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## **Comment on amt-2022-216**

Anonymous Referee #2

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Referee comment on "A High-Resolution Record of Ice Nuclei Concentrations Between  $-20$  to  $-30$  °C for Fall and Winter at Storm Peak Laboratory with the autonomous Continuous Flow Diffusion Chamber Ice Activation Spectrometer" by Anna L. Hodshire et al., Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2022-216-RC2>, 2022

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To allow for long term deployment, the chamber material of the CSU type CFDC was changed from copper to aluminium to avoid the need for chamber maintenance. No change in performance of the aluminium chamber in compared to a copper version was found in a laboratory intercomparison. A 4-month time series of high frequency INP concentration observations at Storm Peak with the aluminium CFDC running autonomously, are presented together with a preliminary analysis of correlations to auxiliary data to learn about potential sources and characteristics of the INP.

### **General comment:**

The scope of the manuscript is unclear. It is neither an instrumentation paper nor a complete scientific analysis of the data. The instrument should be more thoroughly described, its operation summarized, and performance characterized to give confidence in measurements with it. From a technical aspect, novelty is lacking as the operation and automation of the CFDC has been introduced previously in Bi et al., 2019 Sec. 2.2 and the working principle of the CFDC itself in Rogers et al., 2001. Instead of using the 4-month time series to characterize the instrument performance in depth (evolution of background over time and its dependence on measurement conditions or ambient conditions, stability of flows and wall temperatures, optimization of measurement sequences, etc.), the dataset is analysed to obtain scientific results.

I suggest major revision of the manuscript to put emphasis on quantifying the performance of the CFDC running autonomously and to characterize the limits of detection. Such information would add confidence in future investigations with the instrument.

## **Specific comments:**

The title is describing a dataset not a measurement technique. A more concise title would be better. Something like "Autonomous CFDC for high-resolution INP measurements".

Line 19: It seems exaggerated to call switching from a copper to an aluminium chamber a "significant update for the CFDC to run autonomously".

Line 38ff: Elaborate how the mentioned aspects "globally point to the importance of INP measurements".

Line 46: point out that the modifications that allow autonomous operation were previously reported in Bi et al., 2019 and that the latest modification was made to enable operation without annual maintenance.

Methods: Some paragraphs in the Method section (e.g. line 108-116) read very similar to parts of Sec. 2.2. in Bi et al., 2019. In addition to the description of steps for chamber preparation and functional principle of the chamber, a complete summary of the mechanical parts that allowing automatization should be included. A technical drawing could be informative. Also, information on sample and sheath flow rates are needed, for example to estimate the range of measurable INP concentrations. How sheath flows are conditioned could be of interest.

Line 59ff: What chillers are used to cool the walls? Give model. How is the temperature in the lower region controlled? Did the cooling fluid in the chillers need replacement during the 4-month campaign?

Line 62: Specify the temperature of the lower region. Is it kept colder or warmer than the lamina temperature in the upper region?

Line 63: Elaborate based on what criteria ice crystals are detected.

Line 74: How can pulling a vacuum before filling the chamber with water help to prevent frost formation after icing? Where in the chamber would the frost form?

Line 79: How does the chamber switch between water pump, vacuum pump, OPC and

sample pump?

Line 84: More detail than available in the Bi et al., 2019 paper should be included here. Currently the description in Bi et al., 2019 is more complete.

Figure 1: scaling the data with surface area is unnecessary as the data is not compared to literature. Reporting the activated fraction would be better.

Line 97ff: The background counts should be compared to the ice counts during sampling by plotting two histograms (using the same x-axis) showing the frequency of ice counts during e.g., 1 min (equivalent to your lambda) for sampling and background. I would expect two, slightly overlapping distributions. I suggest making 3 figures like that, one for each temperature.

From these figures it could be shown how background counts change with temperature, if a Poisson distribution is justified for ice counts during sampling, if higher background counts at lower temperatures masked the increase in INP concentration, etc.

Line 123: Wouldn't the 2 inlet dryers mentioned above evaporate small droplets and ice crystals so that also their residuals are in the aerosol sampled into the CFDC (and not only interstitial aerosol)?

Line 125: plot the background increase over time for the -30°C-only measurement periods. Elaborate how the background counts effect the upper and lower detection limit. A theoretical determination of the upper and lower detection limit as function of background counts could be an instructive figure. An evaluation of the influence of the length of sampling intervals would be interesting too.

Line 129: How long does it take to refresh the ice layer and what are the limiting factors?

Line 132: How long does the transition from sample to filter take? What kind of valve is used? How is the transition period treated in the data analysis?

Line 132: How long does it take to change sampling temperature? From Fig.3 it seems reaching the lowest temperature takes over an hour, indicating that the inner wall chiller is undersized.

Line 139: Explain how the nitrogen gas is used during icing. Is nitrogen also used for the sheath flow?

Line 166ff: For ambient data where the size of INP was not measured,  $n_s$  is an arbitrary scaling of the data containing more information about the aerosol size distribution than the INP concentration. Justify the conversion to  $n_s$ .

Figure 2: (a) data seems to be covered up by white square around the 16/1/2021. (c), (d) use different colour scheme to improve readability. (d) upper limit in SMPS diameter changes after 9/1/2021.

Line 178: In Bi et al., 2019 6.5% supersaturation was used at all temperatures. Explain why the supersaturation was set to different values at different temperatures for the Storm Peak campaign.

Figure 3: The offset in temperature scale for the inner and outer wall is confusing. Set both to the same scale. During icing, filling the chamber with water and latent heat from ice formation should increase the wall temperature close to 0°C but only a small temperature increase is shown, and target temperature is reached fast after icing. Explain how this is possible. Are the plotted temperatures measured at the chamber wall? How many and where are temperature measurements taken that are averaged?

Line 181, 182: Based on what data are deviations in temperature and supersaturation from the target determined? At how many and which locations are wall temperatures measured?

Line 183: the 15°C temperature difference is not visible in Fig.3

Line 184: Present what was observed during -30°C measurements.

Line 187 ff.: The same INP concentrations at all 3 temperatures is remarkable and should be investigated in depth. This should be done by comparing -20°C, -25°C, -30°C observations in single measurement segment (4-6h) instead of averaging all the data. It needs to be convincingly excluded that the instrument is insensitive to measure the change with temperature because of too low ambient INP concentrations, too high instrument background (higher background at low temperature masking the increase in INP) or the method of analysing the data.

Line 206ff: Looking at Fig.16 in Murray et al., 2012 the concentration of biological INP can be expected to be on the order of  $10^{-3}L^{-1}$ . Also considering the measurements here were taken during winter, makes this interpretation highly unlikely.

Line 212f, 264: Richardson et al., 2007 measured below water saturation, also most of the data in DeMott et al., 2003 is below water saturation. The cited articles are not valid for the comparison. The only data at comparable supersaturation is at  $-35^{\circ}\text{C}$  in DeMott et al., 2003 showing  $1-100\text{L}^{-1}$  (their Fig.2).

Line 216: How do concentrations compare to Brunner & Kanji, 2021 or Brunner et al., 2022?

Line 217: Explain how sampling frequency and duration affect the observed INP concentrations.

Line 219: This seems to contradict lines 194, 195 where measurements are listed to identify free-tropospheric conditions.

Line 221: Note the temperature at which the mentioned INP concentrations were measured.

Line 224: Elaborate how drought severity affects the INP concentration.

Line 226ff: The measured, ambient particle size distributions are bulk and not for dust, making such a comparison questionable.

Line 237ff: As suggested above, periods where all 3 temperatures were measured consecutively should be analysed to see if there is no detectable change in INP concentration between the 3 temperatures.

Line 239: What measure was used to identify that  $n_s$  follows more closely the marine fit?

Line 240f: Comparing  $n_s$  to pollen is misleading. Atmospheric concentrations of pollen must be considered (especially in winter).

Line 241: For atmospheric INP,  $n_s$  is arbitrary scaling with bulk aerosol surface and does not carry the information suggested here.

Line 244: The cited Niemand et al., 2012 parameterization is not shown in Fig.5 or discussed anywhere. It can be removed.

Line 257: It should be made clear that the automatization is not novel but previously described in Bi et al., 2019.

Line 262: Mention that measurements at -30°C make up almost half of the observation period. Specify which stable conditions you refer to.

### **Technical corrections:**

For consistency, use either "ice nuclei" or "ice nucleating particle" throughout the manuscript.

Line 49: "Technical note" looks like a leftover from the previous submission.

Line 50: remove open bracket in front of Niemand

Line 57 is a repetition of line 46.

Line 64: give OPC model, give impactor model.

Eq.1 : define  $x_i$ ,  $n_i$

Line 161-163: give model of SMPS and APS. The Gannet Hallar et al., 2016 citation should be Hallar et al., 2016. In the references double check names in Hallar et al., 2011.

Figure 5: Adapt y-axis to  $10^{-2}$ - $10^2$  to show -20°C data in b).

Update references. Brunner & Kanji, 2021 and Möhler et al., 2021 are missing.

## References:

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