In the following, the reviewer comments are written in bold and our answers in italics. Text passages from the revised manuscript are in quotation marks, modified or newly added passages are marked in green.

Author's response to Reviewer 4

First of all, we would like to thank the reviewer for reading our revised manuscript and our answers to the review comments.

The authors have addressed most of my comments. It would be helpful if the authors can comment on the follow minor issue. In your response to my last comment. You mentioned that "Certainly, these particles can also be counted as INPs in the real sample, which means that contamination particles can be included in the characterization of the atmospheric aerosol. However, since the wafers are cleaned in advance to keep the background freezing at a low level, the influence of potential contamination particles can be assumed to be minimal." Is there any number to show that the background contribution is minimal? For example, with same experimental procedure, what are the ice crystal numbers on the substrates in general after cleaning or before sampling? What are the ratios when compare to the atmospheric samples?

In general, all wafers are cleaned before they are used as sample substrates. After the cleaning process, the wafers are subject to random checks in FRIDGE (see Section 2.1). Typically, even cleaned wafers show low ice formation activity. For tropospheric measurements with typical INP concentrations (measured with FRIDGE) ranging from 0.1 to 10 L^{-1} at -30°C (Schrod et al., 2020b) and up to 100 L^{-1} in dust plumes (Schrod et al., 2017), a set of ten wafers is considered as clean if three randomly selected wafers from the set have an ice crystal number of less than ten during the -30°C measurement in FRIDGE. Background counts can subsequently be converted into a volume-dependent limit value (e.g., 0.1 L^{-1} for a sampling volume of 100 L).
Basically, background counts are always subtracted from the raw INP counts of each sample before calculating the INP concentration.
This background value can be adjusted by setting an individual threshold for defining a set of clean wafers.

Since particularly low INP concentrations were expected in the free troposphere at JFJ in advance, particular emphasis was paid to the purity of the sample substrates. The blank value for the campaign was therefore demanded to be particularly lower than ten (less than 3 ice crystals per wafer at -30°C). This results in a maximum background concentration of 0.03 L^{-1} for a collection volume of 100 L. As the collection volumes were generally larger than 100 L, the value decreases accordingly (ranging from 0.026 L^{-1} for the minimum collection volume of 115 L to 0.001 L^{-1} for the maximum collection volume of 306 L). The INP concentrations determined with FRIDGE at -30°C varied between 0.1 and 1 $stdL^{-1}$ for the JFJ samples, the background values are therefore 1 to 2 orders of magnitude smaller.

As a result, for the campaign at JFJ we assume that an average number of 3 contamination background particles per sample is activated during the FRIDGE measurement at -30°C. Of course, these particles can subsequently contribute to the physico-chemical characterization of the INPs with SEM. But assuming that only 30% of the ice crystals can be assigned a unique INP, the number of contributing background INPs would be reduced from 3 to 0.9 per sample.

Martin (2019) found indications, that the group of C-rich particles appears to be primarily affected by this contamination. However, the number of identified particles was low. Results from a recently ongoing experiment show similar results, but the number of samples still needs to be increased in order to obtain a statistically significant statement.

Martin (2019): Eiskeim-Charakterisierung von Proben des Stormpeak Laboratory in Colorado, bachelor thesis, TU Darmstadt, 2019