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Direct evidence for secondary ice formation at around -15 °C in mixed-phase clouds

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Abstract. Ice crystal numbers can exceed the numbers of ice-nucleating particles (INP) observed in mixed-phase clouds by several orders of magnitude also at temperatures that are colder than required for the Hallett-Mossop process (-3 °C to -8 °C). These observations provide circumstantial evidence of secondary ice formation. Attempting a more direct observational approach we made use of the fact that planar, branched snow crystals (e.g. dendrites) grow within a relatively narrow temperature range (about -12 °C to -17 °C) and can be analysed individually for INP using a field-suitable drop freezing assay technique. During February and March 2018, we analysed 190 dendritic crystals (an average of ~3 mm in size and between 1.3 to 7.6 mm) deposited within mixed-phase clouds at the High Altitude Research Station Jungfraujoch (3580 m a.s.l.), Switzerland. Overall, one in eight of these crystals contained an INP active at -17 °C or warmer, while the remaining seven of eight most likely resulted from secondary ice formation within the clouds. The ice multiplication factor we observed was small (8), but relatively stable throughout the course of the experiment. These measurements show that secondary ice can be observed at temperatures around -15 °C in the atmosphere and thus advance our understanding of the extent of secondary ice formation in mixed-phase clouds, even where the multiplication factor is smaller than 10.

1 Introduction

Ice-nucleating particles (INP) are required to catalyse primary ice formation in clouds at temperatures above -36 °C via heterogeneous freezing (e.g. Vali et al., 2015). This freezing pathway is expected to generate ice crystals in mixed-phase clouds and is supported for instance by Kumai (1951, 1961) and Kumai and Francis (1962) who found an insoluble particle of 0.5 to 8 µm in size in the centre of almost every of about 1000 snow crystals they had analysed. However, in-cloud observations report large discrepancies between measured INP and ice crystal numbers (e.g. Hobbs and Rangno, 1985; Beck et al., 2018) the latter being several orders of magnitudes higher than the former. These observations suggest that not only heterogeneous and homogeneous freezing, but also other processes such as secondary ice production mechanisms (i.e.

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production of ice particles requiring the prior presence of other ice particles (Vali, 1985)) must generate the observed ice particles.

Secondary ice crystals can for instance result from rime splinters that are released upon riming of ice crystals at temperatures between -3 °C and -8 °C (Hallett and Mossop, 1974). Other than the well-known Hallett-Mossop process, mechanisms proposed for secondary ice production include ice-ice collisional breakup (e.g. Vardiman, 1978), droplet shattering or fragmentation upon freezing (e.g. Takahashi and Yamashita, 1970; Lauber, 2018) and sublimation fragmentation (e.g. Bacon et al., 1998). These processes and indications for their occurrence in the atmosphere are summarised in Field et al. (2017). Sullivan et al. (2018a) have recently studied three of those above-mentioned secondary ice formation processes in terms of their thermodynamic and primary ice requirements in a parcel model. They showed that INP concentration can be as low as 2 m^{-3} (0.002 L^{-1}) to initiate ice multiplication by ice-ice collisional breakup. Furthermore, the number of INP is less important than a warm enough cloud base temperature and modest vertical updraft velocity for frozen droplet shattering and rime splintering (Sullivan et al., 2018a). When droplet shattering and ice-ice collisional breakup were implemented into a large-scale weather model, secondary ice contributed as much to ice crystal number concentration than ice from primary nucleation, even though high ice crystal numbers remain underestimated by the model (Sullivan et al., 2018b).

Modelling studies accounting for secondary ice production can explain to some extent the observed ice crystal numbers (e.g. Sullivan et al., 2018b). However, in-situ evidence of secondarily produced ice has been difficult to obtain to date, one difficulty being crystal fragmentation due to the sampling instrument (Schwarzenboek et al., 2009). Furthermore, it probably is not possible to estimate the number of primarily nucleated ice crystals in clouds from the INP spectrum of precipitation samples (e.g. Petters and Wright, 2015) because INP data from bulk precipitation samples cannot be disentangled to the level of individual hydrometeors. Riming (i.e. supercooled cloud droplets that freeze upon contact with a solid hydrometeor) can affect the spectrum of a bulk sample by potentially adding INP immersed in supercooled cloud droplets. Further, ice-nucleation active microbes can be scavenged by raindrops below cloud and alter the spectrum (Hanlon et al., 2017). The first experiment in which individual hydrometeors were analysed for INP, and the only one we are aware of, was conducted by Hoffer and Braham (1962). The hydrometeors were large, frozen water drops that had grown through riming ("snow pellets" or "ice pellets"; Braham, 1964) and collected with an aircraft within summer clouds. They all (n = 301) re-froze at temperatures substantially lower than the expected minimum temperature of the cloud (i.e. the cloud top temperature estimated by radiosonde data) from which they were collected, and the authors presumed them to be of secondary origin.

In this study, similar to the one by Hoffer and Braham (1962), we collected in-cloud hydrometeors to obtain in-situ evidence of secondary ice formation. The main difference to Hoffer and Braham (1962) is that we collected vapour grown snow crystals in winter clouds at a mountain-top station and estimated the temperature of their formation from their habit (shape). Our idea to observe secondary ice at around -15 °C is based on previous studies. First, Westbrook and Illingworth (2013)

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observed a long-lived supercooled cloud layer with a cloud top temperature around -13.5 °C, which continued to precipitate ice crystals well beyond the expected exhaustion of its INP reservoir. Second, laboratory investigations revealed ice-ice collision to be most effective in producing secondary ice particles at around -16 °C (Takahashi et al., 1995), or in collisions involving dendritic crystals (Griggs and Choularton, 1986). Third, the habit of ice crystals forming in super-saturated conditions between -12 °C and -17 °C is well and distinctively defined. It is a planar, sector-type or dendrite-type (branched) habit (Nakaya, 1954; Magono 1962; Magono and Lee, 1966; Takahashi, 1991, 2014) that grows by vapor diffusional growth into a diameter of several millimetres (e.g. forming snow crystals or dendrites) during a vertical fall of a few 100 m (Fukuta and Takahashi, 1999). This in turn restricts their expected nucleation temperature accordingly. These observations suggest that direct evidence for secondarily formed crystals might be obtained by collecting planar, branched snow crystals from supercooled clouds and testing them individually for the presence of INP that might have catalysed their formation. Absence of such an INP indicates that these crystals are not formed by heterogeneous freezing and would thus be strong evidence for the crystal being a product of secondary ice formation.

2 Experimental

2.1 Location and meteorological conditions

Between 15 February and 22 March 2018, we collected and analysed a total 229 planar, sector- and dendrite-type snow crystals (i.e. ice crystals of a size larger than 1.3 mm) during cloudy conditions at the High Altitude Research Station Jungfraujoch (3580 m a.s.l.) in the Swiss Alps. During the collection, cloud base height as measured by MeteoSwiss with a ceilometer located 5 km northwest of Jungfraujoch (Poltera et al., 2017) was on average 950 m below the station (zB, Table 1). Based on air temperature measured by MeteoSwiss at Jungfraujoch, cloud base height and an assumed moist adiabatic lapse rate of 7.5 °C km^{-1} (plausible for approximately 650 hPa and -10 °C) we estimated that cloud base temperatures (CBT) were between 1 °C and -12 °C. The mean air temperature at the station during the sampling periods was -11.0 °C and the mean wind velocity was 9.1 m s^{-1}. On three days air masses arrived from south-east (SE) or east (E), and on seven days from north-west (NW).

2.2 Single crystal analysis

We collected snow crystals on a black aluminium plate (40 cm x 40 cm) at about 1 m above the floor of the main terrace of the Sphinx Observatory at Jungfraujoch and analysed the crystals inside a small, naturally cold (-1 °C to -7 °C) anteroom between the terrace and the laboratory. Among a usually wide variety of shapes and sizes collected on the plate we selected what we considered to be planar, branched snow crystals. Selected crystals were documented by macro (1:1) photography (camera: OM-D E-M1 Mark II, pixel width: 3.3 µm; objective: M.Zuiko ED 60mm f2.8; flash: SFT-8; all items from Olympus, Tokyo, Japan) stabilised by a focusing rack (Castel-L, Novoflex, Memmingen, Germany) propped up on the aluminium plate. Images were later analysed more exactly on a larger screen for the habit, including the degree of riming

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both categorized according to the 'global classification scheme' by Kikuchi et al. (2013). The size of the crystals was determined by using ImageJ (Rueden, 2017; Schindelin, 2012).

After selecting the crystals, we tested them on freezing using a custom-built cold stage (Figure 1; more details in supplement). The cold stage is a modified drop freezing apparatus, a methodology that is commonly used today and initially described by Vali (1971a). The device is meant to be taken into the field, can be set up within minutes and operated without additional infrastructure (i.e. no cooling water or lined power is required). It consists of a gold-plated copper disk with a surface diameter of 18 mm, which is large enough to easily accommodate two dendrites and two control droplets at once (roughly 1 cm apart from each other).

With a fine brush, we transferred two crystals onto the cold stage thinly covered with petroleum jelly and at a temperature between 1 and 5 °C (Fig.1a). Upon contact with the cold stage these crystals melted into droplets. To aid visual detection of freezing during testing, we increased the size of the melted crystal droplets by adding 3 µL of ultrapure water (Molecular Biology Reagent, Sigma-Aldrich) with a pipette. The melted crystals potentially contained the suspected INP and have a rather small volume compared to the added water. For each crystal a control droplet (3 µL) of the same ultrapure water was placed next to the melted crystal droplet and served as control (blank) (Fig. 1b). Then we ramped the temperature of the cold stage down to -25 °C. Shortly after the cold stage temperature reached a value below the surrounding air temperature, we covered it with a transparent hood to prevent contamination and condensation of the drops. From -12 °C and below we limited the cooling rate to 3 °C min^{-1}. The freezing of the droplet and thus the presence of the most efficient INP was detected visually, and the corresponding temperature was recorded manually (Fig. 1c). After a test was complete, we cleaned the cold stage carefully with isopropanol. In total, the procedure (i.e. collecting and analysing two samples) takes ~15 minutes.

2.3 INP concentration in rime

We collected rime to estimate how much INP are included in it and thus how riming affects the INP numbers in rimed single crystals. Rime was collected with a plexiglass plate (Lacher et al., 2017) suspended on the railing of the terrace at Jungfraujoch for a few to several hours (~1-13h). In total, 30 samples were taken during 15 February and 11 March and they were prepared and analysed with the NOAA Drop Freezing Cold Plate following the description in Creamean et al. (2018). To summarize, 300 droplets of molten rime of ~2.5 µL each are tested on a cold plate that was cooled towards a temperature where all droplets were frozen. The cumulative number of INP active at a certain temperature is calculated by taking into account the observed numbers of frozen droplets at a temperature, the total number of droplets and the analysed volume of sample (Vali, 1971b).

3. Results and discussion

Of the 229 crystals analysed in the field 39 had to be excluded retrospectively because a closer inspection of the enlarged photographs showed that they were either not planar or not branched. Most of the excluded crystals were spatial or radiating assemblages of plane-type crystals (P6 or P7, according to Kikuchi et al. (2013)). The remaining 190 crystals were confirmed as planar and branched, i.e. having a habit that typically forms between -12 °C and -17 °C. A large fraction of them were rimed (31%) or densely rimed (51%) dendrites (R1c or R2c, according to Kikuchi et al. (2013)); while the remainder belonged to other categories (in order of decreasing frequency: graupel-like snow of hexagonal shape, hexagonal graupel, composite plane-type crystals, dendrite-type crystals, sector-type crystals or R3a, R4a, P4, P3, P2, respectively, according to Kikuchi et al. (2013)). Their greatest length in the a-axis (outer diameter) ranged from 1.3 to 7.6 mm, with a median of 2.8 mm and a mean of 3.1 mm.

We found no INP active above -12 °C present in the crystals, which corroborates the crystal classification and the temperatures at which they form (Takahashi, 2014). In 24 of the 190 crystals an INP active between -12 °C and -17 °C was present (Figure 2). In the other 166 crystals no INP was found between -12 °C and -17 °C. They either refroze below -17 °C (95) or stayed supercooled until -25 °C (71). Blanks that froze above -17 °C were limited to one count, occurring between -16 °C and -17 °C. Between -17 °C and -25°C, 40 control droplets froze; the rest (149) stayed supercooled until -25 °C.

Throughout the observation period of 10 days the daily fraction of primarily nucleated ice versus ice crystals without an INP was relatively stable (Figure 3). From these results, we conclude that about one in eight (24/190) planar, branched crystals found in mixed-phase clouds at Jungfraujoch during winter months in 2018 resulted from primary ice formation and seven of eight were generated through a process of secondary ice formation at temperatures between -12 °C and -17 °C.

Our preliminary conclusion is based on the following four assumptions: The first assumption is that INP embedded in natural snow crystals can be repeatedly activated at the same temperature. Second, that the analysed crystals did not grow from aerosolised parts of hoar frost growing on surrounding surfaces (Lloyd et al., 2015; Farrington et al., 2016; Beck et al., 2018). Third, that initial ice formation leading to the growth of the analysed crystals likely did not occur at a temperature colder than -17 °C. And, fourth, that the detected INP active at -17 °C or warmer (INP_{-17}) were not scavenged during riming of a secondarily formed crystal.

We are confident that the first condition (i.e. that INP stable over many refreezing cycles) for our preliminary conclusion is met. Although substantial fractions of bacterial INP active above -7 °C are deactivated after a single freeze-thaw cycle, those active below -7 °C are typically unaffected even after three freezing cycles (Polen et al., 2016). Further, experiments with INP from soils show a remarkable stability of the ice nucleation temperature over tens of repeated melting and freezing

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cycles, with standard deviations of 0.2 °C (Vali, 2008). Furthermore, Wright (2013) saw similar results for rain water samples. Since we analysed the collected crystals within minutes after melting, we can also exclude changes due to storage (i.e. aging), which has been observed with bulk snow samples over the course of days or weeks (Stopelli et al., 2014).

Surface frost can be a strong source of very small (~100 µm), secondary ice crystals at Jungfraujoch (Lloyd et al., 2015) and at other mountain stations (Beck et al., 2018). Surface frost grows in saturated conditions, breaks when windy, and broken-off segments can become ingested into clouds and commonly mistaken for secondary ice (Rogers and Vali, 1987). During 7 of 10 sampling events air masses approached from northwest. The terrain falls off steeply in this direction and reaches the average observed cloud base (~1000 m below Jungfraujoch, Table 1) within a horizontal distance of about 2 km. At an average wind velocity from this direction of 8 m s^{-1} the distance is covered within less than 5 minutes, which is too short for small, broken off frost crystals to grow to the average size of the crystals we have analysed (average of 3.1 mm). Even in most favourable conditions a dendrite would not grow to 1 mm diameter within that time (Takahashi et al., 1991). Therefore, it seems unlikely that dendrites which were not associated with an INP_{-17} had grown from particles of hoar frost emitted by surfaces in the vicinity of Jungfraujoch.

According to Furukawa and Takahashi (1999) a dendrite falls about 400 m while growing to a diameter of around 3 mm. Given a diabatic lapse rate of 7.5 °C km^{-1} an initial ice crystal may have been generated in 3 °C colder conditions than where its growth into a 3 mm dendrite was completed. However, as the deposition velocity of a tiny initial ice crystal is small, the initial ice formation will unlikely have occurred at much higher altitudes than where the main growth into dendrites occurred. Even if we consider all crystals which contained an INP active between -12 °C and -20 °C a large fraction of them (81%) remains to be considered the product of secondary ice formation.

The presence of INP active at temperatures colder than -17 °C associated with the collected crystals might be explained by riming, i.e. the collection of cloud droplets containing such particles not activated as INP because ambient temperatures were not cold enough (Table 1). A majority of our crystals were rimed or densely rimed. The median concentration of INP active at -25 °C or warmer (INP_{-25}) in the collected rime at Jungfraujoch was about 1100 ml^{-1} during the period from 15 February to 12 March. Since 41% (background subtracted) of our crystals contained an INP_{-25}, the average mass of rime associated with a single crystal (m) must have been about 4.9 x 10^{-4} g (Eq. 1):

$$m\,[g\ rime\ crystal^{-1}] = \frac{\ln((1-0.41)^{-1})}{1100}\,[INP_{-25}\ crystal^{-1}/INP_{-25}\ g^{-1}\ rime] \quad (1)$$

This is about twice as much as the difference in mass (~2 x 10^{-4} g) between rimed and un-rimed dendrites of 3 mm diameter found at Mount Tokachi, Hokkaido (Nakaya and Terada, 1935). The median of INP active at -17 °C or warmer in rime was 16 ml^{-1}. Therefore, less than 1% of the crystals we have analysed might have scavenged an INP through riming that was active at -17 °C or warmer (16 [INP_{-17} g^{-1} rime] x 4.9 x 10^{-4} [g rime $crystal^{-1}$]).

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Conclusion and outlook

The temperature dependent habit of a planar, branched snow crystal enables the verification of whether it derived from primary or secondary ice formation, as long as certain conditions are met. Although the required experimental procedure including refreezing of dendrites using a drop freezing assay has a low throughput (~15 minutes for two snow crystals) it can provide robust results that cannot be obtained with other approaches. From the single, relatively large dendrites, we found direct evidence for secondary ice formation between -12 °C and -17 °C in winter clouds. The ice multiplication factor we observed was much smaller than the 'several orders of magnitude' sometimes inferred from circumstantial evidence. We presume that the ice multiplication factor close to one order of magnitude was most likely found because we had focused on relatively large crystals which are unlikely the product of local surface sources or instrumental interferences, the latter causing mainly small or broken ice fragments. To learn more about the occurrence of secondary ice formation in moderately supercooled clouds, we think it would be valuable to repeat these experiments in other meteorological conditions or in other locations such as those where most crystals were previously found to contain an insoluble particle in the centre.

Data availability

The data are available from the authors upon request.

Competing interests

The authors declare that they have no conflict of interest.

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Table 1. Sampling periods, numbers of analysed crystals (n), air temperature (T), wind velocity (u) and wind direction (dd) at Jungfraujoch; height of the station above cloud base (zB) and estimated cloud base temperature (CBT).

Date	Time span	n	T	u	dd	zB	CBT
dd/mm/yyyy	UTC	-	°C	m/s	-	m	°C
15/02/2018	07:37 - 21:40	38	-7.0	13.5	NW	944	0.1
16/02/2018	09:30 - 16:20	29	-8.7	9.0	NW	1239	0.6
17/02/2018	09:40 - 23:33	42	-8.5	5.8	NW	693	-3.3
23/02/2018	10:30 - 21:10	20	-14.8	11.9	SE	365	-12.1
06/03/2018	12:20 - 19:10	14	-13.1	5.5	NW	1284	-3.4
07/03/2018	08:00 - 16:30	23	-15.7	4.5	NW	1001	-8.2
10/03/2018	09:30 - 12:40	11	-6.8	5.1	E	196	-5.4
11/03/2018	15:40 - 16:50	6	-9.8	13.1	SE	1485	1.3
12/03/2018	09:10 - 11:00	12	-11.4	6.2	NW	878	-4.8
22/03/2018	15:50 - 22:20	34	-15.2	12.4	NW	1079	-7.1

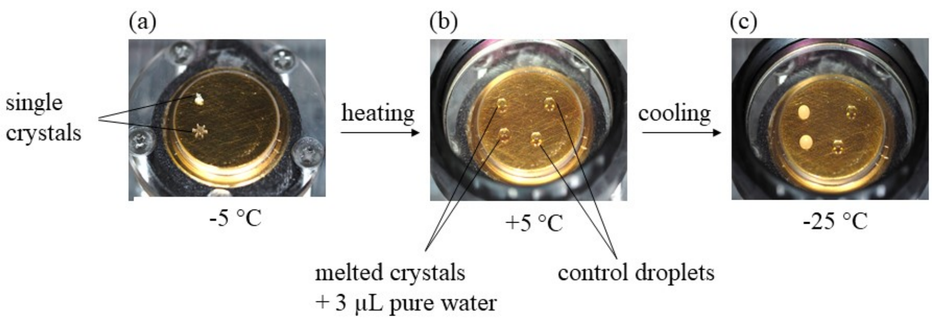

Figure 1. Illustration of a single snow crystal droplet freezing experiment. (a) two crystals on the cold stage (note: the cold stage was set to below 0 °C for this image). (b) melted ice crystals with addition of 3 µL ultrapure water to increase the detection volume (left) and two 3 µL control droplets of the same ultrapure water (right), (c) the sample and control droplets after cooling to -25 °C. Here the samples (left) were frozen, and the control droplets (right) were still liquid.

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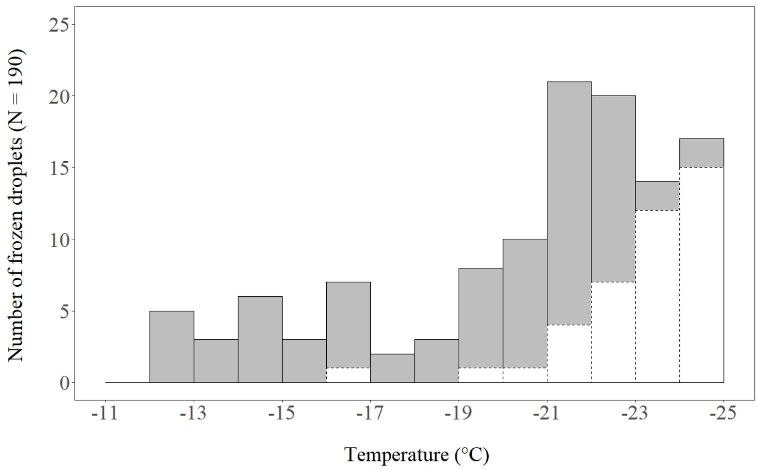

Figure 2. Number of planar, branched snow crystals that re-froze on a cold stage after having been molten (grey bars with solid contour), thereby confirming they contained an INP active within the respective 1 °C temperature step. Of 190 crystals analysed, 24 re-froze at -17 °C or warmer (INP_{-17}). The white bars with dashed contour indicate the number of frozen control droplets. The total number of control droplets was 190 as well.

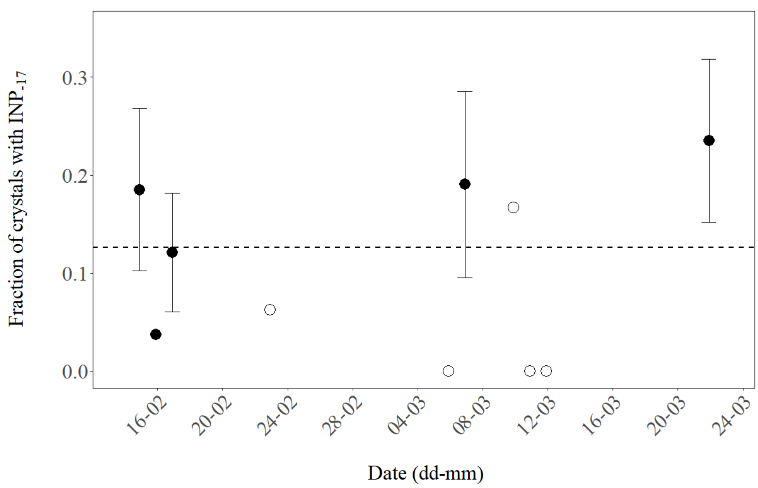

Figure 3. Daily fraction of snow crystals with INP active at -17 °C or warmer (INP_{-17}) observed for 10 days during February and March 2018. The number of crystals analysed per day was between 21 and 34 (closed symbols) or less (3 to 16, open symbols). Error bars indicate an estimate of the standard deviation (proportional to $\sqrt{INP_{-17}}$) for days when at least four crystals with INP_{-17} were found. The dashed line shows the mean value of the pooled data (190 analysed crystals).