

We would like to express sincere gratitude for the referee's helpful comments. Below, we provide our point-by-point responses (in blue colour).

2nd Referee report on "Ice-nucleating particle concentration measurements from Ny-Ålesund during the Arctic Spring-Summer in 2018" by Rinaldi et al.

The authors have addressed most of the previous comments and improved the manuscript. However, I would like to suggest some further revisions to improve the manuscript before it can continue the review process. We appreciate these general remarks, reconsidered referee comments and revised the manuscript accordingly. Below, we provide our point-by-point responses.

Main comment

The number of samples this study is based on is small and might not allow to characterize the population of INP. It needs to be highlighted in the abstract and conclusion that results are preliminary because of the small sample size. Findings in the abstract and conclusion should be limited to the strong signals that are expected to be reproducible in future investigations. For less clear results it should be stressed that more observations are needed. The limitations of the applied analysis due to the size of the available dataset should be stated clearly in each section. This includes stating the number of measurements used and the assumption on the structure of the data, e.g. normal distribution of nINP (not typical) to compare spring to summer concentrations or contribution of coarse and fine particle fraction to nINP.

The number of samples considered for each study is now offered in revised Tables and text. Our datasets are made based on a compilation of 33 DFPC samples (16 in spring and 17 in summer) and 28 WT-CRAFT samples. These numbers are not by far the smallest published in Arctic INP studies as can be seen in the Table below. Thus, we respectfully believe that our results are not "preliminary".

In the text, we have highlighted all the results that may be considered not fully robust because of the low number of samples. For instance, the results of the spatio-temporal correlation analysis with CHL and the output of the CWT algorithm (even though examples of applications of CWT with similar dataset dimension exist in literature (Hsu et al.; 2003)). We have also re-scaled our considerations of the different responses of Arctic aerosol particles to the ice nucleation mode, not because of the number of samples, but because more detailed intercomparisons would be necessary to address this point. As for the rest, we believe that our data are worth to be presented without any caveat on the sample dimension.

Table. Number of samples analysed for previous Arctic INP publications

Reference	Location	Number of samples
Bigg (1996)	High Arctic (cruise)	>50
Bigg and Leck (2001)	High Arctic (cruise)	>50
Conen et al. (2016)	Halde observatory, Norway	4
Mason et al. (2016)	Alert, Canada	9
Creamean et al. (2018)	Oliktok Point, Alaska	17
Si et al. (2018)	Lancaster Sound, Canada	1
Irish et al. (2019)	Multiple (cruise)	28
Santl-Temkiv et al. (2019)	Villum, Greenland	35
Si et al. (2019)	Alert, Canada	16
Tobo et al. (2019)	Mt. Zeppelin, Svalbard	13
Wex et al. (2019)	Alert, Canada	~40
	Utqiagvik, Alaska	~40
	Ny-Ålesund, Svalbard	13
Welti et al. (2020)	Villum, Greenland	11
	High Arctic (cruise)	>50
Schrod et al. (2020)	Mt. Zeppelin, Svalbard	>50
This study DFPC	Ny-Ålesund, Svalbard	33
This study WT-CRAFT	Ny-Ålesund, Svalbard	28

Hsu, Y. K., Holsen, T. M., and Hopke, P. K.: Comparison of hybrid receptor models to locate PCB sources in Chicago, *Atmospheric Environment*, 37, 545-562, 10.1016/s1352-2310(02)00886-5, 2003.

Because correlation analysis is prominent throughout the paper, I suggest to add a subsection detailing the statistical analysis, including how significance in dependence of number of samples is determined (t-test), to the Method Section. This would help to understand, e.g. why correlation coefficients are high and why 0.5 is significantly non-zero for DFPC but 0.8 can be not significant for WT-CRAFT (Tabs.3 and 4).

The required sub-Section has been added. Please see the revised **Sect. 2.4. Statistical data treatment**.

Specific comments

Line 27f, 101f, 609ff.: The difference in $nINP$ due to two different ice nucleation modes does not emerge from the data. To make this conclusion, filter samples from one sampling setup should be analysed with both DFPC and WT-CRAFT. More plausible are differences in the samples used by the two methods.

We agree with the reviewer on the importance of intercomparing the two measurements. Unfortunately, it was not possible to operate the suggested comparison using our 2018 samples for the following reasons:

(1) we cannot use an identical substrate for the two INP analysis techniques. We have assessed the applicability of the cellulose membrane (optimal for DFPC) in WT-CRAFT, and we found notable background artifacts from the blank substrate below -22°C . Thus, the same filter sample cannot be shared for both INP measurement techniques.

(2) Our aerosol samplers employed different sampling periods for optimized flow conditions and estimated detection limits. The aerosol sampler for DFPC employed high flows, which were needed for the desired cut-size (see Sect. 2.1). If we conducted sampling with a longer period than what we employed, DFPC would likely suffer from filter overloading issues. Likewise, reducing the sampling time for WT-CRAFT would have created detection limit issues. As described in Sect. 2.1, the filter sampling flow for WT-CRAFT was ~ 5 LPM.

Nevertheless, we fully understand the concerns of the reviewers about relying on ice nucleation modes for explaining the observed difference between $nINP_{DFPC}$ and $nINP_{WT-CRAFT}$. As we clarified in the revised manuscript, it is not conclusive, and we hope to be able to further investigate this issue in future follow-up

studies. To allay further misgivings, we softened our tones and revised the text to present the particle sensitivity to different ice nucleation modes only as a potential reason. In the revised manuscript, we also discussed other potential factors for the observed discrepancy (please refer to the revised Sect. 4.1). Overall, the addressed potential factors include (1) differences in measurement uncertainties, (2) sampling apparatus, (3) sample storage protocols, (4) substrate types, (5) sampling durations and (6) ice nucleation paths (condensation vs immersion freezing).

Nonetheless, we would like to clarify here that explaining the gap is not a major point of the study. Our work extends the INP observations at GVB, contributing to filling the present lack of observations in the Arctic. Furthermore, it provides information on the AF, which was never calculated before at GVB and which was only rarely addressed at Arctic sites. One of the major findings of our work is that the seasonality of n_{INP} can be significantly different from what was observed in previous studies. This finding is supported by both the INP datasets, notwithstanding the concentration gap and all the sampling differences. Our results evidence potentially a great interannual variability and the necessity for further data coverage to better understand INP dynamics over the Arctic. Furthermore, we present and discuss for the first time the seasonality of the AF, intended as a proxy of the overall ice nucleation ability of the particle population at the study site. Finally, our study reports information on the ice behaviour of fine vs coarse aerosol particles, which was addressed only in a few more Arctic studies so far. This was clarified at the end of the Introduction Section.

David, R. O., Marcolli, C., Fahrni, J., Qiu, Y., Perez-Sirkin, Y. A., Molinero, V., Mahrt, F., Brühwiler, D., Lohmann, U., and Kanji, Z. A.: Pore condensation and freezing is responsible for ice formation below water saturation for porous particles, *Proceedings of the National Academy of Sciences*, 116, 8184–8189, <https://doi.org/10.1073/pnas.1813647116>, 2019.

Wagner, R., Kiselev, A., Möhler, O., Saathoff, H., and Steinke, I.: Pre-activation of ice-nucleating particles by the pore condensation and freezing mechanism, *Atmos. Chem. Phys.*, 16, 2025–2042, <https://doi.org/10.5194/acp-16-2025-2016>, 2016.

Line 29: name the “several important indications”

This sentence was reformulated for major clarity. Please refer to the revised Abstract.

Line 29f: This is inconsistent with what is reported on line 419ff

In line 29, we say that scaling n_{INP} to the total particle number concentration, i.e., deriving the AF, it is possible to extract more information from the dataset. This is a true statement both in general and regarding this particular case. Further on in the abstract, we explain that the AF shows a clearer seasonal trend than n_{INP} . Line 419 and following lines, treats the correlation between n_{INP} and particle number concentration (which is qualitatively observable, but not statistically significant). These topics are mostly unrelated and we do not see how these parts can be contradicting each other.

Line 31: specify “subset of our data”

We specified the following: “(WT-CRAFT, between -18 and -21°C)”. We have conducted the temperature-resolved correlation analysis, and this particular range of temperatures was the one showing a significant difference.

Line 33: Explain how higher AF can be interpreted as larger freezing efficiency of large particles.

This sentence was completely reformulated: “This seasonal AF trend corresponds to the overall decrease in aerosol concentration towards summer and a concomitant increase in the contribution of super-micrometre particles. Indeed, the AF of coarse particles resulted markedly higher than that of sub-micrometre ones (2 orders of magnitude)”.

Line 34: inconsistent to line 417 where it is stated that no clear relation emerges between n_{INP} and meteorology.

We do not think this is inconsistent: we intended to point out that no meteorological parameter is a major driver of the n_{INP} time trend. This supports our choice of using the n_{INP} vs CHL correlation as a tool for identifying potential marine sources of INPs. In this respect, the meteorological analysis is an important step of the process that leads to the conclusion that: “the summertime INP population is influenced both by terrestrial (snow-free land) and marine sources”.

Line 48: what other mechanisms beside Bergeron-Findeisen play an important part?

The transformation of water between vapour, liquid and ice phase in mixed-phase clouds can occur in different ways. The Wegener-Bergeron-Findeisen (WBF) mechanism occurs only if $e_w > e > e_i$, where e is the in-cloud vapour pressure, e_w the vapour pressure of supercooled liquid droplets, and e_i is the ice vapour pressure. Nevertheless, additional situations can be present in mixed-phase clouds. For instance, if $e > e_w > e_i$, both droplets and ice particles grow simultaneously and compete for water vapour. Korolev and Mazin (2003) showed that this condition may occur in ascending mixed-phase clouds when updraft velocity exceeds a fixed value. Fan et al. (2011) found that in the Arctic, in both single and multilayer mixed-phase clouds, the WBF process occurs in about 50% of the mix-phase regime, prevalently in the downdraft region, and in the other half of the cloud both liquid and ice grow simultaneously.

For these reasons, we believe it is correct to refer to the WBF as one of the mechanisms responsible for the relationship between lifetime and ice crystal concentration in mixed-phase clouds.

Fan et al., 2011. Representation of Arctic mixed-phase clouds and the Wegener-Bergeron-Findeisen process in climate models: Perspectives from a cloud-resolving study *J. Geophys. Res.* 116, D00T07, doi:10.1029/2010JD015375.

Korolev, A. V., I. P. Mazin (2003), Supersaturation of water vapor in clouds, *J. Atmos. Sci.*, 60(24), 2957–2974, doi:10.1175/1520-0469.

Line 56, 57: Clarify the difference between water vapour deposition and water vapour condensation.

We have reformulated the description of the deposition mode ice nucleation, making it more adherent to that by Vali et al. (2015).

“Ice formation by deposition occurs when the ambient is supersaturated with respect to ice in water-subsaturated conditions so that ice forms on an INP without prior formation of liquid”.

Line 60f.: If condensation and immersion are the same process how can the difference in n_{INP} between DFPC and WT-CRAFT be explained by it? To make this case, provide evidence that the two mechanisms can be different.

Here, we only intended to introduce what has been studied for condensation vs. immersion by referring to the two cited studies. We did not mean to conclude that condensation = immersion in the introduction section. Further assessments from the laboratory and field settings are needed to understand the similarity of ice nucleation modes and processes. Whether condensation and immersion are the same processes or not highly depends on the aerosol properties. We discuss this point in the revised Sect. 4.1.

We also rephrased the sentence in the introduction section to: “However, the recent inter-comparison study with two different organic fiber samples shows a difference between condensation freezing and immersion freezing measurements (i.e., ice nucleation efficiency of the former is higher than the latter (Hiranuma et al., 2019)). Further laboratory and field assessments are therefore necessary to understand the similarity of ice nucleation modes and processes”.

Line 96: Why are Santl-Temkiv et al. and Wex et al. cited for the interpretation of data in Tobo et al. and not the Tobo et al. paper itself?

This sentence was not related to Tobo et al. (2019) specifically. We refer here to the enhancement of *n*INP reported by all the cited papers (Wex et al., 2019; Santl-Temkiv et al., 2019 and Tobo et al. 2019). In this sense, the citation is not wrong as:

- Wex et al. (2019) state: “In summer, the higher bioaerosol concentrations compared to spring indicate contributions from local and regional terrestrial and marine ice-free areas”; “these INPs can originate from both terrestrial and marine sources in the Arctic. These sources are strong in summer and weak or absent in winter, depending on the conditions on the ground”.
- Santl-Temkiv et al. (2019) state that: “In summer, the higher bioaerosol concentrations compared to spring indicate contributions from local and regional terrestrial and marine ice-free areas”; “Based on this observation, we suggest that the high concentrations of INPs in the air during summer may be partially related to strengthened local terrestrial sources, in particular soil dust”
- Tobo et al. (2019) state: “This suggests that significant local sources of INPs other than marine organic materials might exist in and/or around the Svalbard region in the summer”

After all, these three papers attribute the summertime enhancement of *n*INP to local sources. We have added the reference to Tobo et al. (2019) according to the reviewer comment.

line 104f: Explain why INP measurements are the key to verify that immersion freezing is the most relevant ice nucleation mechanism in Arctic mixed-phase clouds and how such measurements can be used to do that. We admit that this paragraph lacked logical consistency. This sentence has been removed. We have revised the entire paragraph to:

“Our study aims to add to the still scant INP observations in the Arctic environment, investigating *n*INP and potential INP sources during spring and summertime at the ground-level site of GVB. In particular, we extend the INP observations at GVB, previously only 13 samples (Wex et al., 2019), presenting the results of 61 samples investigated with two offline INP measurement techniques. We also analyze the ice nucleation efficiency of Arctic aerosol particles by calculating their activated fraction (AF). To date, only a limited number of studies provide information on INP trends scaled to the total aerosol concentration over the Arctic (Si et al., 2018). AF estimation can be understood as a simple metric indicating the ice-nucleating efficiency of particles within a specific aerosol sample (Schrod et al., 2020). In our specific case, it provides further insight, over and above the *n*INP data, into INP characteristics over the Atlantic sector of the Arctic”.

Line 117ff.: state the number of samples collected in spring, summer, PM1, PM10.

The required information was added in Sect. 2.1.

Line 128ff.: on line 131 it says the 5.4lpm are a subset of the airflow through the TSP inlet. Specify the total flow through the inlet and describe more clearly how the 5.4lpm are extracted from the higher total flow. Was there a pressure drop in the flow from which the subset flow was taken? Was this considered to determine the sample flow? Was there an online measurement of the flow through the membrane filter (how are flow rates in Tab.S1 measured) and why did the flow vary from 4.8-5.6lpm between samples?

The total flow of the central sampling stack inlet was reported in the manuscript (150 LPM). Further, as described in Sect. 2.1, an 8 mm OD stainless steel tube was used as a pickup inlet for a filter sampler. All excess amount of air was drawn through other instruments or a central pump. Below we provide our sampling system schematic as well as the sampling flow measured at the beginning and end of each sampling activity in the figure and table, respectively. A TSI4100 flow meter is placed in the sampling line to check and make sure we have consistent flow on the filter cross-section. We acknowledge onsite operators who typically check our flow daily (even several times per day) to make sure the airflow is consistent. In 2018, our flow measured at the beginning and end of each sampling activity deviated on average only <2% and never

exceeded 5%. Thus, we used an average flow rate measured during individual sampling activity as a representative flow for each sample. While the pressure drop was not measured for each sampling activity, we have measured a pressure drop in the flow of our sampling system to be approximately 15 mb and 200 mb with 1.5 LPM and 9.0 LPM flow. Because we observe such a consistent airflow throughout individual samplings and no physical damages on filters after sampling, we consider our sampling is successful and valid. The observed variation in sampling airflow between samples is presumably due to air valve control. Nevertheless, we estimate a total flow through each filter by accounting for this, and we echo that the airflow was consistent for each sampling activity.

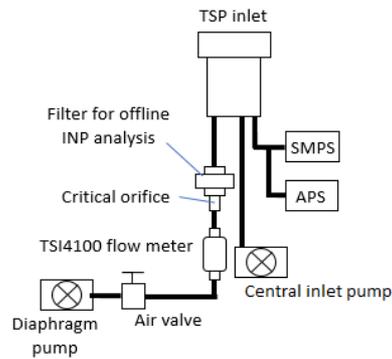


Figure. A schematic of aerosol particle sampling system at GVB. Aerosol particles were collected on a 47 mm Nuclepore filter for offline INP analysis by WT-CRAFT.

Table. Sampling airflow measured at the beginning and end of individual sampling activities plus the averaged flows.

ID	Start_Date	Start_Time	Start_Air_Flow_LPM	End_Date	End_Time	End_Air_Flow_LPM	Average_Flow_LP M
1	2018-04-16	17:00:00	5.10	2018-04-20	10:00:00	5.10	5.10
2	2018-04-20	14:40:00	5.10	2018-04-24	14:40:00	5.04	5.07
3	2018-04-24	18:20:00	5.40	2018-04-28	16:00:00	5.50	5.45
4	2018-04-29	13:30:00	5.60	2018-05-02	16:15:00	5.50	5.55
5	2018-05-02	16:20:00	5.60	2018-05-06	14:37:00	5.34	5.47
6	2018-05-06	14:45:00	5.34	2018-05-10	13:00:00	5.50	5.42
7	2018-05-10	13:10:00	5.70	2018-05-14	11:05:00	5.58	5.64
8	2018-05-14	11:15:00	5.58	2018-05-18	07:50:00	5.35	5.47
9	2018-05-18	08:00:00	5.50	2018-05-22	08:28:00	5.50	5.50
10	2018-05-22	08:30:00	4.80	2018-05-26	11:33:00	4.80	4.80
21	2018-05-26	11:45:00	5.57	2018-06-03	18:30:00	5.46	5.52
22	2018-06-03	18:35:00	5.36	2018-06-07	17:20:00	5.60	5.48
23	2018-06-07	17:24:00	5.40	2018-06-11	17:35:00	5.46	5.43
24	2018-06-11	17:40:00	5.41	2018-06-15	16:24:00	5.30	5.36
25	2018-06-15	16:28:00	5.39	2018-06-19	19:05:00	5.35	5.37
26	2018-06-19	19:09:00	5.26	2018-06-23	19:16:00	5.25	5.26
27	2018-06-23	19:20:00	5.36	2018-06-27	13:55:00	5.33	5.35
28	2018-06-27	14:00:00	5.39	2018-07-01	16:40:00	5.43	5.41
16	2018-07-01	16:50:00	5.43	2018-07-05	17:20:00	5.48	5.46
17	2018-07-05	17:25:00	5.44	2018-07-09	17:22:00	5.39	5.42
18	2018-07-09	17:27:00	5.39	2018-07-13	18:24:00	5.38	5.39
19	2018-07-13	18:33:00	5.34	2018-07-17	16:43:00	5.33	5.34
20	2018-07-17	16:52:00	5.39	2018-07-21	15:55:00	5.30	5.35
11	2018-07-21	16:02:00	5.43	2018-07-25	16:31:00	5.36	5.40
12	2018-07-25	16:38:00	5.40	2018-07-29	15:07:00	5.46	5.43
13	2018-07-29	15:14:00	5.49	2018-08-02	18:39:00	5.41	5.45
14	2018-08-07	15:55:00	5.46	2018-08-11	14:05:00	5.43	5.45
15	2018-08-11	14:12:00	5.42	2018-08-15	17:36:00	5.43	5.43

Line 165: what is meant by “reasonably matches”? The 95% confidence interval is inherent in the cited formula.

Thanks for catching this. We now clarify the previously reported uncertainty estimation method of WT-CRAFT technique as:

“The uncertainty of ice nucleation efficiency estimation in WT-CRAFT was previously estimated based on the average standard deviation at $T_s > -25^\circ\text{C}$ for a known composition (microcrystalline cellulose). Alternatively, as demonstrated in Vepuri et al. (2021) and Schiebel (2017), our experimental uncertainty in estimated $n\text{INP}$ can be evaluated using the 95 % confidence interval method”.

Line 178: point to sec. 2.2.3. for details on how $n\text{INP}$ are estimated

Done.

Line 179: mention already here that half of each filter was used and explain how the used water volume was calculated. Volumes given in Tab.S1, row 2-6 are off by -0.1 to 0.2ml from calculated values.

To incorporate the reviewer’s suggestion, we rephrased L179 to: “Prior to each WT-CRAFT experiment, we suspended particles on an individual filter sample in a known volume of ultrapure High Performance Liquid Chromatography (HPLC) grade water, in which the first frozen droplet corresponded to $\approx 1 \text{ INP m}^{-3}$ (in the range of $0.93 - 1.02 \text{ m}^{-3}$; Table S1). The HPLC water volume was determined according to Eqns. 1-2 in Sect. 2.2.3. Half of each filter was used for each WT-CRAFT experiment, the other half was saved for other and future uses”

We also decided to provide more decimal points for our data provided in the last few columns of Table S1. Note that our mL pipette used in this study has 2 decimals. Our reported $n\text{INP}$ values from WT-CRAFT scale to this initial number (ca. 1 INP m^{-3}), so our $n\text{INP}$ data at across the measured temperatures are valid. While the initial $n\text{INP}$ determines our INP detection sensitivity and low detection limit, the variation between 0.93 m^{-3} and 1.02 m^{-3} does not substantially impact our other experimental parameters/capabilities.

Table S1. Summary of sampling conditions for filters collected for WT-CRAFT.

Sample ID	Filter Sampling Ref Start Time	Filter Sampling Ref End Time	Flow Rate	Air Volume	Suspension water volume	$n\text{INP}$ corresponding to the first frozen droplet
	DAT.UTC	DAT.UTC	LPM	L	mL	m^{-3}
NYA_GVB_01	4/16/2018 17:00	4/20/2018 10:00	5.100	13617.000	2.84	1.00
NYA_GVB_02	4/20/2018 14:40	4/24/2018 14:40	5.070	14601.600	3.08	1.01
NYA_GVB_03	4/24/2018 18:20	4/28/2018 16:00	5.450	15314.500	3.10	0.97
NYA_GVB_04	4/29/2018 13:30	5/2/2018 16:15	5.550	12445.875	2.42	0.93
NYA_GVB_05	5/2/2018 16:20	5/6/2018 14:37	5.470	15471.895	3.26	1.01
NYA_GVB_06	5/6/2018 14:45	5/10/2018 13:00	5.420	15325.050	3.26	1.02
NYA_GVB_07	5/10/2018 13:10	5/14/2018 11:05	5.640	15890.700	3.29	0.99
NYA_GVB_08	5/14/2018 11:15	5/18/2018 7:50	5.465	15179.037	3.10	0.98
NYA_GVB_09	5/18/2018 8:00	5/22/2018 8:28	5.500	15917.000	3.32	1.00
NYA_GVB_10	5/22/2018 8:30	5/26/2018 11:33	4.800	14263.200	2.98	1.00
NYA_GVB_21	5/26/2018 11:45	6/3/2018 18:30	5.515	32883.188	6.86	1.00
NYA_GVB_22	6/3/2018 18:35	6/7/2018 17:20	5.480	15576.900	3.25	1.00
NYA_GVB_23	6/7/2018 17:24	6/11/2018 17:35	5.430	15668.265	3.27	1.00
NYA_GVB_24	6/11/2018 17:40	6/15/2018 16:24	5.355	15218.910	3.18	1.00
NYA_GVB_25	6/15/2018 16:28	6/19/2018 19:05	5.370	15887.145	3.32	1.00
NYA_GVB_26	6/19/2018 19:09	6/23/2018 19:16	5.255	15152.792	3.16	1.00
NYA_GVB_27	6/23/2018 19:20	6/27/2018 13:55	5.345	14525.037	3.03	1.00
NYA_GVB_28	6/27/2018 14:00	7/1/2018 16:40	5.410	16013.600	3.34	1.00
NYA_GVB_16	7/1/2018 16:50	7/5/2018 17:20	5.455	15792.225	3.30	1.00
NYA_GVB_17	7/5/2018 17:25	7/9/2018 17:22	5.415	15587.078	3.25	1.00
NYA_GVB_18	7/9/2018 17:27	7/13/2018 18:24	5.385	15662.273	3.27	1.00
NYA_GVB_19	7/13/2018 18:33	7/17/2018 16:43	5.335	15071.375	3.15	1.00
NYA_GVB_20	7/17/2018 16:52	7/21/2018 15:55	5.345	15241.268	3.18	1.00
NYA_GVB_11	7/21/2018 16:02	7/25/2018 16:31	5.395	15602.340	3.26	1.00
NYA_GVB_12	7/25/2018 16:38	7/29/2018 15:07	5.430	15391.334	3.21	1.00
NYA_GVB_13	7/29/2018 15:14	8/2/2018 18:39	5.450	16254.626	3.39	1.00
NYA_GVB_14	8/7/2018 15:55	8/11/2018 14:05	5.445	15382.126	3.21	1.00

Line 202ff: I couldn't find the size-range in Kanji et al., 2017. Pruppacher & Klett, 2010 section 9.2.3.2 suggest 0.1 μ m as a lower size limit, which I would recommend. The choice needs to be motivated better. How does the lower size limit effect the results in sec. 3.3?

This is a valid question. Our choice (i.e., comparing *n*INP with particles larger than 500 nm) is arbitrary and motivated by different considerations. First, we followed the approach by DeMott et al. (2010), which report: "The choice of 0.5- μ m diameter as the lower limit for summing number concentrations of "large" particles is a relatively arbitrary one of convenience, selected to limit the influence on derived relationships of high concentrations of non-IN particles in the range 0.1–0.5 μ m, while retaining sufficient number concentrations of particles to reference to IN concentrations". Furthermore, we already adopted this choice in previous DFPC papers (Rinaldi et al., 2017; Rinaldi et al., 2019), so we aimed at maintaining the possibility of comparing with previous DFPC results.

We agree with the reviewer that there is no consensus in the literature on which aerosol particle size range would be more appropriate for the AF estimation. For instance, Kanji et al. (2017) report: "As the sites appear with a finite probability (Niedermeier et al. 2015), smaller particles (e.g., ,500 nm) are less likely to act as INPs". Nevertheless, they also add: "However, in Mertes et al. (2007) 200-nm diameter particles were inferred to make up the majority of all INPs, based on the mode size in atmospheric ice residual number distributions". For previous Arctic studies, Si et al. (2018) shows the presence of INPs also in the 200-300 and 400-500 nm size classes. Therefore, a lower particle size limit could be appropriate, and the reviewer's comment (How does the lower size limit impact the results in sec. 3.3?) is also plausible.

Using the AF data calculated using particle number concentration from 100 nm (AF₁₀₀ hereafter) instead of AF₅₀₀ did not change our findings and conclusion. We confirm our result regarding the higher ice nucleation efficiency of coarse atmospheric particles, enhancing the difference between AF_{fine} and AF_{coarse} (almost 2 orders of magnitude difference), remains valid. The spring to summer increase of the AF also remains valid. We have updated the manuscript by introducing the AF₁₀₀ values.

Line 215ff: Clarify what size-range was used to calculate AF. From line 203 it would seem that only APS data (0.5-10 μ m) was used.

We now use AF₁₀₀, and the associated text has been revised accordingly to:

"The number concentration in the resulting overlapping range was taken from the SMPS data. Finally, commutative aerosol particle counts of SMPS and APS were considered as a total aerosol particle number concertation. In order to compare with *n*INP and to calculate the AF, the particle number concentrations at 10 minutes time resolution were averaged over each filter sampling period. AF was calculated using the size range 0.1 – 10 μ m for DFPC_{PM10} and WT-CRAFT data, 0.1 – 1 μ m for DFPC_{PM1} data and 1 – 10 μ m for DFPC data in the super-micrometre regime."

Line 246: provide a reference for the marine boundary layer height in the Arctic.

We provided the requested reference: DAI Cheng-Ying, GAO Zhi-Qiu, WANG Qing, and CHENG Gang, Analysis of Atmospheric Boundary Layer Height Characteristics over the Arctic Ocean Using the Aircraft and GPS Soundings, ATMOSPHERIC AND OCEANIC SCIENCE LETTERS, 2011, VOL. 4, NO. 2, 124-130.

Line 270: It could be already mentioned here that *n*INP at -15°C, PM1 are used for this exercise.

We believe that adding this information without explaining why would be confusing. Our reasons are explained later on in the text.

Line 280: to be consistent throughout the paper, consider using *n*INP instead of Ct for the INP concentration. Done.

Line 295: It could be explained here how the CHL correlation analysis and CWT were overlaid (Fig.8b).
We believe the text is clearer as it is.

Line 303: Can you provide an interpretation of the difference in slope? It could suggest that there is a specific type of high temperature INP, that was only detected in the DFPC measurement. Could storing the filters at different conditions cause such an effect?

The difference in sample storing methods cannot be a sole factor to explain the discrepancy. As we kept the samples for DFPC at the room air T (and the WT-CRAFT samples at 4 °C except during transportation), the suppression of INPs is expected to be more obvious for DFPC than WT-CRAFT, which is not supported by the observed trend of $nINP_{DFPC} > nINP_{WT-CRAFT}$.

We are aware that Beall et al. (2020) recently demonstrated that storage of precipitation samples can lead to losses of INPs (especially the heat-labile ones). Storage at room temperature and at 4°C resulted basically in INP losses, with the former conditions showing somewhat higher losses than the latter. Nevertheless, the results by Beall et al. (2020) are not necessarily valid for particle samples collected on filters. Furthermore, there is not yet a consensus in the literature (and within the scientific community) about the effect of sample storage on INP measurements, so answering this question quantitatively is very hard at present. For instance, Wex et al. (2019) stated that “it is not yet known with certainty how the temperature during storage will affect INP concentrations”. Bigg (1990) reported no significant deterioration in INP concentration after 5 years, for filters sealed in a dry-sealed container. Conen et al. (2015) commented that storage of filters at -20°C may not have been necessary, because re-analysis of filters analyzed previously showed no effect of storage. Wex et al. (2015) and Polen et al. (2016) showed that storage at temperatures above 0°C or even under freezing conditions has been found to reduce the ice activity of biogenic INPs. Stopelli et al. (2014) studied INP concentrations in a snow sample stored at 4°C and observed a decrease in the concentration of INPs active at -10°C over 30 days (factor of 2).

Regarding the effect of sample storage on the $\Delta nINP/\Delta T$ slope, we evidence that DFPC samples were not refrigerated during storage, while WT-CRAFT ones were kept at 4°C. Therefore, based on the limited literature on this topic, degradation of INPs during storage would have caused higher losses in DFPC samples, resulting in the opposite effect on the $\Delta nINP/\Delta T$ slope. For this reason, we believe that losses during storage cannot be considered a valid explanation for the different slopes, at least based on the current knowledge of the topic. We now discuss this point in the revised Sect. 4.1.

Bigg, E.K., 1990. Measurement of concentrations of natural ice nuclei. *Atmos. Res.*, 25, 397-408.

Conen et al., 2015. Atmospheric ice nuclei at the high-altitude observatory Jungfraujoch, Switzerland. *Tellus B*, 67, 1-10.

Conen et al., 2015. Atmospheric ice nuclei at the high-altitude observatory Jungfraujoch, Switzerland. *Tellus B*, 67, 1-10.

Polen, M., Lawlis, E., and Sullivan, R. C.: The unstable ice nucleation properties of Snomax (R) bacterial particles, *J. Geophys. Res.-Atmos.*, 121, 11666–11678, <https://doi.org/10.1002/2016jd025251>, 2016.

Stopelli, E., Conen, F., Zimmermann, L., Alewell, C., and Morris, C. E.: Freezing nucleation apparatus puts new slant on study of biological ice nucleators in precipitation, *Atmos. Meas. Tech.*, 7, 129–134, <https://doi.org/10.5194/amt-7-129-2014>, 2014.

Line 304ff: Consider referring to Tab.2 instead of listing the concentrations.

We would like to report this key information in the text as well.

Line 307: give number of samples instead of “<50%”.

The requested information was added in the revised manuscript: “(1 sample, at T of -9°C , and 13, at T of -13.5°C , over 28 total samples)”.

Line 311: give T -range for the $n\text{INP}$, see comment on Tab.1

The T range was specified as follows:

“The range of $n\text{INP}$ from Table 1 is roughly between 10^{-2} and 10^3 m^{-3} in the T range between -9 and -25°C , in which we detected ice nucleation activity in our samples. This $n\text{INP}$ range covers the majority of our measurements”.

Line 312ff: Can the higher $n\text{INP}$ from condensation mode measurements compared to immersion mode be confirmed from the studies listed in Tab.1?

Comparing $n\text{INP}$ from various studies to discuss condensation vs. immersion modes may be misleading as ambient conditions, aerosol particle abundance and experimental parameters vary from one study to another. We cannot provide a quantitative response to this comment.

Line 320: give number of samples that span the $3\text{-}200\text{m}^{-3}$ $n\text{INP}$ range.

Added as follows: “First, while Wex et al. (2019) report a very narrow concentration range ($27\text{-}33 \text{ m}^{-3}$) at -22°C , having only three samples, our 61 DFPC plus WT-CRAFT data points span a much wider range (ca. $3\text{-}200 \text{ m}^{-3}$)”.

Line 323ff: The difference in $n\text{INP}$ depending on the sensitivity of methods indicates that the INP population is highly variable and the variation is underestimated at the detection limits of methods.

The reviewer is right. We just wanted to point out that our WT-CRAFT’s lower detection limit ($\approx 1 \text{ INP m}^{-3}$ in the range of $0.93 - 1.02 \text{ m}^{-3}$ as mention above) is not as good as what is reported in Wex et al., and this difference should be noted to the reader. We rephrased this sentence to clarify our point as:

“The difference in $n\text{INP}$ towards a lower bound is due to different sensitivities and detection limits of the two methods: WT-CRAFT (ca. 1 m^{-3}) and the immersion freezing measurement technique used by Wex et al. (2019, ca. 10^{-1} m^{-3})”.

Line 325: Clarify for what the $n\text{INP}$ range is substantial. Below what concentration would it be negligible?

Line 328: give $n\text{INP}$ at -22°C to compare to the range at -22°C on line 325. Line 333ff: add $n\text{INP}$ at -22°C measured at the cited locations.

The Paragraph associated with these comments was removed upon request of the other reviewer.

Line 344: As I understand a normal distribution is assumed to obtain the difference at $p < 0.005$. I doubt that the dataset (< 20 measurements per season) is large enough to obtain a valid distribution to perform statistics.

We have now presented our statistical approach to testing differences between datasets more in detail in the new Sect. 2.4, as presented above. We have used both the t-test (assuming normal distributions of the data) and the non-parametric Wilcoxon-Mann-Whitney test (not requiring normally distributed data). We have considered statistically significant only differences that resulted significant for $p < 0.05$, according to both tests. We point out that this (same result from both parametric and non-parametric tests) resulted true for the large majority of the tested cases, which suggests that the normal distribution assumption was not so far from reality in many cases. For homogeneity, we now report through the text only the indication of the minimum tested significance level ($p < 0.05$) even in cases that resulted significant for higher confidence levels.

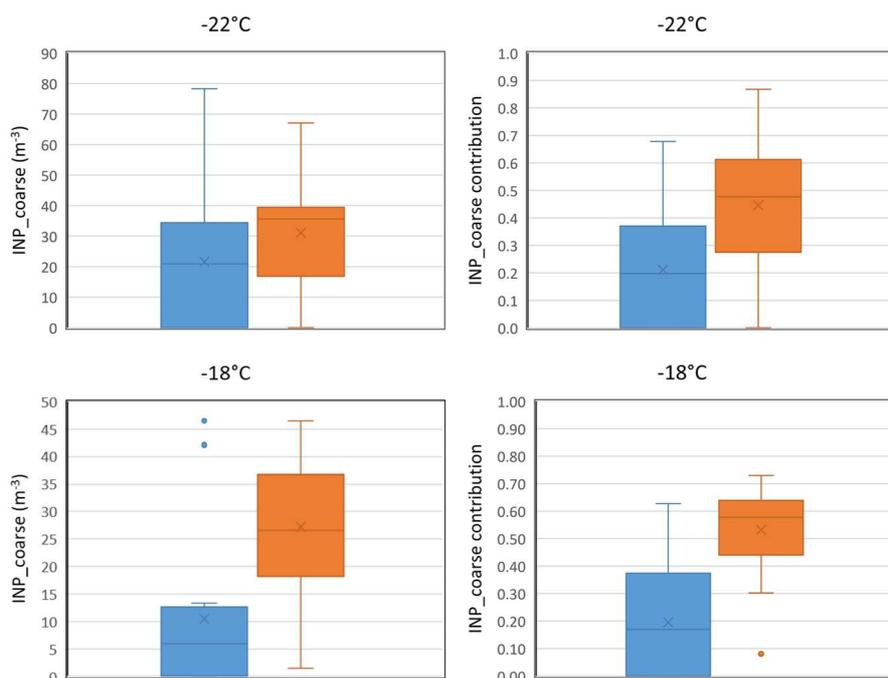
Line 346: From Tab.2 it can be seen that the median $n_{\text{INP,PM10}}$ is the same in summer and spring. The $n_{\text{INP,PM1}}$ is lower in summer thus contradicting the conclusion that coarse INP from exposed surface cause the difference. The higher $n_{\text{INP,PM1}}$ in spring could indicate INP from the arctic haze.

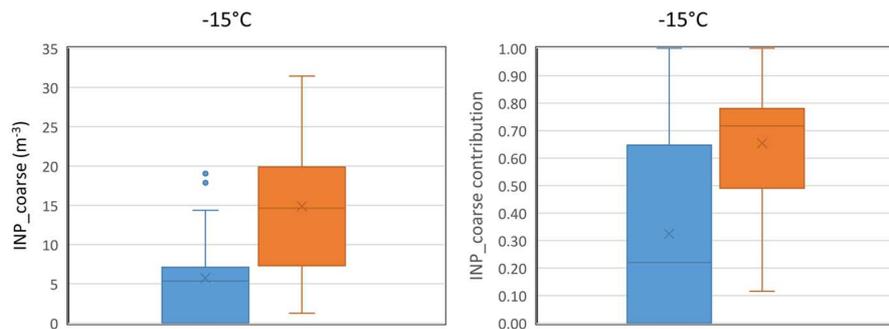
Yes. This is exactly our interpretation, and we do not find any contradiction between Table 2 and our conclusions. The fact that $n_{\text{INP,PM10}}$ is not statistically different between spring and summer, in the DFPC dataset, at T_s of -18 and -15°C , is discussed in detail in the manuscript. But, what is important is the fact that the coarse INP fraction is larger in summer at the same T_s than in springtime. Our data-driven conclusion is that the spring and summer-time aerosol particle populations are different: the spring one is mostly comprised of sub-micrometer INPs, likely transported from lower latitudes (Arctic haze), while the summer one is comprised of both fine and coarse particles (with coarse particles often exceeding a 50% contribution). There is a clear consensus in the literature (e.g., Udisti et al., 2016, Browse et al., 2012) on the fact that the Arctic haze period does not extend into the summer season. Considering the short atmospheric lifetime of aerosol particles, it is reasonable to assume that the spring and summer time aerosol populations are different and due to different sources (distant PM_{10} -dominated sources in spring; local sources providing a high contribution of coarse particles in summer).

Browse, J., Carslaw, K. S., Arnold, S. R., Pringle, K., and Boucher, O.: The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol, *Atmos. Chem. Phys.*, 12, 6775–6798, <https://doi.org/10.5194/acp-12-6775-2012>, 2012.

Line 347: on line 344 the contribution of coarse INP is described as significant to a high level of certainty. How were the “substantial uncertainties” considered to estimate the significance of an increase in coarse INP contribution?

The consideration regarding the “substantial uncertainties” refers to the fact that particle number measurements are generally affected by lower uncertainties than INP measurements. The increase in the contribution of coarse INPs in summer is clear and confirmed by all the performed statistic tests (discussed above). It is also true that the absolute concentration of coarse INPs is generally higher in summer than in spring, with a statistically significant difference ($p < 0.05$, $n_1 = 15$, $n_2 = 17$) for T of -18 and -15°C . This can be assessed from the plots below.





Line 349: On the PM1 filters not only particles larger than 0.5 μ m are sampled, as was assumed for the fractions shown in Fig. S2. I suggest using the full size-range below 1 μ m for the fine particle concentration to calculate the change in the particle population.

We took the reviewer's word for it. We now use the aerosol particle size range of 0.1-10 μ m for our AF estimation.

Line 350: The coarse INP fraction doesn't dominate. Looking at Tab.2, the fine fraction makes up 80% of nINP in spring and 50% in summer.

The "coarse fraction dominated" refers to the summer season, where coarse particles are averagely from 45% (-22°C) to 65% (-15°C). Anyhow we admit that "dominated" may not be the most appropriate term. We have clarified it as follows:

"An INP population similar to summer values at GVB, with a significant coarse fraction contribution, was reported by Mason et al. (2016), from the Alert Arctic station. The authors conducted INP measurements from 29 March to 23 July 2014 and observed an increasing contribution of coarse INPs as a function of the activation T".

Line 352: As mentioned on line 350, Mason et al. also reported a spring to summer dataset, making the results not "unique".

Mason et al. did not distinguish between spring and summertime INPs. From the paper, it is not possible to assess if they observed a seasonal increase of the coarse fraction contribution at Alert. In that sense, our publication is "unique" as it presents this finding for the first time for the Arctic atmosphere.

Line 360: Repeat here how coarse and fine AF are calculated, i.e. particle size-range used and nINP coarse as difference of PM10-PM1 nINP. Also mention it in the caption of Fig.3. What particle size range was used for the WT-CRAFT (sampling through a TSP inlet) AF?

The requested information have been added (Sect. 2.3.1).

For WT-CRAFT, we used the same size range used for PM10 DFPC samples. Our size distribution measurements were made through the TSP inlet, under which the WT-CRAFT filter sampler was deployed. Our APS data shows negligible contribution of > 10 μ m particles in terms of number concentration in 2018. The aerosol particles measured by APS for > 10 μ m accounted for on average only 0.07% of supermicron particles in the range of 1-20 μ m during our sampling period in 2018.

Line 360, 414, 544, 573, 579, 585, 587: Ice nucleation efficiency/ability/activity/capability was not measured in this study, but concentrations of INP. Ice nucleation efficiency requires knowing the concentration of the ice active species. To be more consistent replace them throughout the paper with nINP or AF where appropriate.

The AF can be understood as a simple metric that indicates the ice-nucleating efficiency of particles within a specific aerosol sample (Schrod et al., 2020). We have added this "definition" at the end of the Introduction

Section and used it through the document. We agree with the reviewer that we did not measure the AF, but only n_{INP} and aerosol number concentration; therefore, we have amended the text from any misleading expressions referring to “measurements of AF”.

Line 362: the difference would be much larger if fine particles would not be limited to particles larger than 0.5 μm .

We confirm it. By calculating the AF using particles from 100 nm, the AF of coarse particles results in almost 2 orders of magnitude higher than that of fine (PM_{10}) particles.

Line 363ff: instead of listing AF, point to Fig.3. Disentangle if the AF is governed by changes in particle concentration or n_{INP} .

We thank the reviewer for the suggestion. In this Section, we have provided more details on the variation of AF as a function of the particle size. The higher AF of coarse particles results from the significantly lower particle number concentration in the coarse mode (relative to the fine one), combined with the comparable n_{INP} in fine and coarse modes. Sub-micrometer particle number concentration during our study was of the order of 10 to 200 cm^{-3} , while in the coarse mode it was less than 1 cm^{-3} .

“Examining the size-segregated DFPC data (Fig. 3a and b), substantially higher ice nucleation efficiencies were found in coarse compared to sub-micrometer particles. The enhanced AF of coarse particles is due to the significantly lower number of particles in the 1-10 μm compared to the 0.1-1 μm size range (about two orders of magnitude), coupled with the comparable n_{INP} observed in both size ranges (see Sect. 3.2). As a result, the AF of coarse particles was more than 2 orders of magnitude greater than that of fine particles. In other words, the AF for coarse particles was estimated to be in the order of 10^{-6} to 10^{-3} at T_s between -18 and -22°C, while the AF of sub-micrometer particles was in the order of 10^{-8} to 10^{-5} at the same T_s ”.

In the following Section (3.4), we added more details on the seasonal variation of AF.

“Both the DFPC and WT-CRAFT datasets showed a general increase of the aerosol particle AF from spring to summer as shown in Fig. 5. This increase in the AF is mainly due to a significant reduction of the particle number concentration in the 0.1-10 μm range ($p < 0.05$; $n_1 > 10^3$; $n_2 > 10^3$; Fig. S4), combined with similar or slightly higher n_{INP} (depending on the T). DFPC showed a statistically significant AF increase ($p < 0.05$; $n_1 = 16$, $n_2 = 17$) going from the spring campaign to the summer period for all the probed activation T_s . The seasonal increase in the AF was more evident at higher T_s : the summer to spring mean ratio was 6.2 at T of -15°C and 2.5 at T of -22°C. Fairly consistent results can be observed in the WT-CRAFT dataset. Comparing the samples collected before June 3 with those collected after that date, an AF enhancement (from 1.1 to 3.7 fold) can be estimated for all the activation T_s . This difference was statistically significant ($p < 0.05$, $n_1 = 11$, $n_2 = 15$) for all the activation T_s between -17 and -22.5°C. Unlike the DFPC data, the spring to summer AF increase from WT-CRAFT data peaked at $T = -20^\circ\text{C}$ (3.7; Fig. S5)”.

Line 373: mention what particle size-ranges Si et al. used.

The paragraph was updated as follows:

“In particular, Si et al. (2018) reported average AF at -25°C of $\sim 10^{-4}$, 2×10^{-3} and 6×10^{-2} for the 0.56-1.0, 3.2-5.6 and 5.6-10 μm size ranges, respectively”.

Sec.3.4: This section needs to be structured better. It is currently unclear what the important results are.

The Section was reformulated for major clarity. Please refer to the revised text.

Line 387, 391ff: factors of 1.5 or 1.6 are very small differences to draw conclusions. Additionally, the number of samples could be too small to determine the underlying n_{INP} distributions on which the comparisons in this section are based on.

That is our point: we do not see any important (and significant) increase. Increase or decrease factors of 1.5 and 1.6 indicate an almost flat *n*INP trend. In brief, our results are more in line with those by Schrod et al. (2020), presenting a flat trend, than with the previous publications showing a sharp increase of *n*INP in summer.

The number of samples has been already commented on above.

Line 395: State the number of June samples and explain how it was determined that they represent a significant peak.

During June, 7 WT-CRAFT samples have been collected. At all $T_s < -18^\circ\text{C}$, more than 50% (from 57 to 71%) of the June *n*INP data points are higher than the whole campaign median. For T_s between -19.5 and -21.5°C , 57% of the data points are also above the 75th percentile. This percentage varies between 14% and 43% for the remaining T_s . Furthermore, at all $T_s < -18^\circ\text{C}$, the average and mean values of the June samples are higher than the campaign median.

The requested information was added as follows: "Of the 7 samples collected in June, more than 50% (57-71%) were higher than the whole campaign median at this T range. In addition, the average *n*INP during June was up to ~ 3 ($T = -20^\circ\text{C}$) times higher than the average for the rest of the observation period".

Line 409ff: The effect of particle concentration and *n*INP need to be disentangled. The absence of a change in *n*INP with season points to the particle concentration causing the change in the AF.

This issue is now addressed as explained above. Anyhow, we point out that whatever the cause (increasing *n*INP or decreasing particle number concentration) the significant increase in AF demonstrates that the summertime aerosol population is generally more ice active than the spring one.

Line 430: the 50-120nm size range mentioned here provides additional evidence against the chosen lower size limit of 500nm for this study.

Addressed above.

Line 462, Fig.6: The trajectories with land contact are not visible in Fig.6. Removing the not used grey trajectories could help.

They are barely visible because land contact are sporadic. Removing grey lines does not improve the plots. We would like to retain this figure as it is.

Line 468f: specify that the -15°C , PM1 dataset was used. It is unclear why the coarse and fine data is an advantage for DFPC. There is no comparison between coarse and fine *n*INP shown here, except Figs. S7 and S8. Do these maps confirm Mansour et al. and McCluskey et al.?

Done. Indeed, the maps confirm the results of Mansour et al. and McCluskey et al. Similarly to the cited papers, correlation is evident only for PM1 samples. Using PM10 or TSP data results in no correlation; from that derives the advantage of DFPC data.

Line 472f: Has the analysis been tried for -18°C and -22°C ? It is mentioned on line 494ff that experiments showed INP active at -22°C are generated.

Yes. The analysis was performed also on *n*INP datasets at $T = -18$ and -22°C . The resulting correlation maps were less clear, even though some of the features observed at T of -15°C also appear in these T_s . We interpret it as the effect of background land INPs, presuming the influence of dust, which is more active at low temperatures. We notice that in a previous deployment of the technique, we obtained significant correlation maps for Mace Head station using *n*INP at T of -22°C (Mansour et al., 2020b). Nevertheless, in that case the sampling occurred in carefully selected clean marine air masses, which limited the contribution of land sources virtually to zero. Unfortunately, such a technical solution was not applicable at GVB.

Line 483: Provide an interpretation of significant, negative correlated areas. Are they caused by elevated CHL without the expected nINP response?

The basic assumption for the time-lagged correlation approach (Rinaldi et al., 2013; Mansour et al., 2020a; b) is that marine biological aerosols (in this case, organic-enriched sea-spray particles acting as INPs) should follow the evolution of marine biological activity (traced by CHL). For this reason, we target positively correlating sea regions: in our interpretation, these regions have a higher probability of being related to the observed aerosol properties. An inverse correlation with CHL cannot be explained by a physical mechanism: if we assume the INP are biogenic, their concentration can only increase with increasing algal activity (positive correlation) and decrease with decreasing algal activity. Therefore, we excluded negative correlating regions from the analysis as all the non-correlating regions. They are certainly not associated with INP emission, even though we cannot assess their level of productivity.

Line 487: see comment on Fig.7.

Line 534ff: There could be an increase in biological, high T INP from growing biota on CFPC filter during storage at room T.

This hypothesis is very hard to address at present. Maybe additional analysis of heat treated samples could provide some indications in this sense. Unfortunately, the DFPC technique is not suitable for this kind of tests. Actually, we do not know either if the INP properties of our samples are related to the presence of entire cells or cellular exudates or fragments. The PM₁ size range is unlikely to host entire cells, submicron sea-spray organic matter is mostly composed of exudates (e.g., Facchini et al., 2008; Orellana et al., 2011). This is also confirmed by Wilson et al. (2015), which showed that filtering sea-surface microlayer samples by 0.2 μm pore size does not modify the INP properties. This points to INP properties not related to entire organisms, but more likely to molecules, molecular aggregates (the so-called nanogels) or small cell fragments. This suggests that the biological material collected on the filters (and particularly on the PM1 samples) should not be able to “proliferate” during storage.

Facchini M.C., et al., Primary submicron marine aerosol dominated by insoluble organic colloids and aggregates, *GEOPHYSICAL RESEARCH LETTERS*, 35, L17814, doi:10.1029/2008GL034210, 2008.

Orellana M., et al., Marine microgels as a source of cloud condensation nuclei in the high Arctic, *PNAS*, 108, 33, 13612–13617, 2011.

Line 538: It could be added that similar air volumes were collected on the filter for CFPC and WTCRAFT. This is a very useful suggestion. Thank you and it has been added.

Line 549: It could be expected that soluble compounds suppress ice nucleation. Why would the opposite be observed for these samples?

We note that the effect of soluble compounds on the ice nucleation process is a complex problem and partially unsolved question. As a matter of fact published experimental measurements don't give a unique answer. Reischel and Vali performed a study to determine the influence of 22 different salts dissolved in water on four different nucleants. The responses were complicated, with enhancements and suppressions of ice nucleation much larger than what would be expected from changes in water activity only. More recently, Boose et al. (2016) found that the presence of a soluble salt ion leads to an improved ice nucleation ability of dust particles. INP concentrations were measured in the deposition and condensation mode at temperatures between 233 and 253K. Paramonov et al. (2018) examined surface dust collected from three different locations around the world with respect to its ice nucleation activity in deposition and condensation freezing modes. In order to remove the soluble material, the dust samples were washed. Some of the soil

dusts results in an increase or decrease of ice nucleation activity after the washing procedure. Kumar et al. (2018), in laboratory experiments, found an increase of ice nucleation efficiency for K-feldspar microcline only in dilute NH_4^+ solutions, and a decrease in more concentrated solutions. This result was confirmed by Whale et al. (2018), who found that droplets of suspended particles of feldspars and quartz in ammonium solutions, nucleated ice up to around 3°C warmer than pure water case. Belosi et al. (2019) investigated, by means of the DFPC, the ice nucleating effectiveness of Arizona Test Dust (ATD), bare and coated with NaCl. Results showed a decrease in the AF and n_s between bare and ATD coated with NaCl both at $Sw=1.02$ and $Sw=0.96$.

The cited papers confirms that it is difficult to predict the effect on the measured INP number of removing the soluble fraction from ambient aerosol particles (as in immersion freezing WT-CRAFT procedure) or analyzing particles mostly undisturbed (as in the DFPC procedure). This makes our hypothesis reasonable.

The text was updated treating briefly the above discussion:

“The literature offers diverse results and data interpretations, evidencing both increase and suppression of the ice nucleation ability by soluble aerosol components (Reischel and Vali, 1975; Boose et al., 2016; Paramonov et al., 2018; Kumar et al., 2018; Whale et al., 2018)”.

Belosi F., Santachiara G., Laboratory investigation of aerosol coating and capillarity effects on particle ice nucleation in deposition and condensation modes, *Atmos. Res.*, 230, 2019.

Reischel M.T., G.Vali, 1975. Freezing nucleation in aqueous electrolytes *Tellus*, 27, 414–427.

Boose et al., 2016. Ice nucleating particles in the Saharan Air Layer *Atmos. Chem. Phys.*, 16, 9067–9087, 2016.

Kumar et al., 2018. Ice nucleation activity of silicates and aluminosilicates in pure water and aqueous solutions – Part 1: The K-feldspar microcline. *Atmos. Chem. Phys.*, 18, 7057–7079.

Paramonov et al., 2018. A laboratory investigation of the ice nucleation efficiency of three types of mineral and soil dust. *Atmos. Chem. Phys.*, 18, 16515–16536.

Whale et al., 2018. The enhancement and suppression of immersion mode heterogeneous ice-nucleation by solutes. *Chem. Sci.* 9, 4142-4151.

Line 552: A detailed comparison is not needed. To claim sensitivity of arctic nINP on the ice nucleation mode, exemplary DFPC filter need to be analysed with the WT-CRAFT method and vice versa. Otherwise it is speculative.

Yes. This suggestion is valid. The reason why we could not share the filter for both techniques was provided in a previous answer. In addition, in the revised manuscript, we limited to present the sensitivity of Arctic nINP on the ice nucleation mode as a possibility. Please, refer to the revised Sect. 4.1 for more details.

Line 566: It seems plausible that due to small number of samples, the INP population was not well characterized in aforementioned studies, leading to misinterpretation.

We also think the lack of long-term Arctic nINP data is problematic, and we clearly suggest the necessity of future long-term INP monitoring with an online instrument in the same paragraph. Nevertheless, we do not feel comfortable mentioning the number of samples used for previous works as a misleading factor and prefer to avoid discussing it.

Line 578: state which results

Done, in the following way:

“Our AF estimates support the hypothesis that...”.

Line 591: where is shown that land sources have this potential? PM10 nINP do not change much even when more land and open sea are along trajectories and PM1 nINP decrease.

We have reformulated this part, also adding the appropriate reference, to support our statement:
 “Our analysis points out that both marine and terrestrial sources may contribute to the INP population in the study area. Land sources may be potentially important given the higher ice activity of mineral dust and soil particles in comparison to marine particles (McCluskey et al., 2018). On the contrary, marine sources may be significant because of the extension of ice-free sea waters during the Arctic summer”.

Line 591, 592: at what T have mineral dusts a higher activity and at what T are marine INP less active?
 This information is already addressed in the Introduction section.

Line 598: specify the results, e.g. “location of hot spots for marine INPs”
 Added: “In particular, the approach adopted highlights the sea waters to the southwest of Svalbard, those immediately to the east of Greenland and to the northeast of Iceland as potential INP hotspots during our summer campaign”.

Line 610f: The presented analysis does not support this conclusion. A more plausible reason could be differences in the DFPC and WT-CRAFT samples. As mentioned above, to arrive at this conclusion the same samples must be analysed with both methods. Replace “undeniable” by “potential”.
 We have reworded the text as: “We considered many factors that could potentially explain the discrepancy observed (Sect. 4). While differences in the sampling approach and overall measurement uncertainties have certainly contributed to the offset, a different response of aerosol particles to the ice nucleation mode could be also considered as a potential contributing factor. All future investigations into Arctic INP compositions and the ice nucleation process employing both the condensation and immersion freezing approach will provide further understanding of this issue”.

Line 616: The dataset is not unique, other studies have collected Arctic INP covering different seasons e.g. Manson et al., Schrod et al.
 Reworded as: “This study also examined the seasonality of INPs in the Arctic with respect to n_{INP} and AF....”.

Line 624: how did the back-trajectories show this separation? They were done for summer only.
 All the BTs are presented in Figure 6.

Line 629: repetition of line 625
 Modified to avoid repetition.

Tab.1: Consider making 3 columns showing the n_{INP} at -15°C , -18°C , -22°C , relevant for this study, instead of the current last two columns. A minus sign is missing in Bigg, 1996 T range.
 Not all the papers present n_{INP} data at these precise T_s , so we preferred the presented option to provide a wider overview.

Tab.2: Subtracting 1-3 standard deviations from the average values gives negative concentrations. n_{INP} can not be negative, the real variation is clearly asymmetric. Consider reporting the range and average instead and refer to the table instead of listing these values in the text.
 The fact that the standard deviation is higher than the mean value depends on the data distribution and is fairly common in atmospheric data. In the revised version, we reported the standard error, instead. Furthermore, we provided the data range, together with the median value, for completeness.

Tab.3, 4: Scatterplots for all 39 significant correlations reported in these tables could be shown in the supplement and investigated for outliers to exclude false positives.

Scatter plots for all the correlations (significant and not) have been added in the Supporting (Fig. S7-S10).

Fig.3: mention the particle size range used to calculate AF in a), b) c)

Done: "Particle size ranges used to calculate AF are 0.1-1 μm , 0.1-10 μm and 0.1-10 μm for DFPC PM1, DFPC PM10 and WT-CRAFT samples, respectively".

Fig.4: indicate what is shown in a), b), c) in the caption. Specify that uncertainties can be found in sec.2.2.1 (CFPC) and 2.2.2 (WT-CRAFT)

Done.

Fig.6: consider removing grey trajectories.

Fig.7: specify which region belongs to which time lag. Consider overlaying the trajectories as thin lines to indicate upwind locations and clarify the choice of regions.

The regions corresponding to the different time lags are now indicated in the revised caption.

BTs are considered by merging the correlation maps with the CWT plots (which are BT-based). No region can be evidenced in Figure 8b if it is not upwind to the sampling station. So there is no reason to make these plots more complex to present redundant information.

Fig. 8 b): consider showing the trajectories corresponding to the 14 INP samples as thin lines

Only areas covered by the BTs (i.e., upwind to the sampling location during the sampling periods) can be evidenced in this plot as the CWT is BT-derived. So there is no reason to overlap the BTs.

Tab. S1: Column headers "Total Flow (optimized for 50% of filter)" and "Suspension water volume (First frozen drop=0.001 INP L-1)" are unclear. Give a description in the legend and rename the columns, e.g. "air volume" and "suspension water volume". Check the calculation of the suspension volumes.

Done – see above.

Fig.S3: consider using the same y-axis scale for subplots showing the same temperature. Even if there are only 3 spring datapoints in Wex et al. at this T, add -22°C for DFPC and WT-CRAFT as the seasonal change is discussed in the main text. Give a description of the subplots a)-f) in the figure legend.

The seasonal trend at T=-22°C is already in Figure 3a.

Fig. S4: Describe y-axis in the figure legend. Is PM1 or PM10 shown for DFPC? Mark the increase in AF due to the decrease in particle concentration from spring to summer (= particle conc. summer/particle conc. spring) as horizontal line. Spring to summer AF increase close to this line indicate no change in nINP.

Respectfully, we believe that this way of presenting the plot would suggest wrong conclusions. The particle populations are not the same in spring and summer, so even though the AF variation was due only to particle number concentration decrease (being nINP constant), this would imply nonetheless a seasonal modification in the general ice nucleation properties of the particle population (i.e., particles are more efficient in forming ice in summer than in spring).

Technical corrections

Provide more links/references between sections and to tables and figures to navigate the paper.

Line 27: remove "trustful"

Done.

Line 79: "suggested them to be..."

Done.

Line 200: remove “now”

Done.

Line 210: replace “side-by-side” with “in parallel”.

Done.

Line 413: replace “point sin” with “points in”

Done.

Tab. 4b: remove “l” before nINP in the caption.

Done.

Fig. S1: change (a) -18°C to (b) -18°C in the second figure.

Done.

We would like to express sincere gratitude for the referee's helpful comments. Below, we provide our point-by-point responses (in blue colour).

Review of "Ice-nucleating particle concentration measurements from Ny-Ålesund during the Arctic Spring-Summer in 2018" by Rinaldi et al.

General comment:

The authors incorporated several changes in the revised version taking into account most of the listed comments from my initial evaluation. The readability of the manuscript was improved, in part by the new structure and the new additions (text, figures/tables, and data, etc.). Given that several parts of the revised version are completely new, additional concerns appear to me. Although I appreciate the efforts made by the authors, I cannot recommend the publication of this manuscript until the following points are carefully addressed.

We appreciate these general remarks. Following the referee's advice, we revised our manuscript structure and contents to improve the readability and conciseness of this paper. Below, we provide our point-by-point responses.

Major comments:

1. Although English is not my mother tongue, I think that it still needs to be improved. Some examples related to the Introduction only (this apply to the entire text) are provided below.

In the revised version, the text has been language-checked by a native speaker with expertise in the editing of scientific texts.

2. In the first round of comments I asked the following major point:

What is different or what is novel in the present study compared to Wex et al. (2019) and Hartmann et al. (2019)? The answer was that "In this study we present parallel observations of immersion and condensation INPs, which was never achieved in the Arctic. One of our major findings is indeed related to the different behaviour of aerosol particles sampled at GVB under different ice nucleation modes".

We believe this is still a valid uniqueness. We compiled past studies of *n*INP in the Arctic in Table 1, and we see no studies reported *n*INP from multiple ice nucleation measurement techniques. We invite the reviewer to read below for further considerations on the novelty of our manuscript.

However, the authors stated in the manuscript and in the reviewer's response (citing a paper from one of the coauthors) that both heterogeneous ice nucleation mechanisms "might be the same process". Therefore, they are contradicting themselves.

Here, we only intended to introduce what has been studied for condensation vs. immersion, by referring to the two cited studies. We did not mean to conclude that condensation = immersion in the Introduction section. Further assessments from the laboratory and field settings are needed to understand the similarity of ice nucleation modes and processes. Whether condensation and immersion are the same processes or not highly depends on the aerosol properties. We discuss this point in the revised Sect. 4.1.

For major clarity, we rephrased the sentence in the introduction section to: "However, the recent inter-comparison study with two different organic fiber samples shows a difference between condensation freezing and immersion freezing measurements (i.e., ice nucleation efficiency of the former is higher than the latter (Hiranuma et al., 2019). Further laboratory and field assessments are therefore necessary to understand the similarity of ice nucleation modes and processes".

If they authors would like to state that they are reporting the INP concentrations from two different mechanisms, a more in depth analysis is needed where the ice nucleation efficiencies of different aerosol “standards” (e.g., SNOMAX, illite, ATD, etc.) from both instruments are reported. Such experiments need to be run in parallel and using the same aerosol type and particle size. Currently, it is unclear if the observed differences are related to the used methods.

We agree with the reviewer on the importance of understanding more in detail the ice nucleation modes and processes. Nevertheless, this is not the focus of the present paper. In our paper, we present the results of two different INP quantification techniques which provide complementary information. These techniques were developed to represent different ice nucleation modes. Independently on the fact that immersion freezing and condensation freezing may be or may be not the same process in the atmosphere, it is undeniable that aerosol particles are exposed to different conditions during the measurements in the DFPC and WT-CRAFT. For instance, soluble components remain over the sampled particles in DFPC analyses, while they are diluted in the bulk of the extraction water in the WT-CRAFT. Furthermore, in condensation mode measurements, water vapours condense on the surface of sampled aerosol particles, possibly triggering the pore condensation freezing (David et al., 2019; Wagner et al., 2016). In the case of immersion freezing measurements, the pore condensation freezing is not assessable because all particles are scrubbed in the bulk suspension water. For these reasons, it is reasonable to assume that ambient aerosol particles may respond differently to the different analytical techniques. However, we stress that this is not a central point of our work (see below for further discussion). Considering that our INP detection techniques are within an acceptable agreement, according to previously published INP intercomparisons (DeMott et al., 2017; Hiranuma et al., 2019), and that the observed concentration gap does not affect the conclusions presented in this study (given the consistency of the DFPC and WT-CRAFT time trends) we believe that our results do not need the support of an in-depth intercomparison study with aerosol “standards” to be published.

However, we considered the reviewer suggestion and conducted additional immersion freezing experiments of Arizona Test Dust (ATD) with WT-CRAFT, comparing the results to previously published ATD result by another immersion freezing technique, Colorado State University Ice Spectrometer (CSU-IS; Perkins et al., 2020). The ATD used for WT-CRAFT is the same sample used in Möhler et al. (2006) and Niemand et al. (2012) (Powder Technology Inc.; 0-3 μm). As ascribed in Perkins et al (2020), the A2 ATD (Powder Technology Inc.; $\sim 22\%$ $< 2.75 \mu\text{m}$, $\sim 58\%$ $< 11 \mu\text{m}$, and $\sim 90\%$ $< 44 \mu\text{m}$) was used for the CSU-IS analysis. For the WT-CRAFT measurements, we prepared an original stock suspension of 0.1 wt% ATD in ultrapure water and two diluted suspensions (x100 and x1000). The diluted spectra and original spectrum were merged with a method introduced in the main manuscript Sect. 2.2.2. The figure shown below presents the estimated INP concentration per unit mass (nm, g^{-1}) with WT-CRAFT and CSU-IS in the temperature range of $-5 \text{ }^\circ\text{C} \geq T \geq -25 \text{ }^\circ\text{C}$ and $-2 \text{ }^\circ\text{C} \geq T \geq -24 \text{ }^\circ\text{C}$, respectively. As seen in the figure below, we see a reasonable agreement between the two techniques within uncertainties for the assessed temperatures. Thus, we believe that the applicability of WT-CRAFT for immersion freezing experiment is validated within its detection limits and capabilities (Sect. 2.2.2).

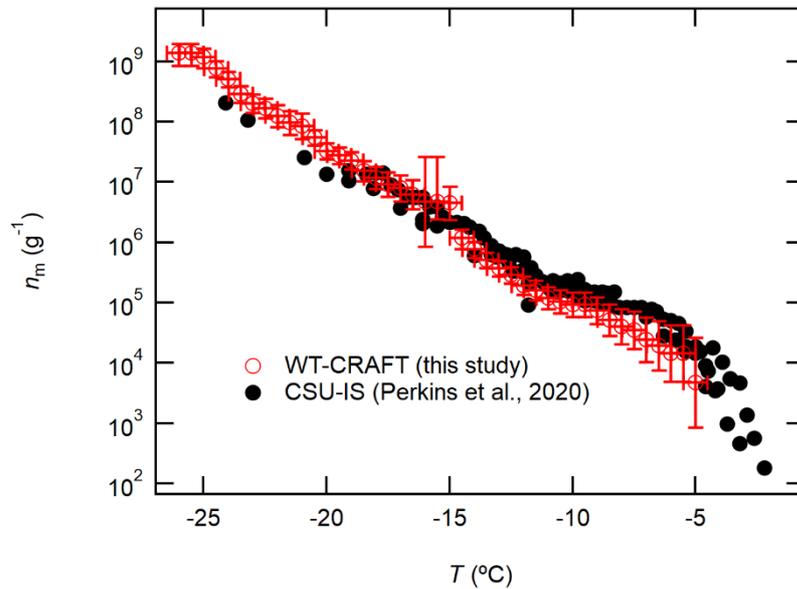


Figure. Indirect comparison of immersion freezing efficiencies (n_m) of Arizona Test Dust estimated by WT-CRAFT and CSU-IS.

In addition, condensation freezing mode with the same ATD dust was investigated by means of the DFPC (Belosi F. et al., 2018). The obtained ice-active surface site density n_s at T of -22°C and S_w of 1.02 (ca. 10^{10} m^{-2}) was comparable with the results by Niemand et al. (2012) (compare Figure 1b of Belosi et al. with Figure 6 of Niemand et al.). Furthermore, an intercomparison between DFPC and FRIDGE (FRankfurt Ice Nucleation Deposition freezinG Experiment) was carried out with different dust types, including Illite, and the obtained n_s values were in agreement between the two techniques (Belosi et al., 2016; see Figure below).

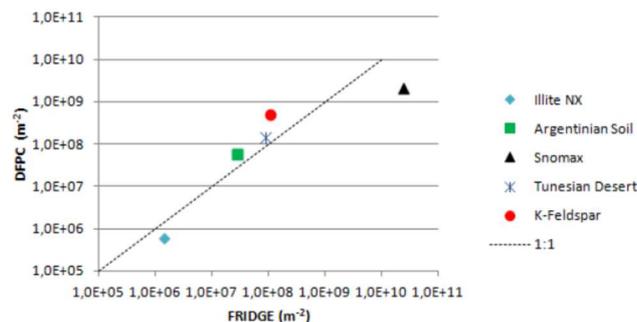


Figure. Comparison of n_s of standard aerosol types obtained by DFPC and FRIDGE (from Belosi et al., 2016)

Although the above results cannot replace a systematic intercomparison study, they suggest that for simple aerosol standards, DFPC and WT-CRAFT respond consistently to other well characterized INP quantification techniques (and therefore, we can assume, consistently to each other). This suggests that the complexity of ambient aerosol particles may be a potential reason to explain the observed discrepancy between the two techniques at GVB.

Nevertheless, we fully understand the concerns of the reviewers about relying on ice nucleation modes for explaining the observed difference between $n_{\text{INP}_{\text{DFPC}}}$ and $n_{\text{INP}_{\text{WT-CRAFT}}}$. As we have made clear in the revised manuscript, we cannot be conclusive about this, and we hope to be able to further investigate this issue in future follow-up studies. To allay further misgivings, we softened our tones and revised the text to present the particle sensitivity to different ice nucleation modes only as a potential reason. In the revised

manuscript, we also discussed other potential factors for the observed discrepancy (please refer to the revised Sect. 4.1). Overall, the addressed potential factors include (1) differences in measurement uncertainties, (2) sampling apparatus, (3) sample storage protocols, (4) substrate types, (5) sampling durations and (6) ice nucleation paths (condensation vs immersion freezing).

Unfortunately, we could not reduce the potential discrepancy factors during our 2018 sampling campaign, for the following reasons:

(1) we cannot use an identical substrate for the two INP analysis techniques. We have assessed the applicability of the cellulose membrane (optimal for DFPC) in WT-CRAFT, and we found notable background artifacts from the blank substrate below -22°C . Thus, the same filter sample cannot be shared for both INP measurement techniques.

(2) Our aerosol samplers employed different sampling periods for optimized flow conditions and estimated detection limits. The aerosol sampler for DFPC employed high flows, which were needed for the desired cut-size (see Sect. 2.1). If we conducted sampling with a longer period than what we employed, DFPC would likely suffer from filter overloading issues. Likewise, reducing the sampling time for WT-CRAFT would have created detection limit issues.

Finally, we would like to clarify here that explaining the gap is not a major point of the study. Our work extends the INP observations at GVB, contributing to filling the present lack of observations in the Arctic. Furthermore, it provides information on the AF, which was never calculated before at GVB and which was only rarely addressed at Arctic sites. One of the major findings of our work is that the seasonality of $n\text{INP}$ can be significantly different from what was observed in previous studies. This finding is supported by both the INP datasets, notwithstanding the concentration gap and all the sampling differences. Our results evidence potentially a great interannual variability and the necessity for further data coverage to better understand INP dynamics over the Arctic. Furthermore, we present and discuss for the first time the seasonality of the AF, intended as a proxy of the overall ice nucleation ability of the particle population at the study site. Finally, our study reports information on the ice behaviour of fine vs coarse aerosol particles, which was addressed only in a few more Arctic studies so far.

Belosi F., Schrod J., Nicosia A., Santachiara G., Prodi F., Weber D., Bingemer H., Off-Line measurements of ice nucleating particles, European Aerosol Conference, Tours 4-9 september, 2016.

Belosi F., Piazza M., Nicosia A., Santachiara G., Influence of supersaturation on the concentration of ice nucleating particles, *Tellus B: Chemical and Physical Meteorology*, 70:1, 2018.

David, R. O., Marcolli, C., Fahrni, J., Qiu, Y., Perez-Sirkin, Y. A., Molinero, V., Mahrt, F., Brühwiler, D., Lohmann, U., and Kanji, Z. A.: Pore condensation and freezing is responsible for ice formation below water saturation for porous particles, *Proceedings of the National Academy of Sciences*, 116, 8184–8189, <https://doi.org/10.1073/pnas.1813647116>, 2019.

Möhler, O., Field, P. R., Connolly, P., Benz, S., Saathoff, H., Schnaiter, M., Wagner, R., Cotton, R., Krämer, M., Mangold, A., and Heymsfield, A. J.: Efficiency of the deposition mode ice nucleation on mineral dust particles, *Atmos. Chem. Phys.*, 6, 3007–3021, <https://doi.org/10.5194/acp-6-3007-2006>, 2006.

Niemand, M., Moehler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., and Leisner, T.: Parameterization of immersion freezing on mineral dust particles: An application in a regional scale model, *J. Atmos. Sci.*, 69, 3077–3092, 2012.

Perkins, R. J., Gillette, S. M., Hill, T. C. J., and Demott, P. J.: The labile nature of ice nucleation by Arizona Test Dust, *ACS Earth Sp Chem.* 4, 133–141, 2020.

Wagner, R., Kiselev, A., Möhler, O., Saathoff, H., and Steinke, I.: Pre-activation of ice-nucleating particles by the pore condensation and freezing mechanism, *Atmos. Chem. Phys.*, 16, 2025–2042, <https://doi.org/10.5194/acp-16-2025-2016>, 2016.

Note that the size of the aerosol particles analyzed by both instruments are not the same as the pore size of the filters used by the DFPC (0.45 μm) and WT-CRAFT (0.2 μm) are not identical. Although it is clear that supermicron particles are likely the most efficient INPs, small particles (between 0.2 μm and 0.45 μm) can be soluble and change the composition of the droplets and their freezing points. Note that the used filters in both systems are neither the same. Although the authors argued in their answer that the pore size is not important, I disagree with them and I think that the combination of different filter type and different pore size can result in the capture of different aerosol particles.

The difference in pore sizes might not be a substantial factor to explain the gap between DFPC and WT-CRAFT measurements. While we cannot rule out the possibility that DFPC misses ice nucleation active aerosol particles in the size range between 0.2 and 0.45 μm , this difference might not substantially contribute to the gap as $n_{\text{INP}_{\text{DFPC}}}$ is generally higher than $n_{\text{INP}_{\text{WT-CRAFT}}}$. This point is now discussed in the revised Sect. 4.1.

Also, given that the sampling time of the samples analyzed in the WT-CRAFT was much longer (4 days) than the samples analyzed in the DFPC (4 h) it is very likely that the aerosol particles analyzed in both instruments are completely different. It would have been desirable that the same filter was analyzed by both methods.

The motivations for which it was not possible to analyse the same samples by the two techniques are enlisted above. The difference in sampling durations (~ 4 hours for the DFPC and ~ 4 days for WT-CRAFT) is a valid concern. Although we cannot exclude the possibility that short episodes of high INPs-containing air masses increased $n_{\text{INP}_{\text{DFPC}}}$, it is unlikely that this factor can explain a systematic difference, like that object of this discussion. This would presume a strong n_{INP} daily trend with the maximum coinciding with the DFPC sampling time. We excluded this by analysing the daily evolution of the particle number concentration, which does not present any evident diurnal trend both in spring and summer (not shown).

3. (1) How can the authors confirm the absence of the liquid phase in the DFPC? (2) Is it really possible to have ice crystal growing on individual aerosol particles? From my personal experience it is very difficult to avoid that several particles enter in contact when collected in a filter. (3) Therefore, It would be nice if the authors can provide evidence of how the aerosol particles were distributed in their nitrocellulose filters and a picture on how an ice crystals forms on a single particle.

(1) The air enters the DFPC chamber through a perforated plate, spreads into an ice bed, and becomes saturated with respect to ice, but undersaturated with respect to water. Afterward, the air flows into the second chamber, grazing the filter located on a metal plate cooled by a Peltier device. Only in this small surface air becomes supersaturated with respect to water. The following Figure presents a scheme of the DFPC.



Figure. 1: air inlet; 2: minced ice; 3: slit and air temperature thermocouple; 4: filter; 5: filter temperature thermocouple; 6: Peltier cooling device; 7: thermocouple; 8: air outlet; 9: plexiglass cover; 10: observation slit.

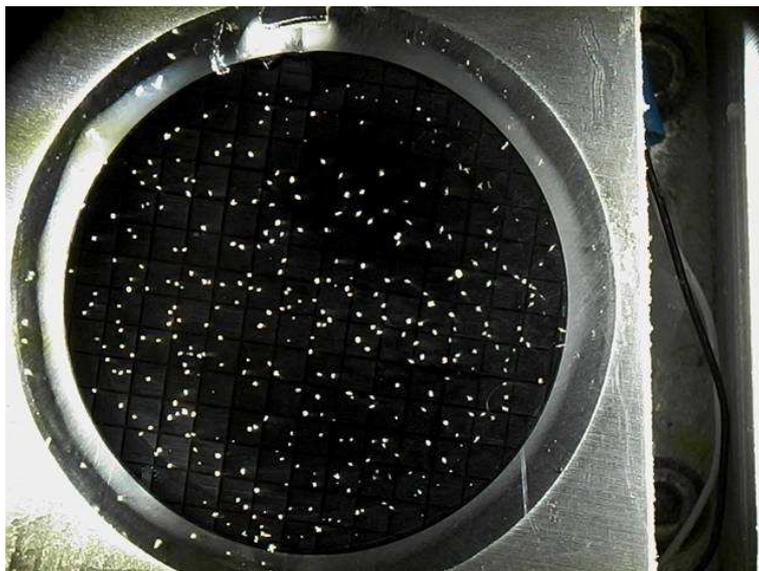
The manuscript was modified as follows:

“Particle-free air entered the DFPC chamber through a perforated plate, spreading to an ice bed to become saturated with respect to ice but undersaturated with respect to water. The air then proceeded to the filter, cooled by a Peltier device in contact with the supporting metal plate. Only at this point, did the air become supersaturated with respect to water. By controlling the T_s of the filter and surrounding air, the samples could be exposed to different T_s while keeping the water saturation ratio (S_w) above 1. The supersaturation ratio was calculated theoretically from vapour pressures of ice and water at the T_s considered (Buck, 1981). More details of the DFPC working principle can be found in the supplement of deMott et al. (2018)”.

DeMott P.J., Möhler O., Cziczo D.J., Hiranuma N., Petters M.D., Petters Sarah S., Belosi F., Bingemer H.G., Brooks S.D., Budke C., Burkert-Kohn M., Collier K.N., Danielczok A., Eppers O., Felgitsch L., Garimella S., Grothe H., Herenz P., Hill T.C.J., Höhler K., Kanji Z. A., Kiselev A., Koop T., Kristensen T. B., Krüger K., Kulkarni G., Levin E. J. T., Murray B.J., Nicosia A., O'Sullivan D., Peckaus A., Polen M. J., Price H. C., Reicher N., Rothenberg D. A., Rudich Y., Santachiara G., Schiebel T., Schrod J., Seifried T. M., Stratmann F., Sullivan R. C., Suski K. J., Szakáll M., Taylor H. P., Ullrich R., Vergara-Temprado J., Wagner R., Whale T. F., Weber D., Welti A., Wilson T. W., Wolf M. J., and Zenke J., " The Fifth International Workshop on Ice Nucleation phase 2 (FIN-02): Laboratory intercomparison of ice nucleation measurements", *Atmos. Meas. Tech.*, <https://doi.org/10.5194/amt-2018-191>, Vol. 11, 6231-6257, 2018

(2) Avoiding the overlapping of particles on the filter is the main reason for the low sampling times of DFPC samples. By considering the sampled air volume (about 5.5 m³) and the aerosol particle concentration in the range 0.1-10 μm (about 80 cm⁻³), the probability of particles overlapping on the filter surface (47 mm diameter) is very low. More in detail, considering also the average aerosol particle size distribution at the sampling site, we estimated a total surface occupied by particles on the filter of the order of 0.02 cm² on a total filtering surface of ~10 cm². This means a low particle density on the filter.

(3) We attach a picture of ice crystals grown on one of the analyzed filters to show the sparseness of the ice crystals on the filter. Analysis conditions are the following: $T(\text{filter})=-22^\circ\text{C}$; $T(\text{air}):-19.6^\circ\text{C}$; $S_w: 1.02$; PM1 fraction.



4. “nINP measured in condensation mode (DFPC) resulted generally higher than those measured in immersion mode (WT-CRAFT) and the deviation became even more apparent towards higher T”. The authors need to clearly discuss this. How realistic is this observation and what is the reasoning for more aerosol particles to act as INPs via the condensation freezing? Also, why at higher temperatures condensation freezing becomes a more efficient pathway to catalyze ice formation compared to immersion freezing?

In the revised version, we considered a different response of aerosol particles to the different INP measurement techniques as a possibility, among others, mainly related to differences in the sampling approaches and measurement uncertainties. We admit that our observations are not conclusive and that further studies would be necessary to address this point. Please see our answer to comment #2 above.

5. The overall uncertainties of individual ice nucleation measurements cannot explain entirely the observed discrepancy”. How the authors reached this conclusion. What type of analysis was it performed? How the different uncertainties were calculated and combined?

We compared DFPC and WT-CRAFT results during periods of parallel sampling to reduced the sources of variability in the results. To do this, we averaged multiple DFPC samples to match with the corresponding WT-CRAFT sample collected over the same period. For all the samples, $nINP_{DFPC}$ was higher than $nINP_{WT-CRAFT}$ and the difference was:

$$\Delta_nINP = nINP_{DFPC} - nINP_{WT-CRAFT}.$$

For each sample, we calculated:

$$\Delta_nINP' = (nINP_{DFPC} - \sigma INP_{DFPC}) - (nINP_{WT-CRAFT} + \sigma INP_{WT-CRAFT}),$$

where σINP_{DFPC} and $\sigma INP_{WT-CRAFT}$ are the absolute $nINP$ uncertainties for DFPC and WT-CRAFT respectively, calculated according to the overall relative uncertainties presented in Sect. 2.2.1 and 2.2.2. The assumption is that both DFPC and WT-CRAFT uncertainties contributed to their maximum to generate the observed $nINP$ gap. In other words, we considered the maximum possible underestimation error for DFPC and the maximum possible overestimation error for WT-CRAFT, to check if this was enough to close the $nINP$ gap.

Comparing Δ_nINP with Δ_nINP' , we estimated to what extent the measurement uncertainty can explain the observed concentration gaps.

The results of this comparison are reported in the revised text as follows:

“The uncertainties of individual ice nucleation measurements cannot entirely explain the discrepancy observed. Even considering the largest error contribution, uncertainties can on average explain up to 50, 66 and 76% of the observed $nINP$ offset at -18, -22 and -15°C, respectively. These percentages were calculated by assuming that the measurement uncertainties combined with each other to determine the maximum possible reduction of $nINP$ difference (i.e., assuming the maximum possible underestimation of $nINP_{DFPC}$ and the maximum overestimation of $nINP_{WT-CRAFT}$) and considering only periods of parallel sampling (to minimize sources of discrepancy unrelated to measurement uncertainty)”.

6. “As we do not observe any strong indications of these influence within given uncertainties, it is at least conclusive that the size-dependant collection efficiency of aerosol particles is not a solo-dominating factor causing the difference between $nINP_{WT-CRAFT}$ and $nINP_{DFPC}$.” This is in indirect conclusion without any clear robust evidence supporting it.

We admit that our evidence cannot be fully conclusive. Please, see the above considerations.

7. “the suppression of INPs due to the sample storage difference does not explain the observed general trend of $nINP_{DFPC} > nINP_{WT-CRAFT}$.” Why not? How was this evaluated and confirmed?

The difference in sample storing methods cannot be a sole factor to explain the discrepancy. As we kept the samples for DFPC at the room air T (and the WT-CRAFT samples at 4 °C except during transportation), the

suppression of INPs is expected to be more obvious for DFPC than WT-CRAFT, which is not supported by the observed trend of $n\text{INP}_{\text{DFPC}} > n\text{INP}_{\text{WT-CRAFT}}$.

We are aware that Beall et al. (2020) recently demonstrated that storage of precipitation samples can lead to losses of INPs (especially the heat-labile ones). Storage at room temperature and at 4°C resulted basically in INP losses, with the former conditions showing somewhat higher losses than the latter. Nevertheless, the results by Beall et al. (2020) are not necessarily valid for particle samples collected on filters. Furthermore, there is not yet a consensus in the literature (and within the scientific community) about the effect of sample storage on INP measurements, so answering this question quantitatively is very hard at present. For instance, Wex et al. (2019) stated that “it is not yet known with certainty how the temperature during storage will affect INP concentrations”. Bigg (1990) reported no significant deterioration in INP concentration after 5 years, for filters sealed in a dry-sealed container. Conen et al. (2015) commented that storage of filters at -20°C may not have been necessary, because re-analysis of filters analyzed previously showed no effect of storage. Wex et al. (2015) and Polen et al. (2016) showed that storage at temperatures above 0°C or even under freezing conditions has been found to reduce the ice activity of biogenic INPs. Stopelli et al. (2014) studied INP concentrations in a snow sample stored at 4°C and observed a decrease in the concentration of INPs active at -10°C over 30 days (factor of 2).

We also evaluated the effect of sample storage on the $\Delta n\text{INP}/\Delta T$ slope, we evidence that DFPC samples were not refrigerated during storage, while WT-CRAFT ones were kept at 4°C. Therefore, based on the limited literature on this topic, degradation of INPs during storage would have caused higher losses in DFPC samples, resulting in the opposite effect on the $\Delta n\text{INP}/\Delta T$ slope. For this reason, we believe that losses during storage cannot be considered a valid explanation for the observed gap, at least based on the current knowledge of the topic. We now discuss this point in the revised Sect. 4.1.

Bigg, E.K., 1990. Measurement of concentrations of natural ice nuclei. *Atmos. Res.*, 25, 397-408.

Conen et al., 2015. Atmospheric ice nuclei at the high-altitude observatory Jungfraujoch, Switzerland. *Tellus B*, 67, 1-10.

Conen et al., 2015. Atmospheric ice nuclei at the high-altitude observatory Jungfraujoch, Switzerland. *Tellus B*, 67, 1-10.

Polen, M., Lawlis, E., and Sullivan, R. C.: The unstable ice nucleation properties of Snomax (R) bacterial particles, *J. Geophys. Res.-Atmos.*, 121, 11666–11678, <https://doi.org/10.1002/2016jd025251>, 2016.

Stopelli, E., Conen, F., Zimmermann, L., Alewell, C., and Morris, C. E.: Freezing nucleation apparatus puts new slant on study of biological ice nucleators in precipitation, *Atmos. Meas. Tech.*, 7, 129–134, <https://doi.org/10.5194/amt-7-129-2014>, 2014.

8. “results from higher ice activation at the conditions of DFPC analyses rather than from higher aerosol particle number concentration in DFPC samples”. I sort of agree with the authors; however, how about the chemical composition? No drastic changes in the aerosol particle concentration does not mean that the chemical composition along 4 days is constant.

This comment is pertinent. Nevertheless, we believe that a systematic aerosol particle chemical composition difference occurring only during DFPC sampling time, so that it can determine the observed difference, is highly unlikely.

9. “A detailed intercomparison of techniques is not under the scope of this study”. I disagree with this statement. Given that the authors are claiming that they report the INPs concentration via two different ice nucleation modes, they need to provide convincing evidence and not unsupported statements such as “suggest that the unexplained concentration gap might stem from other factors, and it is plausible to consider a different sensitivity of Arctic INPs to different ice nucleation modes”.

Discussed above – see our response to comment #2.

10. “we conclude that a different sensitivity of Arctic INPs to different ice nucleation modes explains the observed discrepancy.” and “it seems conclusive to address there is ice nucleation mode dependent INP propensity at GVB in 2018 at least.” I disagree with both statements.

Both statements have been removed, and other factors contributing to the observed discrepancy between DFPC and WT-CRFAT measurements are discussed in Sect. 4.1.

Minor comments:

1. I am not sure how useful is to use “T” and “Ts” instead of “temperature” and “temperatures”.

The temperature word appears numerous times in our manuscript, and we decided to use an abbreviation for it to increase the conciseness and readability of our manuscript. We had a previous suggestion of doing this for another ACP manuscript. However, we understand that the ACP standard may have been updated and are willing to comply with the current preference/rule by the editorial team.

2. “We hypothesized that the n_{INP} variability at a single T can be explained by differences in freezing modes.” What does it mean?

We admit that it was not intuitive, and the sentence was removed from the revised Introduction. The potential factors of the observed n_{INP} variability are now discussed in Sect. 4.1.

3. It is mentioned that the Back-trajectories were “simulated for an altitude of 100 m above mean sea level (amsl) over the GVB”. Given that the long-range transport of aerosol particles was evaluated in the present study, why higher altitudes were not taken into account as the long-range transport of air masses does not take place that close to the surface?

For clarity, 100 m was the height of the arrival point of air mass evaluated by the back-trajectory analysis. It was assumed to be close to the ground because we sampled at the ground level. This does not mean that the trajectory always travelled at that same height.

4. “The range of n_{INP} from Table 1 is roughly comprised between 10^{-2} and 10^3 m^{-3} ”. I would restrict this to the temperate range of the present study.

We considered the range between -9 to -25°C which covers the range in which we observed some ice nucleation activities in our samples. Restricting it only to the DFPC temperatures would not be representative of our full dataset. The temperature range is now clarified in the revised text:

“The range of n_{INP} from Table 1 is roughly between 10^{-2} and 10^3 m^{-3} in the T range between -9 and -25°C , in which we detected ice nucleation activity in our samples. This n_{INP} range covers the majority of our measurements”.

5. “suggesting that the dominant INP sources may be located at long distances (scale of the order of 100s-1000s km)” How the authors reached those numbers?

We removed those numbers: we inserted this specification upon request of the other reviewer. Long-range transport is a common expression in the field, indicating sources located far from the sampling location. Usually, this expression is used in contraposition to “local sources”, when it is not possible to provide more details. We believe in this case this general indication is enough.

6. Last paragraph Section 4.2. I suggest to expand this a bit more including previous studies where the Arctic aerosol composition and sources are discussed including new particle formation.

This is a good suggestion. The paragraph was extended as follows: “The higher AF of summertime (local) aerosol particles may be related to the enhanced contribution of super-micrometer aerosol particles, which

we have shown to be markedly more ice active than sub-micrometer particles. Nevertheless, we cannot exclude or quantify, the contribution of other physico-chemical properties of aerosol particles, which may vary between spring and summer (e.g., chemical composition).

It is worth considering that changes in the estimated AF are influenced not only by variations of n_{INP} but also by variations of the concentration of non-ice-active aerosol particles, including secondary aerosols formed through new particle formation (NPF) mechanisms. Secondary aerosol particles may not contribute to INPs (Kanji et al., 2017), but they can lower the estimated AF. Recently, Beck et al. (2021) evidenced that different mechanisms, precursors and formation rates characterize spring and summertime NPF events at GVB. Dall'Osto et al. (2019) evidenced that the production of fresh particles is frequent during the period from May to August at GVB, while April is characterized by the presence of aged, accumulation mode particles. These aspects may influence the seasonal variation of the estimated AF. Dall'Osto et al. (2017; 2018; 2019) linked NPF frequency in the Arctic atmosphere to the fast-decreasing sea ice extent, probably via increased phytoplankton productivity. This leads to the hypothesis of increasing NFP impact in the future. By the same token, the predicted shrinking of snow and sea-ice coverage in the Arctic is likely to increase the ambient n_{INP} from sea spray and terrestrial sources, such as mineral and soil dust particles (Tobo et al., 2019). Predicting future n_{INP} and aerosol particle AF over the Arctic in such a rapidly changing scenario is challenging. It, however, provides the motivation for further investigation of INP processes in the Arctic region”.

7. The authors showed the clear difference in the ice nucleating abilities of submicron and supermicron particles; however, little was mentioned about their composition. I suggest to add a little discussion about this based on the available literature for the Arctic trying to link the particles composition with their size.

In principle, this is a good suggestion and we already considered adding such considerations while working on the manuscript. Previous studies (e.g., Giardi et al. 2018, Zangrando et al. 2012, Young et al., 2016, to remain close to the study location) have shown that sub-micrometer aerosol particles in the Arctic are dominated by ammonium sulfate and nitrate, together with organic matter. These components can be both biogenic or anthropogenic, with a dominance of the former with respect to the latter which varies according to the period of the year (Arctic Haze). Coarse fraction aerosol particles are instead contributed by sea salt and mineral/soil dust particles, even though organic and biological particles can be also important in this size range. These differences certainly contribute to the different ice nucleation efficiencies of fine and coarse aerosol particles discussed in this work. Nevertheless, the bulk aerosol chemical composition can hardly be related to INP properties in a quantitative way, considering that only about 1 particle over 10^5 can act as an INP. The bulk chemical composition depends necessarily on the rest of the particle population. Therefore, we preferred to avoid presenting considerations on the link between chemical composition and INP properties in the manuscript, which could be seen as speculative.

We clarified the necessity of identifying the chemical identity of the Arctic INPs in the revised Sect. 3.6.1: “It should be noted, however, that our tracer analysis only infers the aerosol properties, with the result that further analysis of INP identities and properties (e.g., ice crystal residual analysis) would be necessary to reveal the source of INPs”.

Giardi et al., *Rend. Fis. Acc. Lincei*, DOI 10.1007/s12210-016-0529-3

Young et al., *Atmos. Chem. Phys.*, 16, 4063–4079, 2016

Zangrando et al., *Atmos. Chem. Phys.*, 12, 10453–10463, 2012

Technical Comments:

Line 41: Add a reference after “quantify”.

Added (Schmale et al., 2021)

Schmale, J., Zieger, P. & Ekman, A.M.L. Aerosols in current and future Arctic climate. *Nat. Clim. Chang.* **11**, 95–105 (2021). <https://doi.org/10.1038/s41558-020-00969-5>

Line 57: Change “condensation nucleus” by “INP”.

Done.

Line 57: Change “a nucleus” by “INP”.

Done.

Line 58: Add “at temperatures above 0C” after “water droplet”.

Done.

Line 58: Change “in” by “via”.

Done.

Line 58: Add “when T is decreased” after “immersion freezing”.

Done.

Line 58: What do the authors mean with “extramural”?

Changed in: “In contact freezing, an INP promotes freezing when it comes into contact with a supercooled droplet from the outside”.

Line 65: Remove “according to Fig. 13”.

Done.

Line 66: “an exception of K-feldspar”. Do the authors mean that K-feldspar it not a mineral?

We mean that it is an exception as it “can facilitate ice nucleation at much higher T_s compared to other mineral compositions (Atkinson et al., 2013; Harrison et al., 2019)”.

Line 67: “biogenic INPs”. It was called biotic above.

We say that INPs can be of “biotic origin”, which means they are “biogenic”. We do not see any inconsistency in the terminology here.

Line 67: “to support”. To favor?

Changed into favour.

Line 75: Delete “next”.

Done.

Line 76: “The Ocean was considered to be a prevalent source of INPs”. What do the authors mean?

Reworded: “The Ocean was identified as a major source of INPs,…”

Lines 76-77: “based on the high negative correlation between n_{INP} and the time since the sampled air masses have been over the open ocean”. Unclear.

We have rephrased the sentence as follows:

“The ocean was identified as a major source of INPs since n_{INP} fell as a function of the length of time that had elapsed since the air masses had left the open sea”.

Line 79: “probable submicron fragments”. Unclear.

Reworded in “fragments of…”.

Lines 82-83: “oceanic air tripled after about one day of passage over land”. Unclear.

Reworded: “During the summer, the authors observed that n_{INP} (T of -15°C) in air masses from the ocean increased three fold after about one day of passage over land”.

Line 94: Remove “six samples” and “seven samples” as I found it useless.

Done.

Line 96: Should “Santl-Temkiv et al., 2019; Wex et al., 2019” be “Tobo et al. (2019)”?

This sentence is not related to Tobo et al. (2019) specifically. We refer here to the enhancement of n_{INP} reported by the all the cited papers (Wex et al., 2019; Santl-Temkiv et al., 2019 and Tobo et al. 2019). In this sense, the citation is not wrong as:

- Wex et al. (2019) state that: “In summer, the higher bioaerosol concentrations compared to spring indicate contributions from local and regional terrestrial and marine ice-free areas”; “these INPs can originate from both terrestrial and marine sources in the Arctic. These sources are strong in summer and weak or absent in winter, depending on the conditions on the ground”.
- Santl-Temkiv et al. (2019) state that: “In summer, the higher bioaerosol concentrations compared to spring indicate contributions from local and regional terrestrial and marine ice-free areas”; “Based on this observation, we suggest that the high concentrations of INPs in air during summer may be partially related to strengthened local terrestrial sources, in particular soil dust”
- Tobo et al. (2019) state that: “This suggests that significant local sources of INPs other than marine organic materials might exist in and/or around the Svalbard region in the summer”

In brief, all the three papers attribute the summer time enhancement of *n*INP to local sources. We have added the reference to Tobo et al. (2019).

Line 99: “to fill the present gap of INP observations in the Arctic environment”. The lack of measurements?

Reworded: “Our study aims to add to the still scant INP observations in the Arctic environment...”.

Lines 100-101: “by two INP quantification techniques”. This is very awkward.

Reformulated: “In particular, we extend the INP observations at GVB, previously only 13 samples (Wex et al., 2019), presenting the results of 61 samples investigated with two offline INP measurement techniques”.

Lines 102-104: Delete “Recent modeling simulation and remote sensing studies suggest immersion freezing is the most relevant heterogeneous ice nucleation mechanism in mixed-phase clouds, which are prevalent in the Arctic (Hande and Hoose, 2017; Westbrook and Illingworth, 2011).”

Done.

Line 175: Delete “Next, we briefly explain our experimental procedure”.

Done.

Lines 214-215: “The number concentration in the resulting overlapping range was taken from the SMPS data as SMPS provides more size bins”. Unclear.

“The number concentration in the resulting overlapping range was taken from the SMPS data”.

Line 228: “operating flow rate of 2.3 m³ h⁻¹” for how long?

24 h, added.

Lines 325-235: Delete.

Deleted.

Line 510: “bio INP”. It was not defined before.

“Bio” was removed.