Authors' response to Anonymous Referee #2
Review received and published: 11 September 2018

For clarity and easy visualization, the referee's comment is copied here in black. We have divided his/her text into numbered sections to facilitate discussion. 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.

Authors present the experimental work where they collected the snow crystals, melted the crystals and visually observed the freezing of the crystal droplet. These results were used to understand more about the secondary ice formation and ice multiplication factors. These questions are challenging, and the community needs an understanding of these cloud processes for better representation in the cloud model. However, this study lacks appropriate experimental technique/methodology to answer these questions, and for this reason, the paper is not ready for the publication. I'm not sure if the major review could improve the paper further as substantial experimental work is involved. There are a number of issues in the present experimental study.

We thank Anonymous Referee #2 for openly sharing his/her ideas on our recent manuscript. We agree that the questions about secondary ice formation and multiplication factors are challenging and that the community needs to answer them to improve cloud parametrizations in models. There are different approaches to answer these questions. The work presented here addresses them by applying an unconventional, new method. The study combines the growth temperature encoded in the habit of snow crystals with a drop freezing assay and thus complements previous observations of secondary ice formation. Our experimental technique is appropriate for detecting insoluble ice nucleation particles (INPs) in single crystals and enables us to estimate with an uncertainty of about 20% the lower bound of the ice multiplication factor in clouds during our sampling campaign at Jungfraujoch. Herewith, we would like to dispel the referee's doubts and elucidate how we will make use of the referee's comments in a revised version of the manuscript. Furthermore, we are confident that our manuscript constitutes a valuable contribution to ACP and we appreciate the opportunity to openly stand up for and constructively discuss our work.

Section1

If no INP was observed within the crystal, it does not mean that crystal was formed through secondary ice formation mechanism. It is possible that a INP may have induced nucleation of ice, and still while INP is floating within the atmosphere may have detached from the ice crystal because the crystal evaporated or through some turbulent process. Now, this crystal when sampled had no INP.

We are not aware of any literature describing the mechanisms to which this statement could refer to. Does Anonymous Referee #2 have supportive evidence for ice crystals losing their INP through evaporation, sublimation, or through "some turbulent process" in the atmosphere that are resulting in ice particles without INP?

In the atmosphere, ice nucleation has been observed at temperatures warmer than that of homogeneous freezing (Ansmann et al., 2005). Four main pathways of heterogeneous freezing have been identified: contact, deposition, condensation, and immersion freezing (Pruppacher and Klett, 1997). In our study, we investigated freezing through the immersion freezing mechanism. Immersion freezing refers to the initiation of ice nucleation by a solid and insoluble INP immersed in a water droplet. To our understanding, the immersed INP will catalyse an initial crystal, in which the INP is embedded. This initial crystal then grows through vapour deposition. In this process, the INP in the initial crystal will increasingly become encased in ice that grows thicker around it. If this crystal then begins to sublimate, the ice covering the initial droplet surrounding the INP will become thinner again, which we expect to evolve rather uniformly from the outside (i.e. edge of the crystal) towards the inside (i.e. initial droplet that froze by immersion). The INP will be released from the ice only once the ice of the very initial frozen droplet has sublimated, resulting in an INP without ice, but not in ice without an INP.

Besides that, we are also not aware of observations that show how "some turbulent processes" may detach the INP from a crystal. How should the INP get out of the crystal structure? Is there at all relevant turbulent friction at the submillimetre-scale in the free atmosphere? We would however be happy to discuss such mechanisms in our manuscript if they have a theoretical or observational basis.

Section 2

It is also possible that INP is present, but was deactivated while it went transformation (change in physical and chemical properties) during sampling, heating or droplet preparation. There are numerous studies in the literature that discusses the deactivation of INP. Such discussion is missing.

Indeed, studies exist that discuss the deactivation of INP during transformation. In our experiment, the crystals were sampled below melting temperatures, and melted or "heated" to between +1 °C and +5 °C (page 4, line 12) before being analysed within the next minutes. It is not unusual to store INPs in water at +4 °C for several hours before analysis (e.g. Wilson et al., 2015). Studies reporting deactivation through heating typically refer to heating temperatures close to the boiling point of water (e.g. Christner et al., 2008).

There is also convincing evidence in the literature that INPs, which are active at temperatures relevant for our study, can be repeatedly activated, going through multiple cycles of freezing and melting. We have discussed and referred to these studies on page 5 line 30 to page 6 line 3. Furthermore, we have clearly formulated that our findings are based on the assumption that the cited evidence also applies to our samples, see page 5 line 23.

Several laboratory studies have investigated the role of coating of mineral dust particles and the related changes in ice nucleation efficiency (e.g. Knopf and Koop 2006, Cziczo et al., 2009; Kanji et al., 2018). Soluble coating or soluble INPs could be altered through melting or droplet preparation. However, the work presented here is not investigating the effect of soluble coating and neither of soluble INPs. Soluble INPs probably do not play a role at temperatures warmer than about -27 °C (Knopf et al., 2018, see their Fig. 5). Based on the referee's comment, we will emphasize in a revised version, that we are focusing on insoluble INPs in dendrites that can be activated through immersion freezing at temperatures above -17 °C for at least two freezing cycles (one when forming the crystal and one when doing the measurement).

Section 3

Experiments are needed that investigate the ice nucleation efficiency of crystal melted droplets up to -37 degC (below this temperature homogeneous freezing is the dominant mode of ice nucleation) to understand more about the insoluble INPs, but for soluble INPs experiments should be investigated at homogeneous freezing temperatures too. Without such results, the conclusions regarding secondary ice formation cannot be inferred.

Heterogeneous freezing at temperatures below -25 °C and homogenous freezing at even colder temperatures are certainly important topics of research, especially when investigating cold mixed-phase clouds or cirrus clouds. Observations have shown that an overwhelming majority of ice particles originate from supercooled liquid clouds at temperatures > -27 °C, which strongly suggests that the initial process of ice formation in mixed-phase clouds > -27 °C occurs through immersion freezing (Westbrook and Illingworth, 2011). Therefore, we assume that homogeneous freezing does not play an important role in mixed-phase clouds surrounding Jungfraujoch during our campaign where temperatures were clearly higher (see Table 1). Further, every experimental study has a limited parameter space. We set the frame for our study in the second part of the introduction. Briefly, our objective was to detect the presence of INPs active at around -15 °C in dendrites, which typically grow around that temperature. By investigating ice nucleation down to -25 °C we already expanded our measurements well beyond the necessary to answer the question to what proportion dendrites are the result of primary ice formation.

Section 4

Supporting experiments are needed to say why there was no INPs present (page 5 line 14). It would be just that the limitation of the experimental setup. In this study, the sample collection onto the cold stage is not done in clean air conditions. It is possible that crystals were contaminated with room air particles. Further, it is possible that these particles may have induced nucleation of ice but not the primary INP (the first INP that was responsible for freezing the droplet in the atmosphere before sampling). Without knowing the composition of residue it is difficult to infer which INP (primary or room air particulates) was responsible for freezing.

Indeed, it is very important in a first step to avoid contamination as much as possible and in a second step to quantify it. We examined contamination with control droplets of molecular grade water (blanks). If contamination, including deposition of INP from the room air would have been a problem, we would have seen it in the freezing of control droplets. As shown in Fig. 2 and discussed in the text, of 190 control droplets only one froze within the temperature range where the analysed crystals may have formed (-12 °C to -17 °C). Deposition of “room air-INPs” is only one out of several possible reasons why this control droplet may have frozen. Another reason could have been surface contamination of the cold stage. Please note that the control droplets were exposed to the same room air during the same time as were our sample. Thus, even without knowing the composition of residue, we can show, with the results of the control droplets, that INPs deposited from room can not have been responsible for the freezing of the crystal droplets.

It is not a limitation of our experimental setup that no INP active around -15 °C was found in a large proportion of the analysed dendrites. A possible explanation for the absence of INPs are crystals formed through secondary ice formation processes. Our results are consistent with findings and conclusions from other studies (page 1 line 26). Several studies measured much lower INP concentrations than ice crystal number concentrations in clouds by using different approaches and measurement techniques from ours.

Section 5

It is not clear how section 2.3 supports the secondary ice formation analysis. Details such as validation and performance calibration of the cold stage (shown in Fig 1) under different temperature and humidity conditions are missing.

The majority of analysed crystals were rimed. Rime could have added INPs active at around -15 °C to initial crystals (page 2 line 23-24). Therefore, we analysed not only (rimed) crystals but also rime itself (method in section 2.3). Our results show that riming had only a very minor influence on our results (page 6 line 23-34).

The cold stage was used to test for INPs in immersion freezing mode. Details of the cold stage as well as calibration can be found in the supplement, including the result of tests at a range of temperatures. We are not sure why we should perform validation and calibration at different humidity conditions. These would play a role only, if we would study deposition or condensation freezing.

Section 6

Any results from previous studies who had attempted to study secondary ice formation should be shown in Figure 2 and 3.

It would make sense to compare our results with previous studies. However, the results of previous studies are based on completely different approaches. Their results are not directly comparable to ours. One of the main differences is that we have analysed relatively large snow crystals (several millimetres in diameter) to make sure our results are not influenced by local surface sources of secondary ice formation. We will discuss differences regarding results and methodology between previous studies and this study in more detail in a revised version of our manuscript.

Section 7

Discussion regarding nature of INP is missing. What are their composition and size? One should use Ice-CVI (Mertes et al 2007) to sample only ice crystals, sublimate/evaporate these crystals, count the residues and investigate the ice nucleation propensity of a single residue. By comparing inlet ice crystal and residue concentrations one can infer some understanding regarding secondary ice formation.

Mertes et al. (2007) sampled very small ice particles, between 5 and 20 micron (aerodynamic diameter). Lloyd et al. (2015) concluded for Jungfraujoch that “hoar frost crystals generated at the cloud enveloped snow surface could be the most important source of cloud ice concentrations.” The same may apply to other mountain stations (Beck et al., 2018). Therefore, repeating the experiments of Mertes et al. (2007) would tell us mainly about ice residues in hoar frost particles generated by local surfaces. This is not what we are interested in. We would like to know more about secondary ice formation in mixed-phase clouds themselves. This is the reason why we have sampled larger crystals with a regular shape that are unlikely to have resulted from surface processes and tested these crystals for the presence of INPs active within the temperature range they typically form.

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