Dear Reviewer,

We thank you for doing this review and for your suggestions that helped to improve our manuscript. Below, please find your original comments in blue and our responses in black. When referencing page and line numbers, we are always referring to the old version of the manuscript.

General Comment

In the current manuscript, aerosol particle samples were collected close to Punta Arenas, Chile, on Cerro Mirador at the southernmost tip of South America between May 2019 and March 2020. The objective was to evaluate the capability of these particles to act as ice nucleating particles (INP) and to identify their possible sources. Also, the INP data were complemented with meteorological data, air mass backward trajectories, and lidar observations. I consider that this study is relevant for the ice nucleation community and it is especially important for the Southern Hemisphere due to the lack of studies in this region. Also, I think that the database generated in this study is valuable because of the long-term sampling time. The present manuscript is within the ACP scope and it can be accepted for its publication once the following minor comments are considered.

Minor comments:

Line 6 Why do the authors state that 90% and 80% of the INP are proteinaceous material. I mean, how did you calculate these percentages? I could not find any chemical analysis to determine the quantity of proteinaceous material within the samples.

It is explained in P8 Line 255 that protein-based biogenic INPs will lose their ice nucleating ability when heated at 95 °C for 1 hour, so that the remaining INPs were labeled as heat-resistant INPs. By using equation (6), i.e., by relating the original INP concentration with that obtained after heating, we can calculate the proteinaceous-based INP number fraction.

Line 58 In this part the authors mention that it is important to perform INP measurements in the Southern Ocean; however, the samples were collected in a mountain close to the coast and not in

the open ocean. Therefore, it is very likely that the present results were influenced by continental aerosol sources.

How far away is Punta Arenas from Cerro Mirador? Can anthropogenic particles emitted in Punta Arenas influence the measurements performed in Cerro Mirador? Also, are there fertile soils close to your sampling site? Or why did you compare your results with O'Sullivan et al. (2014)? The description of the sampling site needs to be improved.

The Cerro Mirador measurement station is southwest of the Punta Arenas city, about 11.6 km away. We modified the description of the measurement site and added the following figure in the supplement to show the location of three measurement stations. We also added, to the sentence you refer to here: "and in the Southern Hemisphere in general."

The anthropogenic particles are not expected to influence the INP measurement for the following two reasons. (1) The backward trajectories show that the air parcel mainly came from the west to the Cerro Mirador station. The land cover on the west of Cerro Mirador is mainly forest and herbaceous vegetation, and the open ocean. (2) Anthropogenic pollution is not a major source of INP at temperatures above -20 °C, even in a very polluted megacity (Chen et al., 2018).

We modified the measurement site description and added one paragraph at the beginning of Sect. 2.1 Measurement sites to introduce the landscape of Punta Arenas and Cerro Miradors.

"Punta Arenas is one of the largest cities in the Patagonia region, with a population of more than 100000. It is about 1419 km from the coast of Antarctica. To the west of Punta Arenas is a

mountain area and the landscape is mainly forest and herbaceous vegetation, whereas to the east of Punta Arenas there is relatively flat terrain, with a landscape of herbaceous vegetation."

Considering the geographical feature of Punta Arenas and air parcel origins, the conventional thinking previous to our study was, that INPs at Punta Arenas are from the marine aerosol in the Southern Ocean. Nevertheless, Punta Arenas is indeed surrounded by at least 150 km of mountainous land interspersed with fjords in all directions, and we clearly saw higher INP concentrations than reported for the Southern Ocean, biogenic in origin, likely originating from terrestrial sources. This is the main reason why we compared our results with O'Sullivan et al. (2014) and Tobo et al. (2013). Our results also point out that already the tip of South America can be an important land source of INP in the Southern Ocean, at least in the vicinity of the continent.

Line 76 I suggest changing the word "aerosols" to aerosol particles. Also, it is mentioned that the used filters contain a pore size of 800 nm; however, afterward it is mentioned that the analysis was performed for particles >500 nm. Please explain this discrepancy.

We exchanged it to "aerosol particles".

As for the pore size, it is important to note that 800 nm pore size does not mean that particles smaller than 800 nm are not collected. There is a size-dependent collection efficiency of different pore sizes. We based our choice on the study by Soo et al (2016), who, for the here used type of filter and flow, give a collection efficiency of 96%. Moreover, we compared results for the use of different filter pore sizes (200 nm and 800 nm) on different occasions and got the same results wrt. INP concentrations. This was presented e.g., at EAC 2020 (Abstract title: "Variations in off-line filter sampling and analysis for Ice Nucleating Particle measurements") and will be included in a publication which is currently in preparation.

So it can be assumed that we collected particles down to much smaller sizes than 800 nm. However, INP may still mostly be larger particles (as e.g. found in Mason et al., 2016, Creamean et al., 2018, and Gong et al., 2020). That is at the core of using particle number concentration for particles > 500 nm for INP parameterizations, as originally suggested by DeMott et al. (2010)

(cited in our text). Therefore, for testing this parameterization by DeMott et al. (2010), we needed to use particle number concentration for particles > 500 nm as an input parameter. Therefore, it is also useful to compare particle number concentration for particles > 500 nm as derived from our in-situ ground-based measurement with those from lidar measurements, which happen to have the means to derive particle number concentrations for this size range.

Line 78 What was the used aerosol sampler?.

The sampler used in this study is a 47 mm Single Stage Filter Assembly from Savillex (https://www.savillex.com/en/product/filtration-assemblies/47-mm-single-stage-filter-assembly-14-x-14-tefzel-clamp--401-21-47-30-21-2?pageid=19).

Line 85 Can the authors please elaborate more on the sample storage protocol? Is there any chance that the samples could have experienced a memory effect when stored at -20C? For how long were the samples stored prior to the ice nucleation analysis? Did the authors evaluate the impact of long-term storage?

A memory effect due to frozen storage to our knowledge has not been reported in the literature, nor been observed by any colleague doing these kinds of studies. Instead, our past (unpublished) experience (and that of others) showed that samples degraded over the course of a couple of weeks when they were not stored frozen (both, at room temperature and also in a refrigerator at +4°C). Related issues were described by a publication by Beall et al. (2020), who recommend storage at -20°C as the method causing the least changes in INP concentrations. We mention this in our text now.

Concerning the lengths of storage, the samples were collected from May 2019 to March 2020. The samples were measured from 26 May 2020 to 18 August 2020 at TROPOS, Germany. Once the samples were taken out of the freezer, the filter sample washing and INP measurement is completed within 2 hours. Based again on Beall et al. (2020), we do generally not expect an effect from our storage on our samples.

Line 94-95 I am wondering if the PCR-tray with the sample was heated or if the heat was only applied to the sample?

We pipetted the samples into the PCR tray, sealed the PCR tray with a foil, and then heated the PCR tray together with the samples.

Line 121 I believe that "V" corresponds to the volume of the liquid deposited on each well instead of the air volume. Please clarify this.

If the "V" represents the volume of the liquid deposited on each well, then the calculated N_{INP} is the INP number concentration in the water samples. That can be used when samples such as precipitation or sea water is examined, to obtain concentration per liter of water.

Here we use the "V" as the air volume which, literally speaking, was collected into each well, so that the calculated N_{INP} is the INP number concentration in the ambient air.

Line 149 - 150 Describe in more detail how ns was calculated.

We think that equation (5) together with the text here provides the full description of what was done.

Line 175-178 This classification should be in the methods section.

The classification does not belong to the method section. The idea of the classification only starts to make sense after one has seen the measured spectra of N_{INP} in Fig. 1. Mentioning this part of the further evaluation already in the method section will disturb the logical flow. So we think it is better to keep it as it is.

Line 203 I do not understand why the authors compare their results with those from Tobo et al. (2013) and the former study was performed on aerosol particles from a forest while the present study focuses on coastal particles.

Please see our reply for the 2nd minor comment.

Line 228-229 Why did you choose these temperatures or do they refer to a range of temperatures?

These temperatures were chosen, as they cover a large temperature range and as for each of them, a considerable amount of data points exist.

We did not choose temperatures below -18 or above -8 °C because, for a large number of samples, all or no examined droplets were frozen, i.e., the related INP concentrations are outside of the detection range of our instruments, so that no data exists.

Figure 3b I did not completely get the purpose and meaning of panel b.

The boxplot in Fig. 3b shows which fraction of INP originated from biogenic particles at different temperatures (based on results from equation 6). The red line, corresponding to the right axis, shows how many samples contribute to this statistical analysis. As mentioned in our answer to your previous remark, at the very low and high temperatures, the INP concentrations of some samples are out of instrument detection limits. Therefore, the red line shows low values at very low and high temperatures.

In panel b, we show that (1) most of the INPs are biogenic particles in origin, and (2) the fraction of biological INPs is getting lower at lower temperatures.

We think we explain this clearly in the text L241-246. We however added at the end of the figure caption: “(relating to the right axis)”.

Line 269 According to Figure 4, the black dots correspond to the heated samples and the red dots to the unheated samples; however, in line 269 the authors state the opposite. Also, this is not consistent between the text in the figure caption and the figure legend.

We corrected the color in Fig.4.

Line 324 and 375 Why Samples 11 and 12 were selected for the case study? It is unclear to me what was the motivation to have the Case study and also the cluster analysis performed in Section 3.5. Is it not better to remove the Case study and to add those 2 samples to the cluster analysis shown in Section 3.5?

Sample 11 is the only sample that shows low N_{INP} during the cold season, this is why choose sample 11 for the case study. Sample 12 is just sampled close to 11, so while we expected similar conditions, the N_{INP} of Sample 12 is much higher. We have explained this in Lines 324-326.

Examining these two samples in detail is highly valuable, as sample 11 is very special. Adding this to the cluster analysis would take away the here gained information, and we think it is worth going into such detail on the comparison between these subsequent Samples 11 and 12.

Line 328 Please add the used software to get the air mass the backward trajectories in the Methods.

We added the following in P14 L329:

"Backward trajectory analyses were performed with the HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Stein et al., 2015), based on GDAS (Global Data Assimilation System) meteorological data."

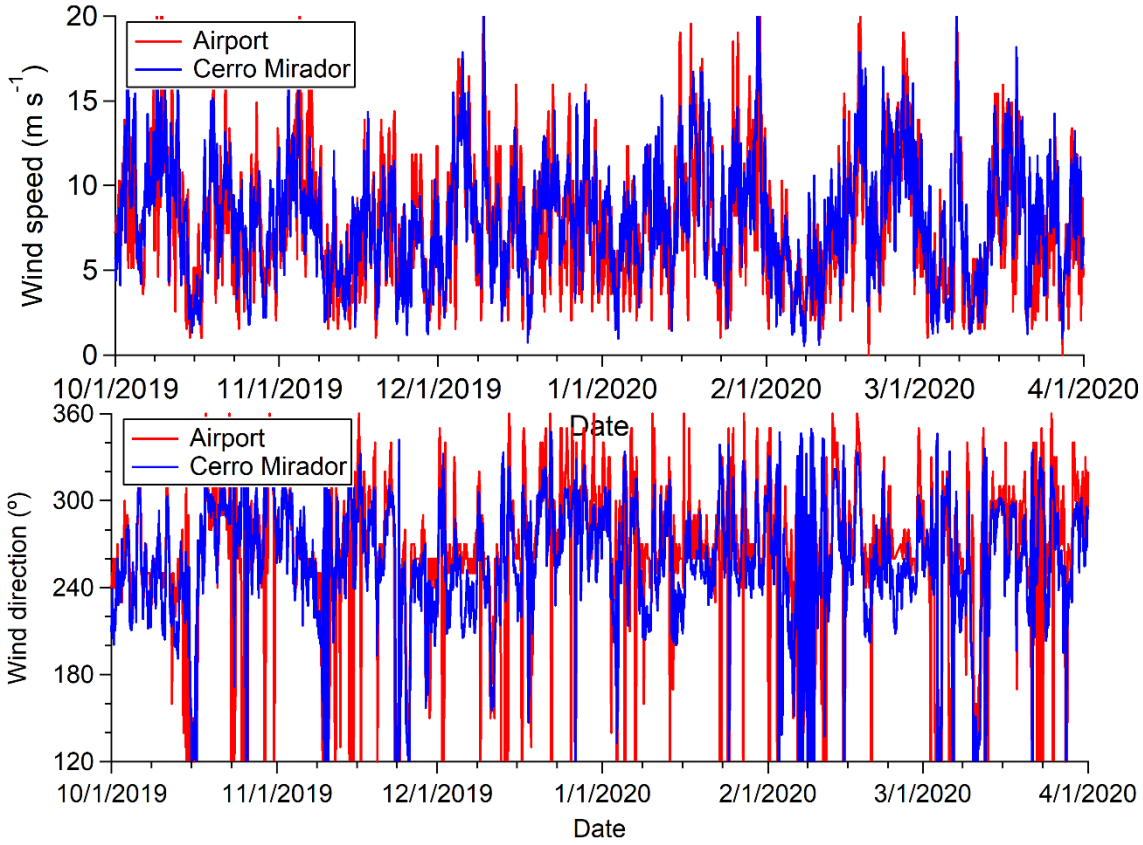
Line 330 I am wondering how valid it is to correlate the meteorological data from the airport with the INP data from the Cerro Mirador.

We performed wind direction and speed measurements at the Cerro Mirador station, but the measurement only started at end of September 2019. The figure below shows the time series of wind direction and speed at Cerro Mirador station (in red) and the airport (in blue). Wind direction and speed are comparable at both stations. When doing the comparison of meteorology parameters, we always show the probability density function over 7-14 days.

Moreover, we used the cumulative precipitation along the backward trajectory (P_{BT}). Considering the spatial resolution (1-degree latitude-longitude) of the HYSPLIT model, P_{BT} should not show a difference between the two stations. Temperature and pressure are considered

to be uniform on a larger scale. Therefore, we assume that meteorology data at the airport can be used for this study. At the end of Sect. 2.1, we included the following:

“These data were used as there was no meteorological data available at Cerro Mirador at the time of sampling. However, based on more recently installed meteorological measurements at Cerro Mirador we know, that wind speed as well as wind direction there are similar to those at the airport.”



Line 401 Based on the heat treatment analysis the authors state that some INPs were of biogenic origin; however, it is unclear what is the source of bulk INPs, i.e., those that are not of biogenic origin?

This is a good question. We assume the heat-resistance INPs in this study are mainly from soil dust, with little contribution from the sea-spray aerosol. Unfortunately, we can not test the particle composition after the heating process. As we can not give any more information on these

particles, we did not change anything in the text. If you prefer us to muse about this in the text, please let us know, but we would prefer to not do this.

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