleation on solid particles depend on many details, a general nucleation threshold (as e. g. for homogeneous nucleation, but see minor comment below) cannot be determined. This problem is already reflected in this scheme by the use of 2 different parameterisations and their difference of about 20-30%. Therefore, the definition of the class $HET_{in/out}$ is quite arbitrary, since the lower bound is very fuzzy.

(b) For cloud free air the class might be useful, since then the possibility of heterogeneous nucleation cloud be estimated. But again the arbitrary thresholds of heterogeneous nucleation make it very difficult to use this information in a meaningful way.

(c) For cloudy data, this class might lead to severe misinterpretation of the data. In the text it is suggested that for data points of HET_{in} heterogeneous nucleation takes place or even ice crystals in this category stem from heterogeneous nucleation. This suggestion is not correct because of the problem stated in (a): The nucleation threshold is not well-posed, thus it might be that using a low threshold no heterogeneous nucleation takes place (since the IN need higher saturation); thus the interpretation of ongoing nucleation would be wrong. In the case study the lower threshold is used, but it is not clear if this is really the right one.

These problems weaken the classification scheme in a serious way; therefore I recommend either to remove the class HET_{in} completely or even to refine the representation using different heterogeneous nucleation threshold as standard. Perhaps additional information cloud be given in addition to the coarse classification HET. If the class HET is kept in the scheme, its use, benefits and problems should be described carefully.
There is another issue regarding the interpretation of the classification. The scheme is based on measurements, i.e. on an Eulerian viewpoint, since the time evolution cannot be seen. If ice crystals were found in the class HET_{in}, they are not necessarily formed by heterogeneous nucleation. The classification just can tell some information of the actual state of possible nucleation, but not about the particles, which are already in the air mass. For instance, sedimenting ice crystals could be found in the air mass, but they were formed at higher altitudes under completely different conditions. The authors should mention this problem, since confusing Lagrangian and Eulerian viewpoint could lead to completely wrong results.

Author's response

We thank Referee #2 for his comment. ML-CIRRRUS provides an extensive data set captured during 16 research flights in the mid-latitudes with both in-situ and remote sensing instruments. One scientific aim of ML-CIRRUS was to investigate optical, microphysical and radiative properties of cirrus in different stages of evolution. Our method shows that it is possible to identify life-cycle stages from remote sensing humidity data and temperature. It may provide one consistent way to set data from in-situ and remote sensing into perspective of cirrus evolution. We added corresponding phrases to the introduction.
Referee #2 is right, in pointing out the limitations and problems of the HET classification. However, it has been shown that heterogeneous freezing is one important freezing mechanism in the mid-latitudes (Cziczo et al. 2013). Therefore we prefer to keep the class HET_{in}. It adds more information to the classification and helps to characterize investigated cirrus clouds in more detail.
In Section 3 we discuss the problems associated with HET parameterizations, the simplifications that were made, and consequent limitations. In response to this review, we added another explicit mention of the fuzzy lower bound of HET regions and the possibility of sedimentation from higher levels to this discussion.
In order to prevent possible confusion about the Eulerian and Lagrangian view point and to underline the strength of our method a new paragraph was added in the end of Section 3.

Changes in manuscript

1. Introduction: … A classification scheme that reveals the spatial distribution of evolution stages would facilitate the investigation of possible dependencies on cirrus evolution.

…

The classification scheme that we present is based on atmospheric Lidar cross-sections and therefore facilitates the detailed investigation of evolution stages, their vertical and horizontal order, the impact of atmospheric dynamics, and their specific

optical properties.
…
By setting in-situ and remote sensing data in perspective to cirrus evolution it facilitates the study of the specific optical, microphysical and radiative properties of evolution stages.

3. Cirrus evolution classification scheme: … However, due to the limitations mentioned above, the border of HET towards lesser supersaturated areas must be interpreted with caution. Also ice crystals found in HET must not necessarily be formed by heterogeneous freezing, as sedimentation from higher levels featuring different nucleation conditions may take place. Still, heterogeneous freezing is an important freezing mechanism in mid-latitudes (Cziczo et al. 2013) and the class HET_{in} adds more information to the classification leading to a more complete characterization of cirrus clouds.
…
During the life-cycle of a cloud, nucleation, growth and sublimation events may occur more than once, e.g. when atmospheric dynamics cause renewed updrafts and a second freezing event on top of pre-existing ice takes place. As described, our method is able to identify nucleation, growth, sublimation regions and pre-stages of cloud formation. However on its own, it does not yield any information about earlier developments of those regions. Its very strength is to reveal the actual atmospheric state with regards to cirrus evolution at the time of measurement. This is done on a high spatial resolution that exceeds typical resolutions of GCMs, enabling the detailed study of individual cloud parts.

Comment 2 from Referee (Major comment)

Analysis of case study
The demonstration of the classification scheme was carried out using a very special case of orographic cirrus clouds. In general this is ok, but the interpretation of the case could be more specific.

(a) Probably weak sedimentation
Since the cirrus cloud was obviously formed by a (strong) wave, probably sedimentation was not a big issue, since many small ice crystals were formed. The region at the top of the cloud showing very high backscatter ratios is a hint into this direction. Maybe the authors could use the analysis of the trajectory in order to estimate the vertical velocities which might be interesting for homogeneous nucleation.

(b) Descent of the cloud
The authors claim that the descent of the ice cloud is probably triggered by large-scale downdrafts. However, this could be corroborated using ECMWF wind data, which are available; this would also strengthen the argument for the occurrence of region DEP and SUB. In addition, they should estimate sedimentation velocities of ice crystals for typical sized in order to rule out the case of sedimenting ice crystals leading to this cloud descent.

(c) High supersaturation without ice nucleation

In the measurement time 14:34-14:36 high ice supersaturation occurs (at least higher than $RH_{i,het}$) without ice nucleation. This might point to the possibility that either heterogeneous nucleation at high thresholds or even only homogeneous nucleation are the preferred nucleation types in this situation. Again, this points to the weakness of the definition of HET regions without a concise threshold for heterogeneous nucleation. What about measurement errors in relative humidity (of order of 10-15%)? Might it be possible to reach higher values of RHi?

Author's response

We thank Referee #2 for his suggestions regarding a more specific interpretation of the presented case study. We implemented them as follows.
After reviewing 48h backward trajectories of cloud masses at 8500m, 9000m and 9500m we changed the HET parameterization to coated soot, as we found no mixing with lower, dust polluted air masses. This eliminates HET_{out} regions in front of the cloud, showing that only freezing at higher relative humidity is relevant in this case. Like mentioned by Referee #2, we also deduce that homogeneous freezing might be the dominant freezing mechanism here, as no isolated HET regions exist and HOM also sets in at the cloud edge.
From the initial updraft (14:31-15:36 UTC), we estimated an average vertical velocity of 50 cms^{-1} and therefore a crystal number density of 3-10 cm^{-1} and a mean crystal radius of under 10 µm (Kärcher and Lohmann, 2002). The estimated sedimentation velocity of such small ice particles is 1.5 cms^{-1} (Gasparini et al., 2016; their Fig. 1). The descent velocity of the cloud top on the other hand is 30 cms^{-1}. This shows that sedimentation may contribute but that the main effect comes from large scale dynamics also apparent in ECMWF.
An analysis of the trajectory reveals peak vertical velocities of up to 120 cms^{-1}. That may lead to even more and smaller crystals during nucleation.
Of course, the Uncertainty in the calculated RHi may also lead to false interpretations. We added a specific discussion to Sect. 4.2.

Changes in manuscript

Please see our revised Sect. 4.2 and 4.3.

Comment 3 from Referee (Minor comment)

1. Page 2, lines 8-12: in situ vs. liquid origin ice crystals

The discrimination between these two types is based on thermodynamics

- Liquid origin: freezing of existing water droplets at water saturation
- In situ: freezing of solution droplets or heterogeneous nucleation at ice supersaturation but below water saturation

Maybe this could be mentioned in the text, Please also add the reference Wernli et al. (2016), since the classification (in situ/ liquid origin) is also used in this study.

Author’s response

We thank Referee #2 for this comment and changed the paragraph to include the reference Wernli et al. (2016) and a more detailed explanation of the classes liquid-origin and in situ clouds.

Changes in manuscript

Recently a more general classification was introduced that distinguishes the groups of ``liquid origin'' and ``in situ'' clouds that describe whether the cirrus formed by freezing of existing water droplets at water saturation, or by freezing of solution droplets or heterogeneous nucleation at ice supersaturation but below water saturation (Krämer et al., 2016; Wernli et al., 2016).

Comment 4 from Referee (Minor comment)

2. Page 2, line 19-25: vertical structure of ice clouds
The description is probably only valid for stratiform cirrus clouds, formed by in situ formation mechanisms. For liquid origin ice clouds and for clouds with dynamics (wave or instabilities) the structure might be different. This should be mentioned in the text.

Author’s response

In response to a comment from Referee #3, we changed the corresponding paragraph to specify the contributions of the cited scientists and to clarify the mentioned vertical structure of ice clouds. Possible differences in clouds influenced by certain dynamics were also added to this paragraph.

Changes in manuscript

Heymsfield (1975) first illustrated and documented the vertical and dynamical structure of ice generating cirrus uncinus clouds. This early work and following in-situ measurements (Heymsfield and Miloshevich, 1995) indicate that there is a vertical order of cirrus evolution stages with ice nucleation near cloud top level, deposition of water vapor onto ice crystals and thus particle growth in the middle, and sublimation and sedimentation at cloud base level. A more recent, statistical study by Comstock et al. (2004) evaluated an extensive data set of ground-based Lidar measurements taken at the ARM Southern Great Plains site (Oklahoma, USA) over a time period of one year. Vertical profiles of determined relative humidity with respect to ice (RH_i) inside of cirrus clouds were divided into the upper most 25 %, the middle 50 % and the lower 25 % of total cloud depth. The frequency distribution of RH_i of the upper 25 % show a considerable amount of supersaturated regions with high RH_i values up to 160 %, associated with ice nucleation. The distribution of the lower 25 % is shifted towards subsaturation with a maximum between 70 % and 80 % and values down to 10 %, clearly dominated by crystal sedimentation and sublimation. Therefore they showed that the generally accepted vertical order of evolution stages dominated the majority of measured clouds while individual clouds, depending on cloud type, generation mechanism, cloud age, and atmospheric dynamics, may show strongly differing distributions (Comstock et al., 2004; Groß et al., 2014).

Comment 5 from Referee (Minor comment)
3. Use of ECMWF data
Which kind of ECMWF data is used and how? Is there a mixture of analysis data (available every 6 hours) with short term forecasts? Please explain this in more details.

Author's response
We added corresponding details about ECMWF data usage.

Changes in manuscript
We use ECMWF analysis data (available every 6 hours), with a horizontal resolution of 0.25° and 91 vertical levels that we interpolate linearly in time and bi-linearly in space onto the Lidar measurement cross-section.

Comment 6 from Referee
4. Measurement of temperature during ML-CIRRUS
As far as I remember; during ML-CIRRUS temperature profiles were measured with the MTP instrument. Why do you not use these measurements instead of coarse resolution ECMWF data?

Author's response
Referee #2 is correct: MTP was a part of the ML-CIRRUS payload. However, the retrieval for this instrument is still under development and improvement. Therefore up to now only preliminary data is available. For that reason we opted to use ECMWF data, until quality checked MTP data is available.

Comment 7 from Referee (Minor comment)
5. Page 4, line 32-33:
A suitable reference for the occurrence of sufficient solution droplets, i.e. sufficient soluble aerosol particles would be Minikin at al. (2003).

Author's response
We thank Referee #2 for this recommendation and added the reference Minikin et al. (2003).

Comment 8 from Referee (Minor comment)
6. Page 5, lines 1-5: Representation of homogeneous nucleation
The representation of homogeneous freezing of solution droplets and the derivation of freezing thresholds is very short and misleading for non-experts; it should be expanded. The volume nucleation rate depends on water activity, i.e. $J=J(\Delta a_w)=J(RH_i,T)$ and the nucleation rate ω is composed by using the volume of a solution droplet $V = \frac{4}{3}\pi r_0^3$ with size $D=2r_0$, i.e. $\omega=JV$. Koop et al. (2000) made the (arbitrary) setting of one minute (Δt=1 min) with a probability of $P = 1-\exp(-\omega\Delta t) \approx 0.63$. However, the choice of ω is quite arbitrary and should be mentioned, while $D = 2r_0 = 0.5$ μm might be a reasonable choice of a typical size. This should be mentioned in the text.

Author's response

Upon review, we decided to construct the threshold parameterization directly from the experimentally determined value $\Delta a_w = 0.305$ for homogeneous freezing (particle size ~1-10 µm; Koop et al., 2000), as nucleation rates are of interest, primarily from the modelers point of view and are not accessible from remote sensing data. But we like to mention that "The parameterization […] is not very sensitive to this value [i.e. nucleation rate]" (Kärcher and Lohmann, 2002). We added a description of the water-activity criterion introduced by Koop et al. (2000) and of our new derivation of the freezing thresholds.

Changes in manuscript

Koop et al. (2000) found that the freezing temperatures of droplets of numerous aqueous solutions (~1-10 µm) would fall on one single solute-independent curve, when plotted in terms of water activity a_w. They suggested that this curve could be constructed by shifting the melting-point curve by Δa_w. From experiments on homogeneous freezing, they determined a shift by $\Delta a_w = 0.305$. For atmospheric applications, water activity is equal to relative humidity, when the droplet is in equilibrium with the water vapor pressure of the surrounding air (Kärcher and Lohmann, 2002). We use these findings to extract a parameterization of the temperature dependent onset humidity for HOM (see Table~1, $RH_{i,HOM}(T)$).

Comment 9 from Referee (Minor comment)

7. Page 9, line 4: Gravity waves are not really small-scale dynamics.
The statement of gravity waves as small-scale dynamics is a bit weird and should be rewritten; maybe mesoscale dynamics is a better classification, since small scale is more associated with turbulence.

Author's response

We thank Referee #2 for pointing this out and changed the respective passages.

Comment 10 from Referee (Technical comment)

1. The colour bars in almost all figures are not easy to read. Especially for figures 4 and 6, colour bars with more colours and/or clearer increments should be used.

Author's response

We added clear increments to the color bars in Fig. 4 b) and Fig. 6
For the Water vapor mixing ratio in Fig. 4 a), we prefer to keep the continuous color map, because we want to show the variability and overall distribution of humidity rather than highlight specific humidity values. For that purpose we also believe that a single hue color map is best fitted (as opposed to e.g. the jet color map).

Comment 11 from Referee (Technical comment)

2. In figure 5, the difference between regions HET and HOM cannot be seen, since the colours are too similar.

Author's response

We changed the color map for our classification.

Comment 12 from Referee: (Technical comment)
3. In figure 6 the trajectory could also be shown for clarification of the derivation.

Author's response
We thank Referee for this hint and have added the contour line of BSR=1.2 as well as the trajectory line that is the contour line shifted by 200 m.