Authors' response to Anonymous Referee #1
Review received and published: 28 August 2018

For clarity and easy visualization, the referee's comment is copied here in black. The authors' replies are in blue font with an increased indent below each of the referee's statements. Page and line numbers refer to online ACPD version.

General comments

The manuscript shows results of cold stage tests from samples taken at Jungfraujoch with the aim of illustrating secondary ice formation at an "individual hydrometeor level". These analyses could yield quantitative estimates of ice crystal enhancement, but the data are too few to make a publication-worthy conclusion in my opinion. The authors note that Hoffer and Braham attempted a similar per-particle analysis more than 50 years ago. Their sample size was 300 snow pellets, 150% that presented here, and they note in their abstract that "a firm statement could not be made as the number of observations is limited." The burden is on the authors to explain why it is sufficient to show ground-based data from only 10 days.

> We thank Anonymous Referee #1 for his/her assessment and valuable suggestions. In a 'short comment' we have already clarified the valid point about the sample size. We will discuss that the number of snow crystals found to have formed through heterogeneous freezing determines how firm a conclusion can be drawn and not the total number of snow crystals analysed and we will add the uncertainty of the multiplication factor during revisions. The uncertainty associated with the observed multiplication factor is about 20% (square root of 24 divided by 24). The detailed answer can be found in the short comment posted on 5th September 2018.

Thereafter, the introduction needs to be expanded in my opinion. Right now, there is not a thorough discussion of existing literature. Approximate values and measurement techniques for INPs and IRs in mixed-phase clouds should be mentioned, in particular the abundance of measurements from Jungfraujoch (e.g. with the Ice Selective Inlet (Kupiszewski et al. 2015), the Ice Counterflow Virtual Impactor (Mertes et al. 2007), and the Horizontal Ice Nucleation Chamber (Lacher et al. 2017) as discussed in Cziczo et al. Measurements of Ice Nucleating Particles and Ice Residuals).

> We will expand our introduction and refer to the mentioned studies to make clear already in the introduction the novelty of our approach. By sampling small ice particles (few tens of micron in aerodynamic diameter) at Jungfraujoch earlier studies were not able to separate ice which had formed in clouds from aerosolised parts of hoar frost growing on surrounding surfaces (Lloyd et al., 2015; Farrington et al., 2016; Beck et al., 2018). By sampling larger, regular ice particles (e.g. dendrites) we minimised the influence of local surfaces on our results (page 6 line 10-14) and can draw a conclusion regarding secondary ice formation within mixed-phase clouds.

The analyses also need to be fleshed out. A more complete picture of the meteorology could be given by including the range and variability of air temperatures and wind velocities during the sampling periods. If photos of all the crystals were taken with a high-quality camera, some of these should be shown. Is there a more rigorous means of classifying the crystals than what is "considered to be planar, branched"? If the size of the crystals was measured with ImageJ, could some of these statistics also be presented? In Section 2.3, there is also a mention of rime analyses with a second cold plate, but it was not clear to me how this fit in. The results shown in Figures 2 and 3 are from pristine, unrimed dendrites, right?

We will provide the range and variability of air temperatures and wind velocities in a revised version. We are ready to show representative pictures of the crystals that were taken in the main paper or all pictures as supporting information. As mentioned on page 3 line 31, we classified the crystals by habit and riming degree using the 'global classification scheme'. There are some variations in crystal shape. We considered as planar, branched crystals, crystals that can be classified into the following classes: R1c, R2c, R3a, R4a, P4, P3, P2 according to Kikuchi et al. (2013) (see first paragraph in Section 3; please note that the paper by Kikuchi et al. contains representative images of all the crystal classes mentioned in our paper). Figures 2 and 3 show the results of the 190 crystals. A large fraction of them are 'rimed dendrites' or 'densely rimed dendrites' (see page 5 line 6). Rime itself was analysed to determine the fraction of analysed crystals that possibly had scavenged through riming an INP active at -17 °C or warmer. This fraction was smaller than 1% (page 6, lines 32-33)

With additional data and stronger analysis, more could be gleaned from this study. If the cold plate measurements are subject to any contamination, then 12.6% of the droplets refreezing is actually an overestimation. And a limited crystal geometry has been used to define secondary ice; at -15°C and lower supersaturations, other geometries are possible. So perhaps the multiplication factor of 8 is more of a lower bound. Quantitative estimates of this factor are needed for models, and field measurements at the hydrometeor level, rather than the bulk cloud level, are a new, if labor intensive, technique.

With the same number of control droplets as droplets from crystals we assessed potential contamination. For temperatures at which the analysed crystals had formed (-12 °C to -17 °C) only 0.5% (1 in 190) of the droplets were contaminated. Indeed, at lower supersaturations other crystal geometries are possible at around -15 °C. However, as we were sampling within mixed-phase clouds, we were always within highly supersaturated conditions. We would like to recall that our aim was to find reliable evidence for secondary ice formation at around -15 °C in clouds. For this reason, we had to exclude as rigorously as possible the influence of secondary ice formed and aerosolised from local surfaces (e.g. hoar frost). This requirement called for selecting crystals with a regular shape that forms in clouds and a size large enough to tell they have not grown from splinters emitted locally (see page 6, lines 5-14). We agree that the estimated ice multiplication factor may therefore be a lower bound, a point we will make clear when revising the manuscript.

Specific comments

Page 1, Lines 18-20: The conclusion that "secondary ice can be observed at temperatures around -15°C" is not an especially compelling one, given that many previous studies have already shown this. Is there a hypothesized mechanism? Or was observed multiplication factor higher under certain conditions?

As far as we know, no previous study has directly observed secondary-produced ice at around -15 °C in natural mixed-phase clouds. What has been reported were large discrepancies between number concentrations of ice crystals and INPs. We could only speculate which mechanisms is responsible for the secondarily produced ice by referring to the papers by Field et al. (2017) and Sullivan et al. (2018), both cited in the manuscript. As shown in Figure 3 the daily fraction of primary crystals was relatively constant and varied around the mean value of the pooled data. When considering the uncertainty of those days where we had at least four primary crystals (black dots), their means are not distinguishable from the pooled data (mean +/- standard deviation of the pooled data).

Page 1, Line 23 – "These freezing pathways" as there can be contact or deposition or immersion freezing.

Correct, thank you.

Page 1, Line 26 – A few additional, more recent observations might be cited. For example, Lasher-Trapp et al. JAS [2016], Ladino et al. GRL [2017], and Jackson et al. ACPD [2018].

We will add them.

Page 2, Line 19 – For completeness, you could mention the correction of such shattering artifacts in more recent data by inter-arrival time algorithms and K-tip probes.

Thank you for mentioning; we will do that.

Page 2, Line 22 – I would define rime when you first discuss rime splintering above in Lines 4-5.

O.k., we will define rime there.

Section 2.2 and Figure 2 – The authors have taken a number of concerns about cold-stage measurements into consideration with their setup, which I appreciate. I would cite Tobo 2016 for the use of a semi-sold, hydrophobic substrate, and you might mention the possibility that INP settle out or aggregate within your large-volume droplet [e.g., Emerstic et al. 2015 ACP]. I am still concerned, however, that 20% of the control droplets have frozen by -25°C, almost 10°C above the threshold temperature for homogeneous freezing. Could the estimated enhancement factor be adjusted to account for these "false positives"?

The estimated enhancement factor relates to the temperature window in which the collected ice crystals were likely to have formed (-12 °C to -17 °C). In this temperature window we had only one false positive in 190 tested controls. The number of droplets frozen by -25 °C only plays a role when estimating the average mass of rime associated with a single crystal. In this estimate we have accounted for the false positives (subtracted frozen controls from frozen droplets; please see page 6, line 27).

Page 4, Line 1 – I would add a sentence that summarizes what this 'global classification scheme' is because it is not so widely used, as far as I know.

We will add a sentence that summarizes the global classification scheme.

Page 4, Line 27 – Is there a reason that the "custom-built cold stage" used for single crystal analysis was not also used for the rime?

Unlike traditional cold plate systems, the custom-built cold stage is mainly made to be easily field transportable to remote locations. It was however not used for the rime samples as it has a rather small surface (surface diameter of 18 mm, page 4 line 5). Analysing rime with it would have led to less measurement time for the single crystals. Our goal was to get as much measurement time for the single crystals as possible. This requires a cold stage which is ready to be used when the specific type of crystals precipitate. The second cold stage i.e. the NOAA Drop Freezing Cold Plate has a larger surface, therefore more droplets can be placed on it, which is convenient for the rime analysis. Please note that the NOAA Drop Freezing Cold Plate requires an external refrigerated circulation bath, lined power and is relatively large. We could not put it into the anteroom and analyse single crystals. We used the most suited cold stage type available for each sample type. We will add the reason why we used two different cold plate systems in the revised manuscript and discuss whether the results of both plates are comparable

Page 4, Lines 28-29 – I am not sure what is meant by "droplets of molten rime". You are melting the aggregation of frozen droplets and then refreezing them upon a cold plate? Or somehow separating the droplets within a single aggregate? Please clarify here.

Indeed, we are melting the aggregation of the frozen rime droplets and then refreezing them. We will clarify this in a revised manuscript.

Page 5, Lines 19-21 – Measurement uncertainty and / or variability for this estimate needs to be included.

Measurement uncertainty will be included on page 5, lines 19-21.

Page 5, Line 32-Page 6, Line 1 – The mention of INP from soils does not seem particularly relevant to me, as those will not be the INP source at Jungfraujoch.

This sentence presents one of three examples from the literature that illustrate the ice nucleation temperature stability during repeated melting and freezing and therefore we think that it is worthwhile to mention it. Besides that, we think that aerosolised soil particles, or soil dust, potentially emitted from fields in northern Italy, southern France, southern Germany, and the Swiss Plateau might make a relevant contribution to INPs active at relatively high temperatures (i.e. > -17 °C) at Jungfraujoch. Note that the most prominent particle classes (reflecting particles in the size range between 0.5 um and 5 um) determined at Jungfraujoch were carbonaceous particles (Hinz et al., 2005). Furthermore, most of the fields within the fetch of Jungfraujoch are not covered by snow during winter and wind blown dust emissions are relatively high during that season in Europe (Korcz et al., 2009).

Page 6, Lines 5-14 – Blowing snow is a very important consideration here, given several existing studies on this mechanism at Jungfraujoch. You are considering pristine dendrites here, right? Otherwise, there is the potential for riming growth, not just depositional growth.

We are considering planar, branched crystals including rimed crystals. This is the reason why we have also analysed the INP spectra of rime itself. Our results show that less than 1% of the analysed crystals may have scavenged an INP active at a similar temperature as the INP which might have catalysed the formation of a dendrite (page 6 lines 23-34).

Table 1 – For periods that last as much as 14 hours, it would be more rigorous to give mean and standard deviation for values like air temperature / wind velocity since a single value will not be characteristic. Are there are any vertical wind measurements?

O.k., we will add standard deviations for air temperature and wind velocity. No vertical wind measurements were taken though.

Authors' response to interactive comment from Anonymous Referee #1
Interactive comment received and published: 10 September

Thanks very much for your response concerning the uncertainty in the ice multiplication factor estimate given your sample. My concern is more related to the uncertainty in the representativeness of your sample for the population. Assuming that "the population" here is ice in a mixed-phase cloud, then you can estimate the population size ..say conservatively that the cloud is 2 km deep and has an equivalent radius of 3 km, then it already has a volume on the order of 10ˆ10 cubic meters. Even if the ice crystal concentration in the cloud is only a crystal per cubic meter, you have sampled a very small portion of the population for which you are making a conclusion. This is how I am thinking, but I understand that there are all sorts of subtleties related to representativeness and that your collection process is laborious, so let us see what other reviewers say.

The number of crystals we have sampled and analysed is indeed a very small fraction of all crystals in all clouds that have passed Jungfraujoch between 15 February and 22 March 2018. If we had sampled the crystals from a small fraction of a cloud volume and would extrapolate our findings to a much larger volume in which primary and secondary crystals are very heterogeneously distributed, we would face a problem. However, we have sampled crystals on 10 days in different clouds spread over a period of 36 calendar days. To estimate the total path of clouds crossing Jungfraujoch during our sampling events, we can multiply the sampling duration of an event with the mean wind speed (values in Table 1). By doing so and taking the sum of all the sampling events, we get a total path of 2368 km within clouds along which we have taken our samples. We think that this is a representative distance of cloud passage and thus a representative sample for this year's winter clouds at Jungfraujoch. Figure 2 shows that the ice multiplication factor for individual days is similar to the mean of the pooled data, considering the larger uncertainty of daily estimates as compared to the estimate for the whole period. Hence, the average of the period is not subject to bias of a single day with substantially different multiplication factor from the rest of the days.

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