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Cloud chamber experiments on the origin of ice crystal complexity in cirrus clouds

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Abstract

This study reports on the origin of ice crystal complexity and its influence on the angular light scattering properties of cirrus clouds. Cloud simulation experiments were conducted at the AIDA (Aerosol Interactions and Dynamics in the Atmosphere) cloud chamber of the Karlsruhe Institute of Technology (KIT). A new experimental procedure was applied to grow and sublime ice particles at defined super- and subsaturated ice conditions and for temperatures in the -40 to -60 °C range. The experiments were performed for ice clouds generated via homogeneous and heterogeneous initial nucleation. Ice crystal complexity was deduced from measurements of spatially resolved single particle light scattering patterns by the latest version of the Small Ice Detector (SID-3). It was found that a high ice crystal complexity is dominating the microphysics of the simulated clouds and the degree of this complexity is dependent on the available water vapour during the crystal growth. Indications were found that the crystal complexity is influenced by unfrozen $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ residuals in the case of homogeneous initial ice nucleation. Angular light scattering functions of the simulated ice clouds were measured by the two currently available airborne polar nephelometers; the Polar Nephelometer (PN) probe of LaMP and the Particle Habit Imaging and Polar Scattering (PHIPS-HALO) probe of KIT. The measured scattering functions are featureless and flat in the side- and backward scattering directions resulting in low asymmetry parameters g around 0.78. It was found that these functions have a rather low sensitivity to the crystal complexity for ice clouds that were grown under typical atmospheric conditions. These results have implications for the microphysical properties of cirrus clouds and for the radiative transfer through these clouds.

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1 Introduction

Cirrus clouds are situated in the cold upper troposphere at temperatures typically below -35°C , and, therefore, they are completely composed of ice particles (Guignard et al., 2012; Baran et al., 2012). With their persistent coverage of about 30 % in the mid-latitudes and up to 70 % in the tropics, these clouds are an important component of the Earth's energy balance (Guignard et al., 2012). The underlying net radiative effect is expressed by the difference between a cooling effect due to the reflection of incoming shortwave solar radiation (albedo effect) and a warming effect by the absorption of outgoing longwave terrestrial radiation (greenhouse effect). Both effects depend on the macroscopic properties of the clouds, like the ice water content (IWC) or the cloud optical thickness τ , as well as the size, shape, and crystal complexity of the constituent ice particles (Zhang et al., 1999).

The mean global net radiative effect of cirrus clouds is highly uncertain, because their solar albedo cannot easily be quantified due to the unknown scattering properties of the complex-shaped ice particles. The crystal complexity can alter the net radiative effect of a cirrus cloud (with otherwise fixed macroscopic parameters) from -40 to $+20\text{ W m}^{-2}$ (Zhang et al., 1999). Airborne in situ measurements of the microphysical properties of cirrus clouds have revealed an increasing wealth of different sizes, shapes, and crystal complexity (see e.g. Baran, 2012 for an overview). This clearly emphasizes that more specific studies are necessary on the link between the ice particle microphysical complexity and the macroscopic cloud radiative properties.

For a better assessment of the cirrus radiative effects, single particle light scattering models for different ice particle shapes and degree of crystal complexity have been developed over the last two decades (Baran, 2009). These calculations have been stimulated by the remote sensing community, which need realistic ice particle light scattering properties to improve their retrieval algorithms. The cirrus ice crystal model used in these algorithms has successively been extended in order to cover the increasing ice cloud remote sensing observations like spectral reflection, polarization and depo-

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larization. With these new observations, there is now increasing evidence that highly complex ice particles (irregular particles or crystals with rough surfaces) are necessary to get a consistent interpretation of the combined satellite observations (Baum et al., 2011; van Diedenhoven et al., 2013; Liu et al., 2014; Cole et al., 2014). The angular scattering function of such ice particles is calculated to be featureless with a flat trend at backscattering angles, so that the ice features observed in the scattering function of pristine ice particles, i.e. the 22 and 46° halos as well as the ice-bow at backscattering angles, are smoothed out. This results in relatively low asymmetry parameters, g , below 0.8 for these particles and in retrieved g values between 0.75 to 0.78 inferred from recent remote sensing polarization observations (van Diedenhoven et al., 2013; Cole et al., 2014).

In-situ measurements of the angular light scattering function in mid-latitude cirrus clouds (Gayet et al., 1998, 2004; Febvre et al., 2009) as well as Arctic mid-level ice (Lampert et al., 2009) and mixed-phase clouds (Jourdan et al., 2010) by the Polar Nephelometer (PN, Gayet et al., 1997) revealed a low asymmetry parameter in the range from 0.76 to 0.79, indicating ice particles with deeply roughened surfaces. Ulanowski et al. (2006) measured the angular light scattering function of smooth and roughened ice analogue crystals in the laboratory. They deduced very low asymmetry parameters of 0.63 for rough hexagonal rosette crystals, which differ significantly from the parameters around 0.8 recorded for smooth rosettes.

Over the last few years, an increasing number of studies have been published that apply the environmental electron microscopy technique to investigate the surface properties of growing and sublimating ice particles over a wide range of temperatures (Pfalzgraff et al., 2010; Neshyba et al., 2013; Ulanowski et al., 2014; Magee et al., 2014). The common observation of these studies is a prevalent mesoscopic roughness topography of the ice crystal surfaces on the scale of 1–20 μm . Neshyba et al. (2013) derived a roughness parametrization from their micrographs and applied a ray-tracing model to calculate the expected consequences for the angular light scattering function. They calculated a significant reduction of the asymmetry parameter of about 0.04 to 0.06 due

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to the derived surface roughness. Although these investigations indicate a prominent mesoscale surface texture of the ice crystals with the ice saturation ratio as an important driving factor, the relevance of these findings for real atmospheric ice particles are still under debate.

5 Yet, if ice crystal surface roughness is a prevalent feature of atmospheric ice particles, the reduced asymmetry parameters of these particles has significant consequences for the cloud radiative effect as it was recently shown in a climate modelling study by Yi et al. (2013). They used the National Center for Atmospheric Research
10 Community Atmosphere Model (CAM, version 5.1) and calculated the global net radiative effects of clouds consisting of smooth and severely roughened ice particles. A global-averaged difference of -1.46 W m^{-2} was calculated between the runs with smooth and roughened ice particles, indicating that ice particle surface roughness has the potential to force the energy balance in a similar magnitude as the greenhouse
15 gases but opposite in sign. However, the underlying assumption that all atmospheric ice particles have a similar degree of crystal complexity and, therefore, a similar low asymmetry parameter is highly questionable.

In the present study, the development of crystal complexity during cirrus ice particle formation and growth was investigated in controlled experiments at the cloud simulation chamber AIDA (Aerosol Interactions and Dynamics in the Atmosphere). A new experimental procedure was applied to grow and sublimate ice particles at defined super-
20 and subsaturated ice conditions and for temperatures in the -40 to -60 °C range. In these experiments the following questions were addressed:

- i. what is the role of the thermodynamic conditions prevailing during ice particle growth for the crystal complexity?
- 25 ii. does the ice nucleation type, i.e. heterogeneous or homogeneous, has an influence on the ice crystal complexity?
- iii. what is the impact of ice crystal complexity on the angular light scattering function and the asymmetry parameter g ?

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The crystal complexity was deduced from single particle measurements by the latest version of the Small Ice Detector (SID-3), which has been recently applied in an aircraft field study by Ulanowski et al. (2014).

The SID-3 method to deduce single particle complexity is described in Sect. 2.1 followed by an introduction to the AIDA cloud chamber instrumentation and the experimental procedure in Sect. 2.2. It was found that ice crystal complexity is frequently observed in the simulated ice clouds and the degree of this complexity is dependent on the available water vapour during the crystal growth. A parametrisation was developed to link the crystal complexity to the available water vapour. This parametrisation as well as the link between the angular light scattering properties and ice crystal complexity are presented in Sect. 3. The atmospheric relevance of the results is discussed in Sect. 4 and the final conclusions are drawn in Sect. 5.

2 Methods

2.1 Quantification of ice crystal complexity and surface roughness

2.1.1 Measurement of high-resolution angular light scattering patterns

To measure crystal complexity of individual small ice particles we used the Small Ice Particle Detector Mk. 3 (SID-3). Here, it has to be emphasized that the term “crystal complexity” comprises any kind of crystal distortions (surface roughness, polycrystals, aggregates, (stepped) hollowness, etc.) that results in the formation of speckles in coherent light scattering. The analysis of these spatial fluctuations in the scattered intensity of individual particles is the fundamental method of measuring single particle complexity with SID-3 (Ulanowski et al., 2014).

SID-3 is the latest version of a suite of airborne single particle cloud probes that were developed by the University of Hertfordshire, UK. The derivation of ice particle complexity from SID-3 measurements is possible, since the basic concept of the SID

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arc that corresponds to the c axis of the crystals. In the case of roughened or distorted columnar ice crystals (as shown in panels c and d of Fig. 1), speckles emerge in the angular regions outside the diffraction arcs indicating a more diffuse spatial scattering behaviour of the corresponding crystals. A description of how the scattering patterns are analysed to infer a measure of the crystal complexity is given in Sect. 2.1.2. Details on the experimental procedures to grow ice crystals with different degrees of complexity are presented in Sect. 2.2.2.

In addition to the 2-D scattering patterns, the triggering system of SID-3 stores scattering intensity data of all particles that have passed the sensing area of the instrument, i.e. also for those particles that were detected while the ICCD was still busy with the image processing of a previous particle. The maximum acquisition rate for trigger intensity data is 11 kHz. This feature allows us to (i) calibrate the trigger detector by a set of scattering patterns from water droplets and (ii) generate statistically well-defined particle size distributions based on the trigger intensity measurements. In the case of SID-3, the trigger intensity measurements are performed under a relatively narrow solid angle around the 50° direction with respect to the forward direction. Therefore, the measurement is biased by the particle orientation in the case of aspherical ice particles. For this reason, the trigger intensity measurements from the Particle Phase Discriminator (PPD-2K), the laboratory version of SID-3, was used in the present study to determine the particle size distribution of the chamber ice clouds. PPD-2K measures the light scattering intensity of each individual particle over the annulus-shaped main detector aperture which is less biased by particle orientation. In addition, PPD-2K has a specifically designed inlet nozzle that focusses the sampled particles to the sensitive area of the instrument which results in better counting statistics compared to SID-3. Details on the PPD-2K and the size calibration procedure can also be found in Vochezer et al. (2015).

2.1.2 Scattering pattern analysis

For the analysis of the 2-D scattering patterns, a comprehensive image processing and analysis software was developed using the Vision Development Module of the LabView™ (National Instruments Inc., USA) package. A detailed description of the main analysis steps per scattering pattern can be found in Vochezer et al. (2015). These steps comprise

- the computation of the polar integrated azimuthal intensity profile for droplet/ice discrimination
- a Mie fit procedure of the azimuthally integrated polar intensity profile in the case of spherical particles to determine the droplet diameter
- a discrete fast Fourier transformation of the polar intensity profile for the shape classification of ice particles
- a speckle pattern texture analysis to deduce ice crystal complexity.

While the first three steps are explained in detail in Vochezer et al. (2015), the final speckle pattern analysis step is described in the following.

The crystal complexity analysis relies on the grey-level co-occurrence matrix (GLCM) method described in Lu et al. (2006). This method was originally developed in the context of quality control of surface treatment processes, and Ulanowski et al. (2010) transferred it for the first time to the analysis of SID-3 ice crystal scattering patterns. The GLCM method was further applied in a first study on ice crystal complexity of simulated cirrus clouds generated in the AIDA cloud chamber (Schnaiter et al., 2011) and in the analysis of SID-3 data of atmospheric ice particles by Ulanowski et al. (2014).

The GLCM describes how often pairs of grey-levels occur in the texture image for pixels separated by a certain distance and along a certain direction (Haralick et al., 1973). Speckle pattern texture features can then be extracted from the GLCM. Routinely, the five features contrast, correlation, energy, entropy, and homogeneity are computed (see

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Haralick et al., 1973 for details). In addition to these texture features, a “combined roughness measure” as defined by Ulanowski et al. (2014) as well as the normalized energy feature parameter k_e are computed by the image processing software. The latter was introduced by Lu et al. (2006). They found that the energy feature parameter k_e has “... a good relationship with the surface roughness; is more robust to the variations of the setup configuration, the position, and the orientation of the surface to be measured; and is the best feature parameter to characterize the surface roughness”. The robustness of k_e for the SID-3 optical system was also investigated in this work by using analogue ice particles residing on glass plates as test objects (see Appendix A for details). Further, Discrete Dipole Approximation (DDA) light scattering calculations of deformed spherical ice particles were conducted for different deformation degrees in order to determine a relationship between the k_e value and the particle surface modifications. Details of this modelling work can be found in Appendix B. In summary, these tests show that (i) k_e is the most robust feature parameter to characterise crystal complexity with SID-3 and (ii) there is a correlation between the optical feature k_e and the physical surface distortions. The analysis and interpretation of the SID-3 data therefore rely on k_e as the primary measure for ice crystal complexity.

For this work the above listed scattering pattern analysis steps were restricted to images within a narrow mean brightness (grey level) range between 10 and 25. In this way we could minimize remaining image brightness biases on the GLCM analysis results. For this brightness range the relative standard deviation in k_e is only 0.7 % and the fraction of saturated pixels per pattern is always below 1 % even for the brightest images (see Appendix A).

2.2 Cloud chamber simulation experiments

The ice crystal growth experiments were conducted in the aerosol and cloud simulation chamber AIDA (Aerosol Interactions and Dynamics in the Atmosphere) of the Karlsruhe Institute of Technology. AIDA can be operated as an expansion chamber to simulate the atmospheric conditions in ascending cloud parcels in the temperature

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Ice particle replica were generated on 35 mm transparent plastic film strips that were precoated with polyvinyl formal (Formvar). Precoating was produced at room temperature by brushing one side of the plastic film with a solution of 5% Formvar in chloroform which results in a Formvar coating of about 100 μm . For the operation at the AIDA chamber the replicator was modified by a solvent dispensing system composed of $\sim 3\text{m}$ teflon tubing and a syringe pump located outside the insulating housing of the chamber. In this way the softening of the Formvar coating by dispensing dichloromethane to the strip could be externally controlled. We used dichloromethane to soften the coating prior to the impaction of ice particles because of its low freezing point of $-95.1\text{ }^\circ\text{C}$ (Takahashi and Fukuta, 1988).

The airborne instruments PN, PHIPS-HALO, and SID-3 were located underneath the chamber within the temperature controlled environment of AIDA. There, the instruments were installed into specific vacuum sealed canisters in a strictly vertical orientation. These canisters have a 10mm inner diameter stainless steel sampling tube that penetrates the chamber wall and projects into the chamber volume by about 0.2m. Each sampling tube is equipped with a horn-shaped inlet to minimize sampling artefacts. Within the instrument canister, the sampling tube terminates about 10mm in front of the sensing area of the instrument. Each canister is connected via a mass flow controller to the central vacuum system of the AIDA facility. By applying a mass flow of typically 50 standard liters per minute ($\text{standard L min}^{-1}$) cloud particles from the chamber volume can be sampled, accelerated to a speed of about 10ms^{-1} in the sampling tube and measured when exiting the tube close to the sensing area of the instrument. The ice particle replicator was installed in a similar canister as the other airborne instruments but was aspirated by only $20\text{ standard L min}^{-1}$ in order to get a reasonable separation of the ice particle replica on the strip.

The PPD-2K has a vacuum sealed detection cell, so there is no need for installing the instrument in a separate canister. The detection cell is equipped with a focusing particle nozzle that produces a confined particle beam with a cross section of 2.5mm^2 . This cross section is small enough to completely fit into the flat rectangular laser beam cross

section of the instrument. In this way, the forward scattering intensity of all sampled particles is detected and analysed in terms of the equivalent optical particle diameter. Further details on the instrument design and the calibration procedure can be found in Vochezer et al. (2015).

5 Three different aerosol types were used in the cloud simulation experiments to initiate the the ice formation: soot particles from a graphite spark generator (GFG 1000, PALAS) (Möhler et al., 2005b), Arizona Test Dust (ATD) particles that were re-dispersed by a rotating brush generator (RGB 1000, PALAS) in conjunction with impaction stages to size select the $< 2 \mu\text{m}$ particles (Möhler et al., 2006) as well as aqueous sulfuric acid
10 particles generated in a home-built $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ nucleation generator (Wagner et al., 2008).

Aerosol number concentrations prior to and during the cloud expansion runs were measured with a condensation particle counter (CPC3010, TSI). Several modifications have been applied to the CPC in order to ensure reliable operation during the cloud expansion runs, i.e. at differential pressures between the sampling line and the environment of more than 500 hPa (Seifert et al., 2004). The aerosol number size distribution was measured with a scanning mobility particle sizer (SMPS, TSI) and an aerodynamic particle sizer (APS, TSI) (Möhler et al., 2006).

20 The presence of cloud particles inside the chamber is monitored by the laser light scattering and depolarization instrument SIMONE (Schnaiter et al., 2012). SIMONE measures scattered light from particles residing in the center of the chamber from near-forward (2°) and near-backward (178°) directions. The backscattered light is analysed with respect to its polarization state which facilitates the derivation of the linear depolarization ratio δ_1 . Although SIMONE is routinely used to detect phase-transitions
25 in aerosol and cloud particle ensembles, it has recently also been used to investigate the microphysical properties of corona-producing ice clouds (Järvinen et al., 2014).

Water vapour measurements are conducted in two ways: in-situ measurements by using a tunable diode laser (TDL) spectrometer (Ebert et al., 2005) to measure the interstitial water vapour mass and an extractive measurement for measuring the total

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(interstitial plus condensed) water mass. For the latter, we used a fast chilled-mirror frost-point hygrometer (MBW, model 373) that is located outside the insulating housing of AIDA and that is connected via a heated stainless steel tube to the chamber volume. Further details on the water vapour measurements at AIDA can be found in Wagner et al. (2010) and Fahey et al. (2014).

2.2.2 Growth and sublimation cycles

A novel experimental procedure for the AIDA cloud chamber was developed in order to grow and sublimate ice particles at defined super- and subsaturated ice conditions. This procedure consists of the following main steps that are applied to the pre-cooled chamber. In the present work the experiments were conducted at initial chamber temperatures of -40 and -50°C .

1. Preparation: The chamber volume was cleaned by evacuation and flushing cycles. Then water vapour was added in excess to the evacuated cold chamber so that a thin ice coating was formed on the inner chamber wall. This humidification step was followed by refilling the chamber with dry synthetic air until a slight overpressure of about 1 mbar was reached. Homogeneous temperature and humidity conditions throughout the volume were ensured by operating the mixing ventilator of the chamber. After these preparation steps, a saturation ratio with respect to ice S_{ice} of about 0.95 and a background particle number concentration of $\sim 0.1 \text{ cm}^{-3}$ were typically measured.
2. Aerosol addition: aerosol particles were generated by the respective aerosol generator (Sect. 2.2.1) and the aerosol flow was directed to the AIDA chamber. The AIDA particle number concentration was continuously monitored and the aerosol addition was stopped after the desired concentration had been reached. The aerosol size distribution was measured after a homogenization period of several minutes.

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perature. Consequently, the ice saturation ratio S_{ice} changed from supersaturated to subsaturated conditions due to the addition of dry air and the gas temperature increase (panel b of Fig. 2). By controlling the compression air flow, a defined saturation ratio can be adjusted. In the experiment shown in Fig. 2, the saturation ratio was set to about 0.8. As soon as the compression was applied, the ice particles started to sublime as indicated by the decrease in the ice particle size (panel c of Fig. 2). The ice particles were forced to sublime for a period long enough to see a clear change of their shape and surface properties, but short enough not to completely sublime them. A sufficient particle surface processing by sublimation is indicated by the observation of oval SID-3 scattering patterns as shown in Fig. 3.

5. Regrowth period: After the ice particle processing by sublimation had been completed, the regrowth of the ice particles was started. For this purpose, the addition of compressing air was stopped followed by a controlled expansion of the chamber volume (e.g. at 580s experiment time in Fig. 2). Since the basic idea of this experimental procedure was to control the ice particle grow speed, the regrowth should happen at a specific but nearly constant supersaturation (e.g. about 1.05 in case of the experiment shown in Fig. 2). For this purpose, the expansion was manually controlled by tuning the suction flow, while monitoring the saturation ratio measurement. Depending on the target supersaturation, suction flow tuning was applied either by using a mass flow controller (range up to $\sim 100\text{ m}^3\text{ h}^{-1}$) or by changing the suction capacity of the pump. In this way, the regrowth of the ice particles was conducted at a nearly constant supersaturation over periods of at least 200s (indicated by the shaded areas in Fig. 2). Several regrowth periods at different supersaturations can be performed within one ice cloud by repeating steps (4) and (5), provided that the ice particle number concentration, which is decreasing by sedimentation and sampling losses, is still above $\sim 5\text{ cm}^{-3}$.

3 Results

For the present work, a total of 45 sublimation and regrowth experiments have been selected from the AIDA campaigns HALO06 (02/2011), RICE01 (11/2012), RICE02 (05/2013), and RICE03 (12/2014).

As the water vapour mixing ratio is frequently measured inside cirrus ice clouds, the basic idea of the regrowth experiments was to study the dependence of the ice particle complexity on the ice saturation ratio during the growth of the crystals. Therefore, the start and end times for each regrowth period were manually determined by examining the time curve of S_{ice} (blue line in panel b of Fig. 2) and identifying those periods where S_{ice} showed a plateau (as indicated by the shaded areas in Fig. 2). The results of the single particle scattering pattern analysis were then statistically analysed for these regrowth periods. As already mentioned, only scattering patterns within a narrow mean brightness (grey level) range from 10 to 25 were considered as valid patterns in this statistical analysis. Periods that had less than 100 valid scattering patterns were excluded from the study. For the same time periods, S_{ice} was averaged to determine the mean ice saturation ratio, to which the ice crystals had been exposed to during their regrowth.

The result of this analysis is shown in Fig. 4. Experiments that were started at initial temperatures of -40 and -50°C are given as black and grey symbols, respectively. Note that the one sigma error bars given for S_{ice} represent the variations that are due to the manual control of the pumping speed in order to keep a constant saturation ratio. The median complexity parameter k_e clearly increases with increasing supersaturation in the case of the -50°C experiments, and this trend is even steeper for the experiments that were conducted at an initial temperature of -40°C . As for a given supersaturation the available condensable water mass is increasing with increasing temperature, the ice particles will grow faster at warmer temperatures and, consequently, more defects in the crystal lattice are expected. Ice particle habits and growth rates were measured in the laboratory by Bailey and Hallett (2004) for ice particles grown

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on thin glass wires over a broad temperature range from -20 to -70 °C. They clearly observed faster growth rates with higher supersaturations and warmer temperatures. At -50 °C and for supersaturations of 20 and 40 %, growth rates of about 0.035 and $0.07 \mu\text{m s}^{-1}$ were respectively measured for columnar ice particles. These results are in good agreement with the results of our own growth rate measurements by analysing PHIPS-HALO images (see Appendix C). At -40 °C and for the same supersaturation range, Bailey and Hallett (2004) measured growth rates that were higher by a factor of at least two compared to the -50 °C results. The authors also report an increase in crystal complexity with increasing supersaturation, mainly represented by the development of hollowness and the formation of rosettes. At -50 °C these growth instabilities started to appear at an ice supersaturation around 20 % which is in good agreement with the ice crystal habits that were found on the replicator strips (Fig. 5). At -40 °C a significant crystal complexity was already observed at supersaturations around 10 %.

3.1 Correlation between \widetilde{k}_e and the condensable water vapour mixing ratio ξ_v^{acw}

The previously discussed results by Bailey and Hallett (2004) as well as our own results from the optical imaging of laboratory grown ice particles indicate that the available condensable water mass is the driving factor for the crystal growth rate and finally the crystal complexity. Although these studies do not give insight into the ice particle surface properties (except the occasionally observed stepped hollowness, Fig. 5), it is reasonable to assume that a high ice particle growth rate also results in the formation of surface complexity. Therefore, the ice saturation ratio plotted in Fig. 4 for the -40 and -50 °C experiments was converted into the available condensable water vapour mixing ratio ξ_v^{acw} given on the x axis in Fig. 6. In this approach, ξ_v^{acw} was calculated as the difference between the measured interstitial water vapour mixing ratio ξ_v and the saturated mixing ratio ξ_v^{S} for the given pressure and temperature conditions inside AIDA. ξ_v^{S} was thereby calculated based on the parametrization of the ice vapour pressure by Murphy and Koop (2005). ξ_v^{acw} therefore represents the mixing ratio of excess

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water molecules that can deposit to the solid ice face. This way of representing the SID-3 crystal complexity data results in a combined correlation between \tilde{k}_e and ξ_v^{acw} for the -40 and -50°C experiments. Figure 6 confirms the assumption that the ice crystal growth rate also controls the ice particle surface complexity. The correlation can be quantified by fitting a simple linear regression model to the data which gives a least square functional dependency of

$$\tilde{k}_e = 0.088 \times \xi_v^{\text{acw}} + 4.22 \quad (1)$$

with ξ_v^{acw} given in ppmv.

Another conclusion that can be drawn from Figs. 4 and 6 is that the observed correlation does not depend on the type of the used aerosol or nucleation mode (homogeneous or heterogeneous) which means that the thermodynamic conditions during the ice particle (re)growth control the crystal complexity or surface roughness and not the type of ice nucleus. There is one exception however; the regrowth experiments conducted at an initial temperature of -50°C and using homogeneous nucleation in aqueous sulfuric acid particles to initiate the ice cloud (grey triangles in Fig. 6). These experiments result in higher \tilde{k}_e values compared to the corresponding experiments with heterogeneous initial ice formation on soot (circles) or dust (stars) as well as homogeneous initial nucleation at -40°C (black triangles). This indicates a “memory” effect in the ice crystal complexity for homogeneous nucleation at colder temperatures. A possible explanation for this observation could be the formation of concentrated $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ residuals on the ice crystal surface, which affects the regular crystal growth. Such a separation into solid ice and unfrozen residual solution was observed by calorimetric measurements on aqueous H_2SO_4 droplets in the cirrus temperature range $190\text{K} < T < 230\text{K}$ (Bogdan et al., 2006; Bogdan, 2006). Although emphasized by the authors, the formation of a complete $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ coating of the crystal surface was not unambiguously proven by these studies. Further studies with homogeneously nucleated ice particles are necessary to investigate the impact of unfrozen $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ residuals on the ice crystal complexity.

3.2 Discussion of specific ice particle fractions

Table 1 gives examples of the SID-3 pattern analysis results for the regrowth experiments labelled in blue and red in Figs. 4 and 6. The fraction of rough particles is thereby calculated by setting a threshold value k_e^{thr} of 4.6 and classifying all particles below this threshold as smooth (examples given in panels a and b of Fig. 1) and all particles equal and above as rough (examples given in panels c and d of Fig. 1). The fraction of columnar ice particles is calculated from a fast Fourier transformation of the polar integrated azimuthal intensity profile of the patterns according to the description given in Vochezer et al. (2015). While the fractions of rough particles and rough columns are clearly correlated with S_{ice} and $\xi_{\text{v}}^{\text{acw}}$ the fraction of columns stays rather constant at a value around 0.6 up to ice saturation ratios of about 1.15 followed by a significant decrease for higher supersaturations. This observation can be explained by the formation of hollow columns and rosettes at an onset saturation ratio between 1.15 and 1.2 which results in a distorted or changed symmetry in the corresponding light scattering patterns. Another result from Table 1 is the behaviour of the fraction of those crystals that have indications of a 22° halo in the scattering pattern. This fraction is deduced by a visual inspection of each individual valid scattering pattern for intensity spots around 22° that are clearly contrasted from surrounding pixels. Interestingly, this particle fraction is not depending on S_{ice} or $\xi_{\text{v}}^{\text{acw}}$ but is, except experiment 42 from RICE03, rather constant within the narrow range between 30 and 40%. For the interpretation of this result one has to keep in mind that the pixels in the 22° halo spot are always saturated due to the limited dynamic range of the ICCD camera of 8 bit. Therefore, the true intensity in the halo spot is unknown and, thus, it cannot be concluded from the fraction of halo particles whether and how strong a 22° halo would appear in the ice cloud. Even if it is unlikely that an ice cloud with roughened ice crystals would show a halo feature, it has to be kept in mind that there is always a significant fraction of the (small) hexagonal ice columns that show the 22° halo feature in the SID-3 scattering patterns also for those experiments with a very high crystal complexity.

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3.3 Angular light scattering functions

For the three experiments from RICE03 labelled in red in Figs. 4 and 6 and listed in Table 1 polar scattering functions were measured by the polar nephelometers PN and PHIPS-HALO and are plotted in Fig. 7. The individual scattering functions given in Fig. 7 are mean values of all valid particle scattering functions measured during the regrowth periods. A valid particle scattering function has (i) an intensity above the background level for each individual angular channel and (ii) has no saturated channels. Scattering functions that do not fulfil these criteria were removed from the analysis. A background intensity is deduced for each individual channel and is subtracted from the valid particle scattering functions before averaging. In the case of the PHIPS-HALO instrument, channel crosstalk and sensitivity characteristics are taken into consideration by applying a correction matrix to the mean scattering function. See Jourdan et al. (2003) for a description of the channel correction in the case of the PN instrument. After this, the resulting mean particle scattering function is normalised so that its integral over the instrumental angular range equals 1.

Although there are systematic deviations in the normalized scattering functions deduced from the PN and PHIPS-HALO measurements, both instruments agree in the following observations:

- i. the measured scattering functions show only minor changes from experiment to experiment
- ii. the scattering functions are rather flat and featureless with no clear indications of a 22 or 46° halo
- iii. there are some indications of a slight ice bow-like feature in the 130 to 150° angular range

Observations (i) and (ii) can be interpreted as the result of crystal complexity that randomizes spatial light scattering to a sufficient degree so that shape-dependent features, like halo features, are removed from the polar scattering function resulting in a flat

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function with no significant variations for the different experiments. Interestingly, the polar scattering function is rather insensitive over a range of ice particle growth conditions where the SID-3 method still sees significant differences in the complexity parameter \tilde{k}_e . Although the particle scattering functions for very low \tilde{k}_e values have not been measured, these results might indicate that after a certain crystal complexity is induced, the angular light scattering function is insensitive to further increases in this complexity. The corresponding threshold in terms of S_{ice} is quite low and around 1.1 for the ice clouds grown in the -50°C temperature range. In terms of the temperature-independent condensable water vapour mixing ratio ξ_v^{acw} , this threshold is about 4.3 ppmv. However, further analyses of the scattering functions in the different phases of the experiment runs are necessary to come to a final conclusion on the sensitivity of the light scattering functions to the ice particle complexity.

Observation (iii) indicates a minor influence from regular or near-spherical ice crystals on the angular light scattering function of the ice clouds (Baran et al., 2012). A similar featureless scattering function with a slight indication of an ice bow-like feature was measured by the PN in a case study of a mid-level Arctic ice cloud (Lampert et al., 2009). A particle mixture composed of deeply roughened hexagonal columns and smooth ice spheres was necessary to represent this observation. The corresponding angular scattering function of this particle model has an asymmetry parameter g of 0.78 and is plotted as solid line in Fig. 7. A very good agreement between the modelled and measured scattering functions is found, which is astonishing given that the modelled function is retrieved for an Arctic mid-level (not cirrus) cloud at a mean temperature of -24.3°C . This is a first indication that the above discussed complexity threshold for a rather insensitive mean particle scattering function might be of general nature and also valid for natural ice clouds. In conjunction with the main finding of our study, namely a fast developing ice particle complexity with supersaturation conditions, this indicates some important consequences for the light scattering properties of atmospheric ice particles that will be discussed in the following section.

4 Atmospheric implications

In the present study the ice supersaturation with temperature, i.e. the available condensable water vapour ξ_V^{acw} , was identified as the main driving factor for the formation of ice crystal complexity. This factor has a low threshold value of 4.3 ppmv for the growth of ice particles with a sufficient degree of crystal complexity. A strong randomization of the light scattering is the consequence of this complexity resulting in a featureless cloud angular scattering function with a stable asymmetry parameter g that is insensitive to further changes in the growth conditions. So, the question is how frequent such threshold conditions can be found in cirrus clouds in order to draw conclusions on the role of ice crystal complexity for the short-wave radiative properties of these clouds.

In a study by Krämer et al. (2009) in-situ water vapour data from Arctic, mid-latitude, and tropical cirrus were analysed and discussed in terms of the frequency of supersaturated conditions inside cirrus clouds. They found frequency distributions of the ice saturation ratio S_{ice} that are symmetrically distributed around a peak value close to 1 for the $200 < T < 240\text{K}$ temperature range. Although high supersaturations up to the homogeneous freezing level were measured, information of the growth history can hardly be deduced from these data as the supersaturation quickly relaxes after a sufficient amount of ice particles have been nucleated. However, by applying a simple box model and assuming homogeneous nucleation, their modeled ice number concentrations agree well with the measured concentrations for reasonable updraft speeds indicating that the formation and growth pathway of the young cirrus particles could indeed started from high supersaturations.

In the Interhemispheric differences in cirrus properties from anthropogenic emissions (INCA) experiment, ice saturation ratio dependent cloud present fractions were deduced from in-situ particle measurements in northern and southern mid-latitude cirrus (Ström et al., 2003). Cirrus ice particle formation onsets were estimated by the interpretation of local minima in these fractions to be a consequence of the reduced detection efficiency of the cloud particle instruments in case of recently nucleated cirrus

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particles. Following this interpretation, the cirrus clouds were preferentially formed in the S_{ice} range between 1.4 to 1.55 and slightly below 1.3 for the southern and Northern Hemisphere, respectively. It was suggested that this difference in the freezing thresholds is an effect of the different pollution levels in both hemispheres resulting in homogeneous ice nucleation as the dominant freezing mechanism in the relatively clean Southern Hemisphere and heterogeneous nucleation in the more polluted Northern Hemisphere (Haag et al., 2003). By taking into consideration that the frequency distribution of measured in-cloud temperatures during INCA was between -30 to -60 °C with a maximum at about -50 °C (Gayet et al., 2004), the ice particles nucleated and grew in an ice supersaturated environment that forces significant crystal complexity according to the chamber results shown in Fig. 4. In conjunction with the discussion of the light scattering data given in Sect. 3, it can be expected that the cirrus clouds probed during INCA should have angular scattering functions with (i) no indications of a 22° halo, and (ii) an asymmetry parameter g with only little variations in hemispheric as well as interhemispheric comparisons. Both expectations were indeed confirmed by in-situ PN measurements during INCA by Gayet et al. (2004). Based on a statistical analysis of angular scattering functions measured in the northern and Southern Hemispheres a very stable asymmetry parameter was deduced ranging from 0.76 to 0.78 with almost no differences in the mean values for the northern (0.767) and southern (0.770) hemisphere. Only about 3% of all measured scattering functions showed a 22° halo feature, also with no significant differences between the northern and southern measurements. It was concluded that the angular particle scattering properties give no indications on privileged ice nucleation mechanisms in the two hemispheres, although it was indicated from the freezing onset analysis. This is in accordance with the findings of the present paper where also no significant differences in the angular light scattering properties could be identified although significant differences in the complexity parameter \tilde{k}_e were deduced. The cirrus clouds probed during the INCA experiments therefore probably have formed in ice supersaturated conditions that were high enough to produce significant crystal complexity, irrespective of the ice nucleation mechanism. In

addition to the SID-3 laboratory results, there are indications from airborne SID-3 measurements that the GLCM speckle analysis method might be sensitive to the nucleation mechanism (homogeneous or heterogeneous) in ice clouds composed of complex ice particles (Ulanowski et al., 2014).

5 Conclusions

In this paper the origin of ice crystal surface complexity and its consequences for the angular light scattering properties of cirrus clouds were investigated in specific cloud chamber experiments performed in the -40 to -60 °C temperature range. The particle surface complexity was deduced from highly-resolved angular light scattering patterns of single ice particles measured by the latest version of the Small Ice Detector (SID-3). Similar to the previous work by Ulanowski et al. (2014), a speckle texture analysis was developed based on the grey-level co-occurrence matrix (GLCM) as part of the SID-3 scattering pattern analysis package (Vochezer et al., 2015). It was found that the normalized energy feature parameter k_e of the GLCM is the most robust measure of crystal surface complexity. The angular light scattering function of the laboratory generated ice clouds was measured by the two currently available airborne polar nephelometers PN and PHIPS-HALO.

A new experimental procedure was applied at the AIDA cloud chamber to regrow ice particles at defined ice saturation levels after the crystal surface properties from the initial nucleation and growth phases have been removed in a sublimation period. The nucleation of ice particles was initiated by using three different aerosol types; soot particles and mineral dust for heterogeneous ice nucleation as well as sulphuric acid particles for homogeneous nucleation. For 45 stable regrowth periods, the median crystal complexity parameter \tilde{k}_e was deduced from the individual SID-3 scattering patterns. Averaged angular light scattering functions were deduced from the polar nephelometers for three regrowth periods.

From the experimental results the following main conclusions can be drawn:

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Midlatitude Cirrus (ML-CIRRUS) and Aerosol, Cloud, Precipitation, and Radiation Interactions and Dynamics of Convective Cloud Systems (ACRIDICON-CHUVA). The results of these measurements will be the subject of forthcoming publications.

Appendix A: Characterisation of the normalized energy feature parameter k_e

5 According to Lu et al. (2006), k_e represents the coefficient of an exponential fit to the normalized energy feature vs. pixel distance curves that were calculated for the four distinct directions 0, 45, 90, and 135° throughout the grey-level image. In the present analysis algorithm, k_e is deduced based on the normalized energy feature curves that are calculated for the first three directions. Each of the three curves covers the pixel
10 distance range 1 to 31. So, a total number of $3 \times 30 = 90$ are calculated and analysed per scattering pattern. See Figs. 3 and 4 of Lu et al. (2006) for an illustration of how the grey-level co-occurrence matrix (GLCM) is calculated for different directions and pixel distances.

15 The conclusion given by Lu et al. (2006) that k_e is the most robust GLCM feature to quantify surface roughness was checked for the SID-3 set-up by the following procedure. An analogue ice particle aggregate with severely roughened surfaces was placed on a thin polycarbonate window with anti-reflective coating. See Krasinski et al. (2007) for a description on how these ice analogue particles can be produced in the laboratory. After a visual inspection of the analogue aggregate under the optical microscope
20 (left image of Fig. A1), the sample particle was moved into the sensitive area of SID-3 and was fixed at this position. While the instrument was forced to continuously trigger, the laser power was varied from a few mW to its maximum power of 100 mW. In this way a series of SID-3 patterns was acquired for the same particle at the same orientation but for different laser intensities (see right image of Fig. A1 for an exam-
25 ple). The latter variation results in images with different brightnesses. In a final step, the GLCM features k_e , energy, and combined roughness were statistically analysed for different grey-level ranges. The result of this analysis is given in Table A1. The test

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diameter during the initial growth (I) and the regrowth (II) phases of the experiment. The AIDA atmospheric conditions forced the generation of two main habit types (compact particles and columns) which could be classified by the imaging system of PHIPS-HALO as well as the replicator strip analysis (Fig. 5). The temporal evolution of the particle equivalent diameters shows quasi linear increases of the particle dimension during the phases of near-constant ice supersaturation conditions (indicated in yellow in Fig. C1). These periods were later used to calculate the particle growth rate. The regrowth at $S_{\text{ice}} \approx 1.2$ (phase II) resulted in the formation of hollow columns that were observed in this phase (sample images in Fig. C1 as well as Fig. 5). In other experiments at higher S_{ice} even more complex deformations were observed due to the fast growth of the crystals. Finally, panel d shows the column/non-column ratio deduced from the PHIPS-HALO images. It shows that the fraction of columnar particles increased with time, which is due to a size-dependent shape classification for particles in this size range. However, at the final stage of phase II, a column fraction of 0.4 to 0.5 were deduced which is close to 0.58 inferred from the SID-3 patterns analysis. After phase II, there is a period (starting at 1500 s) where the temperature increased and S_{ice} decreased. At this stage the particles started to sublime at ice subsaturated conditions resulting in the formation of rounded particle edges that could be clearly observed in the PHIPS-HALO images of the columnar ice crystals in this stage (see images in panel c and Fig. 3). Interestingly, the sublimation process results in a scatter of the deduced column fraction in panel d.

Along a series of AIDA expansions during this campaign, different S_{ice} values were produced at the same temperature to study the influence of S_{ice} on the particle growth rate at constant temperature. The S_{ice} value was kept constant for certain time in each expansion to allow sufficient crystal growth. The growth rates of the c axis, a axis, and equivalent diameter of the columnar ice particles were deduced for four different S_{ice} regrowth experiments conducted at the same temperature (-56°C). The results are shown in Fig. C2. In agreement with the results from Bailey and Hallett (2004), the growth rates are proportional to the ice supersaturation. However, Bailey and Hallett

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(2004) deduced a linear dependence of the maximum particle dimension with supersaturation while our results rather indicate an exponential dependence. This can be attributed to the difference in experimental procedure where in the case of Bailey and Hallett (2004), the crystals were nucleated and grown on a glass wire while in our case the ice nucleation and growth is contact free, which mimics the growth under natural conditions. It is also noticed that the c axis grows faster than the a axis of the columnar ice particles. The growth rate of $0.047 \mu\text{m s}^{-1}$ deduced for $S_{\text{ice}} = 1.26$ lies within the range given by Bailey and Hallett (2012) for this saturation ratio.

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Table 1. Results of the particle fraction analysis for those experiments of the RICE01 and RICE03 campaign that are respectively labelled in blue and red in Figs. 4 and 6. All experiments were conducted at an initial temperature of -50°C and with soot particles as heterogeneous ice nuclei.

Exp.	S_{ice}	$\xi_{\text{v}}^{\text{acw}}$ [ppmv]	Fraction of rough particles	Fraction of columns	Fraction of rough columns*	Fraction of halo crystals
RICE01						
22	1.02	0.86	0.14	0.63	0.06	0.34
23	1.14	3.97	0.44	0.65	0.28	0.39
24	1.25	6.71	0.60	0.56	0.41	0.37
26	1.38	10.6	0.84	0.39	0.73	0.31
RICE03						
40	1.13	4.48	0.51	0.58	0.24	0.41
42	1.30	7.03	0.72	0.45	0.50	0.22
43	1.20	4.33	0.55	0.53	0.28	0.29

* Ratio of the number of rough columns to the number of all columns.

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Table A1. Relative 1σ standard deviations of the three GLCM features k_e , energy, and combined roughness. The SID-3 patterns were acquired on the analogue ice particle shown in Fig. A1. The GLCM speckle texture analysis was restricted to three different grey-level ranges.

Grey-level range	k_e	Energy	Combined roughness
10–25	0.66 %	1.38 %	0.95 %
10–50	0.49 %	5.48 %	1.43 %
1–100	0.72 %	7.77 %	1.79 %

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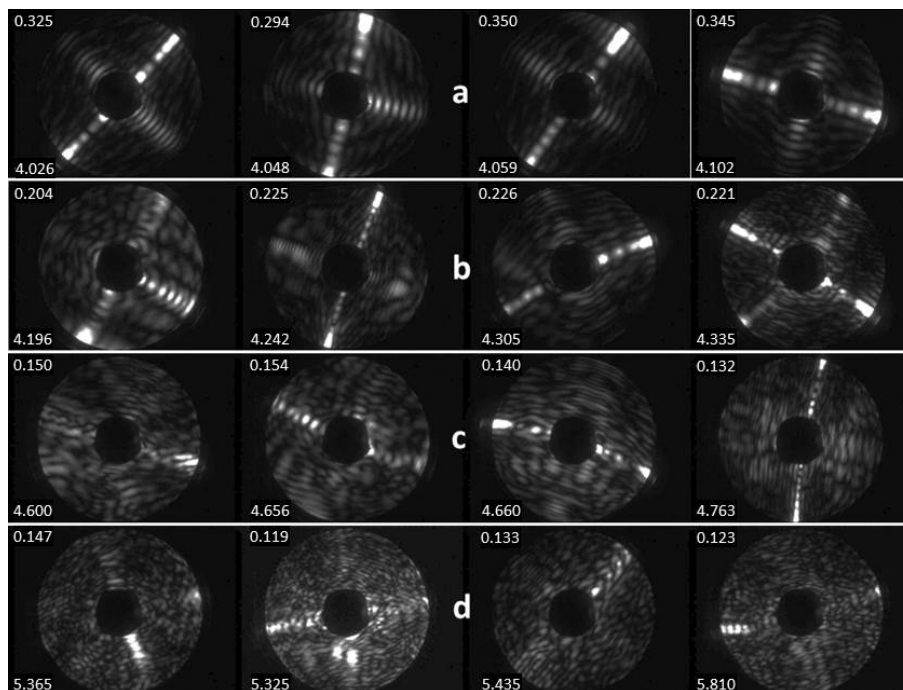


Figure 1. Representative examples of 2-D light scattering patterns of individual small columnar ice particles generated in re-growth cloud chamber experiments at -50°C . The patterns are labelled by the image texture feature energy (upper left corner) and energy feature parameter k_e (lower left corner). The patterns are arranged according to increasing k_e from left to right and from panel (a) to panel (d). Rough particle fractions are deduced by setting a threshold value of $k_e^{\text{thr}} = 4.6$ and classifying all particles equal and above as rough.

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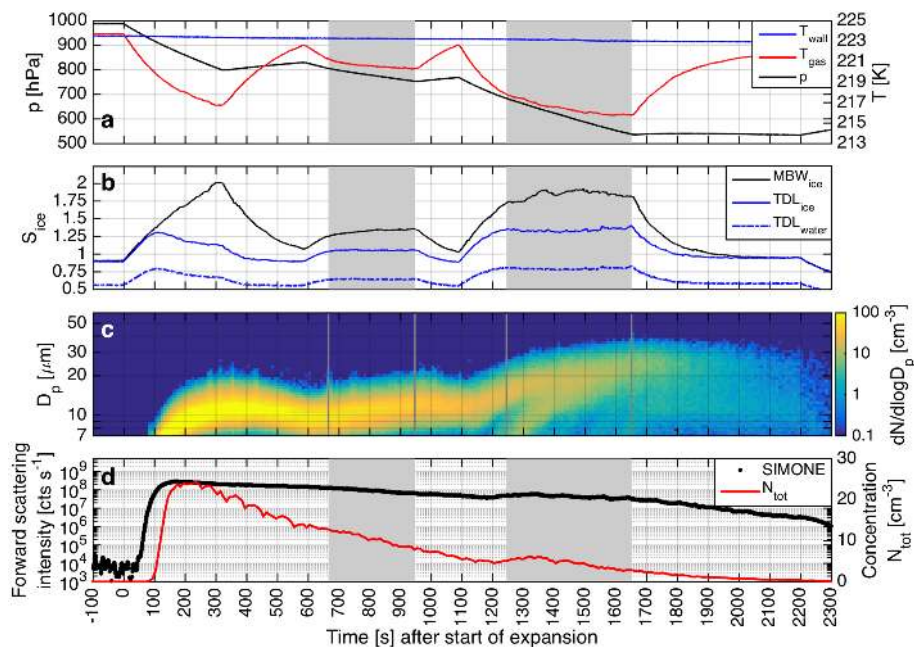


Figure 2. Example of a regrowth experiment conducted in the AIDA cloud chamber at an initial temperature of -50°C . The experimental procedure is specifically designed to grow and sublimate ice particles at defined super- and subsaturated ice conditions (solid blue line in panel **b**). Regrowth periods are indicated by the shaded areas. See text for details.

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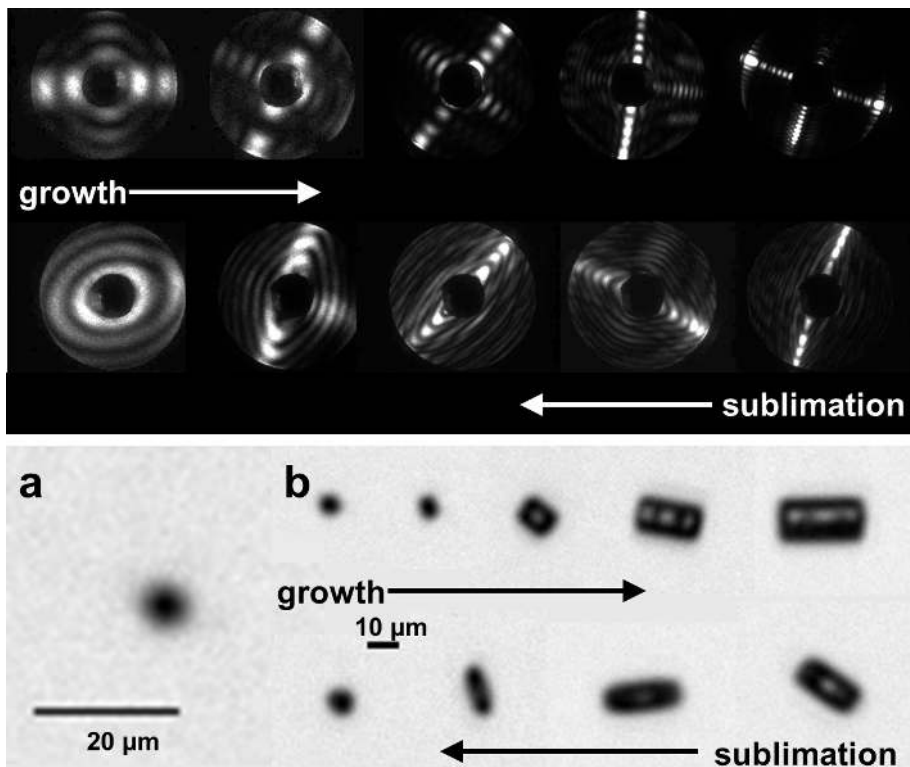


Figure 3. Evolution of SID-3 scattering patterns (upper panel) and PHIPS-HALO micrographs (series b in the lower panel) of columnar ice particles collected during growth and sublimation periods in the -40°C temperature range. Ice particle surface processing by sublimation is clearly visible by a rounding of the ice particle edges resulting in oval SID-3 scattering patterns. Note that SID-3 is capable to determine the particle shape for crystals with sizes of only 2 to 3 μm (patterns on the left side of the upper panel) whereas the imaging method used in PHIPS-HALO cannot give a clear shape information for particles smaller than about 10 μm (image a in the lower panel).

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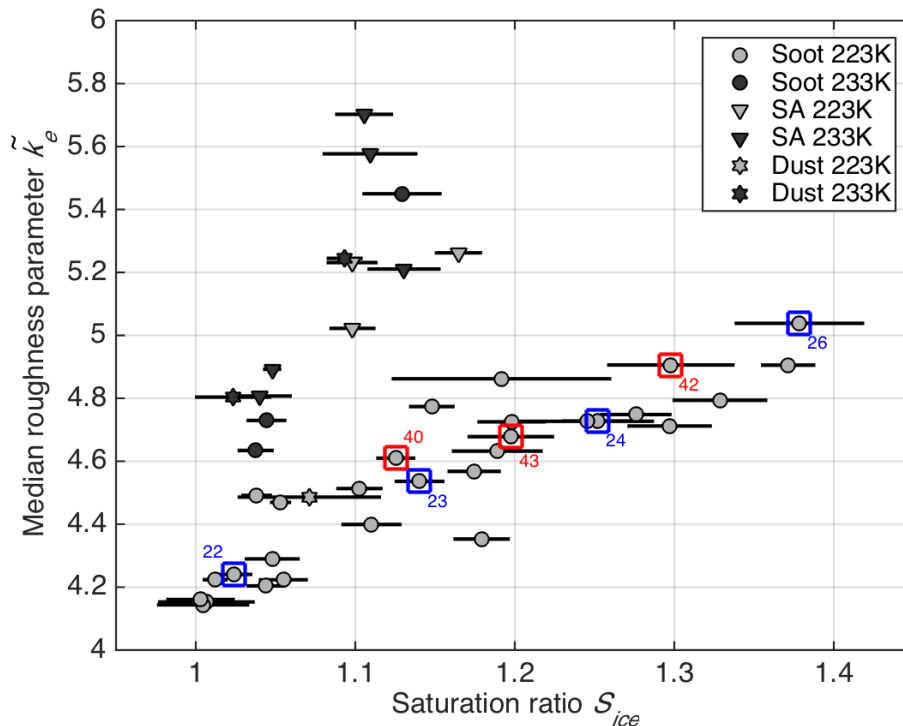


Figure 4. Median ice crystal complexity parameter \tilde{k}_e deduced from SID-3 scattering patterns that were measured in simulated cirrus clouds at the AIDA cloud chamber. The ice clouds were grown at different ice saturation ratios S_{ice} and at initial chamber temperatures of -40°C (black symbols) and -50°C (grey symbols). Three different aerosol types were used to nucleate ice by heterogeneous nucleation on soot (circles) and mineral dust (stars) as well as by homogeneous nucleation in aqueous sulfuric acid particles (triangles).

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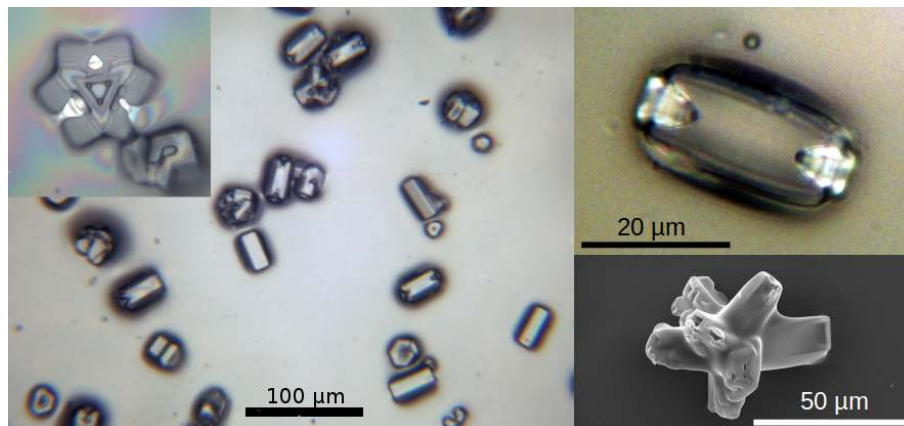


Figure 5. Ice particle replicas generated in the regrowth period of RICE01_27 at a temperature around -55°C and an ice supersaturation of 19%. The particle habit distribution is a mixture of solid and hollow columns as well as small rosettes (left). Hollow rosette with a maximum dimension of about $35\ \mu\text{m}$ (upper left). Stepped hollow column as observed occasionally (upper right). Scanning electron micrograph of a rosette replica showing that in general the ice particle surface properties are masked by the Formvar replication method (lower right).

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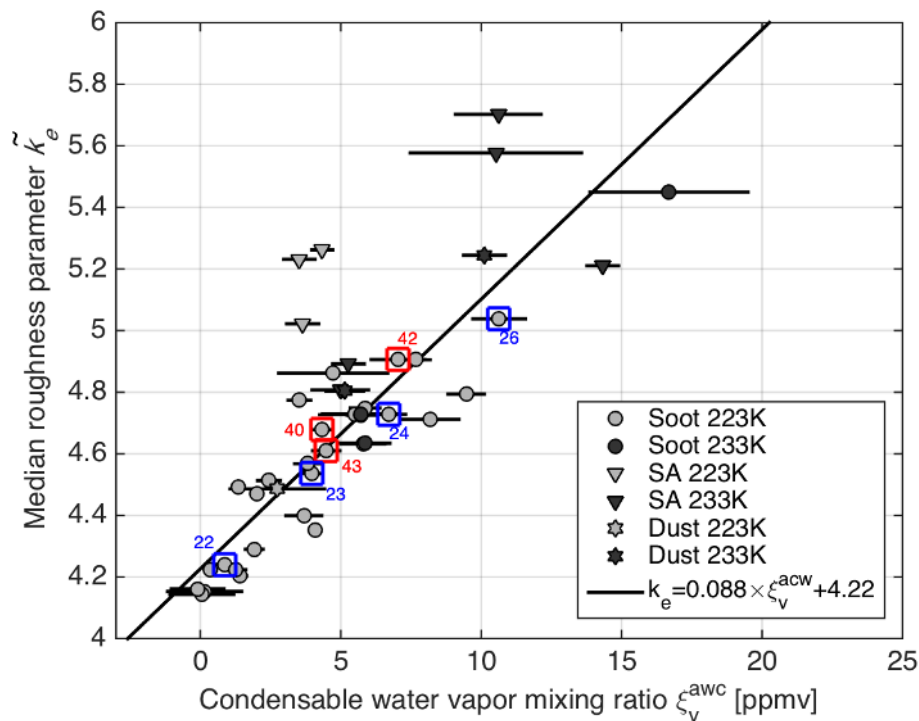


Figure 6. Median ice crystal complexity parameter \tilde{k}_e plotted against the condensable water vapour mixing ratio that was available during the regrowth of the ice particles.

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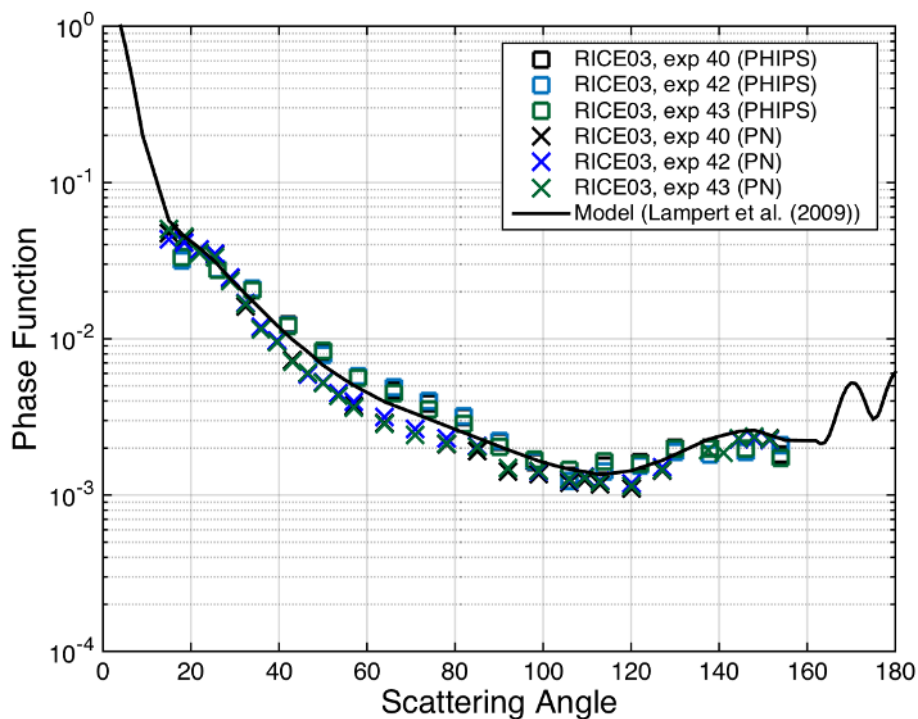


Figure 7. Mean angular scattering functions measured in the regrowth periods of those ice cloud experiments of RICE03 that are labelled in red in Figs. 4 and 6. Two polar nephelometers were deployed in these experiments; the airborne approved polar nephelometer from Clermont-Ferrand (PN, crosses) and the novel instrument PHIPS-HALO from KIT (squares).

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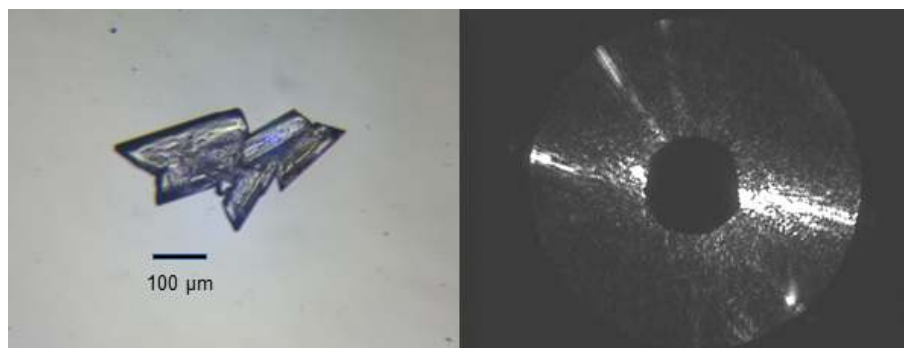


Figure A1. Microscopic image of an ice analogue aggregate with rough surface residing on a polycarbonate window (left). The sodium fluorosilicate crystals can grow to hexagonal morphologies similar to water ice crystals. The fact that their refractive index is close to water ice makes them well suited for light scattering investigations (Ulanowski et al., 2003, 2006). The crystal was moved into the sensitive area of SID-3 where scattering patterns were acquired (right).

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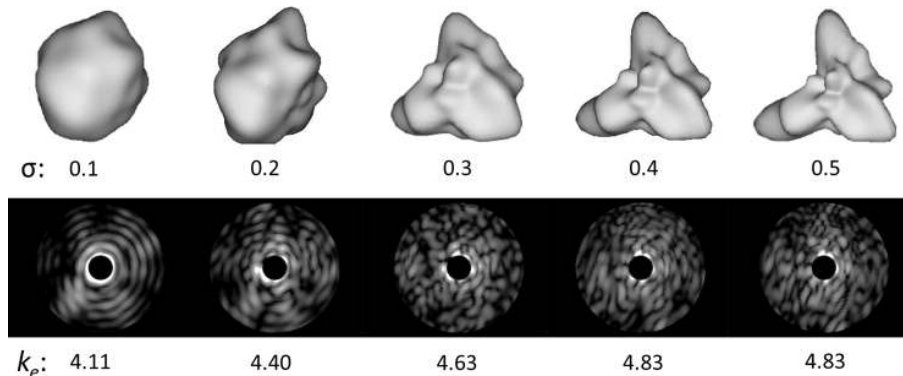


Figure B1. Distorted spherical ice model particles with different deformation parameter σ that were used in the ADDA/GDT-matrix approach (upper panel). All input model particles have a fixed size parameter of 46. Corresponding 8 bit grey-scale output patterns (lower panel). The k_e values of the GLCM pattern analysis are given below each pattern.

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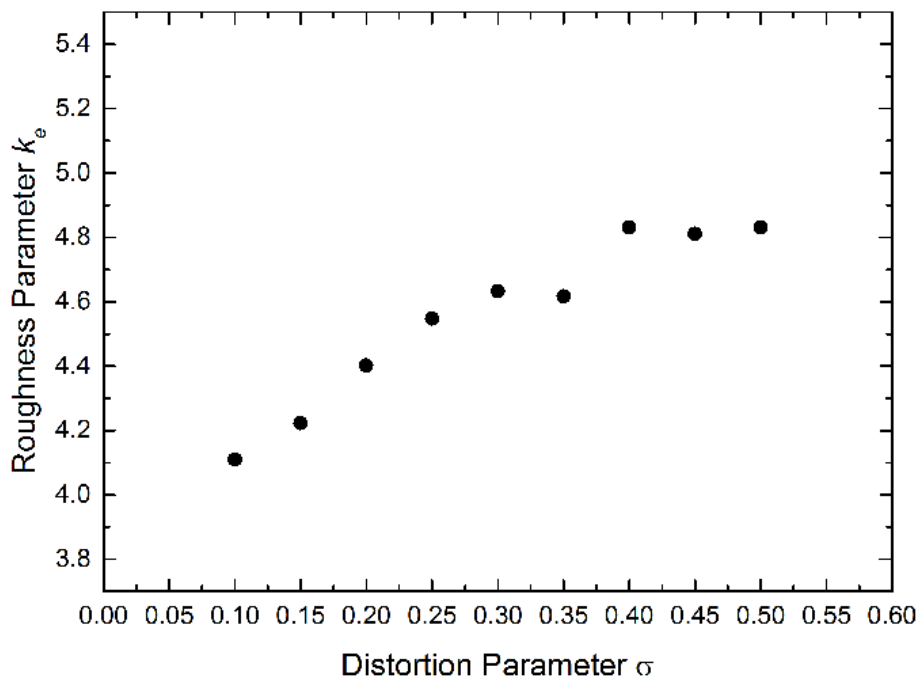


Figure B2. Dependence of the crystal complexity parameter k_e on the distortion parameter σ for the model particles shown in Fig. B1.

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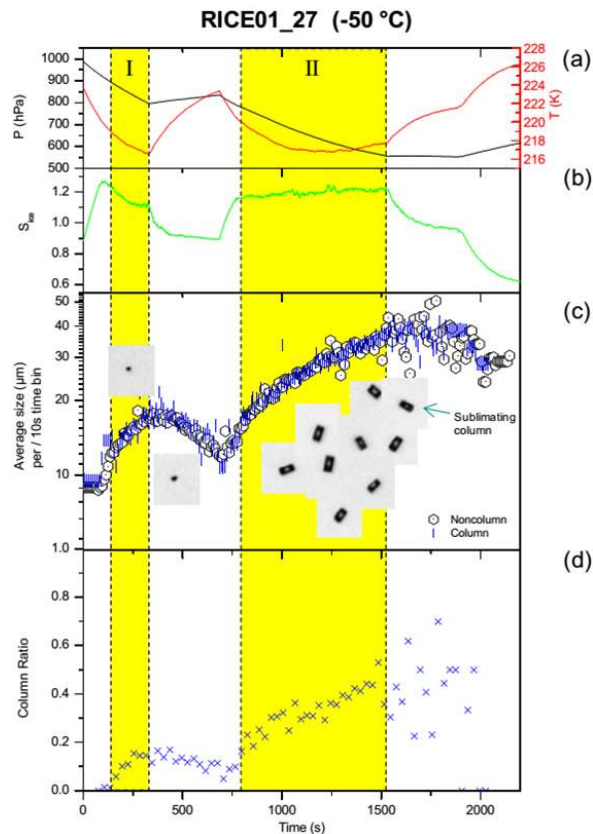


Figure C1. Temporal evolution of the AIDA cirrus cloud experiment RICE01_27. **(a)** Gas temperature and pressure inside the AIDA chamber. **(b)** Ice saturation ratio S_{ice} inside the AIDA volume. **(c)** Geometric particle diameters as measured by PHIPS-HALO. **(d)** Column ratio during the experiment.

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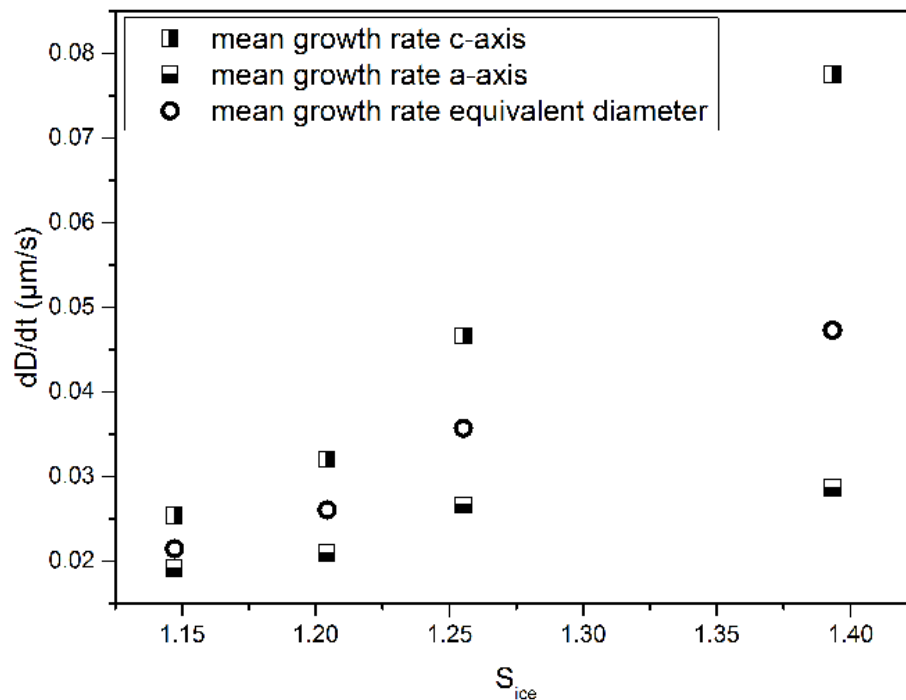


Figure C2. Growth rates of the c axis, a axis, and equivalent diameter of columnar ice particles deduced for four different S_{ice} regrowth experiments. The regrowth periods were conducted at the same temperature of -56°C .