

Response to Referee #2

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.

General Comment:**RC:** In this manuscript the INP concentration in Ny-Ålesund (Svalbard) was evaluated in two different seasons and in two different ice nucleation modes, using two different offline techniques. Given the high importance of the Arctic and the low number of studies focusing on INPs, the present study is useful for the ice nucleation, aerosol, and cloud microphysics communities. Although the present results are very valuable, is not completely clear how this study differs from previous studies conducted at the same Arctic location. The manuscript is poorly written as the authors did not pay attention to several important details as listed below. I encourage the authors to improve the quality of the manuscript taking into account the Major, Minor, and Technical comments.

AR: The authors appreciate these general remarks regarding our manuscript by Referee #2. Below, we provide our point-by-point responses. To reflect our changes and articulate what is truly presented in the revised version paper, 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". We admit that we have made some insufficient discussions, leading some of our data interpretations in an original manuscript to be speculative and unclear. Based on the peer-review comments, we removed/modified them to motivate the research as described below in our individual responses.

Major Comments:

RC: 1. I do not see a clear difference between the content of the Abstract and the Conclusions. These two sections need to be different with the Abstract including more concise and quantitative information.

AR: We have revised our abstract as well as the conclusion to reflect all of our major revisions (please see the Track Changes version paper).

RC: 2. The English needs to be significantly improved. The way the document is written makes it very difficult to read it in some parts.

AR: The revised manuscript has been carefully checked by the authors.

RC: Also, the document seems disorganized, and therefore, I encourage the authors to improve this part. The authors need to be more precise, more quantitative, and improve the statistical analysis.

AR: We have improved the manuscript organization by adding a Discussion Section. Moreover, we have reorganized the Results Section for clarity. Now the Results Section reports only the results of our observations and elaborations, while implications of the results are addressed in the Discussion.

RC: 3. There is key information missing in the text (e.g., technical details, references, units, correlation coefficients, etc.). See below.

AR: Thank you for pointing this out. The authors found it as invaluable guidance. We considered it as addressed below.

RC: 4. Lines 94-96: What is different or what is novel in the present study compared to Wex et al. (2019) and Hartmann et al. (2019)?

AR: The revised Abstract and Conclusions Sect. explain the novelty and significance of this work in a better way. In this study we present parallel observations of immersion and condensation INPs, which was never achieved in the Arctic. One of our major findings is indeed related to the different behaviour of aerosol particles sampled at GVB under different ice nucleation modes:

“We 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 *T*_s (-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 *T*_s (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.

Minor Comments:

RC: Line 43: Add a reference after “amplification”.

AR: The authors agree. Serreze and Barry (2011) is now added.

RC: Line 46: Add a reference after “detail”. How about Abbatt et al. (2019)?

AR: This is a valid suggestion. Murray et al. (2021) is now added.

RC: Line 53: “sufficient numbers”. Please be clear.

AR: The authors clarified the sentence by rephrasing it to, “Thus, the presence of aerosol particles that can trigger heterogeneous ice nucleation (ice-nucleating particles, INPs, hereafter) in the Arctic atmosphere can potentially have substantial impacts on precipitation formation, cloud radiative properties and climate (Solomon et al., 2018; Murray et al., 2021)”.

RC: Line 56: “transport dynamics”. Please be clear.

AR: We meant to say “For these reasons, the current inadequate understanding of INP sources, transport and removal processes in the Arctic region...” After the revision, the authors found this part is irrelevant to our introduction. Thus, we decided to remove this sentence.

RC: Line 57: Add a reference after “budgets”.

AR: deBoer et al. (2014) and Morrison et al. (2012) would be appropriate references. However, for the same reason addressed above, we decided to delete this sentence.

RC: Line 69: Define “T”.

AR: Defined for temperature (T).

RC: Line 77: “tripling INP”. Please be clear.

AR: We have rephrased this sentence to “Conen et al. (2016) measured n_{INP} at a coastal mountain observatory in Northern Norway. During the summer, the authors observed that n_{INP} (T of -15°C) in oceanic air tripled after about one day of passage over land”.

RC: Line 88: “evidencing order of magnitude wise increase”. Please be clear.

AR: The authors decided to use more straightforward language (Line 92-95): “In particular, Wex et al. (2019) observed an increase of n_{INP} of more than one order of magnitude from spring to summer (e.g., ~ 14 times at $T=-15^{\circ}\text{C}$) at GVB in 2012. Tobo et al. (2019) focused on two field campaigns held at Mt. Zeppelin, in July 2016 (six samples) and March 2017 (seven samples). They report n_{INPs} at -20°C of about 0.01 L^{-1} in spring and about 0.1 L^{-1} in summer”.

RC: Line 89: Add a reference after “ice”.

AR: Wex et al. (2019) and Santl-Temkiv et al. (2019) are now added.

RC: Line 99: “The aerosol sampling was performed at the Grubebadet observatory”. Add a map showing it.

AR: This is a good suggestion. It is now offered in our new Fig. 1.

RC: Lines 110-111: “The sampling generally started in the morning, during the spring campaign, while it started typically in the afternoon during the summer campaign”. What is the reason for this?

AR: This is a valid question. The variation in our sampling start time stems from only logistical reasons (e.g., not to disturb other activities taking place in each season). We now clarify this point in the text as “The sampling generally started in the morning during the spring campaign, while it started typically in the afternoon during the summer campaign (in coordination with other scheduled activities at GVB).”

In addition, we have carefully assessed the daily variation of aerosol particle concentrations in Spring and Summer (please see the figure below). As seen in the figure, the aerosol particle concentrations are consistent within the 25-75% percentile range. We hope the referee finds our method reasonable.

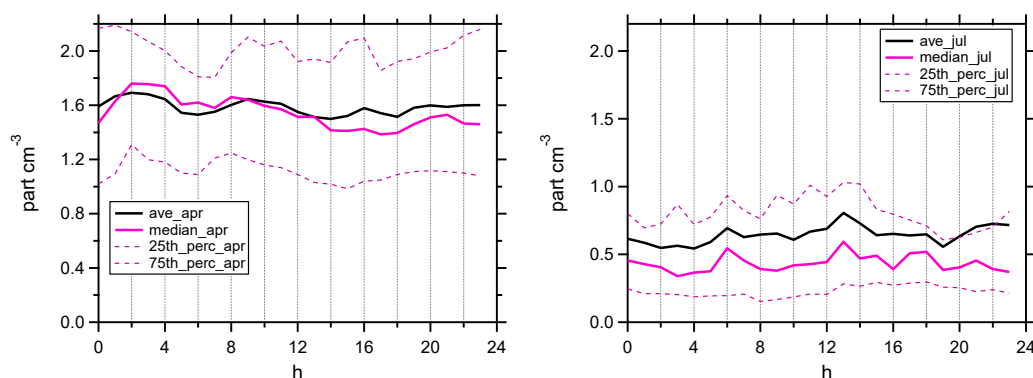


Figure: Daily profiles of particle number concentration at GVB during spring (April) and summer (July) 2018.

RC: Lines 110-111: Add a Table with the details of each sample from both techniques (e.g., initial time, final time, date, etc.).

AR: As suggested, the WT-CRAFT sampling details are now available in Table S1.

RC: Lines 113-119: When where the samples collected with the WT-CRAFT?

AR: Thanks for asking this. 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. We have clarified these in Sect. 2.1; “For the application of West Texas Cryogenic Refrigerator Applied to Freezing

Test system (WT-CRAFT) analysis, a total of 28 samples were collected from April 16 to August 15, 2018. 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)".

RC: Line 114: "0.2 μm pore size". Brand? Model?

AR: Whatman, Track-Etched Membranes, 0.2 μm pore (added in Line 128).

RC: Line 115: Define "TSP".

AR: Defined - total suspended particulate (TSP). Now in Line 129.

RC: Lines 118-119: If the flow rate of the WT-CRAFT is 150 lpm and the flow rate for the DFPC is 38.3 lpm, why the samples from the former one was 4 days and for the later one just 3-4 hours?

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. 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.

RC: Line 121: The authors need to provide a brief description of the method.

AR: Added in Lines 143-148.

RC: Lines 128-129: I would rather add a small paragraph indicating how good is the agreement of the DFPC data compared to other techniques.

AR: As we point out in the revised version manuscript, the agreement depends on the analyzed aerosol particle type. For this reason, instead of addressing generally the agreement of DFPC with other techniques, we discuss in the manuscript the agreement of DFPC with WT-CRAFT in the present and previous deployments. Please, refer to the new Sect. 4.1 for details.

RC: Lines 141-142: "in a known volume". Specify the volume.

AR: Now provided in Table S1.

RC: Line 141-142. This is not very clear.

AR: The authors now clarified our INP estimation method in Sect. 2.2.3. We optimized our suspension water volume in the way the first frozen droplet correspond to 1 INP m^{-3} .

For the WT-CRAFT analysis, we first computed the $C_{\text{INP}}(T)$ value, which is the nucleus concentration in HPLC suspension (L^{-1} water) at a given T as described in Vali (1971). This $C_{\text{INP}}(T)$ value was calculated as a function of unfrozen fraction, $f_{\text{unfrozen}}(T)$ (i.e., the ratio of number of droplets unfrozen to the total number of droplets) as:

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

in which, V_d is the volume of individual droplets (3 μL). Next, we converted $C_{\text{INP}}(T)$ to $n_{\text{INP}}(T)$. 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 151: No parametrizations were derived in this study.

AR: The referee is right. We only did estimation rather than parameterization. The new heading of this Section reads as “2.2.3 Derivation of INP atmospheric concentrations”.

RC: Line 152: “concentration of ice nucleating particles (nINP)”. This was defined in Line 69.

AR: Corrected.

RC: Line 164: “1.95 g cm⁻³”. Add a reference.

AR: We have added the requested reference (Lines 212-214). Now the text reads: “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”.

RC: Line 167: “air temperature, T”. This was first used in Line 69.

AR: Corrected.

RC: Line 173: “GVB”. Define it.

AR: Defined. Line 15.

RC: Line 173-174: “on filters collected”. The authors need to be clear on what filters and how the particle were collected.

AR: This part was extended as follows: “The chemical analysis of major and trace ion species, used in this work as aerosol source tracers, was accomplished on Teflon filters (PALL Gelman) collected at GVB by means of a TECORA Skypost sequential sampler equipped with a PM10 sampling head and operating at 2.3 m³ h⁻¹ (EN 12341)”.

RC: Line 177: “C2O4-2”. Fix it.

AR: Fixed.

RC: Line 179: “Mg²⁺, Ca²⁺”. Fix it.

AR: Fixed.

RC: Line 211 and along the text: The authors used “at Ny-Ålesund”, “GVB”, and “Gruvebadet”. Please be consistent.

AR: Corrected, we now referred to the sampling location as GVB through the whole manuscript.

RC: Line 217-219: “t”, “L”, “Ct” and “Dijt” should be in italics.

AR: Thanks for noticing, they have been checked and corrected accordingly.

RC: Lines 241-242: “The observed offset may derive from the different time resolutions of the sampling for INP analyses, as well as from uncertainties in sampling activities and/or measurement uncertainties”. How about the particle size analyzed in both techniques? The pore size of the filters used is different in each technique.

AR: The filter pore size does not influence the lower size cut-off of the sampled particles for the reason described below. In the process of particles filtration from air, particles smaller than the nominal filter porosity are retained onto the filters, differently from what happens in fluid. This happens because the filters capture particles by different mechanisms: inertial impact, interception and Brownian diffusion. As an

example Willeke and Baron (1993) show that cellulose acetate/nitrate membranes (0.45 micron porosity) capture particles in all the aerosol size spectrum with an efficiency range of >99.8 - >99.99 at face filtration velocities 1-100 cm s⁻¹, respectively. Therefore, the difference in the filter nominal pore size might not substantially impact our INP results and explain the observed INP concentration difference. The authors appreciate the referee for bringing up this point though. While large particles are typically assumed to act as active INPs for their surface, a potential contribution of small soluble particles cannot be ruled out. People in the INP community should keep this in our mind.

RC: Line 242: In the Hiranuma et al. (2015) paper the 2 techniques were no used.

AR: The authors meant to refer to uncertainties in each technique, which are reported over these two inter-comparison papers (DFPC in both and WT-CRAFT in H19). Nonetheless, the authors agree that H15 is not necessary here. It has been removed.

RC: Line 244: Line 43: Add a reference after “questionable”.

AR: We have extended the discussion on ice nucleation modes, mainly in the Introduction. Now the manuscript reads (Lines 59-61): “The distinction between condensation-freezing and immersion-freezing is still matter of debate (Dymarska et al., 2006). Nevertheless, the recent results of Wex et al. (2014) and Hiranuma et al. (2015) suggest that they might be the same process”.

RC: Lines 250: Are you sure the authors clearly distinguished between the 2 modes in Wex et al. (2014)?

AR: Yes. We have double-checked the referred publications:

In Wex et al. (2014) we read: “The above-described results support the hypothesis that condensation and immersion freezing (i.e., the ice nucleation of an insoluble core immersed in a haze particle or in a diluted droplet) might basically be the same process, with the only distinction that a freezing point depression has to be accounted for in the subsaturated regime (i.e., for the haze particles).”

In Hiranuma et al. (2015): “Two types of immersion freezing experiments are presented. One set of experiments was designed to fully activate droplets before ice formation (that is, $T_{\text{droplet}} > -10$ °C), whereas another set was aimed to examine immersion mode freezing at or during droplet formation (that is, $T_{\text{droplet}} \sim T_{\text{IN}}$). The good agreement between the two approaches (see Fig. 2) demonstrates that the ice-nucleating efficiency is similar for immersion and condensation freezing for MCC, supporting the idea that those two mechanisms are in fact the same.”

RC: Line 252: “different aerosol types yielded different results”. Again, is it not the size measured by both techniques? Depending on the aerosol type, their size distribution changes, and therefore, the particle collection efficiency of each technique.

AR: Discussed above.

RC: Lines 257-259: “median 115”. From Figure 1, the value seems to be close to 90 m⁻³ instead of 115 m⁻³.

AR: The old version of Figure 1 referred to the whole DFPC dataset, while in the text we presented the data divided in spring and summer campaign. The revised Figure, Figure 2, presents the DFPC data divided by season. So everything should be consistent now. Our apologies for inconsistency and confusion.

RC: Line 259: “33-135 (median 77), 18-107 (45) and 6-66 (20) m⁻³”. This is not shown in Figure 1. Please add them to the Figure.

AR: In the revised version, all the single data points are presented in the Figure (Fig. 2).

RC: Line 264: “24-9082”. What is the reason of such large variability?

AR: It is due to the outlier data in the last sample of the campaign. This is discussed later on in Sect. 3.4.

RC: Line 274: “range 5-10, 10-30 and 30-70”. Units are missing.

AR: Added. This part was removed and merged with the Introduction section (Lines 70-98).

RC: Lines 281-182: “we can conclude that the results of the present study are generally consistent with literature”. Add literature data to Figure 1.

AR: We have decided to extend this discussion – we now provided a summary of past results of Arctic *n*INPs and used instruments and *T* ranges along with all references in our new Table 1. Associated discussion also appears in Sect. 3.1.

RC: Line 282: Add Wex et al. (2019) data to Figure 1.

AR: This is a good suggestion. Added accordingly to Fig.2.

RC: Line 294: “similar”. By how much?

AR: This sentence was removed as discussing the agreement of DFPC and other immersion freezing techniques (apart WT-CRAFT) is beyond the purposes of this manuscript. For reference, the agreement reported in McCluskey et al. (2018), between DFPC and CSU cold stage, is within 2.5 times.

RC: Lines 300-302: I don’t get what is unique here.

Line 304: “shows the bimodal activation with a hump feature at *T* above -15 °C.”. Really?

Lines 304-305: “may be due to marine biogenic aerosols”. Are not you sampling in a marine environment? Why is this surprising?

AR: The authors agree that this part is too speculative. Based on the suggestion made by Referee #1, the part of the text, which the above 4 comments refer to, and our former Fig. 2 were removed.

RC: Line 311: “suggesting that the dominant INP sources may be located at long distances”. This is rather speculative. Make a link with the evidence you show later.

AR: This part was revised, and we believe the clarity is improved (Lines 340-344): “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: Lines 313-314: “likely resulting from the activation of local sources after snow and ice melting”. Why coarse particles can be transported long distances in spring and not in summer.

AR: Perhaps, our previous sentence was not clear enough to infer that aerosol particles derive mainly from long distance sources at GVB in Spring, therefore there are few coarse particles. In summer, on the other hand, the contribution of coarse particles is dominant, which is hardly reconcilable with long-range transport. This is consistent with literature, describing the phenomenon of the Arctic Haze as cited in the text. We have substantially altered our paragraph here: “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). During the summer campaign, a significant ($p < 0.005$) increase of the contribution of coarse INPs was observed (i.e., 65% at $T = -15^\circ\text{C}$), resulting from the contribution of locally emitted aerosol particles (see Sect. 3.6) in part from the surface exposed to the air after snow and ice melting. 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). The increase of coarse INP contribution, from spring to summer time, is progressively more pronounced with increasing activation *T*.”

RC: Line 317: Mason et al. (2016) found a large contribution from the coarse particles with most of the samples collected in Alert during the spring.

AR: Mason et al. (2016) present the results of measurements performed at Alert between 29 March and 23 July 2014. This period covers both spring and summer. Unfortunately, no information on the seasonal evolution of the fine and coarse INPs is provided. In any case, the analogy with the size distribution in our summer samples is noteworthy and we reported it in our manuscript. We have modified the revised version as follows (Lines 351-354), “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 319: The Creamean et al. (2018) samples were collected in spring.

AR: Thank you for this useful comment. We now mention it in our Sect. 3.3; “Analogously, Si et al. (2018) and Creamean et al. (2018) reported a higher ice nucleation efficiency for super-micrometer particles sampled at Arctic stations. The above cited papers report data collected in both summer (Si et al., 2018) and spring (Creamean et al., 2018).”

RC: Lines 321-322: Should “cm⁻³” be “m⁻³”?

AR: cm⁻³ is the correct unit as particle number concentration is several times higher than INP concentration.

RC: Line 325: “at lower temperatures”. Should it be higher?

AR: Thanks for noting this. In any case, the sentence was removed.

RC: Line 332: Add a reference after “melting”.

AR: Santl-Temkiv et al. (2019) is now added.

RC: Line 354: “at GVB (2012)”. Add the corresponding paper.

AR: This part was merged in the new Discussion Section; the proper citation was added.

RC: Line 360: Why wind direction was not included?

AR: The back-trajectories used for the spatial attribution of INP sources fully cover the requested variable (wind direction). For this reason, we decided not to consider wind direction in the meteorology analysis.

RC: Line 361: “were often associated to a reduction”. It is not very obvious from the Figure. This is a qualitative conclusion.

AR: We have added a quantitative consideration on the significance of the correlation. In addition, the Pearson R values and corresponding significance levels (P) were added in each plot of Figure S5.

RC: Line 361: “the exception of precipitation events”. Add the r^2 .

AR: We have added it in Figure S5.

RC: Lines 362-371: I don't think this is really necessary as it adds too little to the discussion and does not help at all to support the data.

AR: The authors agree. This part has been removed as suggested.

RC: Line 372: “covariate”. Add the r^2 .

RC: Line 374: “more accentuated” and “significant correlations”. Add the r^2 .

AR: We added the Pearson's R coefficient as requested.

RC: Line 385: “showed a significant”. Add the r^2 .

AR: Instead of R^2 , we are presenting the results of a T test for two groups of data. This part of the text was removed and was substituted with the new Section on the AF.

RC: Lines 388-389: “mainly related to long-range transport of anthropogenic aerosol particles from lower latitudes (Arctic haze)”. No evidence provided.

AR: This part was substituted with the new Section on AF. We have considered the reviewer’s comments in writing the new AF Section and the new Discussion Section. Proves that spring time aerosol particles are associated to long range transport from outside the Arctic are presented in Sections 3.2, 3.6.1 and 3.6.2. In the Discussion these findings are summarized as follows:

“The chemical tracer correlation analysis, the ground contribution analysis and the above presented considerations on the different contributions of sub- and super-micrometer INPs in spring and summer time suggest that the main sources of spring time INPs measured at GVB may be located outside the Arctic. They are deemed to derive from the lower latitude regions together with anthropogenic aerosols during the Arctic haze (Heidam et al., 1999; Stohl, 2006). Conversely, the summer time aerosol particles population is more related to local (Arctic) sources”.

RC: Line 396: “for all the activation temperatures”. Just 2 temperatures are shown in the Figure.

AR: Now, Fig. 3 shows data for all the temperatures, and Fig. 5 reports data at 3 temperatures.

RC: Lines 396-397: “with the exception of the coldest one ($T = -25\text{ °C}$)”. This information is not provided.

Lines 398-399: “only a minority of samples (<50%)”. 30-40% is a minority?

AR: Based on the comment of Referee #1, we have substantially revised this section (to activated fraction discussion), and these sentences have been removed since no longer relevant.

RC: Lines 399-401: I don’t get it.

AR: The maximum increase was observed at T of -19 °C . This part was rephrased as “Differently from the DFPC data, the spring to summer AF increase from WT-CRAFT data had its maximum at $T = -19\text{ °C}$ (5.7), with the minimum value obtained at $T = -25\text{ °C}$ (1.4) (Fig. S4)” in Lines 407-408.

RC: Lines 409-410: “suggesting that the INP population over the Arctic in summer originates from a combination of mineral dust and marine aerosol particles”. If I understood correctly, this figure contains the data from both summer and spring, therefore, such conclusion cannot be drawn from the data reported in the figure. Please separate the data sets into summer and spring.

AR: This part was removed.

RC: Line 417: “nINPDFPC correlated”. Add r^2 and p .

RC: Line 422 and 424: “anticorrelation” and “significant”. Add r^2 .

AR: These info are reported in Table 2 (and 3).

RC: Figure 2. It seems you are comparing apples with oranges. Why are the y axis scale different? This figure and its discussion seems to be useless.

AR: All this part was removed.

RC: Figure 4. The authors need to separate them between summer and spring.

AR: This was done, but we reported a different metric, the activated fraction (AF) instead of n_s .

Technical comments:

RC: 1. Line 62: Delete the “dot” after “temperatures”.

AR: Corrected.

RC: 2. Line 100, Line 168: “Km” should be “km”.

AR: Corrected.

RC: 3. Lines 116-117: “diaphragm pump”. Please add the details about the pump.

AR: Added (KnF, IP20-T).

RC: 4. Line 272: “immersion mode freezing”. Be clear.

AR: Good point - all details on the technique used and T ranges are now compiled and reported in Table 1.

RC: 5. Line 273: “).” Fix it.

AR: Fixed. We have divided sentences with “.” with some additional information – “Conen et al. (2016) measured n_{INP} at a coastal mountain observatory in Northern Norway. During the summer, the authors observed that n_{INP} (T of -15°C) in oceanic air tripled after about one day of passage over land. Both marine and terrestrial INP sources were identified by Creamean et al. (2018) in the Northern Alaskan Arctic during spring”.

RC: 6. Line 280: “etc...”. It should be one dot.

AR: Corrected.

RC: 7. Line 282: “Both the datasets discussed”. Be clear.

AR: Corrected. both the DFPC and WT-CRAFT datasets

RC: 8. Line 287: “while both our datasets”. Be clear.

AR: Corrected.

RC: 9. Line 293: “ranged 0.4-15 and 2-40 m^{-3} ”. At what temperatures?

AR: Instead of providing individual temperatures, we now enlisted concentrations and T ranges for all previously published data in Table 1.

RC: 10. Line 361: Figure S2 is called before Figure S1.

Fixed

RC: 11. Line 436: Figure S3 is called before Figure S1.

Fixed

RC: 12. The citing format is wrong and needs to be fixed (e.g. missing spaces between references and when multiple references are cited, they are not organized chronologically).

AR: We apologize for not checking on this. It is now all fixed.

RC: 13. The format of the units is not uniform. For example in some cases the authors used “ $X^{\circ}\text{C}$ ” but in other cases “ $X^{\circ}\text{C}$ ” is used. Please be consistent.

AR: Corrected.

RC: 14. Figure S1. Change the color of the “snow lines” as it is not clearly distinguishable from the white background.

AR: Modified (the Figure is the new Fig. 6).

RC: 15. How much time passed from sampling until the actual INP analyses? Add this information to the text.

AR: DFPC analyses were completed within December 2018 (spring samples) and February 2019 (summer ones). All WT-CRAFT measurements were completed by July 5th 2019. The info was added to the text (Sections 2.2.1 and 2.2.2). We note that Beall et al. (2020) recently found a decrease in n_{INP} depending on the storage method/ T_s and suggested correction factors for the T range of -7 to -17°C . As both DFPC and WT-CRAFT analysed n_{INP} beyond that T -range, we did not apply any correction for this study. This discussion is now given in Sect. 4.1.

RC: 16. Figure S2. The n_{INP} in the middle left panel should be in blue. The n_{INP} in the bottom left panel should be in black. Add more details to the Figure caption.

AR: The Figure was corrected and the caption updated.

RC: 17. Figures S2. I am not sure if it makes sense to correlate precipitation and INP concentration when using the WT-CRAFT based on the low time resolution i.e., 4 days.

AR: We agree and in fact we did not derive strong conclusion from that, we just limited to show the data for completeness of information. The correlation is indeed clearer with DFPC data.

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