Response to Referee #1

First of all, the authors thank the referee for submitting helpful and meaningful comments, which lead to improvements and clarifications within the manuscript.
Below, we provide our point-by-point responses. For clarity and easy visualization, the Referee's comments (***RC***) are shown from here on in black. The authors' responses (***AR***) are in blue color below each of the referee's statement. In addition to the responses to referees' comments, we further modified the manuscript to increase its clarity and readability. Abstract and conclusions were mostly rewritten. The Section on the ice nucleation active site density (n_s) was removed; n_s was substituted by the Activate Fraction (AF) parameter in the discussion. The Results section was re-organized for major clarity and separated from the Discussion Section. All the changes can be checked in the track change version of the manuscript, where the new text is highlighted in yellow color. We introduce the revised materials in green color along/below each one of your response (otherwise directed to the Track Changes version manuscript). All references are available in the end of this AR document.

RC: *General comments*
Ambient measurements from Spitzbergen during spring and summer 2018 are reported and analysed. The dataset is a welcome addition to the growing body of ice nucleation concentration measurements from the Arctic region. For the analysis the authors correlate the measured INP concentrations to bulk particle properties, season, meteorology and airmass trajectory. (1) Unfortunately, the analysis is not well motivated by hypotheses, and the results not presented clearly. Additionally, I suspect that (2) the set of measurements is too small to perform a robust analysis and the found correlations might be random. By ignoring this, (3) the authors got mislead to overinterpretations and speculative conclusions. The interpretation of data (e.g. concerning INP size and land vs. marine contributions) agrees with previous studies and (4) no new insights are obtained. The one interesting finding is that there was no seasonal variation observed in 2018. The authors need to be more specific in their descriptions, quantitative within reason for the interpretation and visualize their findings clearer to turn this manuscript into a valuable contribution to the field of ambient ice nucleation measurements.
AR: The authors appreciate these general remarks and constructive criticisms regarding our manuscript by Referee #1. We found these as invaluable guidance. We believe that the hypotheses and analyses in the revised manuscript are robust and insightful. We have limited but very good data, which are statistically valid. We admit that we have made some insufficient discussions, leading some of our data interpretations in an original manuscript to be speculative. Based on the peer-review comments, we removed/modified them to motivate the research. To allay the reviewer's concerns and mitigate any misgivings, the authors have decided to change the title of manuscript to "**Ice-nucleating particle concentration measurements from Ny-Ålesund during the Arctic Spring-Summer in 2018**", reflecting our changes and articulate what is truly presented in the revised version paper. We have also revised our abstract as well as the conclusion to reflect all of our major revisions (please read the Track Changes version paper). Below, we provide our point-by-point responses in hopes of our manuscript being considered for another review by the reviewer.

Specific comments
Major changes are suggested for each section. Minor corrections or clarification requests at specific line numbers are listed below.

RC: **Structure**
The manuscript could be structured better. Adding a Discussion section instead of including the discussion with the results would help the organization of the paper. Currently the Result section has mixed-in discussion, interpretation, literature comparison and some method description. Isolating the Results, visualizing and explaining them more specifically would be helpful to judge the interpretation.
AR: We thank the reviewer for the suggestion. We added a Discussion Section in order to improve the manuscript organization and clarity. The Results section now focus more on describing our data analysis and observation. In our new Discussion Section, we detailed: Interpretation of $n\mathrm{INP}_{\mathrm{DFPC}}$ and $n\mathrm{INP}_{\mathrm{WT\text{-}CRAFT}}$ discrepancy, Interpretation of seasonal variability of nINP, Sources of INPs in Ny-Ålesund etc.

RC: **Citations**
A high number of citations are given, but it is often unspecific what information can be found in which citation or why several citations are listed. Better integration in the text would be helpful instead listing multiple citations at the end of a sentence.
AR: The authors agreed and kept only most relevant references in the revised manuscript.

RC: **Title**
The title does not fit the manuscript. There was never doubt that multiple sources contribute INP at different temperatures. Something along the line of "Concentration of ice nucleating particles measured at Ny-Ålesund during 2018" would be more accurate.
AR: This is a good suggestion. The authors took the referee's word for it, and our title now reads, "Ice-nucleating particle concentration measurements from Ny-Ålesund during the Arctic Spring-Summer in 2018."

RC: **Abstract**
The abstract uses wordy, euphemistic language. In the interest of clarity, the tone should be revised to be strait forward. At the moment, the abstract consists of too many unsupported statements and vague conclusions that are incomprehensive before reading the manuscript in detail.
AR: The authors concur. By incorporating with all review comments, the authors offer concise and straightforward abstract by revising almost entire abstract. Please see the Track Changes version of the manuscript.

RC: Line 15: INP sources, INP concentrations and ice nucleation properties should be distinguished clearly. They are not the same.
AR: The authors agree. We now carefully distinguished INP source, aerosol particle source, INP concentration (*n*INP) and INP properties throughout the manuscript.

RC: Line 15: INP sources are unknown not "inadequately understood"
AR: The authors decided to remove this sentence and offer more detailed discussion in the main manuscript Sect. 4.

RC: Line 18: specify what properties were characterized
AR: We meant INP concentrations (*n*INP), and the text is updated accordingly.

RC: Line 22: why are the temperatures (-15°C, -18°C, -22°C) probed with the DFPC not evenly spaced?
AR: The temperature (*T* hereafter) boundaries of DFPC are determined (limited) by the device operational capability (Rinaldi et al., 2017; Rinaldi et al., 2019). The upper and lower boundary *Ts* of -15 and -22°C are the highest and lowest *Ts* we can be confident for. The *T* point of -18°C was chosen as an arbitrary intermediate step between the two boundary temperatures. We find no scientific reason to consider evenly spaced ice activation temperatures. We rephrased this *T* range to "temperatures (*T*s) of -15 to -22 °C".

RC: Line 24-25: The dependence on ice nucleation mode is not investigated in the manuscript and a sampling issues is a more reasonable cause for the discrepancy.
AR: We respectfully disagree. Perhaps, our inadequate explanation of sampling method has misread the referee. The authors clarified that the flow of 150 lpm represents a total flow through a common TSP inlet, but a diverged flow to our polycarbonate filter sampler was only on average 5.4 lpm in Sect. 2.1. We will address potential sampling issues below in this document.

RC: Line 26: specify how the increase in coarse INP was observed.
AR: The text was modified as follows: "DFPC measured *n*INPs for a set of filters collected through two size-segregated inlets: one for transmitting particulate matter less than 1 µm (PM_1) and another for that of less than 10 µm aerodynamic diameter (PM_{10}). Overall, $nINP_{PM10}$ measured by DFPC ranged from 3 to 185 m^{-3} at temperatures (*T*s) of -15 to -22°C. On average, the super-micrometer INP ($nINP_{PM10}$ - $nINP_{PM1}$) accounted for

approximately 20-30% of nINP$_{PM10}$ in spring and increased markedly in summer. In particular, it contributed 45% at T of -22°C and 65% at T of -15°C). This increase trend of super-micron INP fraction towards summer suggests an important role of super-micrometer aerosol particles as the source of Arctic INPs".

RC: Line 27: Explain why increase in coarse INP fraction suggests local sources.
AR: The authors decided to exclude this part from our abstract and provide detailed discussion in Sects. 3.2 and 3.6 (please see the Track Changes version paper).

RC: Line 28: Speculative. INP active at -15°C are not exclusively biological particles. The source of the particles active at -15°C in this study are unknown.
AR: The referee is right. We decided to exclude this sentence and to avoid similar sentences throughout the manuscript.

RC: Line 30: specify "distinct behaviours of particles"
AR: We admit that this part sounds confusing. Upon reformulation of the manuscript, this part became unnecessary. We have deleted this part from abstract.

RC: Line 31: How was the inter-annual variability of local INP sources previously considered? Specify the evidence for an inter-annual variability based on the current dataset and all available data from Spitzbergen. Consider that the 2012 data set is only 12 measurements and not a strong dataset to compare to.
AR: The authors realize that these information fit better in the main manuscript, and we address it in Sects. 3.4 and 4 (please see the Track Changes version paper).

RC: Line 33: It seems trivial that the INP population can be contributed by terrestrial and marine sources on an island.
AR: The authors agree. We reduced the discussion of it in our abstract.

RC: Line 35: specify nucleation ability. Higher particle load can explain higher INP concentrations without increased ice nucleation activity.
AR: Ice nucleation efficiency would be more appropriate word to be used. We decided to discuss this in detail in Sect. 3.3 and 3.4, and thus removed this part from our abstract.

RC: Line 37-40: requires reading the paper to understand this description.
AR: We agree. We now introduced only general remarks, for which the reader does not need to refer to our main manuscript: "Our spatiotemporal analyses of satellite retrieved Chlorophyll-a as well as spatial source attribution indicates the maritime INPs are expected at GVB from the seawaters surrounding the Svalbard archipelago and/or close to Greenland and Iceland".

RC: **Introduction**
The introduction is not tailored enough to the subject of this study. To make the introduction more effective at explaining and putting into perspective what follows, I suggest to focus on: ice nucleation mechanisms (condensation and immersion mode), INP-cloud interaction in the Arctic without going into detail on radiative effects, previous INP measurements in the Arctic and what has been learned about potential sources, tends, dependencies of nINP in the Arctic. A lot of literature is currently discussed in the Result section. Better to include a concise discussion of relevant literature to the introduction to develop the hypotheses which are then addressed in this study. Currently literature review is used inefficiently in the Result section to point to similar conclusions found elsewhere in literature.

AR: Ice nucleation mechanisms are now discussed in Lines 54-61. The Arctic INP-cloud interaction and its importance is briefly introduced by citing Murray et al. (2021). Previous studies of nINP are now summarized in a tabular form (Table 1). Most articles used in the result section are now merged in the Introduction section as suggested. We clarified our study motivation at the end of the Introduction Section (Lines 99-108): "In the present study, we contribute to fill the present gap of INP observations in the Arctic

environment, investigating *n*INP and potential sources at the ground level site of GVB (Svalbard), through spring and summer time measurements, by two INP quantification techniques, representing immersion and condensation freezing. We hypothesized that the *n*INP variability at a single *T* can be explained by differences in freezing modes. 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). The key to verify this in the Atlantic sector of the Arctic depends on a multitude of ambient INP measurements with a combination of trustful INP measuring systems at wide heterogeneous ice-nucleating conditions. Finally, we investigate the ice nucleation efficiency of Arctic aerosol particles represented by the activated fraction (AF), which provides further insight into the seasonal trend of ice nucleation efficiency besides concentration data".

RC: Line 46: provide reference and explain how aerosol affect cloud properties
AR: Murray et al., 2021 has been added to concisely direct the reader to the negative cloud-phase feedback in the Arctic.

RC: Line49-57: specify how the numerous local processes and feedbacks interact to affect structure, phase and persistence of clouds in the Arctic. Beyond what is generally true for INP-cloud interactions, explain why Arctic clouds are sensitive to INP concentrations. Provide a reference for the uncertainty associated with nINP, eg. DeBoer et al., 2018.
AR: This paragraph is now completely revised with only relevant references. Please see the Track Changes version of the manuscript.

RC: Line 59: the references are not "recently"
AR: The authors agree. We rephrased this sentence to: "Sea water has been identified to be a source of ice active organic matters (Knopf et al., 2011; Wang et al., 2015; Wilson et al., 2015), which are transferable to the atmosphere within sea spray particles (e.g., McCluskey et al., 2017)."

RC: Line 61-62: specify "most" ice nucleation processes. It would be good to introduce ice nucleation mechanism in more detail. Line 63-64: It is not generally true that biogenic INP nucleate ice at temperatures above -15°C. It is a bit of a stretch from ice nucleation properties to rain.
AR: With additional references, we have rephrased this sentence to: "Mineral particles are dominant immersion and condensation mode INPs typically below -20°C according to Fig. 13 in Hoose and Mohler (2012), with an exception of K-feldspar, which facilitates ice nucleation at much higher *T*s when compared to other mineral compositions (Atkinson et al., 2013). Further, biogenic INPs tend to support formation of ice at *T*s relatively higher than abiotic INPs (Murray et al., 2012), even though there is a considerable variation in ice nucleation efficiency within biotic INPs (Kanji et al., 2017)", to clarify our points. In addition, ice nucleation mechanisms are now introduced in the third paragraph of the Introduction section.

RC: Provide a more coherent explanation and provide the link to INP in the Arctic region.
AR: This is a good suggestion. We now compiled 14 previous Arctic *n*INP studies in Table 1 and associated text (Lines 70-98).

RC: Line 67-86: Specify that the literature review is separated into condensation and immersion mode measurements as well as separated into short and year around observations.
AR: This is also a good suggestion. Please see our new Table 1. We have enlisted previous literature according to condensation vs. immersion.

RC: Line 67: specify "short periods of time"
AR: All study time periods are now listed in Table 1. As seen, it is typically in the order of several months. As these time spans vary from study to study, we decided not to use the word of short periods of time in text. Thanks for catching this.

RC: Line 72: add some information how Hartmann et al. confirmed this.
AR: The authors decided to conduct detailed discussion of Hartmann et al. in Sect. 4 (Discussion), Lines 580-581.

RC: Line 74: How did Bigg et al. identify the Ocean was the main source? There was a third Arctic cruise in 2001 and a more recent expedition in 2017. They are reported in Welti et al., 2020.
AR: The authors added the requested details (Lines 76-77). Welti et al., 2020 is now included in the new Table *1*.

RC: Line 76-86: Mention at what temperatures Mason et al., 2016, Si et al., 2018, Creamean et al., 2018, Irish et al., 2019 reported data. This section seems to contradict line 61-64 where it is argued that INP active at T>-15°C are biological and not mineral dust.
AR: The requested information was added in the new Table 1. We have revised any unappropriated statement regarding biological particles.

RC: Line 88: Quantify the increase. Contrast to the fact that often ambient *n*INP measurements scatter within 1-2 order of magnitude within less than a day, highlighting that caution should be used when interpreting variations smaller than one order of magnitude in such dataset.
AR: The authors thank the referee for sharing thoughts, and we agree. An order magnitude discrepancy is not by all means an acceptable margin or any sort of magic numbers. We have carefully removed all of the one order of magnitude discussions from the manuscript.

RC: Line 90: A time-series showing measured nINP at -15°C as function of DOY from all the listed measurements would be a helpful addition to illustrate the discussion and to show if the Arctic region as a whole experiences seasonal variations or if these are local phenomena.
AR: We thank the reviewer, but we believe that we made sufficient discussion in the revised manuscript in comparison to Wex et al. (2019). Specifically, the authors have provided a comparison with the seasonal evolution of *n*INP presented by Wex et al. (2019) at *Ts* of -15 and -18°C in the SI (Fig. S3).

RC: Line 94: As your literature review exemplifies, there is no general "gap" of INP measurements in the Arctic.
AR: We agree and disagree. It is deemed to be inconclusive yet at least. In any case, our *n*INP data for multi-seasons along with a rich set of baseline data from the GVB station (including but not limited to the dataset presented in this work) would be crucial for future verification of more rigorous modeling closure study to examine temporal trends of the Arctic *n*INP. As presented in our work, we find both agreement and disagreement between our *n*INPs from this study and *n*INPs measured in previous studies. Filling that gap completely is beyond the scope of the current work. However, it is an imperative future task, and we believe that our findings of condensation vs. immersion in the Arctic *n*INPs as well as non-substantial seasonal variability in *n*INP are invaluable to report.

RC: **Methods**
The method of how INP concentrations are determined with the WT-CRAFT starting from the air volume sampled through filter to counting the number of frozen aliquots, should be explained in more detail, focusing on how nINP can be derived step by step from sample volume, water volume, droplet volume. How the preparation is done practically is of secondary interest.
AR: This is a valid question. The authors initially intended to include some of these details (omitted concerning the manuscript length). All details regarding WT-CRAFT are now incorporated in our revised manuscript. The authors should have clarified that only a subset of 150 lpm from the common inlet was directed towards our WT-CRAFT filter sampler. We hope these array the referee's misgivings.
Lines 127-137: "Aerosol particles were collected using 47 mm membrane filters (Whatman, Track-Etched Membranes, 0.2 µm pore). Briefly, aerosol particle-laden air was drawn from a central total suspended particulate (TSP) inlet with a constant average inlet flow of 5.4 lpm (± 0.2 lpm standard deviation). We note

that the TSP inlet is custom made, and is designed to operate with isokinetic and laminar flow at 150 lpm. From the central inlet, an 8 mm outside diameter stainless steel tube was directly connected to the filter sampler to intake a subset of air flow. More detailed conditions of our filter sampling, including sampling time stamps, air volume sampled through filter cross section, and the resulting HPLC water volume used to suspend aerosol particles for WT-CRAFT analysis, are summarized in Table S1. Below the filter sampler, the filtered-air was constantly pumped through a diaphragm pump (KnF, IP20-T). A critical orifice was installed upstream of the pump to ensure a constant volume flow rate and control the mass flow rate through the sampling line. A typical sampling interval was approximately 4 days with only one exception (i.e., 8 days for the sample collected starting on 26 May 2018)".

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	Total Flow (optimized for 50% of filter)	Suspension water volume (First frozen drop = 0.001 INP L^{-1})
	DAT_UTC	DAT_UTC	LPM	L	mL
NYA_GVB_01	4/16/2018 17:00	4/20/2018 10:00	5.1	13617.0	2.8
NYA_GVB_02	4/20/2018 14:40	4/24/2018 14:40	5.1	14601.6	3.1
NYA_GVB_03	4/24/2018 18:20	4/28/2018 16:00	5.5	15314.5	3.1
NYA_GVB_04	4/29/2018 13:30	5/2/2018 16:15	5.6	12445.9	2.4
NYA_GVB_05	5/2/2018 16:20	5/6/2018 14:37	5.5	15471.9	3.3
NYA_GVB_06	5/6/2018 14:45	5/10/2018 13:00	5.4	15325.1	3.3
NYA_GVB_07	5/10/2018 13:10	5/14/2018 11:05	5.6	15890.7	3.3
NYA_GVB_08	5/14/2018 11:15	5/18/2018 7:50	5.5	15179.0	3.1
NYA_GVB_09	5/18/2018 8:00	5/22/2018 8:28	5.5	15917.0	3.3
NYA_GVB_10	5/22/2018 8:30	5/26/2018 11:33	4.8	14263.2	3.0
NYA_GVB_21	5/26/2018 11:45	6/3/2018 18:30	5.5	32883.2	6.9
NYA_GVB_22	6/3/2018 18:35	6/7/2018 17:20	5.5	15576.9	3.3
NYA_GVB_23	6/7/2018 17:24	6/11/2018 17:35	5.4	15668.3	3.3
NYA_GVB_24	6/11/2018 17:40	6/15/2018 16:24	5.4	15218.9	3.2
NYA_GVB_25	6/15/2018 16:28	6/19/2018 19:05	5.4	15887.1	3.3
NYA_GVB_26	6/19/2018 19:09	6/23/2018 19:16	5.3	15152.8	3.2
NYA_GVB_27	6/23/2018 19:20	6/27/2018 13:55	5.3	14525.0	3.0
NYA_GVB_28	6/27/2018 14:00	7/1/2018 16:40	5.4	16013.6	3.3
NYA_GVB_16	7/1/2018 16:50	7/5/2018 17:20	5.5	15792.2	3.3
NYA_GVB_17	7/5/2018 17:25	7/9/2018 17:22	5.4	15587.1	3.3
NYA_GVB_18	7/9/2018 17:27	7/13/2018 18:24	5.4	15662.3	3.3
NYA_GVB_19	7/13/2018 18:33	7/17/2018 16:43	5.3	15071.4	3.2
NYA_GVB_20	7/17/2018 16:52	7/21/2018 15:55	5.3	15241.3	3.2
NYA_GVB_11	7/21/2018 16:02	7/25/2018 16:31	5.4	15602.3	3.3
NYA_GVB_12	7/25/2018 16:38	7/29/2018 15:07	5.4	15391.3	3.2
NYA_GVB_13	7/29/2018 15:14	8/2/2018 18:39	5.5	16254.6	3.4
NYA_GVB_14	8/7/2018 15:55	8/11/2018 14:05	5.4	15382.1	3.2
NYA_GVB_15	8/11/2018 14:12	8/15/2018 17:36	5.4	16177.4	3.4

RC: In section 2.3 it should be clarified (by a short explanation at the beginning of each subsection) for what purpose the measurement or analysis is performed or used in the context of this paper. Here it would be helpful to already know from the introduction what the aim of the analysis is or what hypotheses are going to be tested with these data. Give context in the introduction section: why are you investigating ground type, trajectories, chlorophyll,…

AR: We thank the reviewer for the suggestion. We revised the manuscript accordingly. For instance, Sect. 2.3.4 now starts with "In order to investigate the sources that contributed to INPs (i.e., maritime vs. terrestrial), we performed the 5-day back trajectory analysis,…". Sect. 2.3.5 now starts with "Satellite retrieved chlorophyll-a fields were used to track the evolution of oceanic biological activity in the Arctic ocean during the study period".

RC: Line 99: Refer to the location as Gruvebadet station throughout the paper and introduce the abbreviation (GVB) here.

AR: Introduced. Thanks.

RC: Line 100: Point to fig. S1 showing the location on a map.
AR: This is a valid suggestion. A new Fig. 1 is produced, and the GVB location is now shown.

RC: Line 102: Can Longyearbyen in the SE of the GVB station contribute to the aerosol population?
AR: The distance of the sampling site to Longyearbyen, the main settlement of Svalbard with about 2000 people living and working there, is more than 100 km. Dekhtyareva et al (2016) investigated the potential impact of the activities taking place there on the measurements in Ny-Ålesund. Using 3D backward trajectories, they found that during spring NOx values were higher for long range transport cases (defined as those in which the airmasses come from latitudes below 70°N). During summer, mainly due to meteorological patterns, they concluded that it is unlikely that the pollution generated in Longyearbyen reaches Ny-Ålesund. Based on these remarks and considering the distance and the small dimension of the settlement, we would not expect that substantial contributions of this point source in our measurements at GVB.

RC: Line 110: Is filter overloading an issue in the clean Arctic air? The WT-CRAFT filters were sampled longer, with higher flow and on filter with smaller pore size. Where these filters potentially overloaded?
AR: Filter overloading is an issue only for the DFPC technique. DFPC analyzes aerosol particles collected on filters directly; therefore, it was necessary to avoid coalescence of ice crystals while processing condensation freezing experiments. For these reasons, the upper limit of sampled volume through the DFPC filter cross section was optimized to comply with the INP quantification range of ca. 50-1500 INPs per filter.
This overloading concern is not an issue for immersion freezing measurements. If necessary, dilutions of stock suspensions (i.e., aerosol particles suspended in HPLC water) can be assessed. Furthermore, we monitored the flow passing through the cross section of the WT-CRAFT filter while sampling. Between the beginning and the end of each sampling, the flow deviation was <5% for individual samples. With typically <100 p/ccm particle load and ~5.4 lpm of sampling flow rate (See revised texts in Sect. 2.1), we do not expect any particle overloading conditions.

RC: Line 115: Pumping 150lpm through filter with 0.2um pore size creates a huge pressure drop. Can you comment on how sampling was possible without fracturing the filter? How was the flow monitored? In this setup the filter probably acted as flow limiter rather than the critical orifice before the pump. An overestimation of the sample flow would explain the offset between WT-CRAFT and DFPC.
AR: For clarity, 150 LPM is the flow rate of the central laminar flow sampling inlet at GVB. From the central inlet, only a small amount of flow (~5.4 lpm) was bypassed to the WT-CRAFT filter sampler. The text has been substantially modified to clarify what was truly done at GVB in Sect. 2.1. Please see our Track-Changed manuscript.

RC: Line 116: specify pump model
AR: Specified.

RC: Line 118: Is filter overloading (line 110) an issue for 4-day samples? The volume sampled is more than 100-times larger for WT-CRAFT than for the DFPC filters.
AR: Clarified above, and all total sampled air volume for each WT-CRAFT sample is provided in Table S1.

RC: Line 127: Specify how uncertainties in T and Sw convert into uncertainties in nINP.
AR: We considered temperature uncertainties of 0.2°C and 0.1°C for air and filter, respectively. These uncertainties determines an uncertainty of 0.02 on the calculated S_w. We evaluated the effect of such variation in S_w on the final number of counted INPs by extrapolating the results of *n*INP as a function of S_w, obtained by Belosi et al. (2018) for different aerosol particles.
The text was modified accordingly (Lines 151-153).

RC: Line 128f: Has a systematic difference between condensation and immersion mode ice nucleation been observed in these inter-comparisons?

AR: This is a valid question. The authors now clarified the raised point in our revised Sect. 4. Please, refer to sub-Sect. 4.1 for details.

RC: Line 132: The large sample volume of over 800m3 would allow to detect approximately 100-times lower INP concentrations than 1 m-3. Why was the analysis not performed in the full range?
AR: As clarified above, the sampled air volume is much smaller than the said number (see Table S1).
The WT-CRAFT measurement was not conducted below -25°C because we observed that non-negligible amount of field blank and HPLC-grade pure water droplets (>3% of 70 droplets) could freeze at below -25 °C for this study. WT-CRAFT was operated inside of the ventilated fume hood with air flow filtered by HEPA. This effort is to follow the setup of the original NIPR-CRAFT, which is used in a clean booth. Regardless of similar experimental procedures used in both CRAFT systems, this limitation of measureable temperature > -25 °C persisted for WT-CRAFT. While the reason of this limitation is unknown, more insightful description of WT-CRAFT and its capabilities are now available in Vepuri et al. (2021).

RC: Line 133: State how the two CRAFT systems are different. All I could find in Hiranuma et al., 2019 was that they used different sizes of droplets. This is not an instrumental difference.
AR: This is a valid question. Cont'd on our previous response;
Camera: We employ a combination of an Opti-Tekscope OT-M HDMI microscope camera and a Logitech c270 camera to correctly capture the transition of droplet brightness/contrast to opaque ice with 30 fps time resolution with a reasonable pixel resolution as well as magnification (if needed).
Droplet holding plate: We use a thin (<5 mm) polished aluminum plate to warrant an efficient thermal cooling and to make sure the Cryo-cooler system temperature is equivalent to the temperature measured at the surface of the plate within known uncertainties.
Isolation to the lab air: WT-CRAFT is operated in a vertical clean bench (LABCONCO, Purifier®). All droplet preparations (70 x 3µL) were conducted in the clean bench to minimize the chance of contamination from the lab air.
We have clarified these in our revised Sect. 2.2.2. Please see the track change manuscript.

RC: Line 135: Explain how the uncertainty in ice nucleation efficiency is derived.
AR: The uncertainties of temperature, ± 0.5 °C, stems from a sensor manufacturer reported uncertainty (TGK, SN-170N) The uncertainty in ice nucleation efficiency in WT-CRAFT are and ±23.5% according to Hiranuma et al. (2019, i.e., Table S2). Note that our ice nucleation uncertainty was estimated based on the average standard deviation across the examined temperature (T > -25 °C) for known composition (microcrystalline cellulose), which reasonably matches with 95% confidence intervals of individual measurements (i.e., Eqn. 3.21 of Schiebel, 2017).

RC: Line 136: repetition, delete
AR: Deleted. Thanks for catching this.

RC: Line 139: provide camera model specifics
AR: Provided and specified.

RC: Line 140: define how INP concentrations are derived.
AR: Defined in **Section 2.2.3** as follows:
For the WT-CRAFT analysis, we first computed the $C_{INP}(T)$ value, which is the nucleus concentration in HPLC suspension (L^{-1} water) at a given T as described in Vali (1971). This $C_{INP}(T)$ value was calculated as a function of unfrozen fraction, $f_{unfrozen}(T)$ (i.e., the ratio of number of droplets unfrozen to the total number of droplets) as:

$$C_{INP}(T) = -\frac{\ln(f_{\text{unfrozen}}(T))}{V_d} \quad (1)$$

in which, V_d is the volume of individual droplets (3 μL). Next, we converted $C_{INP}(T)$ to nINP(T). The

cumulative *n*INP per unit volume of sample air, described in DeMott et al. (2017), was estimated as:

$$nINP(T) = C_{INP}(T) \times DF \times \frac{V_l}{V_{air}} \quad (2)$$

where DF is a serial dilution factor (e.g., DF = 1 or 10 or 100 and so on). The sampled air volume (V_{air}) and the suspension volume (V_l) are now provided in Table S1.

RC: Line 141f: specify water volumes, for 1 INP per m3 that would be 90mL for 4 day samples and 180mL for 8 day samples.
AR: Now given in **Table S1**.

RC: Line 143f: specify water volume used for soaking
AR: Now given in **Table S1**.

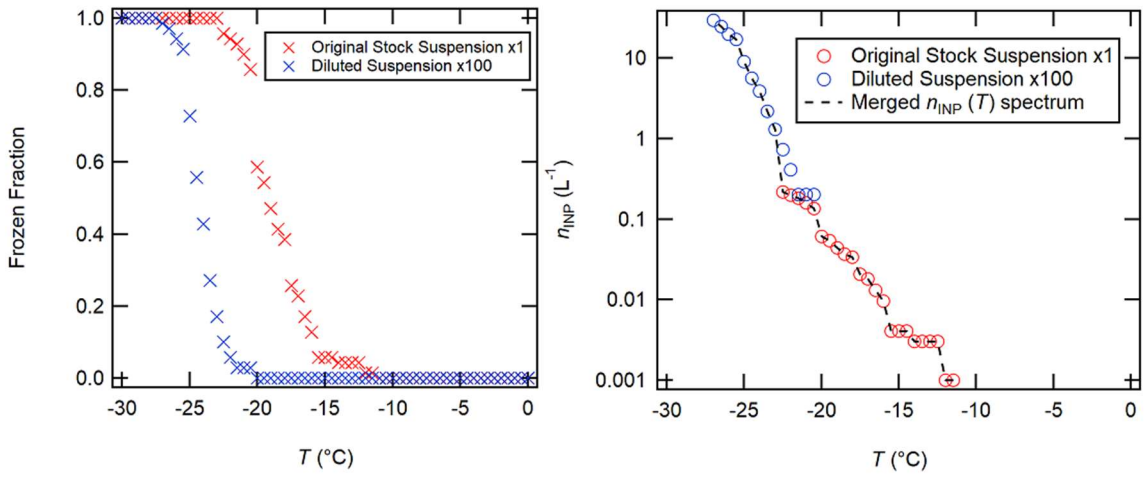
RC: Line 144: specify how mechanical vibration was applied. By sonication?
AR: We soaked each polycarbonate filter in a sterilized falcon tube with HPLC water on the VWR vortex mixer. This point is now clarified in the main text. No sonication was applied not to damage polycarbonate filter as well as particles suspended in water.

RC: Line 145: How were droplets prepared?
AR: Manual pipetting in a clean bench.

RC: Line 146: The method by how much the sample was diluted is not explained clearly. Specify the dilution water volume and how dilution was considered for the derivation of INP concentrations.
AR: Simple serial dilution (x10 and/or x100) as described in Vepuri et al. (2021) was applied. The derivation of INP concentrations including dilution factor is now explained in Sect. 2.2.3.

RC: Line 149: How was the stitching performed? At what temperature were the spectra stitched?
AR: The IN measurements from the undiluted and diluted runs were merged by taking the lower n_{INP} values, which exhibit smaller CL95% error, for the overlapped *T* region (Vepuri et al., 2021). This procedure was employed not to have erroneous jumps – we also attempted to take median or max numbers for overlapping regions, but we found that the proposed procedure gives the least stair case like spectrum. Stitching spectra does not depend on the temperature. Instead, we merge our spectra of an original stock and diluted suspension in the way we make sure the following three criteria are met:
(1). Gap is within a factor of few
(2). Two spectra match within CI95%, within T uncertainty or a combination of both
(3). Two spectra match within 23.5%, within T uncertainty or a combination of both
We now cite Vepuri et al. (2021) in our manuscript.

RC: In Fig.1 a jump in max concentration appears at -23°C. Is this the range of the diluted measurements? To show the "absence of failure" of this technique, it would be helpful to show the individual measurements in Fig.1 and not just the range.
AR: With the procedure explained above for merging spectra, we merged all spectra in a consistent manner. As per request of the review, we show individual spectra for the reviewer mentioned. The authors respectfully wish that the reviewer finds our procedure is reasonable.



RC: Line 153f: Derivation of nINP must be defined clearer. Dividing the number of INP by the total sample volume is incorrect. The concentration is calculated from the filtered air volume, dilution water volume, droplet volume and number.
AR: We clarified this point now in Sect. 2.2.3.

RC: Line 154: As you state on line 379, only a small, T-dependent fraction of ambient aerosol are INP. Therefore, dividing the number of INP by the bulk particle surface has no physical meaning for a heterogeneous aerosol population. I recommend changing this approach to deriving only the fraction of particles that are ice active, by dividing the INP concentration by the particle concentration.
AR: We thank the reviewer for the suggestion. We have decided to follow the reviewer suggestion, limiting to present and discuss activated fraction (AF) data - see our revised Sect. 3.3 and 3.4. Our conclusions did not change.
In the future, long-term *n*INP monitoring by an online instrument (e.g., Möhler et al., 2021) may allow the authors to further evaluate ice nucleation efficiency of the Arctic aerosol particles.

RC: Line 161: specify APS measurement range
AR: The APS measurements range is from 0.5 to 20 micrometers. We modified the text as follows:
"An Aerodynamic Particle Sizer (APS) model TSI 3321 for the diameters between 0.5 and 20 micrometers."

RC: Line 163: what are the references pointing at? How were the size distributions averaged for the sampling interval of the filters?
AR: The citations refer to papers reporting more details on the cited instruments. The average aerosol number concentration, for each INP sample, was obtained by averaging all the aerosol number concentration data points falling within the filter sampling interval. The text was modified adding this detail (Lines 215-219):
"The number concentration in the resulting overlapping range was taken from the SMPS data as SMPS provides more size bins. At the end, commutative aerosol particle counts of SMPS and APS were considered as a total aerosol particle number concertation. 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".

RC: Line 164: What substance is assumed for a density of 1.95 gcm-3? Mineral dust and sea salt have higher densities.
AR: We considered a mixture of different substances, including lighter compounds like methanesulfonic acid and nss-Sulphates, besides sea-salt and dust. Based on the findings from Lisok et al 2016 on the chemical characterization of the aerosol at the same site, we estimated the value of 1.95 gcm^{-3}. We changed the sentence as follows;
"The aerodynamic diameters measured by the APS were corrected to the volume equivalent diameters using an average particle mass density equal to 1.95 g cm-3, assuming a mixture of different substances based on

the findings from Lisok et al. (2016) and a dynamic shape factor of 1. The number concentration in the resulting overlapping range was taken equal to that from the SMPS".

RC: Line 173: Where was this analysis performed? While handling during the analysis is relevant, handling before and after sampling, storage and transport are equally important and could be described.
AR: The authors clarified these in Lines 228-230: "The filters were handled with care (working under a class 100 laminar flow hood by personnel wearing powder free latex gloves to minimize potential contamination) throughout the sampling and offline analysis at the University of Florence. After sampling the filters were stored and shipped at -20°C".

RC: Line 181-196 (Section 2.3.4.): Very similar to the text in Wex et al., 2019. Sentences in line 191-195 are copied from Sec. 2.7 in Wex et al., 2019. It is difficult to understand without consulting the original description. Section 2.3.4. should be rewritten entirely, explaining more clearly how ground types were categorized and how trajectories were merged to the filter sampling intervals. I suggest (instead of the analysis in 2.3.5.) to include high chlorophyll concentration as a fifth ground type in this analysis. Additionally, precipitation along the trajectories should be considered.
AR: The authors apologize for extending the assessment to comment towards raised questions. The relevant text has been substantially updated in Sect. 2.3.4. Please refer to the track change manuscript.
While the introduction of the high CHL class may be valid and one way for analysis, we took an alternative approach following previously published articles as these have been well-established (Rinaldi et al., 2013; O'Dowd et al., 2015; Mansour et al., 2020a; Mansour et al., 2020b). The approach proposed by the reviewer would presume that we can define a CHL threshold associated to emission of INPs. We do not have such a knowledge of the biological processes leading to production of marine INPs. On the contrary, working on the correlation evidences eventually present relations between phytoplankton activity and INP concentration, without arbitrary assumptions. One caveat is that we unfortunately cannot incorporate with the occurrence of precipitations along the considered BTs in our model.

RC: Line 201: state the temporal resolution of the dataset.
AR: Stated as: "The Level-4 product is available globally at ~4 km spatial resolution and daily time resolution."

RC: Line 202: Shift the description of how INP concentrations and chlorophyll maps were merged from the Result section to here. The DFPC summer data consists of only 17 measurement days and 3 are excluded because land influence, leaving 14 data points. Demonstrate that correlations are robust by showing some scatterplots of grid cells with a strong correlation as a supplement.
AR: We thank the reviewer for the suggestion, but we believe that this explanation fits in this particular part and increases a clarity of logical flow.
It is impossible to check visually all the regressions that form the correlation maps discussed in the manuscript as each map is composed of 651,508 pixels, of which between 30,724 (~5%) and 85,829 (~13%) present a positive and significant correlation, according to the considered delay time, from 0 to 26 day. To meet the reviewer's request, which is legitimate, we focused on three evidenced sea regions characterized by systematic high correlation between INP and CHL (Figure 7) and we divided, within each region, the significant and positively correlating pixels into three categories: High, Medium, and Low correlating, according to the distribution of the correlation coefficient. Then we selected randomly 6 pixels within each category, per each region, of which we plotted the results of the INP vs CHL regression analysis, for a total of 54 scatter plots. Careful investigation of the randomly selected scatter plots show a variety of conditions regarding the robustness of the investigated correlation, with generally robust correlations, in the majority of the cases not distorted (or influenced) by one single (or a few) points, which we consider a prove of the robustness of the obtained correlation maps.
The scatterplots have been added as an Appendix to this AR document and to the Supporting Material.

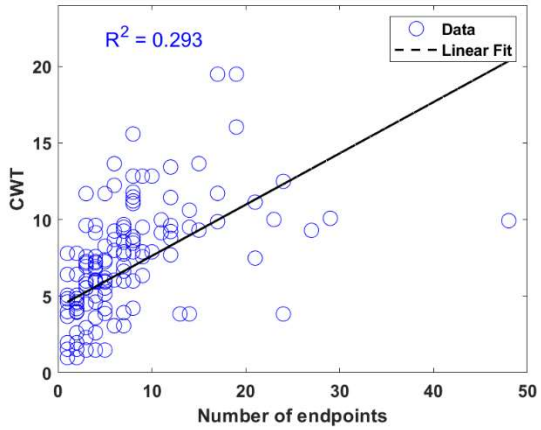
RC: Line 203: Explain why a relationship between INP and chlorophyll concentration is expected.
AR: We have added this lines as explanation: "Recent literature (Wilson et al., 2015; Knopf et al., 2011; Wang et al., 2015) has showed that sea-spray organics can nucleate ice being potentially important INPs in the clean

marine atmosphere. Mansour et al. (2020b) evidenced that *n*INP over the North Atlantic Ocean follows the patterns of marine biological activity as traced by surface CHL concentration".

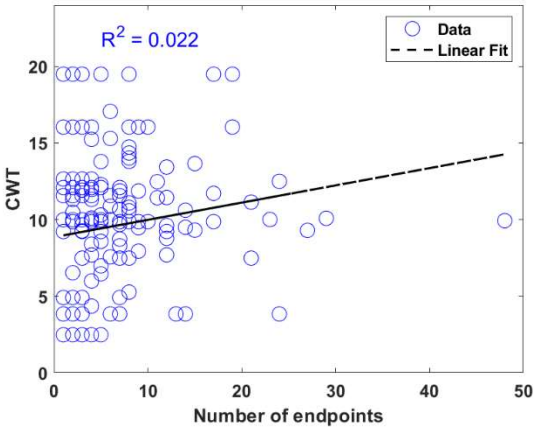
RC: Line 203: Excluding the samples with land input is mentioned several times. Elaborate why this is important.
AR: We are trying to investigate the relation between INPs and marine biological activity: it is reasonable to exclude from the dataset the samples for which we have a clear evidence of a terrestrial influence. This has been made clearer in the text "to focus only on INPs potentially originated from the sea".

RC: Line 212, 213: specify, concentration of INP
AR: This is the general description of a general chemometric approach; it is valid for INPs or for any other atmospheric concentration. We explained in the following lines that we applied it to INP concentrations.

RC: Line 217: Specify how many trajectories were used and demonstrate that this is a large enough sample to draw conclusions. Looking at the figures it seems that higher CWT is found where more trajectories passed.
AR: We thank the reviewer for evidencing the limit of the proposed CWT solution: indeed, we have verified that a significant correlation can be observed between the CWT results presented in the original Figure 7c and the number of BT endpoints in each cell. This is shown in the plot below (left).



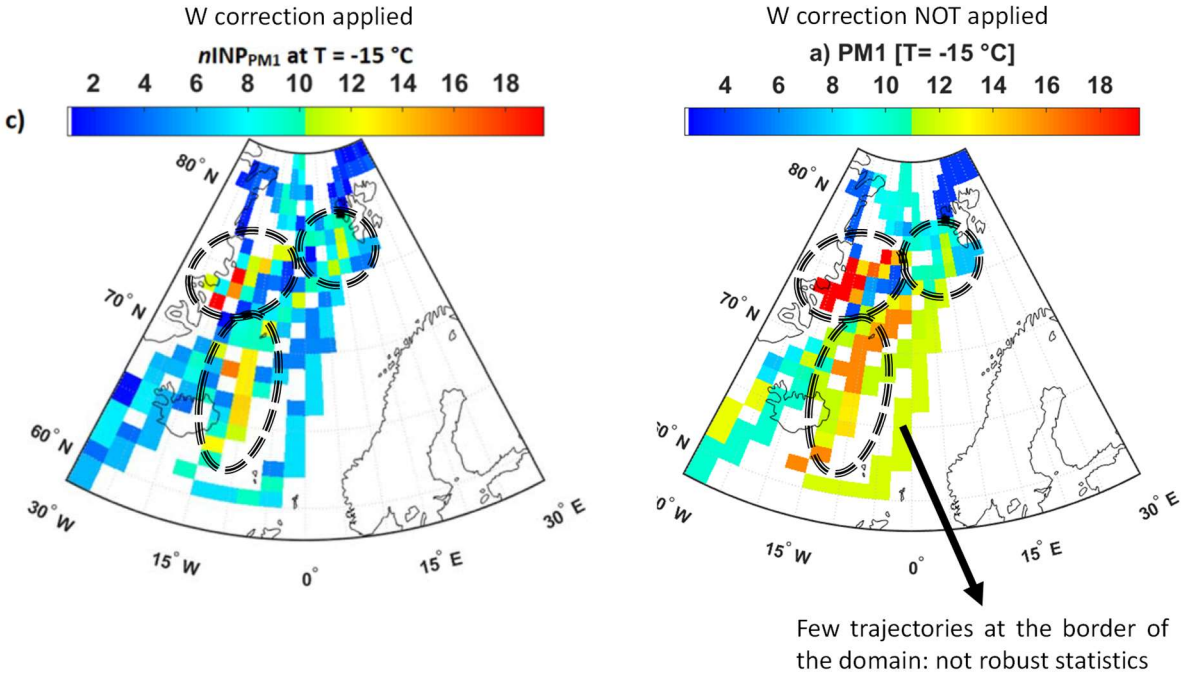
Weighted (Eq. 2)



Not weighted

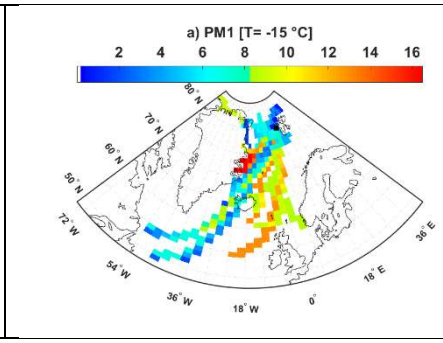
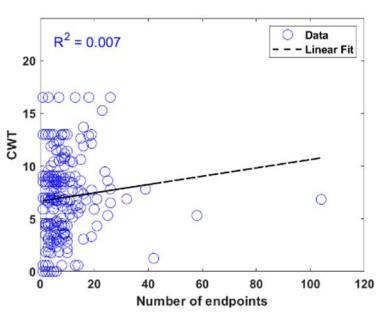
This effect, which should not be present in the outcome of the CWT analysis and requires a correction, is not related to the number of back-trajectories or of samples, rather to the weighting approach described in Equation 2. Indeed, the above (right) plot shows that this correlation disappears if the weighting step is excluded from the CWT analysis.
Weighting based on the number of passages over one cell has the aim of avoiding that cells with a low number of passing back trajectories (typically cells that are at the borders of the domain) are considered of the same importance as cells characterized by many passages, for which the CWT value is statistically more robust. If one cell has only a back trajectory endpoint, its CWT will be determined only by one INP sample. On the contrary, a cell crossed by many trajectories will have a CWT which derives from the weighted averaging of many samples. Weighting the cells by the number of passing endpoints is common practice in applying the CWT method (Cheng et al. 2013; Hsu et al., 2003; Jeong et al. 2011), even though the weighting step may also be excluded (Bycenkiene et al., 2014).
In the original manuscript, we derived the Weighting correction scheme (Eq. 2) from Masiol et al. (2019a, b), selecting it among different examples found in literature, because the correction is based on intrinsic properties of the dataset (i.e., the distribution of the number of endpoints within the cells), which makes the choice of the W scheme less subjective. Nevertheless, we recognize that the weighting criteria are evidently too strong and are responsible of the effect evidenced by the reviewer (high CWT is associated to high number of passing trajectories). To check the effect of the W correction on the overall source location approach, we report below also the unweighted CWT results (i.e., the CWT map before applying the W

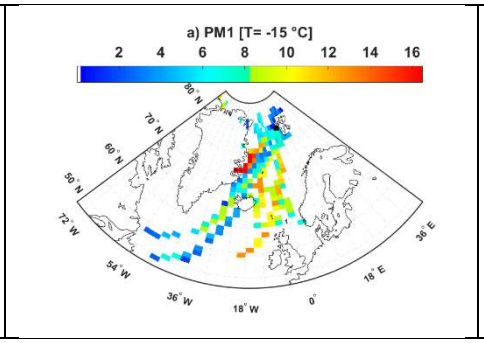
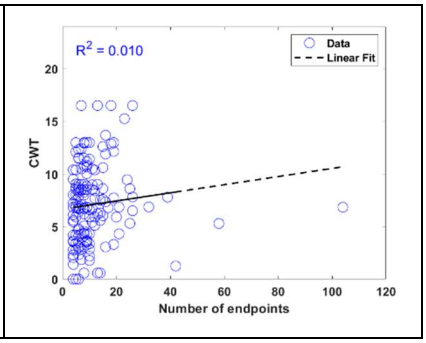
correction). The same main source regions result from both plots (indicated by black circles to guide the eye), while the main difference is observed in the most marginal south-east zone of the domain, where potentially high CWT values (in the uncorrected plot) are down weighted (in the left plot) because of the low number of endpoints determining it. In conclusion, the application of the W correction does not modify substantially the results evidencing the same source regions in both plots.



Considering the evident limits of the solution presented in the original manuscript, we elaborated a new solution for the revised manuscript. In this new solution, we adopted a softer Weighting correction for cells with a low number of endpoints and we increased the number of BTs considering two trajectories per day to be associated to the corresponding INP concentration of the day (total BTs = 28, total end points = 3388 of which 2184 endpoints passing at low altitudes (< 500m)). This doubles the number of BTs enhancing the statistics of the CWT solution. Obviously, we cannot increase the number of INP samples available for the analysis, but we note that Hsu et al. (2003) successfully applied CWT to datasets consisting of 22 and 30 samples, which is not far from the dimension of our dataset.

The following Table presents a comparison between the new UNWEIGHTED and WEIGHTED solutions. As it is clearly shown by the third column, no one of the solutions present a correlation between CWT and the number of endpoints in a cell (N); furthermore, they show a general agreement regarding the major identified sources, demonstrating that the weighting approach does not shape the CWT maps, but it only clears the solution by removing the less representative cells. Finally, the identified sources are generally the same as evidenced in the previous version of the CWT analysis (original manuscript), showing that the solutions are robust and independent on the number of BTs deployed.

No Weighting Total cells = 308		

Weighting: N >= median Weight=1 1st Quart < N < median Weight=0.8 N <= 3 Weight= 0 Total cells = 203		

Finally, we note that, in our work, CWT is used only qualitatively to evidence the location of the most probable source regions of INPs over the selected marine domain (to provide a comparison term for the results of the spatio-temporal correlation with CHL). For this purpose, also a limited dataset as the present one might be sufficient, as shown in the general consistency of the "W corrected" and "not corrected" plots above. On the contrary, to use the same approach quantitatively, for instance by comparing the relative strengths of the evidenced source regions, by comparing their CWT values, a more extended dataset would certainly be necessary.
The text has been modified reporting the new CWT solution (Lines 290-295 and 504-511).

RC: Line 227: Justify that longer residence time in a grid box is related to higher INP concentration. I would expect high windspeed to generate more particles, but also less endpoints at the location because the trajectory moves faster. Discuss assumptions made for this analysis.
AR: Longer residence time does not translate in higher CWT values as the parameter Dij appears both at the numerator and at denominator. For instance, if only one trajectory passes in a grid cell, the resulting CWT for that cell will be equal to the INP concentration measured at the sampling point at time of arrival of that single back trajectory. This will happen independently on the residence time. In fact, in this case the formula will be
CWT = C * Dij / Dij = C (in this case, C = *n*INP).
The residence time is only used to "weight" the relative contribution of each back-trajectory in determining the final CWT value of a cell. In other words, if a cell is crossed by multiple trajectories, the final CWT will be influenced more by the concentration (C) associated to the trajectories that stay over the cell for more time, following a classical "weighted averaging" approach.

RC: Line 227: What uncertainties are avoided by weighting? Motivate the application of a weighting factor. This methodology makes no sense to me and needs a clearer explanation.
AR: Explained above. Thanks for bringing this up – the authors admit that the sentence in the manuscript was not clear; in the new manuscript it was reformulated as follows:

"In order to reduce the impact of grid cells containing a low number of endpoints, for which the calculation of the CWT is statistically less robust, the CWT values were multiplied by a weighting factor (W_{ij}) according to Eq. (4).

$$W_{ij} = 1 \text{ (if } D_{ij} \geq median), W_{ij} = 0.8 \text{ (if } 3 < D_{ij} < median), \text{ and } W_{ij} = 0 \text{ (if } D_{ij} \leq 3) \quad (4)$$

The introducing of the weighing factor reduces the number of considered cells to 203"

RC: **Results**

Line 237-238, 244ff: Discuss different ice nucleation modes in the introduction section. Remove here.
AR: This is a good suggestion. We moved the ice nucleation mode discussion to the third paragraph of the introduction section.

RC: Line 240: specify "sharper"
AR: Corrected using "steeper".

RC: Line 241: Provide a more detailed explanation how "time resolution" and "sampling activities" can explain these differences. Calculate how much of the difference can be explained by the uncertainty introduced by the ice nucleation analysis and how much from uncertainties in sample volume.
AR: The uncertainties involved in WT-CRAFT immersion efficiency analysis and sampling flow rate are ±23.5% and ±3.7%. Those for DFPC are 30% and <10%. These 'systematic errors' would not be able to explain the difference we observed.
More in detail, in the manuscript we enlisted some parameters that may have contributed to the observed discrepancy. For instance, we are comparing samples with different time resolutions (4 days vs 4 hours). This could explain some discrepancy in the resulting INP concentrations by the two techniques, if a strong diurnal gradient was present in the INP concentration. Honestly, we believe that this alone could never explain the observed discrepancy, considering also the absence of a day-night cycle during the Arctic summer. We considered that some uncertainties in the sampling flow rates (see above) could also have contributed a small fraction of the discrepancy, but no flowmeter can be so off as to generate differences of 8 times. Finally, we highlight that the discrepancy is temperature dependent, which could not be justified by sampling volume uncertainties alone.
We have made this clearer in the revised version of the manuscript (please, refer to sub-Sect. 4.1).

RC: Line 242: unclear what the references point at
AR: These references have been removed.

RC: Line 242f: Elaborate based on what it is a valid assumption that the ice nucleation mode generates the observed difference of higher nINP from condensation than immersion mode.
AR: Discussed below.

RC: Line 247: Vali 1975 is a better reference for ice nucleation modes
AR: We used Pruppacher and Klett (2010) and Vali et al. (2015), as reference for the ice nucleation modes.

RC: Line 247-255: This section is speculative. Provide an explanation how the different mechanisms can exert an influence on nINP and why in particular on mixed particles. Much more probable would be an uncertainty in the sample volume.
AR: We echo that a sampling volume uncertainty can hardly explain the observed difference. Furthermore, if a sampling volume would be a source of the issue, it would impact the INP concentration across the assessed temperatures. However, the observed difference varies depending on temperature, which implies that any systematic errors might not be the cause of such a T dependent trend.
We have modified this part, that is now included in the Discussion Sect., as reported above.

RC: Line 257ff (Sec. 3.2): For a field study as this, aiming to learn something about the abundance and nature or source of INP, I would consider the differences in concentration of minor importance. Focus should be on the big picture, on trends while being cautious not to overinterpret the data.
AR: We agree with the reviewer that the inter-comparison between DFPC and WT-CRAFT is not the focus of the manuscript, and we limit to acknowledge its existence and to show that it does not affect any general feature observed by the two measurements (seasonal trend, relative time series). We also now carefully choose the word to discuss agreement with previous measurements.

RC: Line 264: give concentration ranges at -15°C, -18°C, -22°C to compare to DFPC instead. Specify what can be learned from these concentration ranges.
AR: The difference in concentration between DFPC and WT-CRAFT is described in detail in the previous paragraph and now visualized in Fig. 2.

RC: Line 265: repetition from introduction line 67-68.
This part was merged with the Introduction.

RC: Line 272: It is implied that Borys, 1983, Bigg 1996, Bigg 2001 did not measure in the immersion mode. This should be clarified. I recommend merging the literature review here into the introduction.
AR: Indeed, Borys (1983) measured using a dynamic processing chamber, similar to DFPC, while Bigg (1996 and 2001) used a static thermal diffusion chamber. Such information was added in the revised introduction section.

RC: Line 265-290: Consider presenting the comparison to literature in form of a table and to shift it into a Discussion section. Point out and discuss any systematic differences between marine and land influenced data from the Arctic region at specific temperatures.
AR: Now Table 1 reports a compilation of previous INP observations in the Arctic.

RC: Line 279-280: Explain how parameters intervene with INP concentrations. Specify what is meant by "particle activation modality". Quantify the conclusion that the data are generally consistent to literature.

AR: This is a valid suggestion. The sentence was modified as follows: "We note that the comparison to these past studies is only qualitative given the great variability of parameters that could influence *n*INP (e.g., different instruments, locations, season, weather conditions, aerosol particle size distribution, ice nucleation mode, etc.). Regardless, both the DFPC and WT-CRAFT datasets fairly overlap with the *n*INP results reported in Wex et al. (2019), especially for *T*s below -15°C. The authors showed *n*INP previously measured at the same GVB station, during spring and summer 2012. The comparison between the *n*INP data from this study relative to Wex et al. (2019) can be seen in Fig. 2. While this figure provides only a qualitative comparison as two studies examined different aerosol particles collected in different years, we found several interesting agreements and disagreements. First, at *T* = -22°C, Wex et al. (2019) report a very narrow concentration range (27-33 m^{-3}), resulting from only three samples, while DFPC and WT-CRAFT measurements span a much wider range (ca. 3-200 m^{-3}). The upper limit of observable *n*INP in Wex et al. (2019) was roughly 40 m^{-3}, depending on the volume of air sampled onto the analysed filters. On the contrary, the data ranges are in good agreement for *T*s over -18 to -15°C. Finally, the data from Wex et al. (2019) span over a wider range (ca. 10^{-1} – 10 m^{-3}) than WT-CRAFT ones (1-3 m^{-3}) for *T*> -15°C. The difference in the lower limit of the observations is due to different detection limits of WT-CRAFT (1 m^{-3}) and Wex et al. (2019) immersion freezing (ca. 10^{-1} m^{-3}) measurements".

The authors also show a part of our new Fig. 2 for clarity.

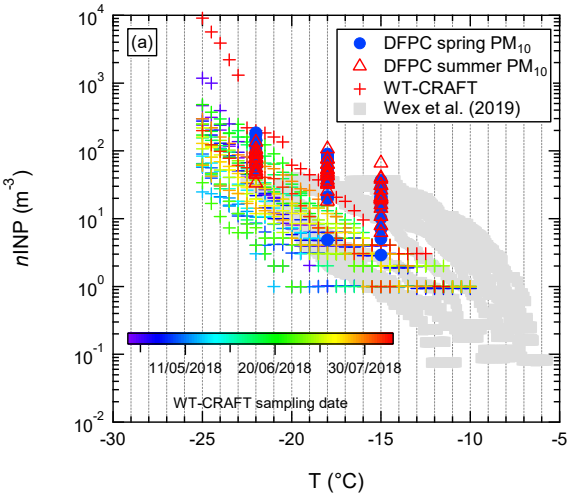


Figure 2: Ambient *n*INP as a function of the activation *T* measured at GVB during 2018 by DFPC and WT-CRAFT. DFPC data are divided in spring (blue) and summer (red) samples, while WT-CRAFT data are color coded according to the sampling date. (a) PM_{10} (DFPC) and TSP (WT-CRAFT) data.

RC: Line 282: Quantify "reasonable agreement"
AR: Discussed above.

RC: Line 285: Quantify "overlaps well"
AR: Discussed above.

RC: Line 286: Quantify "wider range"
AR: Discussed above.

RC: Line 289: What other factors can explain the differences? It would be helpful to specify the upper and lower detection limits of the methods used here for a comparison to Wex et al., 2019.
AR: Discussed above.

RC: Line 291-299: Wide reached and speculative. Sec. 3.7.2. does not provide quantitative evidence on the contribution of continental particles.
AR: We have removed this sentence from the text: "The significantly lower INP concentrations observed over the remote North Atlantic Ocean are likely due to the lack of continental particles, which we will show play an important role in the Arctic atmosphere" The remaining part is just a neutral comparison between present and previous DFPC measurements.

RC: Line 299: Name the locations of the high-altitude and coastal measurements in Rinaldi et al., 2017, 2019.
AR: Done. The revised version of the text is; "If we compare with recent measurements performed at lower latitudes by DFPC, *n*INP over the Arctic was lower than those observed in continental European sites (San Pietro Capofiume, Po Valley, Italy; Belosi et al. (2017) and Rinaldi et al. (2017)), but comparable or even higher with respect to those observed at high altitudes (Monte Cimone, Norther Apennines, Italy; Rinaldi et al. (2017)) or at a Mediterranean coastal location (Capogranitola, Southern Sicily; Rinaldi et al. (2019))".

RC: Line 300: Ice formation is usually observed at -15°C in filter based INP measurements and not unique. It is also present in dust rich environments. Provide references for examples showing otherwise.
Line 303: Specify the "special feature"
Line 304: This is the only reference to Fig.2. The figure is not relevant and can be removed.
AR: We decided to remove the part of the text referred to by the above three comments (L300-306 of the old version), together with previous Fig. 2.

RC: Line 309: Two size ranges do not qualify as "size distribution".
AR: For clarity, we rephrased it to; "…allowed to investigate in fine (< 1 µm) and coarse (>1 µm) INPs".

RC: Line 309: Instead of Table 1, provide a figure showing a scatterplot of INP concentrations measured on PM1 versus PM10 filter in the same time interval (day). All 3 temperatures can be included. Use different colours for spring and summer data.
AR: We have added the requested plot in the supporting material (Figure S1). We would like to keep the Table (now Table 2) in the manuscript. The authors consider these are important data to report in the main text. The Table was updated including WT-CRAFT data sorted by season.

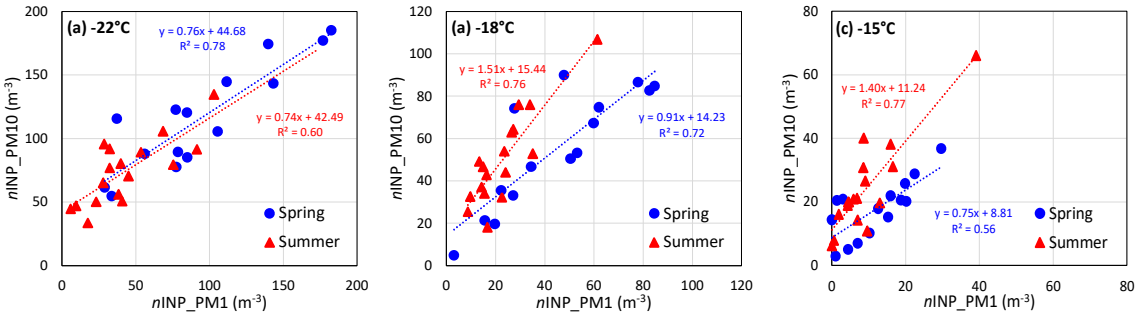


Figure S1. Scatter plot between PM_1 and PM_{10} *n*INP at T of -22°C (a), -18°C (b) and -15°C (c).

RC: Line 311: Specify why long distance to source is suggested. Quantify "long distance".
AR: The authors meant aerosol transport over scales of hundreds to thousands of kilometers, when it is not possible to be more precise on the source distance, by "long distance" or "long range" transport. We have added the explanation in the text; "A small contribution from coarse INPs characterized the spring campaign (~20%), suggesting that the dominant INP sources may be located at long distances (scale of the order of 100s-1000s km), with consequent depletion of the largest particles during transport, due to their higher gravitational deposition velocities. This result is consistent with previous works highlighting the contribution of long range transport from lower latitudes during the Arctic spring (Shaw, 1995; Heidam et al., 1999; Stohl, 2006)".

RC: Line 314: To substantiate this interpretation, compare to by how much the concentration of particles in the coarse fraction change from spring to summer, based on the measured size distribution.
AR: We have added the suggested analysis. The text was modified as follows; "While these coarse INP fraction estimation, presented in Table 2, involves substantial uncertainties, the same trend is inferred by the particle size distribution measurements, which show a significant ($p<0.01$) enhancement of coarse particles contribution in summer (median 30%) with respect to the spring time (median 16%) (Fig. S2)". Moreover, we complementing this statement by presenting the seasonal coarse fraction aerosol particles contribution increase in Fig. S2.

RC: Line 316: Speculative, the coarse particles could be dust particles. It is not clear to what "above considerations" this is liked to.
AR: This part was reformulated to remove speculation; "The increase of coarse INP contribution, from spring to summer time, is progressively more pronounced with increasing activation T. A similar coarse fraction dominated INP population was reported by Mason et al. (2016) for measurements performed between 29 March to 23 July 2014 at the Alert Arctic station, with increasing coarse INPs contribution as a function of the activation T. Our results are unique compared to past studies as our measurements and data support the increase of coarse INP contribution during the meteorological season transition from spring to summer with increasing activation T."

RC: Line 308: PM1 data is not depicted in any figure but used in the analysis. Include DFPC PM1 data to Fig.1 and Fig.3.
AR: PM1 INP data are now reported in the revised Fig. 2 and also in Fig. 4.

RC: Line 308-318: the difference between PM1 and PM10 samples is not obvious from this section. Please provide a figure showing both time series together at -15°C, -18°C, -22°C as well as a scatterplot comparing PM1 to PM10 INP concentrations.
AR: Now provided in Fig. 2, 4 (time series) as well as Fig. S2 (scatter plots).

RC: Line 319-325: INP concentrations from PM1 and PM10 should be compared to particle concentrations <1um and <10um including all smaller sizes instead of only supermicrometre particles. Otherwise the comparison is not objective and only implies that INP concentrations were similar for both cut-offs and the difference is introduced by the choice through what size range was divided.
AR: This is precisely what we did. We compared PM1 INPs with sub-micrometer particle number and COARSE INPs (obtained by difference: PM10 – PM1) with super-micrometer particle number, providing the relative activated fractions. Finally, when discussing the AF of PM10 samples, we have considered the whole range 0.5-10 μm for the aerosol particle number. This part was moved to the new "Activated Fraction" Section (3.3).

RC: Line 327-332: shift to Discussion or introduction. Show how the current data compares to trends found in other studies, e.g., Wex et al., 2019 by plotting the data (at -15°C, -18°C, -22°C) as a function of DOY into the same time-series.

AR: We have added a further plot to the Supporting Material (see below) addressing the referee's point. Unfortunately, Wex et al. measured only three points (all in spring) at T=-22°C, so the only temperatures at which the comparison was possible for the three methods are T =-15 and -18°C.
In calculating the Summer/Spring ratios, we based on the threshold that appears evident from Wex et al. (2019): DOY 150.

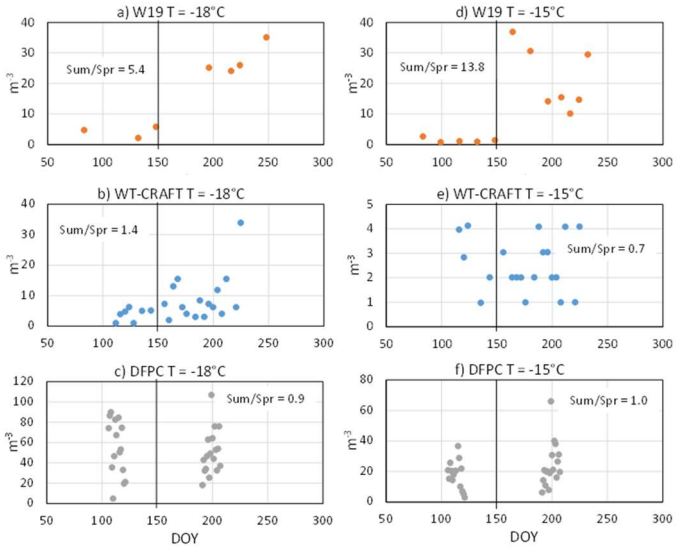


Figure S3. Seasonal evolution of nINP in this study (GVB, 2018) compared to the results by Wex et al. (2019), here indicated as W19, obtained at GVB in spring-summer 2012.

RC: Line 333-343: The main findings need to be worked out clearer in this section. Listing a lot of factors at random temperatures in the text is not helpful to understand the situation. Show that the small dataset can be used to determine robust trends. Factors on the order of 2 are small and should not be overinterpreted. Scattering within a season is much higher.
AR: We would like to retain these discussions as they exhibit statistical significance according to the standard t test. To increase the clarity, we have modified the text as follows; "Interestingly, our 2018 time series data in Fig. 4 do not indicate a clear seasonal increase in ambient *n*INP from spring to summer. A comparison between the seasonal trends in this study and from Wex et al. (2019) can be found in Fig. S3. For the DFPC data, a statistically significant ($p<0.01$) *n*INP reduction (by a factor 1.5) was found at *T* of -22°C, passing from the spring campaign (April) to the summer period (July), while no significant ($p>0.05$) difference was observed for *T*s of -15 and -18°C.

The time series of *n*INP measured by WT-CRAFT agrees with the DFPC one if we consider only the periods in which the two sampling activities were run in parallel: a statistically significant ($p<0.05$) reduction by a factor 1.6 is observed at -22°C and no significant differences can be appreciated at -15 and -18°C. On the other hand, considering the whole WT-CRAFT data extent, a statistically significant ($p<0.05$) increasing *n*INP seasonal trend was observed but only for *T*s between -17.5 and -21.5°C. Even in these cases, the spring to summer enhancement ratios did not exceeded a factor of three. We notice that such variations are smaller than the variability of *n*INP observed within one season. A primary peak in *n*INP was observed by WT-CRAFT during June, at *T*s lower than *T* = -17°C (Figs. 4a and 4b). Further, the increase was visually notable in this case: the average *n*INP during June was up to ~3 (*T* = -20°C) times higher than the average of the rest of the measurement period. As can be seen in Figs. 4a and 4b, a second peak of *n*INP can be observed at the end of the WT-CRAFT measurement period, with the last sample presenting the highest concentrations of all the campaign for many activation *T*s. Further discussion of the *n*INP-AF relationship during this specific period is provided below."

RC: Line 337: What is the reason for limiting the WT-CRAFT dataset to the same period?

AR: The reason is simply to show consistency within the two datasets. By all means, we are not limiting the WT-CRAFT dataset. We first show agreement with the DFPC dataset considering only the overlapping periods, then we present the trends in the whole WT-CRAFT dataset.

RC: Line 340: Why only at -17.5°C and -21.5°C? A plot showing the individual measured Tspectra would be helpful to show how relevant this increase is.
AR: Not only at -17.5 and -21.5°C but between these two temperature boundaries. For clarity, we substituted "within" with "between". Individual WT-CRAFT datapoints are now presented in Fig. 2 (color coded by sampling date), and we state that the Summer/Spring ratio is within a factor of three in the revised text.

RC: Line 340: quantify "clear nINP peak"
AR: We re-worded it as follows; "A primary peak in *n*INP was observed by WT-CRAFT during June, at *T*s lower than *T* = -17°C (Figs. 4a and 4b). Further, the increase was visually notable in this case: the average *n*INP during June was up to ~3 (*T* = -20°C) times higher than the average of the rest of the measurement period".

RC: Line 344: Why was the last sample excluded?
AR: In the revised version we report the difference between the June peak and the rest of the WT-CRAFT observations without excluding any sample. The results do not change. Furthermore we added the following caveat at the end of Sect. 3.4:
We note that the AF data of WT-CRAFT in August is not available due to the lack of SMPS-APS data (maintenance reason). Thus, whether the increase of *n*INP detected by WT-CRAFT in August (i.e., the last two data point sin Figs. 4a and 4b) corresponds to the enhancement of ice nucleation efficiency or absolute aerosol particle concentration remains uncertain.

RC: Line 345: It is an often-misinterpretation of DeMott et al., 2010. The concentration of particles >0.5um are simply used to parameterize INPs of all size, not an actual size fraction of them.
AR: We agree with the reviewer, and this sentence is excluded from the manuscript.

RC: Line 346: This is incorrect. Aerosol were not more ice active, there were only more INP.
AR: We agree with the reviewer - an abundance of INPs and an IN ability/efficiency of aerosol particles are two different things. This sentence is excluded from the manuscript.

RC: Line 347-350: Speculative. Maybe the nINP is higher because more activity at the station towards the end of sampling.
AR: Any activities at the station were carefully recorded, and we do not have any records of suspicious sources of INPs towards the end of our sampling.

RC: Line 352: How was the statistical significance of a seasonal trend determined?
AR: The trend significance was evaluated by checking the statistical significance of the Pearson regression between INP concentration and time.

RC: Line 353: Quantify "peaked mainly"
AR: The magnitude of the June peak is already quantified before; we added the peak/baseline ratio in parenthesis: "(up to 3 times higher concentration that the rest of the measurements)".

RC: Line 355: Speculative. One order of magnitude scattering occurs also on short timescales.
AR: The authors agree. We decided to clarify our points by referring to what the referee pointed out as well as Schrod et al. (2020); "Such results are more in line with the flat trends reported by Schrod et al. (2020). The observed discrepancy between current and aforementioned past studies may be indicative of the inter-annual variability of meteorological conditions and aerosol particle sources determining the ambient *n*INP. Nonetheless, the number of *n*INP observations in the Arctic and their temporal coverage remains limited to derive general conclusions on the *n*INP trends."

RC: Line 360-371: Explain the relevance of scavenging values for the interpretation of measurements here or delete these lines.
AR: We agree with the reviewer on the limited relevance of this part and decided to delete it.

RC: Line 372: Covariance with particle concentration was not shown. This could be an interesting addition to discuss the ice active particle fraction.
AR: We did not report the result of the correlation analysis between *n*INP and particle number concentration as there were mainly non-significant results. Only the few significant results have been included and described in the text.

RC: Line 374: Quantify "even more accentuated".
AR: We modified the text as follows; "During summer, no correlation at all was observed between *n*INP and particle number (R between -0.13 and -0.25)."

RC: Line 375-376: Sentence fragment.
AR: Changed to; "It is, however, important to note that previous studies from different regions report various results about the correlation between INP and particle number concentration: a correlation is often reported with the number concentration of aerosol particles larger than 0.5 µm (DeMott et al., 2010; DeMott et al., 2015; Mason et al., 2015; Schwikowski et al., 1995); in other cases, a complete lack of correlation has been documented (Richardson et al., 2007; Rogers et al., 1998), which is not surprising considering that INPs are only a small fraction of total particles".

RC: Line 376-383: Unclear what this discussion is aiming for. Clarify main point.
AR: This part just aims at setting our results in the perspective of previous measurements. By all means, our results may not be conclusive – that is what we meant to infer.

RC: Line 385ff (Sec. 3.6) Converting nINP to ns doesn't yield new insights. As stated in line 379, INP are only a small fraction of total particles and the total surface area from all different particle types is not related to the number of INP. I recommend deleting the section and Fig. 4 and Fig.5. Instead include the ice active particle fraction at different temperatures and the spring-summer contrast.
AR: The authors agree. We have removed the active site density section and discussed only AF data. But, our overall conclusions did not change. The following comments, therefore, refer to the text that was excluded in part.

RC: Line 389: Repetition of line 72
AR: Yes, thanks for noticing. We believe that this is an important point and worth being echoed.

RC: Line 397: Explain why significance is not found for -16°C. Contradicting results in a narrow T-range could indicate that this analysis is not robust.
AR: This is not a contrasting result; for T=-16°C we have still a positive correlation, but with an R value just below the significance threshold.

RC: Line 399f: The difference indicates that there is no general trend.
AR: The general trend is a slight increase of n_s passing from spring to summer. The fact that we see the maximum increase at different temperatures for the two datasets suggests that aerosol particles may respond differently to the activation modes, as hypothesized previously. Nevertheless, we admit that highlighting this would be speculative.

RC: Line 408: Quantify "substantial good agreement"
AR: The agreement was within a factor of 2.5. Nevertheless, this section was removed as we used AF to describe the ice nucleation efficiency of aerosol particles instead of the ice nucleation active site density.

RC: Line 409f: The aerosol population at GVB is a mixture of many particle types and only a tiny fraction acts as INP. Interpreting ns compared to ns from well constrained particle types is speculative.
AR: This sentence is removed. We now point out "that INPs are only a small fraction of total particles" in Line 427.

RC: Line 417: Show scatterplots of significant correlations in the supplement.
AR: The authors believe that the tabular form of data presentation with our concise explanation is adequate and sufficient. Showing > 30 plots (considering 3 temperatures and two size ranges) for one season would be cumbersome. We provided some representative and meaningful snapshot scatter plots to respond to a later comment (please see below).

RC: Line 421: Explain why and how the results are in line with what considerations.
AR: We thank the reviewer for pointing out this unclear paragraph. The text was modified accordingly:
"In order to investigate the potential sources of the INPs at GVB, a correlation analysis was performed between both *n*INP datasets and the atmospheric concentration of chemical tracers routinely measured at the station. During the spring campaign, *n*INP correlated with tracers of long range transported anthropogenic aerosol particles such as nitrates, non-sea-salt-sulfate and non-sea-salt-potassium (Table 3). Indeed, Udisti et al. (2016) associated spring time non-sea-salt-sulfate at GVB to long range transported anthropogenic sources. The authors also showed that the production of biogenic non-sea-salt-sulfate from the sea is relevant only in summer time. The spring time peak of anthropogenic aerosol transport from lower latitudes is often referred to as the Artic haze (Shaw, 1995). A general tendency to anticorrelation with sodium and chlorine was also observed in both the size classes, though only PM_1 is statistically significant ($p<0.05$)". Further discussion of these findings is provided in the new Discussion Section (4.3).

RC: Line 422: If these are general tendencies they should agree with the PM10 data as well. Explain why the analysis is limited to PM1".
AR: All the INP concentration data anticorrelate with Na and Cl in springtime, but the PM_{10} ones have a Pearson correlation coefficient (R) below the significance threshold corresponding to the 95% confidence interval. In the case of Na (as an examples), R is -0.61 for PM_1 and -0.49 for PM_{10} at T=-22°C; similarly, it is -0.59 for PM_1 and -0.36 for PM_{10}, at T=-18°C; finally it is -0.60 for PM_1 and -0.25 for PM_{10}, at T=-15°C. This shows a general tendency to anticorrelation between the tested variables, even though the result is clearer when considering only the fine fraction. Considering the whole size spectrum (PM_{10}) may include different sources, with different relations with sea spray, resulting in a less clear correlation.

RC: Line 423: Quantify "Less clear"
AR: The text was modified as follows; "The only significant relations observed from the analysis of the summer DFPC data was for T = -15°C…"

RC: Line 427: Explain why these elements are not good tracers for the soil type. What would be good tracers for the local mineralogy of the soil?
AR: This sentence was admittedly speculative and thereby removed. In the future, the measurement by polarization lidar etc. (Mamouri and Ansmann, 2015; 2016) may provide the better insight of dust and *n*INP.

RC: Line 415-429 (Sec. 3.7.1): Add a conclusion, lesson learned from this exercise.
AR: This requested "summary" is now included in the new Discussion Section 4.3.

RC: Line 430-441 (Sec. 3.7.2) Suggest some arguments why a larger land fraction (residence time) of a trajectory should linearly correlate to the INP concentration at -15°C. Why not at lower temperatures as well? The distance of land contact to the receptor, time past, precipitation formation along the trajectory and source strength in different land locations should make a large difference.
AR: We did not address that the effect of land contact is evident only at -15°C. Table 3 and Figure S3 shows that this effect is evident for all the three probed temperatures. Indeed, for the WT-CRAFT dataset, for T=-15°C the correlation is positive but not significant, conversely to the other two reported temperatures.

We are aware that this is a simplified model; nevertheless, we believe that it provides an idea of the broad effect of different land cover types on the INP concentration.

RC: Line 433: Figure S1 is more informative than Fig.6 to show the overpassed ground types. I suggest changing Fig.6 for Fig.S1. In addition, Fig. S1 is referred to more often than Fig. 6 later in the manuscript.
AR: This is a good suggestion. We have merged the two Figures into the new Figure 6.

RC: Line 436: A scatterplot showing fLand versus nINP instead of timeseries would be more helpful than Fig.S3 and Table 3, to show the influence of land sources.
AR: We will address this comment with greater detail and examples below.

RC: Line 435-438: If a fLand effect is found at -15°C it could be evidence against biological INPs dominating nINP at this temperature.
AR: We disagree with this comment. As biological particles or fragments can also derive from soil dust, a snow-free land can act as a source of biological INPs.

RC: Line 440: specify what the "outcome" is and provide an overall conclusion from Sec. 3.7.2.
AR: The following remark was added in the new Discussion Section (Lines 590-594): "This analysis points out that both marine and terrestrial sources may contribute to the INP population in the study area, with land sources showing a potential for dominating the INP pool, due to the higher ice activity of mineral dust and soil particles. On the contrary, marine sources may be significant, even though marine INPs are intrinsically less ice active, because of the extension of ice-free sea waters during the Arctic summer. This has implications also for the future balance between terrestrial and marine INP sources in a warming Arctic (Murray et al., 2021)."

RC: Line 443-482 (Sec. 3.7.3) Specify that this analysis was performed using 14 datapoints from PM1 DFPC. It should be demonstrated that the limited dataset yields robust correlations with CHL. Show some scatterplots. I suggest to include high CHL regions as a ground condition, subdividing the sea category, and include it in the analysis of sec. 3.7.2.
AR: Discussed above. In the new version, we have added the following caveat (Lines 594-602): "The major limitation of our spatio-temporal correlation analysis and of the INP spatial source attribution approach (CWT) is the low number of samples available. This limits the time representativeness of the dataset and increases the uncertainty of the outputs. Nevertheless, the consistency of the two independent approaches (spatio-temporal correlation analysis and CWT source location) provides a certain measure of credibility to the presented results. For this reason, we consider the above as an implication that the marine biota may be a source of INPs in the Arctic. Nevertheless, further studies, based on more robust datasets, are necessary to confirm this result and to achieve a more quantitative understanding of the relative importance of marine vs. terrestrial INP sources over the Arctic. In particular, online INP quantification methods have the potential to provide better suitable data for this kind of statistical approaches and will certainly contribute to clarify INP sources over the Arctic in the future"

RC: Line 443-459: The hypothesis and description of how INP and CHL maps are correlated fits better to the Method section.
AR: Yes, it would. But, as the method itself is stabled and citable (Mansour et al., 2020a and b), we intentionally keep the content close to the Results section for the reader.

RC: Line 451: Why are trajectories with land contact excluded and why only some? A short land contact can have a large impact on nINP.
AR: Here we are trying to show that outside the main INP inputs from land, the background INP concentration has a relation with marine biological activity. Therefore, we have to exclude land influenced samples that would generate a spurious signal in the correlation analysis, being not related to marine sources. We have

excluded samples with fLand>=10% as we did not observe an effect on the INP concentration for samples with lower fLand. Indeed, apart one sample with fLand = 8%, which does not show any anomalous INP concentration, the samples with fLand>0 have values between 1 and 2%, which can be considered negligible. Removing the sample with fLand=8% does not substantially alter the observed correlations with CHL.

RC: Line 460: The time lag doesn't make sense to me. Why would the aerosol generating, biochemical process not change location in 6 days or 16 days? The movement of the surface water should be considered.
AR: The time-lag approach is often used in oceanographic studies (for instance, Volpe et al. (2012), which concluded that phytoplankton biomass and surface heat content anomalies are related with a roughly 5-month time-lag in the Mediterranean Sea). This approach has been derived from oceanography and was applied for the first time on atmospheric studies by Rinaldi et al. (2013) and O'Dowd et al. (2015). They observed a time-lag between CHL concentrations from satellite measurements and organic matter enrichment in the ambient marine aerosol. Later on, a delay time (4 to 10 days) between changes in sea-spray chemical composition and CHL peaks was observed in controlled laboratory experiments conducted in a wave tunnel (Lee et al., 2015; Wang et al., 2015). Furthermore, McCluskey et al. (2017) demonstrated 4-day time-lag between ice nucleating particles (INPs) activation in sea spray aerosol and CHL concentration as a part of the National Science Foundation Center for Aerosol Impacts on Climate and the Environment (CAICE) experiment. These systematic laboratory studies showed short time lag (which may not completely reflect the reality), but demonstrated that a delay exists between the patterns of CHL evolution during a bloom and the observed effects of biological activity on sea-spray aerosol. Finally, Mansour et al. (2020a, b) introduced the use of source identification algorithms (e.g., CWT, as used here, or PSCF) to support the results of the spatio-temporal correlation analysis between in-situ aerosol parameters and surface CHL concentration fields.
In the spatio-temporal correlation analysis between in situ aerosol parameters and surface CHL fields, considering the movement of surface waters is not important. Our analysis evidences a potential relation between the biological activity occurring X days before the aerosol sampling and aerosol properties (in this case INP concentration) at time of aerosol sampling (t_zero), independently on where the bioproducts originating from said biological activity might be at t_zero.
The assumption that the bioproducts of algal activity, responsible for ejection of marine INPs to the atmosphere, do not move substantially from the production region ,within the considered time span, is relevant only when comparing the results of the spatio-temporal correlation analysis with source location results by CWT. This assumption is reasonable: typical surface ocean motion for the Arctic summer is < 5 cm/s (e.g., Lumpkin and Johnson, 2013), corresponding to less than 4 km/day, which is almost negligible considering the resolution of the pixels (~100 Km) in our CWT analysis. Furthermore, Lehahn et al. (2014) showed that an algal bloom can be confined and stable for as long as ca. 30 days in the North Atlantic Ocean (which is characterized by faster currents than the Arctic Ocean).
Hence, the authors believe that our approach is reasonable as supported by previous studies.

RC: Line 464: justify why 6- and 16-day time-lag was selected
AR: The reason is explained in the following lines (484-486): "The maps in Fig. 7 were selected because they clearly show high correlation regions in the seawaters surrounding the Svalbard archipelago (time-lag 6 days), close to the Greenland coast (time-lag 14 and 16 days) and to the northeast of Iceland (time-lag 16 days)."

RC: Line 477-478: Explain how this can be seen in Fig. 7c?
AR: We have added a new plot for major clarity (Figure 8b), showing the intersection between the correlating regions, from the spatio-temporal correlation analysis, and the source regions of marine INPs identified by CWT. This new map confirms substantial agreement in identifying the most likely INP sources in the study domain between the two approaches. We highlight that the two approaches are totally independent and based on different principles; the agreement between the two supports the reliability of the derived conclusions and indirectly also supports the reliability of the correlation analysis, even if conducted with a limited number of samples.

RC: Line 479-482: Consistency is not obvious. There seems to be even more negative correlations. The pattern looks random. I would expect some high productive areas based on ocean currents and biological factors that do not change rapidly.

AR: We have added a new plot showing clearly spatial consistency between positively correlating regions and CWT identified regions. Regarding the existence of negatively correlating regions, we specify that our approach (Mansour et al., 2020a, b) is based on the assumption that if a marine aerosol component is biogenic is should follow the patterns of biological activity (tracked by CHL). For this reason, we focus on positively correlating sea regions in the spatio-temporal correlation analysis. In the lag-correlation approach, an inverse correlation cannot be explained by a physical mechanism: if we assume the aerosol is biogenic, its concentration can only increase with increasing algal activity (positive correlation); therefore, we attribute the observed negative correlations to the relative patterns of CHL in different sea-regions. In other words, if we identify a positively correlating area in Region X (supported by the source spatial location approach, like CWT in this study) and a negatively correlating area in Region Y, we assume that the correlation (negative) in Region Y is due only to the fact that the CHL pattern in Region Y anticorrelate with the CHL pattern of Region X.

RC: **Conclusion**

Avoid euphemistic language.

AR: In the revised manuscript, the authors made sure to put conclusive statements and some future work suggestions in this section.

RC: Line 484-489: It seems that the paper gains little by including the WT-CRAFT dataset. It is only marginally relevant to discuss seasonality in sec. 3.4. In all other sections it is only mentioned that the data agrees with what was seen from analysing DFPC data. It there was a dependency on ice nucleation mechanism (condensation, immersion) it seems not to make a difference on size and source of INP.

AR: The authors now clarify the specifications and capability of WT-CRAFT in the sections above according to the reviewer's inputs. We sincerely hope that our revisions remove misgivings of the reviewer regarding the 150 lpm "stack" flow rate etc. We believe both DFPC and WT-CRAFT results are important to derive our main conclusions, and we keep all INP measurements data as originally presented.

RC: Line 490-493: More, larger INP in summer seems contradictory to the absence of a seasonal trend.

AR: We have revised the Conclusion Section completely. Now we discuss the INP concentration seasonal trend, the AF one and the seasonal evolution of the contribution of coarse INPs. Briefly, we see only a modest seasonal increase in the INP concentration and only limited to a certain T range. The AF, instead, shows a clearer increase at each probed T. Finally, we address the clear and significant increase in the coarse INP fraction contribution. We do not see any contradiction in the above conclusions: INP number concentration is rather constant passing from spring to summer, while the relative contribution of fine and coarse INPs changes with the season.

RC: Line 497: inter-annual variability is a trivial statement. The question is how large the variation is and why it happens.

AR: Given the limited knowledge on INP sources in the Arctic environment, it would unfortunately not be feasible to address the reasons behind the discrepancy between the seasonal trends observed in 2012 and 2018. Regarding the magnitude of the spring to summer concentration increase with respect, for instance to day by day variability, we believe that the new version of the manuscript is more quantitative.

Instead, in Sects. 4.2 and 4.3, we added the following statements as for the future study topic:

"Future application of long-term online INP measurements (e.g., Möhler et al. (2021)) may allow shedding light on the seasonal evolution of *n*INP at GVB and over the Arctic in general."

"In particular, online INP quantification methods have the potential to provide better suitable data for this kind of statistical approaches and will certainly contribute to clarify INP sources over the Arctic in the future."

RC: Line 499: Explain the importance of this study in detail.

AR: The revised Conclusions Sect. explains this in a better way. One of our major findings is related to the different behaviour of aerosol particles sampled at GVB under different ice nucleation modes:
"We considered many factors that could potentially explain the observed difference (Sect. 4) and conclude that the different ice nucleation mechanisms probed by the two techniques (condensation freezing, for DFPC, and immersion freezing, for WT-CRAFT) is an undeniable reason. While differences in the sampling resolution and overall measurement uncertainties have partly contributed to the observed offset, it seems conclusive to address there is ice nucleation mode dependent INP propensity at GVB in 2018 at least. Any future investigations regarding INP compositions and more controlled-study focusing on condensation vs. immersion freezing on identified compositions will lead to further findings to settle this issue".
Another significance of our work is the observation of no substantial seasonal variation in *n*INP accompanied by generally higher ice nucleation efficiency (AF) and a clear enhancement in the contribution of coarse INPs in summer with respect to spring:
"This study also offered unique data examining the seasonality of INPs in the Arctic with respect to *n*INP and AF. Both condensation and immersion INP datasets did not indicate a marked *n*INP seasonal trend. We report a statistically significant spring to summer enhancement in *n*INP only for a narrow range of *Ts* (-17.5 to -21.5°C) and the associated *n*INP enhancement never exceeded a factor of three. On the other hand, the AF of atmospheric aerosol particles from GVB presents a statistically significant spring to summer increase almost independent on the probed *T*, reaching up to ca. 6 times at *T* of -19°C. A clear seasonal evolution of the super-micrometer INP contribution was observed by DFPC. Such contribution was around 20% in spring (with the highest 32% at -15 °C) and increasing markedly in summer and at high *Ts* (45% at *T* of -22°C and 65% at *T* of -15°C)".
Finally, we also provide evidence for the different contribution of local vs. long range, natural vs. anthropogenic and terrestrial vs. marine aerosol particle sources to the INP burden at the study location, contributing to improve our current understanding of INP dynamics over the Arctic.

RC: Line 502-505: This is a weak conclusion. There is no reason mentioned to assume that only one source contributes INP at all temperatures in the spectra.
AR: As evidenced in the revised Introduction, there is not a general consensus in literature on the prevalence of terrestrial or marine sources of INPs over the Arctic. This is partly attributable to the scarce coverage of observational data and may also result from the complexity of the Arctic environment. In this work, we present convincing evidence of the contribution of both source types, even though we are still far from a quantitative understanding of their contributions to the Arctic INP burden. This is probably not the major finding of our work, nevertheless it is a reasonable conclusion worth to be evidenced as it contributes to an open literature debate.

RC: Line 506-508: The relation has not been proven without doubt. It is a speculative interpretation.
AR: In the revised text, the robustness of our approach has been discussed with more detail. Although longer datasets would be desirable for future investigations of the relationship existing between INPs and the marine biological activity, we believe that our results are robust enough to support some connection between the marine biota and atmospheric INPs during the Arctic summer.

RC: **Figures**
The provided figures do not support the content of the manuscript. Fig.2 for example can be deleted is only referred to in a side note and supplementary figures are referred to more often than the actual figures included to the manuscript.
AR: We agree. Our former Fig. 2 was removed.

Helpful figures could include:
1. Temperature spectra with all nINP measured with WT-CRAFT and all datapoints (PM1 and PM10) measured with DFPC, with colour code for DOY of measurement and 3 symbols to differentiate the techniques.
AR: Provided in Fig. 2.

2. Timeseries of nINP data at -15C, -18C, -22C as function of day of year including the 12 data points from Ny-Alesund in Wex et al, 2019,

AR: Thanks for this useful suggestion. We present the DFPC and WT-CRAFT time series in Fig. 4. The suggested comparison with Wex et al. (2019) is now added in the Supplementary Material (Fig. S3).

3. Scatterplot of PM1 vs. PM10 nINP measured with DFPC

AR: Thanks for this useful suggestion. The suggested figure is now added in the Supplementary Material (Fig. S1).

4. Time series showing activated fraction of particles (nINP divided by number of particles in PM1 for DFPC, in PM10 for DFPC and CRAFT) at -15C, -18C, -22C.

AR: Added instead of the n_s time series, including PM1 (Fig. 5).

5. Fig. S1

AR: We merged the old Figure 6 with Fig. S1 obtaining the new Fig. 6.

RC: Fig.1: Showing the individual measurements would be more informative than only median, min, max. Please change the figure accordingly and indicate summer, spring, PM1, PM10 samples in different colours and symbols. Indicate the detection limits of the DFPC and WT-CRAFT.

AR: All incorporated accordingly. To increase visibility for the reader, we have separated Fig. 1 into two panels:

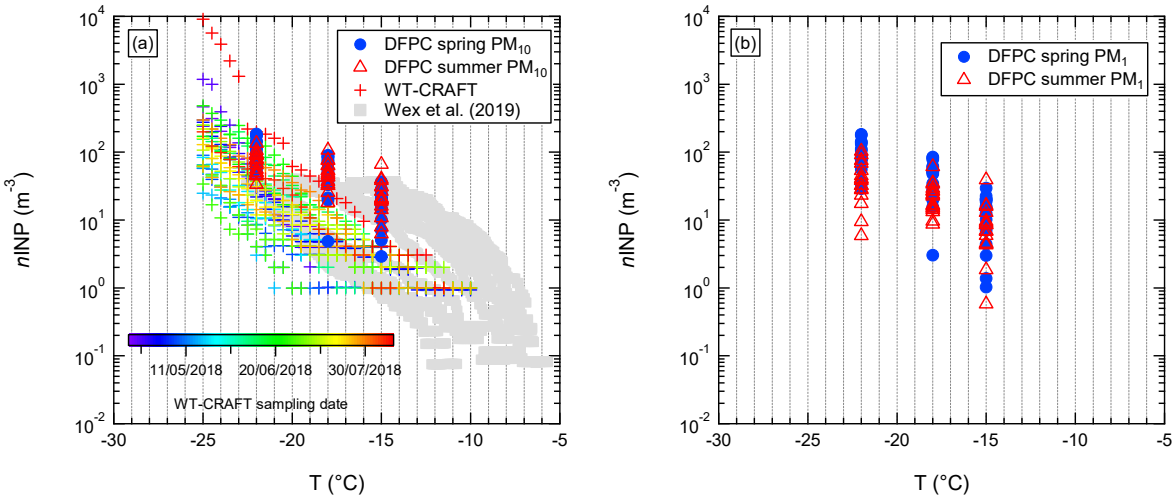


Figure 1. INP atmospheric concentration as a function of the activation temperature measured at GVB during 2018 by DFPC and WT-CRAFT. DFPC data are divided in spring (blue) and summer (red) samples, while WT-CRAFT data are color coded according to the sampling date. (a) PM_{10} (DFPC) and TSP (WT-CRAFT) data. (b) PM_1 data(available only for DFPC). For comparison purposes, the data from Wex et al. (2019), which refer to PM10 samples, are also reported in plot (a). Data for Wex et al. (2019) were downloaded from the repository associated to the publication at….

RC: Fig.2: Comparing it to Fig. 4a in Irish et al., 2017 did not make it obvious how it was adapted. What assumptions are made to overlap the two y-axes (INP in water and INP in air)? This figure can be deleted.

AR: we appreciate the reviewer's suggestion. We agree and removed this figure as suggested.

RC: Fig.3: It needs to be specified how the measurement uncertainty is determined from the sample volume and the analysis. It appears several times throughout the paper and is important.

AR: We thank the reviewer for recapping this point. We addressed this point according to the reviewer's suggestions in the revised version. We added INP_PM1 time series and removed the lines between data points in the WT-CRAFT time series (see Fig. 4 of the revised text).

RC: Fig. 4: It would be more informative to scale the nINP with the total aerosol number.

This would show that INP are not from the bulk aerosol population but rare exceptions.
Summer, spring and PM1, PM10 can be contrasted.
AR: We plotted the AF as requested in a similar way as the revised Figure 2. For major clarity, we divided the Figure in three panels (new Fig. 3).

RC: Fig.5: redundant to Fig.4 no new information in this figure. Remove.
AR: This Figure was substituted with the time series of AF.

RC: Fig.6: The main message from this figure seems to be that sea ice is melting in summer.
This is trivial.
AR: We merged the old Figure 6 with Fig. S1 obtaining the new Figure 6.

RC: Fig. 7: c) It seems the colour bar shows nINP because it is written on top of it. Replace and label colourbar with units. Add a minimal explanation what can be interpreted from the patterns.
AR: Corrected.

RC: Fig. S1: Define ground types in the figure caption.
AR: A reference to the appropriate text Section was added in the caption.

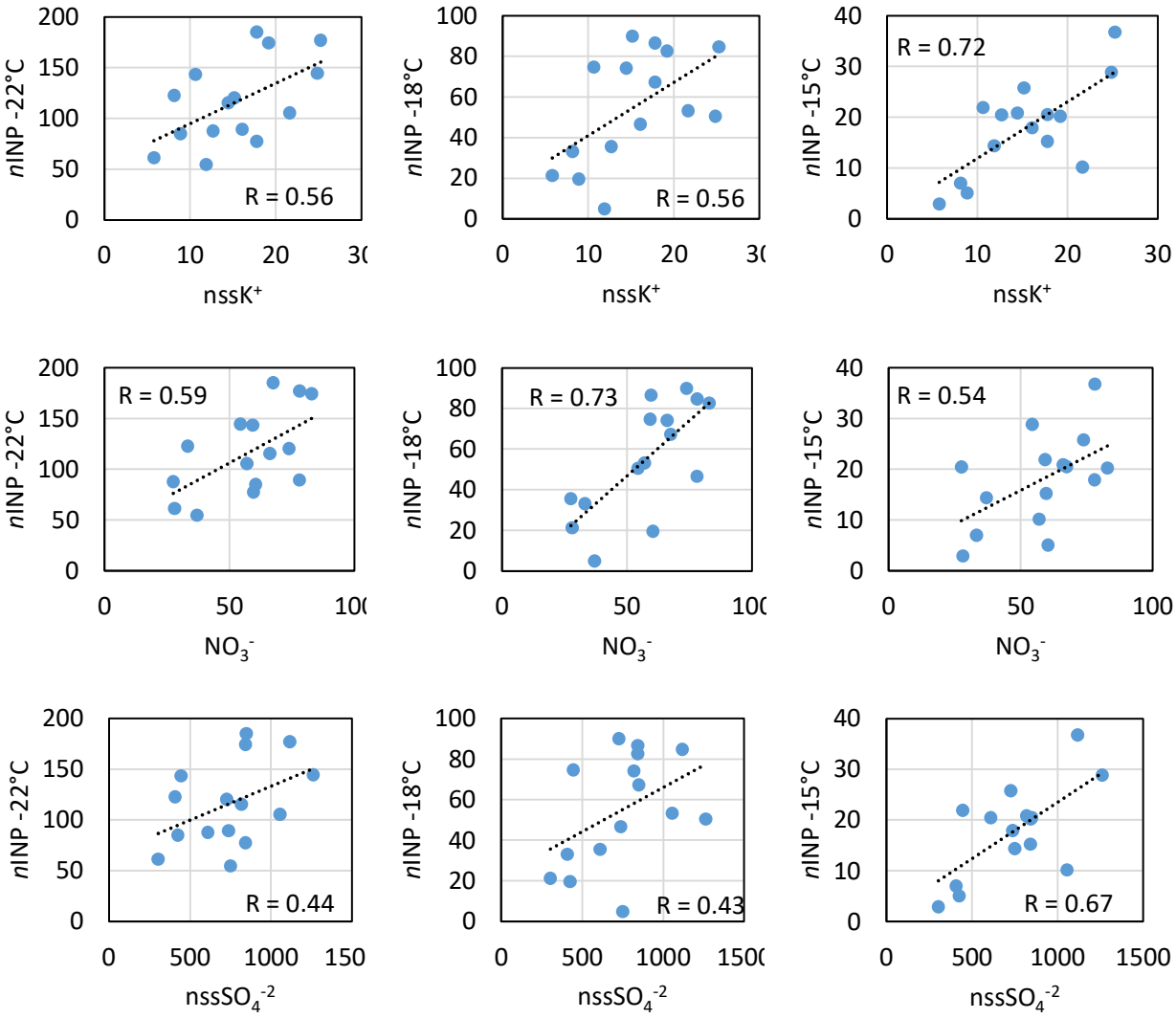
RC: Fig. S2: why are some points connected by lines and others not? Homogenize all precipitation scales and nINP at the same temperatures. -18°C and -22°C plot in first column are switched. Second column last plot DFPC instead of FPC.
AR: Lines between points have the only aim of guiding the eye and evidencing better the similarities between the plotted time series (fLand and *n*INP). Data below detection limit are not represented as markers in the plot and result in the broken line noted by the reviewer.

RC: Fig. S3: use same fLand scale for all DFPC and WT-CRAFT subfigures and same nINP for same temperatures.
AR: We have chosen the scales that makes the plots clearer. Land contribution is much different between DFPC and WT-CRAFT samples because of the different periods covered by the two sampling activities.

RC: Fig S4, S5: Include chlorophyll as fifth land type in S1. Remove figures.
AR: We would like to keep this figure to clarify our points. We have already answered about the reviewer's proposal of changing our approach above.

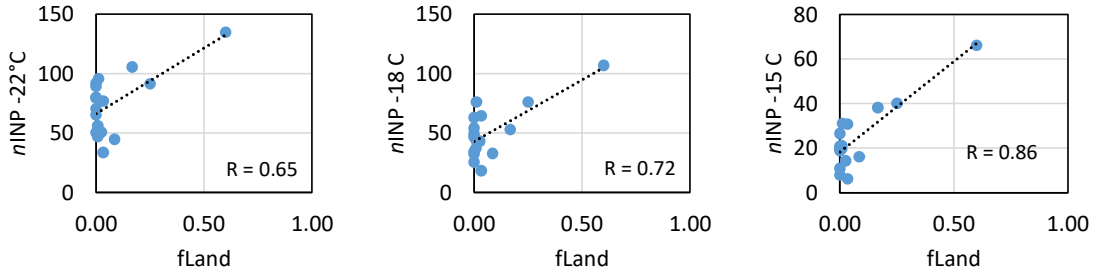
RC: Fig. S6: use same colour-scale range for all subfigures. Use Fig. S1 map design to facilitate comparison. It seems regions where more trajectory points (Fig.S1) pass, also show higher CWT. This points to a problem with the small dataset size for this analysis.
AR: We have answered to these comments above. The new version of the CWT results does not present raised issues.

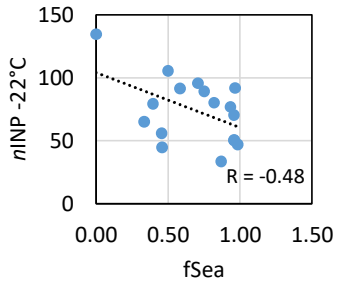
RC: **Tables**
The robustness of correlations in Tables 2 a, b and 3 would be clearer when shown as scatterplots. Due to the small size and structure of the data used, the derived linear correlation coefficients might be strongly biased by few outlier data points and be therefore misleading. Scatterplots help to visually judge correlations. Person's R is sensitive to the data distribution and the R value can be generally misleading. Scatterplots of fLand and nINP would be helpful to investigate these issues.
AR: The authors believe that the tabular form of data presentation with our concise explanation is adequate and sufficient. Showing > 30 plots (considering 3 temperatures and two size ranges) for one season would be cumbersome. Below we report some meaningful examples, taken from the highest correlations we observed in spring time between INP concentrations and chemical tracers of anthropogenic aerosols, on which we based some of our main conclusions on INP sources (INP concentrations are in m^{-3} while chemical species are expressed in $\mu g\ m^{-3}$).

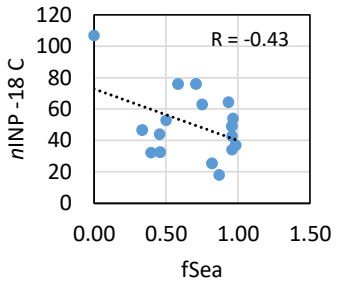


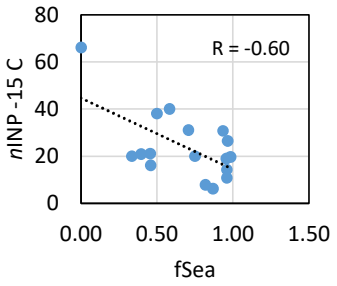
The situation is different for the relationship between INP concentration and ground types along the sampled air mass. In this case, all the more evident correlations are necessarily driven by outliers, which are the few samples presenting non-negligible concentrations of the fLand value (generally resulting in minimum fSea values)! For this reason we decided to present the results in terms of paired time series (Figure S3 of the old version) together with the correlation coefficients of Table 3. Figure 3 shows that each time fLand has a positive peak, this is associated with a *n*INP increase. Below we report scatterplots of the most interesting cases.

DFPC_summer:

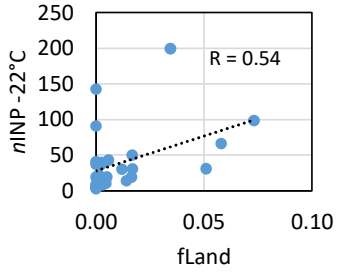


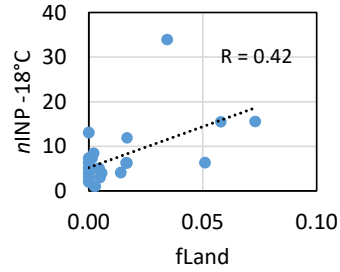


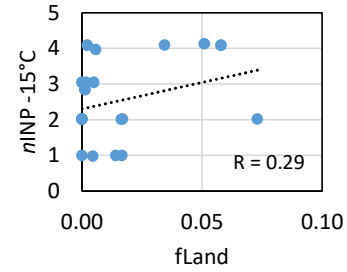




WT-CRAFT_all data:







RC: Technical corrections

Delete “apparently”, “likely”, ”noteworthy”, “worth highlighting” throughout the manuscript.

AR: *removed.*

RC: Line 73: icebreaker ***AR:*** corrected

RC: Line 100, 168: km instead of Km ***AR:*** corrected

RC: Line 104: Section instead of Par ***AR:*** corrected/deleted

RC: Line 115: define TSP, define OD ***AR:*** defined

RC: Line 132, 142: per m3 not per m-3 ***AR:*** corrected

RC: Line 140: replace super-microliter with 3uL ***AR:*** replaced

RC: Line 230: define D^{-} ***AR:*** the symbol does not appear in the updated version as the formulation of the weighting criteria has been changed

RC: Line 239: nINP instead nIPN ***AR:*** corrected

RC: Line 339: p<0.05 instead p<0.5 ***AR:*** corrected

APPENDIX: Scatter plots of randomly selected pixels relative to the spatio-temporal correlation analysis between *n*INP and satellite retrieved surface CHL concentration.

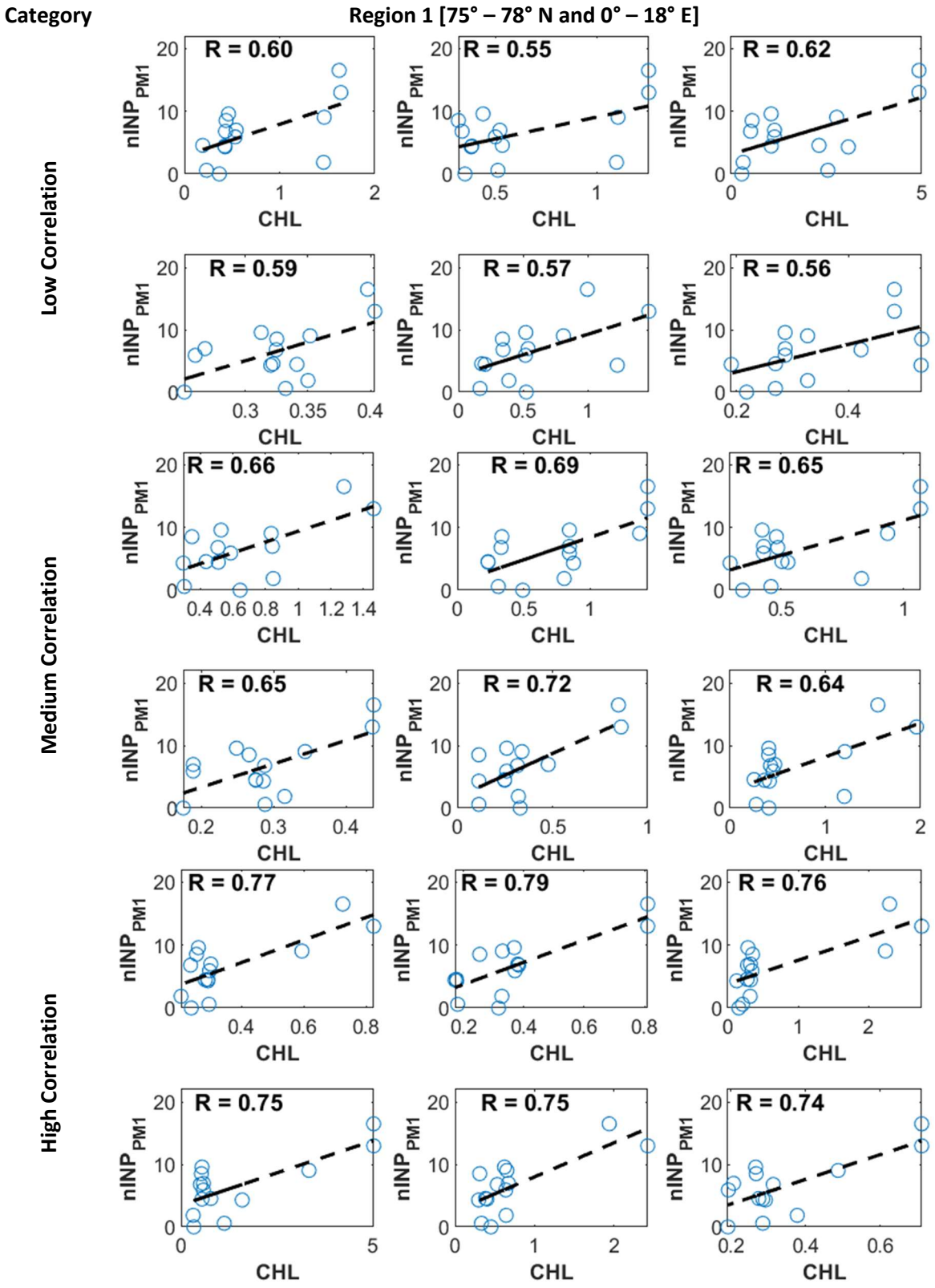


Figure A1: Scatter plots between $n\text{INP}_{PM1}$ sampled at GVB and CHL at pixels selected randomly within seawaters surrounding the Svalbard archipelago.

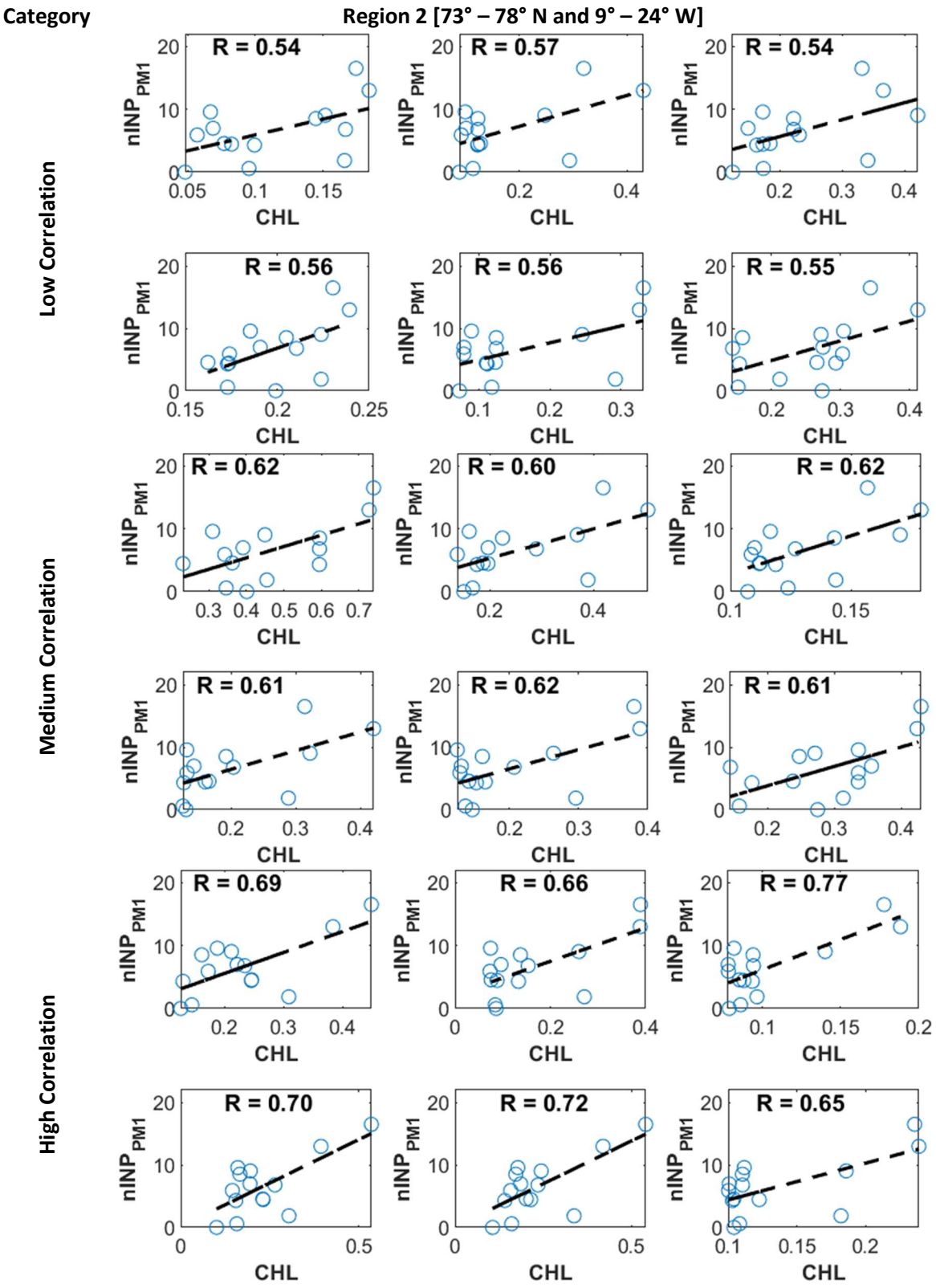


Figure A2: Same as Figure A1, but for seawaters close to the Greenland coast.

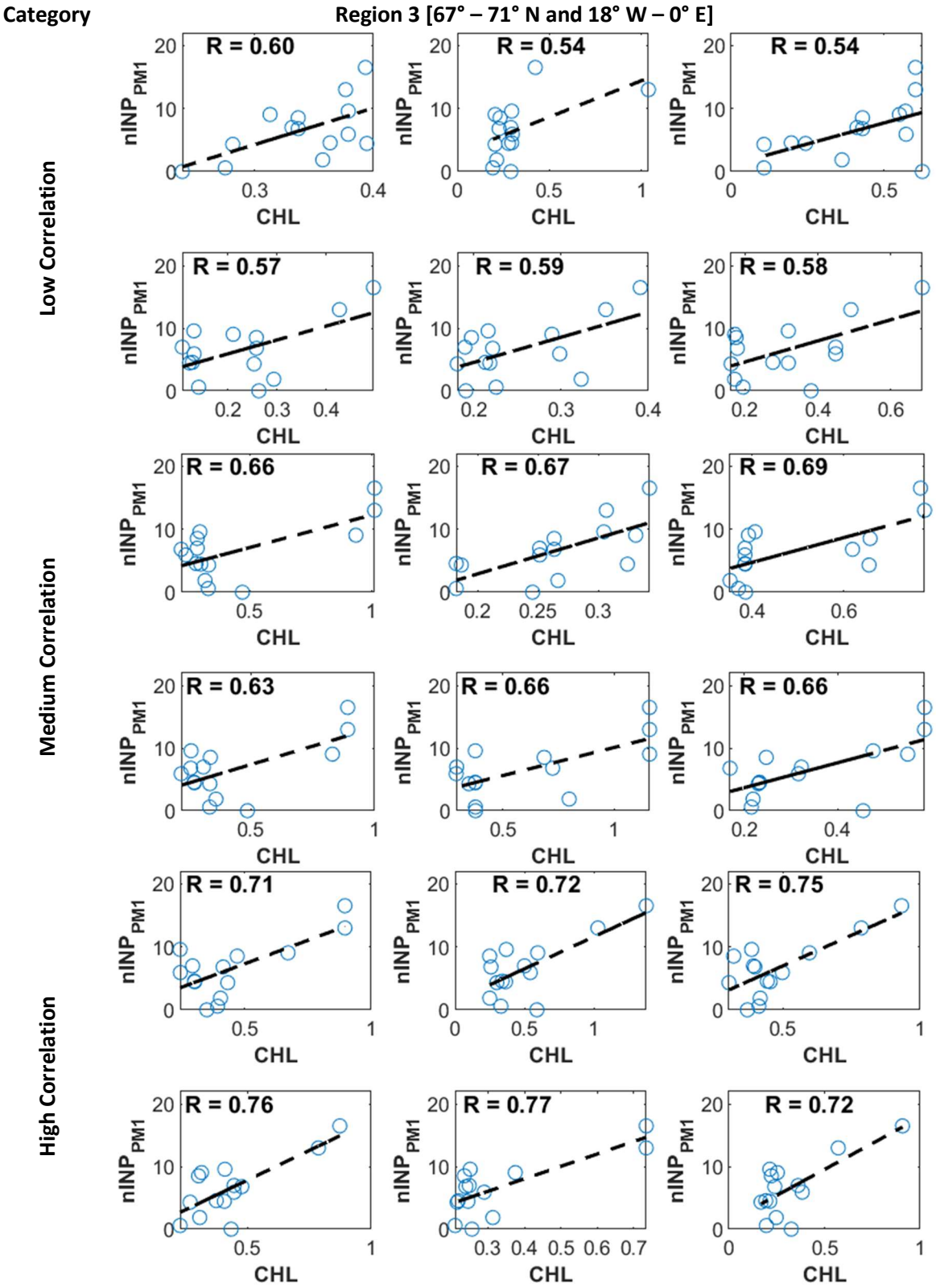


Figure A3: Same as Figure A1, but for seawaters to the northeast of Iceland.

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