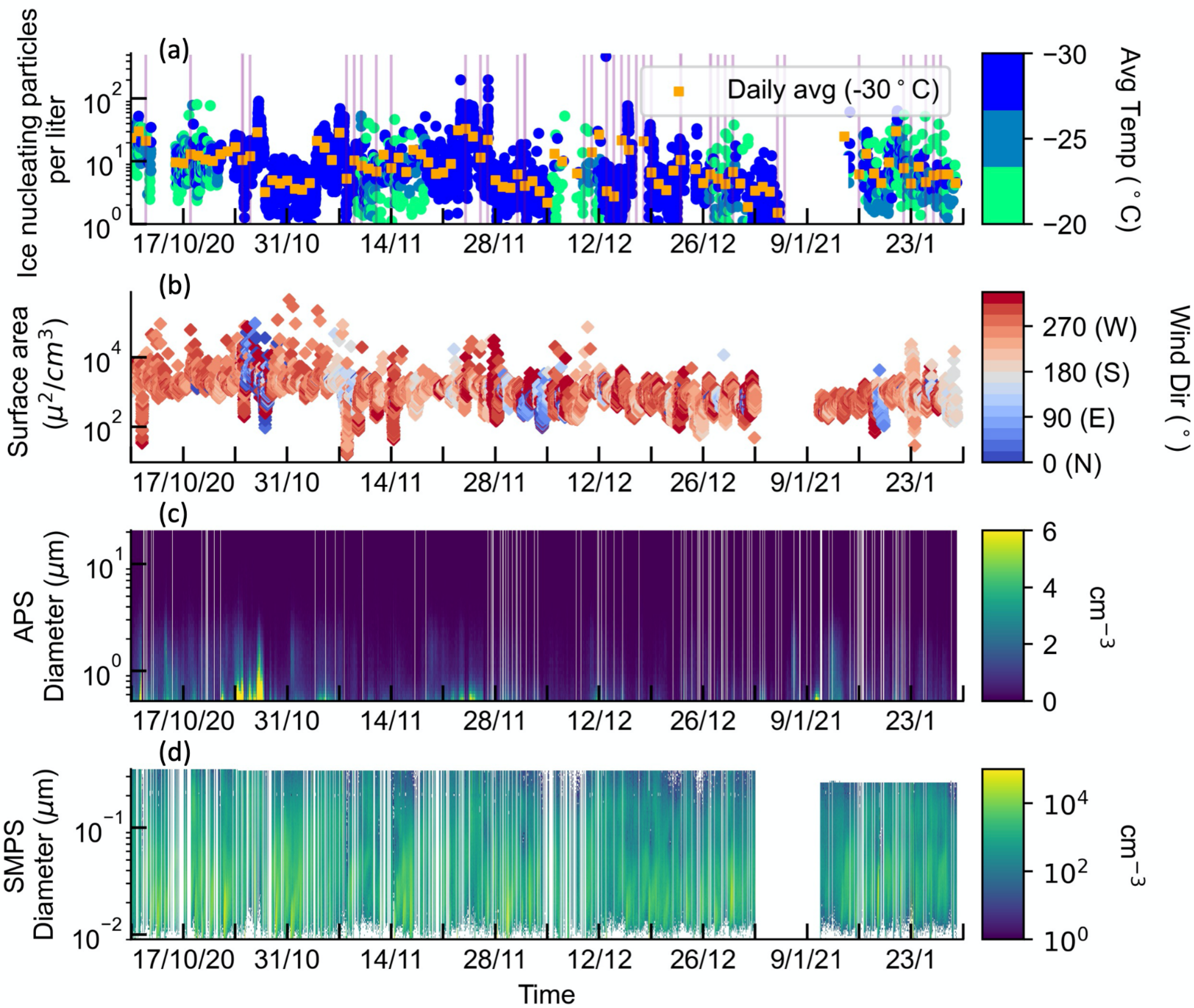
We thank the three reviewers for their comments and have some general corrections and responses before responding to specific comments below.

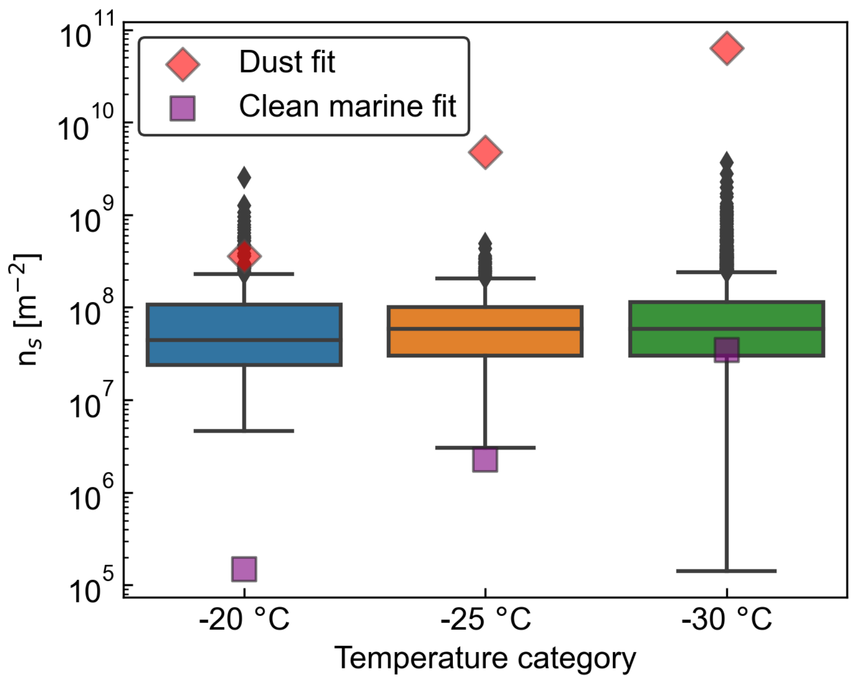
General corrections: While examining reviewer comments, we caught errors in Figure 2a and 2d:

Figure 2a: the orange line for the daily average INP concentration shown is actually a daily average across all temperatures, not just -30 deg C. We have corrected this and now show the daily average for -30 deg C.

Figure 2d: we made a mistake in plotting the SMPS data by plotting the log of the dN/dlogDp data but displaying the colorbar as if it was the log10 of the data. We have fixed this and are now correctly showing log10(dN/DlogDp) - the original colorbar indicated that the particle data went to above 10^{10} particles cm^{-3}, which would be surprisingly high for this semi-remote site in the winter! We also updated Figure 2c and 2d's colorbar labels to 'cm^{-3}' from 'number', which is more descriptive.



As well there was an error in Figure 4b: the x axis temperature categories and dust fit/clean marine fit markers were reversed. We have corrected this:



Style note: all reviewer comments are in dark grey while our discussion is in blue. We include text from the paper in indented black text blocks with new text in **bold black text**.

General response: All three reviews put significant emphasis on the limited results section of this paper. While they all raise interesting scientific questions, the analyses asked for seem more appropriate for a full scientific article in ACP, rather than a Technical note. The ACP definition of a 'technical note' is:

> "Technical notes are peer-reviewed publications that report new developments, significant advances, or novel aspects of experimental and theoretical methods and techniques that are relevant for scientific investigations within the scope of the journal (including assessments/developments of new/improved emission inventories). The title must clearly reflect the manuscript type and start with "Technical note:". For manuscripts focused on the development, intercomparison, and validation of measurement instruments and data processing techniques, we recommend submission to the EGU interactive open-access journal Atmospheric Measurement Techniques (AMT). For manuscripts focused on the development and description of numerical models and model components, we recommend submission to the EGU interactive open-access journal Geoscientific Model Development (GMD)."

Our intended purpose of this Technical note is to introduce improvements made to the Continuous Flow Diffusion Chamber and provide an overview of its measurement capabilities (e.g. several months of semi-autonomous operation to provide a high resolution INP dataset). We will focus our updates to reflect these purposes: another major area of comments from each review was upon the lack of instrument details. We agree that we fell short in this area. The dataset is publicly available and anyone interested in digging in further to provide a robust scientific analysis is welcome to do so. We provide a short list of comparisons to previous analyses (e.g. N_s, D10, D15) to put this data in context to other INP measurements. Finally - the ideas presented by the reviewers provide interesting research directions and if a researcher reaches out directly to us to discuss the dataset, we fully intend to point them to these reviews as areas for further exploration. Along with improvements made to the text based on the reviewers' comments, we end our introduction section with this statement:

> **The purpose of this Technical note is to introduce improvements made to the CFDC-IAS and provide an overview of the data collected in context to previous INP analyses and parameterizations (e.g. (Niemand et al. 2012; DeMott et al. 2010; DeMott et al. 2015; Bi et al. 2019; Lacher et al. 2018).**

In our conclusions we update a statement to say:

> This public dataset may be of interest for **more in-depth scientific analyses beyond the general overview provided here and** model-measurement comparison.

We also note in the acknowledgements:

> **We thank our three reviewers for their comments and encourage anyone interested in doing further analyses upon this dataset to read through their reviews for several excellent suggestions appropriate for a full scientific paper in ACP or elsewhere.**

Review #1

We thank Reviewer #1, Dr. Gabor Vali, for his constructive comments. As many of his comments are presented as paragraphs, we will attempt to address the major ideas presented in each paragraph.

The goal of obtaining INP measurements continuously in time and over the range of temperatures covered by tropospheric clouds is a very desirable one indeed. When accompanied by other physical and chemical data about the atmospheric aerosol such measurements could be very helpful in unraveling the many uncertainties which are obstacles to usable descriptions of ice initiation in the atmosphere. From that perspective the paper is a welcome contribution. The site selected for the measurements here reported is not ideal but has advantages. Past use of the site for similar studies helps to provide some perspective.

This paper is formulated as a Technical Note but it is hard to tell the difference in emphasis between trying to accomplish two things: to demonstrate the accomplishment of continuous operation of a modified CFDC, and to show results obtained during the period of operation. Neither goal is quite accomplished satisfactorily.

We have added more instrument details to the paper, which are detailed below.

The modifications of the CFDC are described in some detail but in a way that is hard to comprehend without intimate knowledge of the design and operation of the instrument. The basic theory of operation of the CFDC instruments is well documented in the literature. However, intimate details of the operation, specifically the cycling needed to maintain the ice coating on the walls, the avoidance of ice shedding from the walls and the drop/ice crystal thresholding are not well explained in this paper and are hard to track down in earlier papers. Thus, the technical details given for various changes are somewhat in the air. More importantly, it would have been very helpful to see more objective measures of assurance that the device was functioning correctly throughout the long sampling period. The comparison with a previously tested device is a good sign but, clearly, those tests were done with operator assistance while autonomous operation is another matter, specially in the case of an instrument that needs periodic rebuilding of the critical ice coatings of the inner surfaces.

The change to aluminum construction was done to provide surfaces that are more suitable for the formation of the ice layers without periodic treatment of the surfaces. This is a good step if it works, but raises questions about how the aluminum surface may have changed during the operating period and how that may have affected the ice coatings. Were the ice layers uniform over the whole surface? Were there patches without ice? Were there any controls, or specifics of the data that could be used to judge the constancy of the required conditions. This is readily done for wall temperature but is more subtle with respect to the saturation value accomplished. Perhaps there is no problem here but the readers should be provided by some assurances.

To address the first part of this comment, we have added the following details about CFDC operation and the modifications we have made to the instrument.

> **The fundamental objective when preparing a CFDC for measurements is to apply a uniform coating of ice on both walls to a consistent height near the top of the chamber while minimizing the formation of frost on the interior surfaces that can cause background counts to be high. When operating, the objective is to maintain the desired measurement conditions while minimizing background counts and thereby maximizing the sensitivity to detecting INP. A major advantage of the software-controlled features developed for the CFDC-IAS is its ability to perform all setup and operational procedures through automatic sequence structures pre-built into the acquisition software. The sequence first prepares the chamber for measurement by pulling a vacuum on the column to remove any liquid water or water vapor that could form frost. It then cools the walls to a set temperature (typically -27 C) and then pumps water into the column, where it freezes and forms the ice layer. The amount of water pumped into the column is monitored with a water level sensor in the supply tank to ensure a consistent fill volume, and thus consistent ice wall length. Water for the ice layer is stored in a 5 L supply tank and while a small amount of water is lost over time to growing droplets and ice crystals, additional supply water was not necessary during the SPL deployment. Once the ice walls have been formed and excess water drained from the column, the instrument automatically adjusts the wall temperatures to achieve the user-set measurement conditions and begins sampling. Over time, typically ~6 hours, the ice walls will begin to deteriorate, leading to higher instrument background and reduced sensitivity. Thus, after a set sampling period, the instrument automatically warms the walls to melt the remaining ice and repeats the cycle.**

The reviewer also raises some valid points about the consistency of the ice layer on the aluminum walls. While it is impossible to know for sure what the ice layer looks like without taking the instrument apart while it is cold, we can offer some reassurances here. First, the icing process which used to be done completely manually has now been automated (Bi et al., 2019). This means that the wall temperatures are always the same for each icing cycle and that the amount of water pumped into the column is also the same. Previously a user had to decide when the walls had reached the correct temperatures and when to turn the water pump on and off to achieve the correct pumped water volume. Automating these processes removes any user error and should lead to more consistent ice walls. Second, we can use the instrument background (ice counts measured when sampling through a HEPA filter) as an indication of ice wall quality. The previous version of the CFDC used copper walls which had to be chemically treated to produce a wettable surface. Over time this surface would break down, leading to patchy and non-stable ice layers. This was observed in the data by higher instrument backgrounds and more frequent "frost spikes", indicating that it was time to retreat the copper. We observed no such degradation in instrument background during this study. Finally, the aluminum column was disassembled after this study and we observed no visible change in the aluminum surface or its wettability, determined by running water over the surface and ensuring that the water spread out into a flat layer with no rivulets or dry patches forming. To address these concerns in the manuscript we have added the following:

> The previous copper column design required chemical treatment (ebonization) to make the surface wettable and ensure a smooth ice layer. **Over time, the surface layer would break down, leading to patchy ice layers that contributed to increasingly higher instrument backgrounds and more frequent "frost spikes", requiring the surface to be retreated**. This process was highly time consuming, requiring the entire instrument to be disassembled yearly, and required use of caustic chemicals. The aluminum column does not require this treatment, **and we observed no background degradation during this study**. To ensure that changing the column material did not alter the instrument's function, we compared the new column to the CSU-CFDC, which uses the copper design, and found good agreement between the two instruments for laboratory measurements (Figure 1). **Additionally, the aluminum column was disassembled after the deployment to SPL and no visible change in the aluminum surface or its wettability was observed.**

Bi, K., McMeeking, G. R., Ding, D. P., Levin, E. J. T., DeMott, P. J., Zhao, D. L., Wang, F., Liu, Q., Tian, P., Ma, X. C., Chen, Y. B., Huang, M. Y., Zhang, H. L., Gordon, T. D. and Chen, P.: Measurements of Ice Nucleating Particles in Beijing, China, J. Geophys. Res. Atmos., 124(14), 8065–8075, doi:10.1029/2019JD030609, 2019.

Regarding the long-term continuous record, the paper presents only a glimmer at the results. Statistics over the whole period are given in Fig. 4. The authors suggest that the 'public dataset maybe of interest ... ". This is likely to be true but the value of the dataset can't be judged from this paper. Would the overall statistics be much different if derived from intermittent sampling, say, on daily basis? How can the reader evaluate the benefits of continuous sampling? It was tempting for this reviewer to actually obtain some of the data and look for answers to the questions posed above. But, it is perhaps best done by someone with direct experience with CFDC instruments.

We agree that we are only presenting the basic data without significant analyses. However, after discussion with the editor, we view this to be appropriate for the level of a Technical note.

The analyses presented are reasonable but add little real substance. The derived surface site density values are speculative because the INP sizes are not known and may be quite different from the assumed values. The comparison with the prior parameterizations is quite short of meaningful correlations and are hard to judge because data for all temperatures are shown in saturated plots (Fig 5).

Due to the scarcity of INP measurements, other published works on INP measurements have provided comparisons to parameterizations for unknown INP types, similar to our approach such as Bi et al. (2019). We cover some basic parameterizations in our Technical note to put our results in context to other studies but leave significant study to other interested researchers.

Bi, K., McMeeking, G. R., Ding, D. P., Levin, E. J. T., DeMott, P. J., Zhao, D. L., Wang, F., Liu, Q., Tian, P., Ma, X. C., Chen, Y. B., Huang, M. Y., Zhang, H. L., Gordon, T. D. and Chen, P.: Measurements of Ice Nucleating Particles in Beijing, China, J. Geophys. Res. Atmos., 124(14), 8065–8075, doi:10.1029/2019JD030609, 2019.

The two points are, in fact, related. Examinations of the detailed data may provide some basis for judging the adequacy of instrument performance through consistency versus erratic behaviors.

Overall, what is presented in the paper does not indicate definite problems but neither does the material show robust reasons for accepting the results. The somewhat surprising lack of INP concentrations on temperature add to justified curiosity about the validity of the data. Does shorter term data show temperature dependence at any time? Or, are all INPs in the dataset activated at temperatures at or above -20°C, the highest temperature at which measurements are made?

We agree that the apparent lack of temperature dependence is surprising and may indeed be indicative of a warm-temperature INP dominating, such as biological INP. This may point to the need for filter sampling along with CFDC sampling to allow for analyses that can determine biological vs non biological INP. We have added the following statements in Sect 3.2

> **The lack of temperature dependence on INP concentrations (Figure 4a) may be indicative of the dominance of warm-temperature INP types such as biological particles (Murray et al. 2012) during the measurement period that generally activate by -20 ℃. The SPL measurement campaign did not include dedicated filter samples that could be analyzed off-line to determine the specific INP types. We suggest that the combination of a high-resolution INP record from a CFDC or similar instrument along with a lower-resolution filter sample record could readily allow for the combined analyses of temporal INP variability and INP speciation.**

It appears that the authors' main focus was making an instrument capable of continuous operation. They are likely to have accomplished that goal, or came close to it. However, for measurements that are highly sensitive to instrumental conditions it is desirable to have some controls monitoring those conditions. If such information is not available and can't be re-created in retrospect, relevant disclaimers or caveats may be necessary.

To demonstrate the nature of high-resolution data (as in the title of the paper) more detail than here given would be beneficial. The title should perhaps also indicate the temperatures for which data have been obtained.

We have included the temperature range of the measurements in the title.

Review #2

Hodshire et al, present results obtained with a newly automated CFDC for the quantification of INPs. Although, the CFDC appears to be working autonomously, which is a major feat, I have serious questions about how the INP concentration observations are reported. The lack of temperature dependence is truly surprising and goes against the previously observed and understood dependence of temperature on the ability of aerosol particles to nucleate ice in the immersion mode. Therefore, I recommend that the authors spend some time to assess the representativeness of the reported values before the manuscript is accepted into ACP. Furthermore, a deeper analysis of the factors controlling the variability in INP concentrations should be presented.

Major comments:

The lack of temperature variability in the observed INP concentrations is truly surprising. The statistical methods to achieve these results need to be discussed and presented. Based on the acknowledged limit of detection (almost 1 L-1) of the CFDC, the presented results are likely not representative of the actual temperature dependence of INPs that are observed at SPL. Please discuss this limitation on the presented results and assess how meaningful the presented values are at -20 and -25 C. With this limit of detection in place, it is clear that only the upper end of INP concentrations occurring at these temperatures will be observable. This limits the meaningfulness of the presented statistics. There are several locations in the paper where this could be discussed/improved as highlighted below.

We agree that the apparent lack of temperature dependence is surprising and may indeed be indicative of a warm-temperature INP dominating, such as biological INP. The other reviewers also point this out. This may point to the need for filter sampling along with CFDC sampling to allow for analyses that can determine biological vs non biological INP. We have added the following statements in Sect 3.2

> **The lack of temperature dependence on INP concentrations (Figure 4a) may be indicative of the dominance of warm-temperature INP types such as biological particles (Murray et al. 2012) during the measurement period that generally activate by -20 ℃. The SPL measurement campaign did not include dedicated filter samples that could be analyzed off-line to determine the specific INP types. We suggest that the combination of a high-resolution INP record from a CFDC or similar instrument along with a lower-resolution filter sample record could readily allow for the combined analyses of temporal INP variability and INP speciation.**

The point about the detection limit being around 1 is a reasonable one - however >75% of the data at each temperature occurs well above the limit of detection (25% quartile at 3, 3.8, and 3.5 sL^{-1} for -20, -25, and -30 C).

The main benefit of having high resolution and continuous INP measurements is to understand the factors that control INPs. Unfortunately, it appears as if there is no dependency on previously established controls (e.g. meteorological factors, aerosols) of INPs. Perhaps this is masked due to the discussion spanning all of the temperatures rather than only the observations at -30 C where background issues are likely less important (e.g. Brunner et al., 2021). Regardless, a deeper analysis controlling the variability of INPs should be conducted. Otherwise, the paper is more of an instrument development/proof concept (e.g. AMT paper) rather than an ACP paper.

As we are aiming for a Technical note rather than a full scientific ACP paper, we view our overview of basic results to be appropriate.

Minor comments:

Line 30: There are now a few automated CFDC measurements that have conducted continuous measurements for longer periods of time (Möhler et al., 2021; Brunner and Kanji, 2021)

We appreciate the reviewer pointing out our oversights and have updated the intro to:

The majority of single-site, longer-term (one year or longer) continuous records of INP require off-line processing and typically have daily to weekly time resolutions (Schneider et al., 2021) or may only capture short periods of each day, such as an hour or a few hours (Schrod et al., 2020b; Müller, 1969a). As discussed in (Schrod et al., 2020b), many successful longer-term records of INP occurred in the 1950s-1970s (Soulage, 1966; Kline, 1963; Bigg and Miles, 1964; Bigg, 1973; Müller, 1969b). **Only recently have high-resolution, longer-term measurements been achieved with INP measurements (Möhler et al. 2021; Brunner and Kanji 2021).** Limited understanding of how rapidly changing global emissions including anthropogenic sources may impact INP and associated radiative forcings (Schrod et al., 2020a; Boucher et al., 2013) along with limited spatial and temporal resolution of INP globally points to the importance of more frequent INP measurements.

Line 42: Again this is perhaps the first of its kind at SPL but definitely not the first automated long-term mountaintop INP measurements (Brunner and Kanji, 2021).

Agreed- we deleted the statement 'first of its kind' as similar types of measurements are being made elsewhere (Brunner and Kanji, 2021) and instead listed the temperature range of the measurements.

The four-month, high-resolution mountaintop time series **provides INP concentrations between -20 to -30 °C** and follows a successful month-long deployment in May-June 2018 of an earlier version of the CFDC-IAS in Beijing, China (Bi et al., 2019).

Line 54: The WRCC climate portal reported snowfall observations are for the town of Steamboat Springs and are not representative of what is observed at SPL. The snow depth sensors and weighing rain gauges around SPL (e.g. Tower Snotel has an average SWE of ~50 inches annually) report a much higher annual snowfall amount. Please double check this.

This is a reasonable point and we instead report the annual snowfall between 2012-2021 as observed midmountain on the ski resort. We also note that the SPL is on the Steamboat Ski and Resort Corporation (SSRC) ski area in the first sentence of this section for clarity.

While snowfall on the SSRC ski area is is highly variable from year to year, observations mid-mountain (455 m below SPL) reported an annual average of 287 inches (7289 mm) between the 2012-2021 seasons (https://www.onthesnow.com/colorado/steamboat/historical-snowfall; (Colton et al., 2008).

Line 90-93: Were the aluminum walls sanded such that they were rough and able to better retain water and subsequent ice? This might be an interesting detail to add for future CFDC development.

The aluminum walls were anodized to increase wettability, but were not sanded. We checked the surface qualitatively by running water over the aluminum pieces and ensuring that it formed a flat water layer without rivulets or droplets and that the surface remained wet for several minutes after the water source was removed. However, other CFDC type instruments have used both sandblasted and anodized aluminum (i.e. Stetzer et al. 2008) and this is a good idea for further development of this instrument.

Line 97: What determined a sampling time of 4 or 6 hours before defrosting and reicing? This is quite a difference in terms of background degradation.

Sampling time was based on the number of temperature setpoints for each measurement cycle, and was not dependent on instrument background. However, the system could be set up to automatically sample until the background reached a certain level and then perform a reicing. To clarify this in the text we have added:

The variability in sampling time was based on the number of temperature setpoints for each measurement cycle, with more setpoints leading to a longer sample cycle.

Line 99-101: A background of 1 L-1 is a significant concentration when considering that typical INP concentrations at -20 C have been previously reported to range between ~0.05 and ~1 L-1 based on precipitation samples (e.g. Petters and Wright, 2015) or between ~ 0.01 and ~100 L-1 in the air (e.g. Kanji et al., 2017). How were sampling periods where the INP concentration was below the limit of detection handled? Is this accounted for in the reported statistics?

We follow the methods excellently outlined in the supplement of Barry et al. (2021), section S.1, to determine statistically significant INP (lines 73-75 of the original manuscript) but it's fair to say that we left out a few crucial details. Specifically, all INP data shown in this manuscript were determined to be statistically significant based on the methods in Barry et al. (2021), using moment based test statistics. We have expanded on this section and removed the statement about confidence intervals, as we do not use them in this work.

We follow the statistical methods laid out in the supplemental material of (Barry et al., 2021), **which models INP counts in filter and sample periods as Poisson distributions. First a Poisson distribution rate parameter (lambda) is found:**

$$\lambda_i = \frac{x_i}{n_i} \qquad \textbf{Eq 1}$$

where x is the ice count, n is the total measuring time (s), and i stands for the filter or sample period. Statistically significant INP values are determined by using moment based

statistics to identify if a sample rate parameter is significantly greater than the filter rate parameters directly before and after the sample period. Finally, the filter-corrected INP concentration (N_{INP}) is determined by

$$N_{INP} = \frac{\lambda_{sample} - \lambda_{filter}}{\bar{v}_{sample}} \quad \textbf{Eq 2}$$

where $\bar{v}_{sample}$ is the average flow rate over the sample time period (SL s^{-1}). Here, we only present INP concentrations from sample periods determined to be statistically higher than background.

Barry, K. R., Hill, T. C. J., Levin, E. J. T., Twohy, C. H., Moore, K. A., Weller, Z. D., Toohey, D. W., Reeves, M., Campos, T., Geiss, R., Schill, G. P., Fischer, E. V., Kreidenweis, S. M. and DeMott, P. J.: Observations of Ice Nucleating Particles in the Free Troposphere From Western US Wildfires, J. Geophys. Res. Atmos., 126(3), 1–17, doi:10.1029/2020jd033752, 2021.

Line 129: What was the target saturation, was it the same for all temperatures?

Good point, we should have stated this:

Most days the instrument was set to measurement cycles of target laminar temperatures of -20, -25, and -30 °C (**target laminar water supersaturations of 3, 4, and 5%;** Figure 3).

Line 136: The lack of dependence of INP concentration on temperature here is astonishing. Furthermore, the lower estimates of the INP concentration are certainly influenced by the limit of detection (the background concentrations). This should be acknowledged here and also how measurements below the limit of detection are handled should be discussed.

We have now discussed how we remove non-statistically significant (above background) INP values from the presented data set in response to the reviewer's comments about Lines 99-101. We remind the reader of this now:

> Concentrations of INPs **found to be statistically higher than background** had a median of 6 sL^{-1} and mean of 10 sL^{-1} across all temperature ranges (-20, -25, and -30 °C) (Figure 2 and 4), but with large variation (~0-470 L^{-1}).

We addressed the reviewer's comment about the temperature dependence previously.

Line 136-138: How efficient is the inlet at sampling precipitation particles e.g. cloud droplets? If these particles are not sampled then does this indicate that the INP measurements during precipitation are of interstitial aerosols? Also, the lack of a diurnal cycle is quite striking considering results on the influence of boundary layer intrusions on INPs at other mountaintop observatories (e.g. Lacher et al., 2018; Brunner et al., 2021). Do the aerosol concentrations have a diurnal cycle?

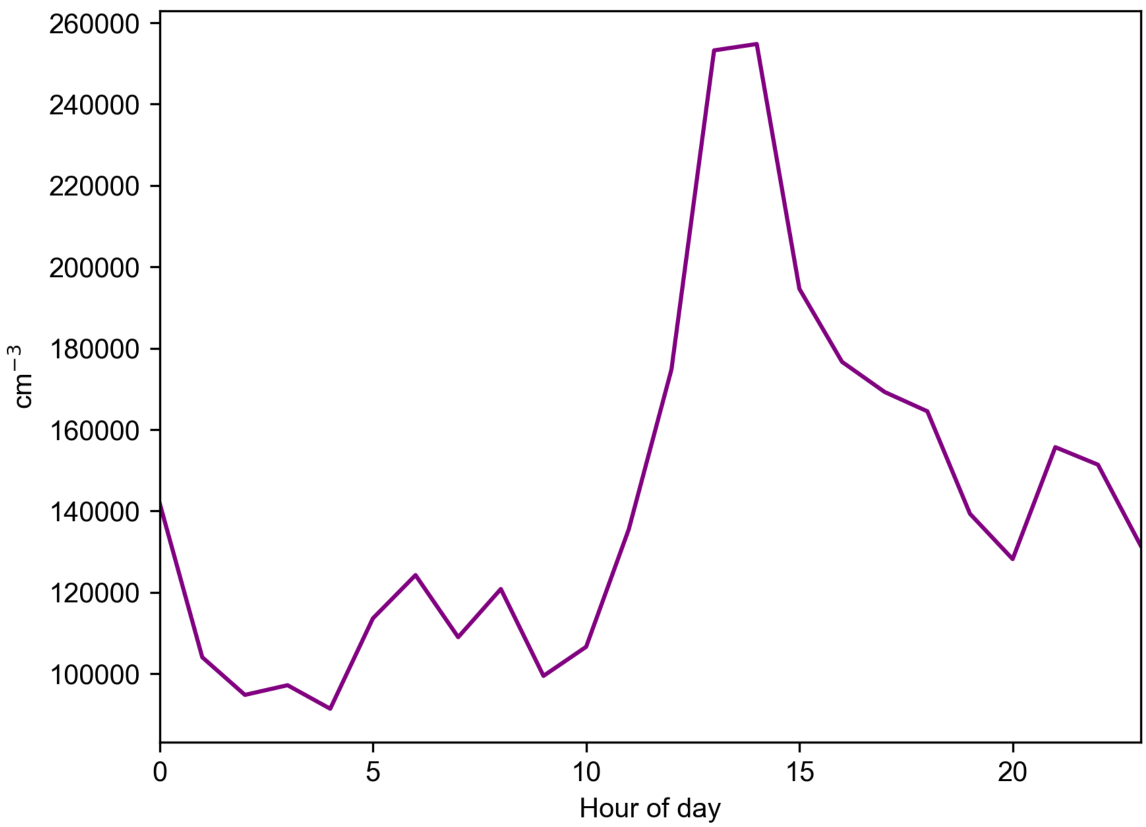
The inlet at SPL can transmit particles with an aerodynamic diameter up to 13 µm (Peterson et al. 2019). Given their low density, this can include small snowflakes or ice crystals. However, these large particles would not make it past the inlet impactors on the CFDC. Thus we assume that we are primarily measuring interstitial aerosol. We have added a line to the paper stating this as follows:

> **Although the SPL inlet can transmit particles with an aerodynamic diameter up to 13 µm, only particles smaller than 2.5 µm can make it through the CFDC-IAS inlet. Thus we assume that during cloud or precipitation events we were primarily sampling the interstitial aerosol.**

We did add additional details about the daytime versus nighttime measurements in response to reviewer 3:

> **Observations taken during the day had a lower mean INP concentration than observations taken versus during the night (9.8 sL^{-1} versus 12 sL^{-1}; Figure S1), but still had similar interquartile ranges (<20% different) between daytime and nighttime observations.**

The aerosol do appear to have a diurnal cycle if we plot the average trace of the combined SMPS and APS data throughout the CFDC deployment time period:



Line 146-148: Was the amount of precipitation along the back trajectories considered? Previous studies have suggested that precipitation can either increase or decrease INP concentrations (e.g. (Stopelli et al., 2015; Huffman et al., 2013; Mignani et al., 2021)

We did not consider the precipitation along the back trajectory in our original analysis. This is an interesting direction for future analyses to take and we note this in the text now:

> **We do not include an analysis of precipitation along the back trajectories here but this could be an area of further research, as previous studies indicate that precipitation can either increase or decrease INP concentrations (Stopelli et al. 2015; Huffman et al. 2013; Mignani et al. 2021).**

Line 170: Again, the lack in variability between -25 and -30 in ns values is truly surprising. This would indicate that the aerosol particles responsible for the observed ice activation would have the same efficiency at -25 as at -30 C. Typically, the INP concentration increases by an order of magnitude every 5 degrees (e.g. Atkinson et al., 2013; Murray et al., 2012)

We agree that the apparent lack of temperature dependence is surprising and may indeed be indicative of a warm-temperature INP dominating, such as biological INP. This may point to the need for filter sampling along with CFDC sampling to allow for analyses that can determine biological vs non biological INP. We have added the following statements in Sect 3.2

The lack of temperature dependence on INP concentrations (Figure 4a) may be indicative of the dominance of warm-temperature INP types such as biological particles (Murray et al. 2012) during the measurement period that generally activate by -20 ℃. The SPL measurement campaign did not include dedicated filter samples that could be analyzed off-line to determine the specific INP types. We suggest that the combination of a high-resolution INP record from a CFDC or similar instrument along with a lower-resolution filter sample record could readily allow for the combined analyses of temporal INP variability and INP speciation.

Figure 2: Based on panel a. it looks like there are occasions where the INP concentration is higher at warmer temperatures than colder ones. This seems unphysical and again raises the issue of the importance of the background on the measurements.

This point was raised by reviewer 3 as well - our response is:

The CFDC was periodically run only at -30 °C and it appears that coincidentally some of the lowest detected INP concentration periods of the campaign were doing those times - e.g. during Jan 2-5. We will note this in Sect 3.1:

Periodically, the instrument was instead set to run at only -30 °C to test instrument performance at one temperature, including between October 22 to November 8 2020, November 19 to December 4 2020, December 13 to December 25 2020, and January 2 to January 5 2021.

We address the apparent contradiction in INP concentrations in Sect 3.2:

Coincidentally, some of the cleanest periods of the campaign happened during times that the CFDC was run only at -30 °C, leading to the apparent contradiction of -30 °C measurements including the lowest INP measurements (Figure 4a).

Figure 4: It would be worth including the number of statistically significant observations used to make the box and whisker plots for each set temperature.

We have updated the figure captions (Figure 4 and Figure S1) to reflect this information:

Figure 4. a) All statistically significant INP observed at Storm Peak Laboratory, by temperature range. **The number of samples (N) for each boxplot is N=385 for -20 °C,**

N=445 for -25 °C , and N= 2993 for -30 °C. b) Surface active site density, n_s, using aerosol surface area from the SMPS and APS. The dust fit is from Ullrich et al., (2017) (their equation 5) and McCluskey et al. (2018) (outlined in their Sect. 3.5). **The number of samples (N) for each boxplot is N=314 for -20 °C, N=380 for -25 °C , and N= 2453 for -30 °C.**

Figure S1. a) All statistically significant INP observed at Storm Peak Laboratory, separated by nighttime and daytime observations (daytime assumed to be between 7 am – 6 pm, MST). **The number of samples (N) for each boxplot is N=1958 for night and N=1865 for day**. b) All statistically significant INP observed at Storm Peak Laboratory, separated by whether or not the data was flagged to be out of cloud (<90% RH) or in cloud (>90% RH). **The number of samples (N) for each boxplot is N=1941 for out of cloud and N=1882 for in cloud.**

Technical comments:

Figure 1: The legend has filled markers yet the figure has open markers. Also, it might be worthwhile to add uncertainties to the reported data points to account for uncertainties in temperature.

Good catch - we have updated the figure to have consistent markers. We have also added x error bars to indicate +/- one standard deviation in temperature measured during the measurement interval (approximately 5 minutes).

We have added the following text to the Figure 1 caption:

> **The x-axis error bars indicate the +/- one standard deviation in temperature measured during the measurement interval (approximately 5 minutes).**

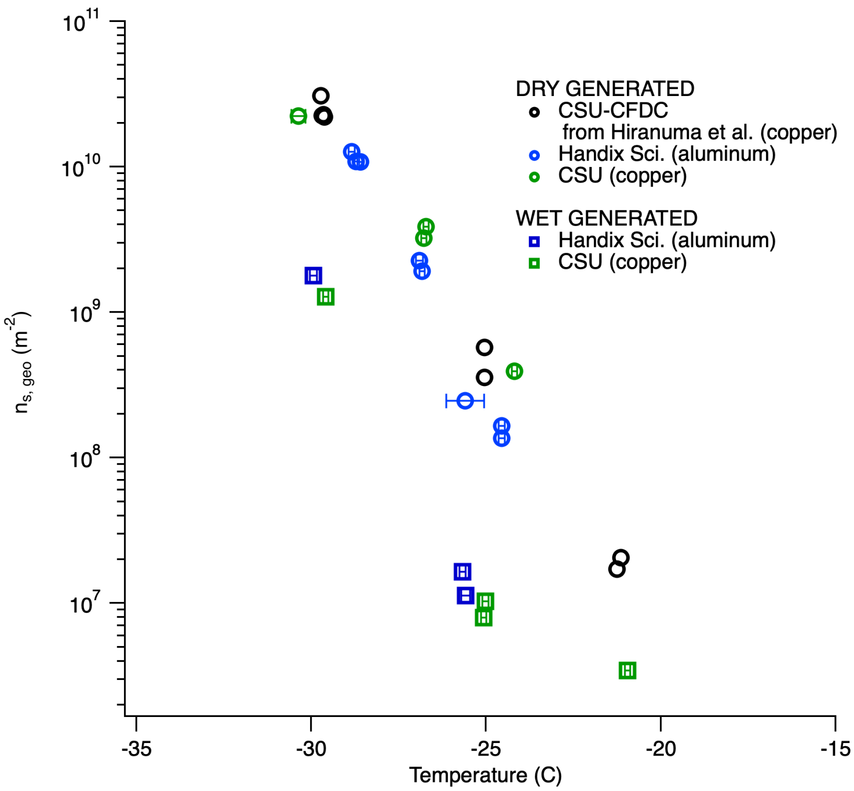


Figure 2: As only three set temperatures were investigated, consider switching to a discrete color bar rather than a continuous one.

Great suggestion: we have updated the figure (provided at the start of this response document).

References:

Atkinson, J. D., Murray, B. J., Woodhouse, M. T., Whale, T. F., Baustian, K. J., Carslaw, K. S., Dobbie, S., O'Sullivan, D., and Malkin, T. L.: The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds, Nature, 498, 355, https://doi.org/10.1038/nature12278, 2013.

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Review of Hodshire et al. (2022): Technical Note: A High-Resolution Autonomous Record of Ice Nuclei Concentrations for Fall and Winter at Storm Peak Laboratory

General comments:

In this technical note, the authors present modifications to an automated, near-continuous INP counter, the Handix Scientific CFDC Ice Activation Spectrometer (CFDC-IAS), and data measured with the CFDC-IAS for four months at the Storm Peak Laboratory (SPL). Specifically, measurements of atmospheric INP concentrations at 10-minute resolution are presented between October 9, 2020, and January 29, 2021, with the chamber in operation for the entire period except January 3-10. Parallel APS and SMPS measurements at SPL allowed estimation of the density of active surface sites. In addition, a period of high and low INP concentration was qualitatively analyzed using NOAA HYSPLIT back trajectories.

The writing (from an editorial standpoint) is to be commended. However, relevant technical details on the design changes are missing for a technical note. Validation of the changes is lean, but what is presented is solid. The discussion of validation is brief and must be done by the reader through study of Figure 1 itself. There is too much information on atmospheric parameters other than INP concentrations (e.g., Fig. 2b-d) for a technical note. It is debatable whether the single design change qualifies for a technical note or whether it would be better included in a manuscript with an in-depth analysis of its measurements. Potentially, the manuscript aims for the latter, but in my opinion misses to qualify, as the manuscript fails to analyze and discuss important elements. An example is the observed lower INP concentrations at -30 °C than at -25 °C and -20 °C, which does not reflect the consensus of previous studies and may indicate an invalid measurement methodology, thus, should be critically reviewed by the authors. The topic of the paper is well suited for ACP. However, I suggest that the manuscript undergo a major revision to reflect the following comments:

Major comments:

1. Please provide more technical information on the design changes. Also, relevant parameters and statistics are missing for an automated near-continuous INP counter. E.g.: how long are the gaps needed to renew the ice layer? What is the percentage of atmospheric measurements within the total time (atmospheric measurements divided by total time including atmospheric measurements, background measurements, cooling, warming, or temperature compensation periods, ice layer renewal, and maintenance)? How does the signal-to-noise ratio change over time (not only qualitatively, but also quantitatively)? What happens with the water needed to form the ice layer and how is it recycled?

The changes to the CFDC to allow for autonomous operation are described in detail in Bi et al. (2019). We have also added more details to this paper in response to Reviewer 1, above.

As discussed in the paper, the main changes between this instrument and the one used in Bi et al. is the change from a copper column to aluminum, which does not require periodic treatment to achieve a wettable surface.

During this deployment the instrument operation time was almost evenly split between atmospheric measurements (31% of the time), filter measurements (37% of the time), and defrosting/reicing intervals (33% of the time). While the amount of time needed to refresh the ice layer is relatively fixed, the split between filter and sample measurement time is a user setting. For this field campaign we chose to set both the filter and sample periods to 5 min as we were interested in both the ambient measurements as well as changes in instrument function over time (the slightly longer percentage of time on filter was because the inlet was set to "Filter" when changing sampling temperatures). However, users could choose to perform ambient sampling for 10 min with 5 min filter periods, for example, to increase the amount of atmospheric measurement time. We added the following text:

The variability in sampling time was based on the number of temperature setpoints for each measurement cycle, with more setpoints leading to a longer sample cycle. During the SPL deployment, instrument operation time was almost evenly split between atmospheric measurements (31%), filter measurements (37%), and defrosting/refreezing intervals (33%). While the amount of time needed to refresh the ice layer is relatively fixed, the split between filter and sample measurement time is a user setting. For this field campaign we chose to set both the filter and sample periods to 5 min as we were interested in both the ambient measurements as well as changes in instrument function over time (the slightly longer percentage of time on filter was because the inlet was set to "Filter" when changing sampling temperatures).

Water recycling is discussed below.

The signal-to-noise question is a reasonable one but maybe not as relevant here. We think that the reviewer is curious how the background degrades over time - in the original text we state "Instrument background concentrations were on average less than 1 L-1 at STP (denoted sL) at the beginning of an operation period and steadily increased, occasionally reaching average concentrations of 2-12 sL-1." We then take the background into account when we are conducting the statistics that determine INP concentrations for a set sampling interval (~5 minutes) (Section 2.2). We have added more details to Sect 2.2. To help make our steps here more clear. For this work, we have removed any INP intervals that are not statistically significant above the background (filter measurement), which is about 34% of the data. We added this note -

From these calculations, 34% of the SPL CFDC-IAS data was determined to not be statistically higher than the background.

2. Are there any indications for the lower INP concentrations at -30 °C compared to -20 °C? This does not reflect the consensus of previous studies and may indicate an invalid measurement methodology, thus, should be critically reviewed by the authors.

   The CFDC was periodically run only at -30 °C and it appears that coincidentally some of the lowest detected INP concentration periods of the campaign were doing those times - e.g. during Jan 2-5. We will note this in Sect 3.1:

   > **Periodically, the instrument was instead set to run at only -30 °C to test instrument performance at one temperature, including between October 22 to November 8 2020, November 19 to December 4 2020, December 13 to December 25 2020, and January 2 to January 5 2021.**

   We address the apparent contradiction in INP concentrations in Sect 3.2:

   > **Coincidentally, some of the cleanest periods of the campaign happened during times that the CFDC was run only at -30 °C, leading to the apparent contradiction of -30 °C measurements including the lowest INP measurements (Figure 4a).**

3. Please provide numbers and discuss the implications in more detail. For example, lines 136-138 state that there was little difference in medians or IQR between observations inside and outside clouds or between daytime and nighttime observations, but lack a more detailed discussion, e.g., of what these results mean and what the causes might be when comparing observed patterns of total aerosol number concentrations or other aerosol quantities. Missing discussions on implications are also true throughout the manuscript. Analysis using back trajectories has been done extensively in numerous previous studies. However, new insights remain missing, and thus, the back trajectory analysis can be left out from the manuscript. In addition, the advantage of continuous, high-resolution INP measurements is not exploited as only two events were analyzed.

   Again we note that this manuscript is intended to be a Technical note that provides some simple overviews and comparisons to previous methods.

Specific comments:

Abstract (lines 15-21): a very concise summary. However, it lacks an introductory sentence or two on why measuring atmospheric INP concentrations is relevant. Since the focus of the manuscript is on autonomous INP measurements, more information should be provided on the frequency of site visits required (e.g., 1/week to replace desiccant).

As mentioned in the paper, the instrument was only serviced once per week by a technician who changed out the desiccant in the diffusion driers and replaced the nitrogen tank when needed (roughly every 3 weeks). The instrument was also monitored remotely via TeamViewer with occasional changes to the sampling routine (i.e. changing the number of temperature setpoints for each sampling cycle) to investigate instrument operation. However, this is not a requirement for autonomous operation. We have added the following sentences to the abstract:

> **Ice nucleating particles (INPs) influence the timing and amount of precipitation in mixed-phase clouds by acting as seeds for supercooled liquid droplets to form ice upon.**

And

> **The CFDC-IAS was served once a week by a technician who changed out diffusion dryer desiccant and replaced the nitrogen tank as needed, and otherwise was operated remotely for desired changes in the sampling routine.**

Chapter 2.1 (line 56): please provide the amount of snow in standard international units (mm).

Updated

Chapter 2.2 (lines 95-98): please provide measurements or estimations of the transmission fraction of particles <2.5 μm through the sample line and the diffusion dryers until entering the CFDC.

The inlet geometry and flow path for this instrument are the same as previous versions of the CFDC. In the lab we have measured losses of ~10% for particles less than 2.5 μm in diameter (e.g. Prenni et al. 2009). We have added this statement to the text.

Prenni, A. J., P. J. DeMott, D. C. Rogers, S. M. Kreidenweis, G. M. McFarquhar, G. Zhang, and M. R. Poellot, 2009: Ice nuclei characteristics from M-PACE and their relation to ice formation in clouds. Tellus, 61B, DOI: 10.1111/j.1600-0889.2009.00415.x, 436-448.

Chapter 2.2 (lines 95-108): I infer that the only supply needed to run the chamber continuously are electric energy, desiccant and nitrogen. How is the water for the ice layer recycled?

As well as electricity, desiccant, and nitrogen, the instrument requires DI water and some coolant fluid (we used ethylene glycol for this deployment but have also used ethanol and silicone oil). For very long deployments additional water or cooling liquid may need to be supplied after the initial setup, but that was not the case during this four month time period. The water for the ice layer is stored in a 5L supply tank. During the icing procedure it is pumped into the column and excess water drains back into the tank. During the melting cycle, the water is again drained back into the supply tank. A tiny amount of water is lost over time, as it is the vapor source to grow droplets and ice crystals, so eventually additional water will need to be added to the supply tank. However, again, this was not needed during this deployment. We have added the following text that also addresses comments from Rev 1:

> **The fundamental objective when preparing a CFDC for measurements is to apply a uniform coating of ice on both walls to a consistent height near the top of the chamber while minimizing the formation of frost on the interior surfaces that can cause background counts to be high. When operating, the objective is to maintain the desired measurement conditions while minimizing background counts and thereby maximizing the sensitivity to detecting INP. A major advantage of the software-controlled features developed for the CFDC-IAS is its ability to perform all setup and operational procedures through automatic sequence structures pre-built into the acquisition software. The sequence first prepares the chamber for measurement by pulling a vacuum on the column to remove any liquid water or water vapor that could form frost. It then cools the walls to a set temperature (typically -27 C) and then pumps water into the column, where it freezes and forms the ice layer. The amount of water pumped into the column is monitored with a water level sensor in the supply tank to ensure a consistent fill volume, and thus consistent ice wall length. Water for the ice layer is stored in a 5 L supply tank and while a small amount of water is lost over time to growing droplets and ice crystals, additional supply water was not necessary during the SPL deployment. Once the ice**

walls have been formed and excess water drained from the column, the instrument automatically adjusts the wall temperatures to achieve the user-set measurement conditions and begins sampling. Over time, typically ~6 hours, the ice walls will begin to deteriorate, leading to higher instrument background and reduced sensitivity. Thus, after a set sampling period, the instrument automatically warms the walls to melt the remaining ice and repeats the cycle.

Chapter 3.2 (line 136): Please quantify "little difference" for both cases (in cloud vs. out of cloud and night vs. day).

Agreed that we didn't do this justice. We have fleshed out this statement :

> Little **(<10%)** difference in medians or interquartile ranges occurred for observations taken in cloud (observations at >90% RH) versus out of cloud **(Figure S1). Observations taken during the day had a lower mean INP concentration than observations taken versus during the night (9.8 sL^{-1} versus 12 sL^{-1} ; Figure S1), but still had similar interquartile ranges (<20% different) between daytime and nighttime observations.**

Chapter 3.2 (line 142): Please quantify "No strong correlation" for all cases.

> No strong correlation **(correlation coefficient > 0.8; [(Evans 1996)])**

Chapter 3.2 (lines 143-148): Food for thought: Given the large amount of data collected over the four months, more than a qualitative comparison of two periods would have been interesting.

We feel the details presented are appropriate for a Technical note in ACP.

Chapter 3.2 (lines 143-148): Whether it is a qualitative comparison of two periods or a quantitative analysis of the entire four months, the limitations of the used tools must be addressed: what are the limitations of the analysis using back trajectories from a 1-degree GDAS reanalysis, since they are unlikely to fully resolve local features? Please explain the degree of uncertainty in the back trajectories used, e.g., by referring to previous studies where this has been analyzed at SPL.

The details asked for here extend beyond the simple analysis we provide here.

Chapter 3.2 (lines 151-153): "Other high-elevation free tropospheric INP measurements…" implies, the presented measurements at SPL were sampling free tropospheric air masses. Please provide quantitative evidence for this statement.

This is a good point - we do not have the available data to definitively state when we are in the free troposphere or not. We have updated this section to reflect this uncertainty:

> Other high-elevation INP measurements also tend to fall within <1-10 sL^{-1}, with some sites reaching ~20-30 sL^{-1} (Lacher et al., 2018). The discrepancy in maximum concentrations observed between our study and other SPL and high-elevation studies may be due to differences in sampling frequency and duration: our measurements provide better temporal coverage than previous measurements at SPL, increasing the likelihood that we would be able to capture short-term, high-concentration events. **As well, the high-elevation measurements in (Lacher et al. 2018) focus on free-troposphere measurements while we do not have available measurements to definitively state when we are in or out of the free troposphere and instead only estimate that nighttime measurements are likely to be in the free troposphere. The SPL nighttime-only median INP concentrations were 17% higher than the campaign median of 6 sL^{-1}, at 7 sL^{-1}, still well within the range of (Lacher et al. 2018).**

Chapter 3.2 (lines 151-156) and chapter 4 (lines 197-198): There are interesting statements within these lines: short-term, high concentration events that were not picked up by previous measurement techniques. Please elaborate on these events. How frequent were they observed? How long did they last? Where there co-located signals in other aerosol parameter such as total number concentration or spikes in aerosol in a specific size bin? Can local pollution be ruled out? To my understanding, such brief events should also be captured during the three times longer sample duration used in, e.g., Lacher et al. (2018) or Brunner et al. (2021). Brunner et al. (2021) also measured continuously for one year, so their measurements should provide near identical temporal resolution. What are other reasons that other studies have missed these short events?

The reviewer provides some excellent ideas for a more in-depth analysis that we hope will someday be explored.

Chapter 3.2 (lines 163-171): As there is much emphasis on surface active site density, I would suggest to show a time series of ns and discuss ns in more detail. E.g., also looking at the number concentration of large particles. Furthermore, the fact that ns for INP at -25 °C and -30 °C are identical is striking and does not align with previous studies. This should be discussed in more detail in addition to the major comment #2.

We present n_s results to allow them to be compared to other INP studies but do not think that a deeper analysis is necessary here for the level of a Technical note. The similarities between n_s for

-25 and -30 C may again be related to the fact that the CFDC was periodically run only at -30 C for several days in a row. We note this now:

> **The lack of a significant difference in the median value of n_s between -25 and -30 °C may again be due to the fact that the CFDC-IAS was periodically ran only at -30 °C for several days in a row, and these days included some of the lowest INP concentrations of the campaign.**

Chapter 4 (lines 198-199): What are the different transport patterns between this and previous campaigns? Please elaborate in Chapter 3.

To do a proper job of addressing this question, we would need to do a day by day analysis of transport patterns for all campaigns discussed, which is more the purview of a review paper than a Technical note.

Figure 3: Please add the year to the title of the figure.

Fixed.

Figure 4: In panel a, the °-symbol is missing and in panel b, the unit of the temperature categories remains missing. For consistency, I would suggest to add the units to the axis labels (e.g., Temperature category [°C]). Also, "temperature category" form the axis labels is once capitalized and once not. Please use a consistent style.

We have corrected these inconsistencies, thank you for pointing them out. As well, as mentioned in the general corrections section, for Figure 4b the x axis temperature categories and dust fit/clean marine fit markers were reversed. We have corrected this.

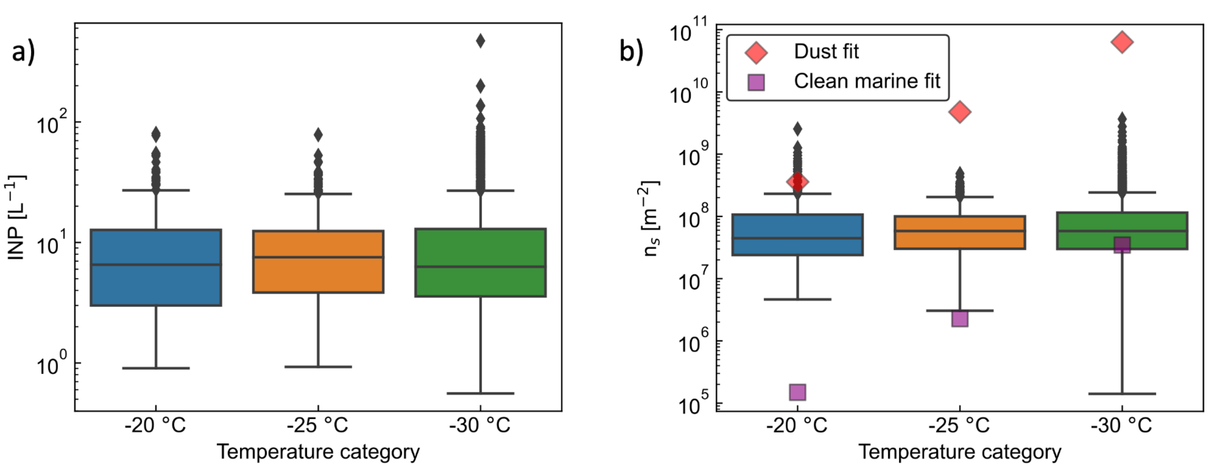
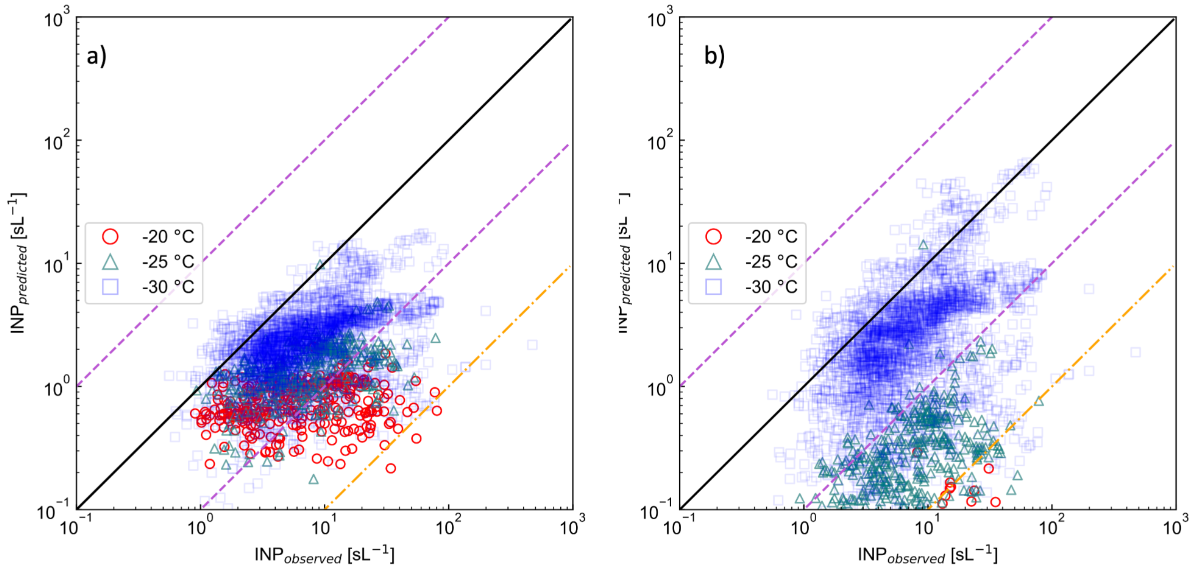


Figure 5: The data points at -25 °C and -20 °C are shadowed by the data points at -30 °C. I would suggest adding transparency to the markers, such that more information is visible.

Good suggestion - we have updated transparency and colors to make Figure 5 more clear.



Literature:

Brunner, C., Brem, B. T., Collaud Coen, M., Conen, F., Steinbacher, M., Gysel-Beer, M., and Kanji, Z. A.: The diurnal and seasonal variability of ice nucleating particles at the High Altitude Station Jungfraujoch (3580 m a.s.l.), Switzerland, Atmos. Chem. Phys. Discuss. [preprint], https://doi.org/10.5194/acp-2021-710, in review, 2021.

Lacher, L., DeMott, P. J., Levin, E. J. T., Suski, K. J., Boose, Y., Zipori, A., Herrmann, E., Bukowiecki, N., Steinbacher, M., Gute, E., Abbatt, J. P. D., Lohmann, U., and Kanji, Z. A.: Background freeâtropospheric ice nucleating particle concentrations at mixedâphase cloud conditions, J. Geophys. Res., 123, 10,506–10,525, 2018.