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Interactive comment on "Ice nucleating particle concentrations of the past: Insights from a 600 year old Greenland ice core" *by* Jann Schrod et al.

Anonymous Referee #2

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The authors have made a great effort in trying to reconstruct from the analysis of an ice core the atmospheric concentration of ice nucleating particles (INPs) in the atmosphere over Central Greenland between the years 1370 and 1990. It is only the second such attempt, after a similar but less comprehensive study published last year by another group. Overall, the manuscript is clearly written. Everything is well explained. The text is easy to follow. Data are arranged in a meaningful way in Tables and Figures. Half of the main text is description of methods. Interpretation of results is cautious, if not hesitant. It is here that I see some room for improvement, apart from a few other, minor issues.

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The most surprising outcome of this study, from my point of view, is the narrow range of INP concentrations in ice and atmosphere during a period in which Earth has seen a tenfold increase in land area used for agriculture (Pongratz et al., 2008). Ploughing of the North American prairies and the Russian steppes during the past two centuries has greatly accelerated wind erosion with drastic consequences, like the harvest faillure of 1891 in the Russian steppes (Moon, 2005) and the Dust Bowl situation in the USA during the 1930s. Also intensive grazing by exploding numbers of domesticated animals has had its share in fostering wind erosion during that time (Neff et al., 2008). Other than desert dust, soil dust from more fertile land carries INPs active at moderate supercooling (O'Sullivan et al., 2014). Therefore, I would have expected to see growing number concentrations of INP active at temperatures at around -15 °C or warmer in samples deposited over the last two centuries. However, this does not seem to be the case. Only 3% of all samples, each consisting of 0.5 mL of melted ice, contained INPs active at -15 °C. There are at least two plausible explanations for this observation. First, it could be that anthropogenically caused dust in the midlatitudes was not transported in detectable quantities to the Arctic and deposited in Central Greenland. The overwhelming majority of dust and INPs deposited in the Arctic probably originates from regions north of 60 °N in America and Eurasia, latitudes not much affected by landuse change in the past. Regions in North America located south of 60 °N probably contribute less than one percent to the total surface dust concentration in the Arctic (Groot Zwaaftink et al., 2016, Table 3 therein). Thus, large-scale landuse change and increased wind erosion of fertile soils in the midlatitudes following the colonisation of North America by settlers mainly from Europe may indeed not have had a marked effect on INPs deposited in Central Greenland, although it clearly increased dust deposition in the midlatitudes (Neff et al., 2008).

Another explanation for landuse change over the past centuries not being reflected in the INP record of the analysed ice core could be a loss of IN-activity, in particular the loss of biological INPs that dominate the spectrum at temperatures warmer -15 °C (Murray et al., 2012). Deactivation might have happened during decades and centuries in ice or during sample preparation, in particular during melting of the core and while the samples were in liquid form. Since Hartmann et al. (2019) found clearly enhanced INP activity between -5 °C and -15 °C even in some older sample (year 1484), sample preparation may be the more relevant issue. It would be interesting to know the temperature on the hot side of the instrument in which the ice core was melted. Further, for how long, in total, were samples in liquid form between the first melting of the core and INP analysis? Evidence pointing at a partial loss of INPs is in Figure 9 of the manuscript in discussion. It shows from -30 °C to -24 °C increasingly larger INP concentrations in the modern, as compared to the older samples. The relative difference between modern and older samples collapses quickly towards the warmer end of the temperatures scale. I would have expected this difference to continue increasing further until the warmest temperature is reached at which INPs are detectable. Maybe there is no difference at warmer temperatures detectable today because INPs active above -22 °C had lost their activity before INP analysis? In my experience, any challenge put to a population of INPs, such as warming or storage in water, always leads first and foremost to a loss of those INPs that are active at the warmest temperature. The warm temperature "bulge" in a cumulative INP spectrum disappears with increasing severity or duration of a challenge, resulting in the cumulative spectrum approaching a linear shape on a log-scale. The same applies to certain mineral INPs (Harrison et al., 2016, their Figure 4a, top panel). Partial deactivation most likely results in the remaining part of the INP population becoming increasingly homogenous, a guess supported by Figure 8 in the discussed manuscript: the distribution of frozen fractions at a specific temperature was much narrower for the older samples (pre- 1960) as compared to the modern samples (1960 to 1990). The majority of fragile INPs, which may have been present at the time of deposition, and still are to some extent in the samples from 1960 onwards, may have been lost, leaving behind a relatively homogenous population of very stable INPs. To summarise, very limited dust transport from the midlatitudes, where most landuse change has happened in past centuries, and deactivation of INPs active at temperatures warmer than -20 °C may explain why the concentration of INPs

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in the ice core is confined to a narrow range and does not reflect the growing human impact on land over the past few centuries. These considerations are of cause speculative, but I hope they encourage the authors to push their interpretation a bit further.

Minor issues

Page 9, lines 19-20: I am always at a loss when told that results "...should be interpreted with care." Is not every interpretation or conclusion based on empirical evidence a preliminary one and absolutely true statements only to be found within closed systems (mathematics, logic)?

Page 9, line 32: Why use the number of frozen droplets and not the number of INPs in the assay (INPs in 195 droplets) as the criterion from which to estimate uncertainty?

Page 15, line 26: The data has a lognormal distribution. Was it log-transformed before the t-test?

Conclusions section: Effects of INPs on cloud radiative properties are mentioned and I wonder whether the very small number concentrations found in the ice core, and the difference between 1960 to 1990 or before, are indeed in a range where they might lead to differences in radiative properties?

Regional sources and geographical differences in INPs may not only be accessible through the analyses of ice cores but also through modelling approaches making use of historical records of land cover.

Figure 1b: Would it be possible to indicate the season for samples with time coverage below one year?

Figure 2: I would like to see more than one background measurement.

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