

Prof. Barbara Ervens

Co-Editor of Atmospheric Chemistry and Physics

Dear Barbara,

Listed below are our responses to the comments from the referees of our manuscript. For clarity and visual distinction, the referee comments or questions are listed here in black and are preceded by bracketed, italicized numbers (e.g. [1]). Author's responses are offset in blue below each referee statement with matching numbers (e.g. [A1]). Page and line numbers refer to the online ACPD version. We thank the referees for carefully reading our manuscript and for their helpful comments!

Sincerely,

Allan Bertram,  
Professor, Department of Chemistry  
University of British Columbia

## Anonymous Referee #1

Si et al. investigated the concentrations of ice nucleating particles (INP) at Alert, Canada for three weeks in 2016. This study measured INP concentrations in the immersion freezing using the droplet freezing technique. The presented INP concentrations are within the ranges of previous INP measurements in the Arctic. Complementary measurements of tracers for mineral dust, sea spray aerosol, and anthropogenic aerosols were also conducted. The correlations of INPs with these tracers are also investigated. The subject of this manuscript is within the scope of this journal. There are some minor issues and comments that the authors may want to address or consider in the revision.

[1] P4, L17-18, the particles were exposed to RH above water saturation, to what extend? Is there an estimation?

[A1] A humidified gas flow with a dew point of approximately 3 °C was passed through the flow cell. When the flow cell was at 0 °C, the particles were exposed to a RH with respect to liquid water of approximately 115 %. To address the referee's comments, this information has been added to Sect. 2.2.2 in the revised manuscript.

[2] P4, L28 to P5, L11, it is not very clear how the upper and lower limits were calculated and how the freezing of non-spot droplets was considered in the INP calculation. It may be easy for reader to understand or to use such method if example can be provided in the supplement.

[A2] To address the referee's comment, the description of the calculation of INP concentrations and their uncertainties has been improved in the revised manuscript. Specifically, the following equation has been added to illustrate how the freezing of non-spot droplets was considered in the INP calculation:

$$\#INP(T) = \left( -\ln \left( \frac{N_{us}(T)}{N_s} \right) \right) N_s + N_{ns}$$

where  $N_{ns}$  is the number of frozen non-spot droplets. Please see Sect. 2.2.2 for details.

[3] The spots of particles may contain more than one INPs. When cooling the droplet, the first or most efficient INP will trigger the freezing. Once the droplet freezes, it won't freeze again at lower temperature. This could lead to the underestimation of INP concentration. Please comment on this issue.

[A3] Equation (1) does not underestimate the INP concentrations due to the issue raised by the referee. Equation (1) represents the cumulative nucleus spectrum or the concentrations of INPs active at all temperatures  $\geq T$ . The use of Eq. (1) to quantify the concentrations of INPs active at all temperatures  $\geq T$  from droplet freezing experiments has been justified using Poisson's law and Monte Carlo simulations. To address the referee's comments, this information has been added to the revised manuscript.

**[4]** P6, L30-31, although the equation 3 is the standard method to calculate nss-so4, how the Cl depletion in sea salt particles will affect these calculations.

**[A4]** To address the referee's comment, we have calculated  $[\text{nss-SO}_4^{2-}]$  using a method that does not rely on  $[\text{Cl}^-]$ . Specifically, we calculated  $[\text{nss-SO}_4^{2-}]$  based on  $\text{Na}^+$ , where  $\text{Na}^+$  comes solely from sea salt and is not affected by Cl depletion (Balasubramanian, 2003). The difference between the concentrations of  $\text{nss-SO}_4^{2-}$  based on  $\text{Cl}^-$  and the concentrations based on  $\text{Na}^+$  is less than 5 %. This difference has been added to Sect. 2.4 in the revised manuscript.

**[5]** P8, L31 to P9, L2, the manuscript states this assumption, it would be useful if the authors can add a short discussion on the potential bias this assumption may lead to the correlation analysis.

**[A5]** In the revised manuscript, we have commented on the potential implications of this assumption. Specifically, the following statement has been added to Sect. 3.3 in the revised manuscript:

“...If this assumption is not correct, correlation coefficients between INPs and aerosol tracers will be smaller than expected. Shown in Fig. S1 are air mass back trajectories initiated every 2h during each 24h quartz filter sampling period. The back trajectories suggest that for a large fraction of the samples, the source of the air masses during INP sampling was similar to the source of the air masses measured during quartz filter sampling.”

**[6]** How the non-INP affect the freezing data when dissolve in the droplets, this is due to sampling method (collecting a spot of impacted particle vs. individual particles), such as salts including NaCl? This may need additional discussion.

**[A6]** The referee is correct that the method of collection and formation of droplets results in the soluble material, such as salts including NaCl, being mixed with mineral dust particles within the same droplets. However, in the atmosphere, the soluble material, such as NaCl, may not be internally mixed with mineral dust. Studies of the mixing state of soluble material with mineral dust at Alert during the same time of the year are needed. To address the referee's comments, additional discussion has been added to Sect. 3.4 and Conclusions in the revised manuscript.

## **Anonymous Referee #2**

Review for “Concentrations, composition, and sources of ice-nucleating particles in the Canadian High Arctic during spring 2016” by Meng Si and co-authors, submitted to ACPD:

The manuscript reviewed herein is a well written, up-to-date and informative piece of work. Impactor samples were taken in the Arctic and analyzed wrt. atmospheric concentrations of ice nucleating particles. The results were brought into context with information on the chemical composition and on air mass origin. INP that were ice active at -25°C were shown to correlate with mineral dust particles, with the Gobi desert being a possible source. For INP ice active at higher temperatures, no correlation was found, although sea spray and anthropogenic tracers were tested.

There is a number of (mostly smaller) issues listed below. Other than that, the manuscript well merits publication in ACP once these issues have been tackled.

[7] Abstract: p1, line 17: Add “at that temperature” behind “INP population”, as this is likely not the case at higher temperatures.

[A7] To address the referee’s comment, we have specified the temperature after “INP population”.

[8] Also, mention that you examine immersion freezing – best do that already in the abstract, and/or maybe again later.

[A8] To address the referee’s comment, we have indicated earlier in the abstract that we examined immersion freezing.

[9] And either here or maybe in the methods section, mention how many separate samples you analyzed for INP concentrations. I did not know what to expect until I saw Fig. 2.

[A9] To address the referee’s comment, the total number of INP samples reported in current study has been mentioned in the Abstract and again in the Methods.

[10] Introduction: p2, line 31 ff: It should become clear that different ice active substances are ice active at different temperatures. Sea spray, anthropogenic aerosol and mineral dust all are a bit intermingled, here. Please make this clearer.

[A10] To address the referee’s comment, the introduction has been modified to make it clear that different ice active substances are ice active at different temperatures. Specifically, the sentences have been modified to the following:

“These data were used to determine if mineral dust, sea spray aerosol, and anthropogenic aerosol are a major contributor to the INP population at -15, -20, and -25 °C in the Canadian High Arctic during spring. Studies as a function of temperature are necessary since different types of aerosols are ice-active at different temperatures. Although a few studies have identified mineral dust particles as an

important contributor to the INP population in the Arctic during spring, additional studies are needed to determine how often mineral dust is an important contributor. The measurements reported here together with particle dispersion modelling were also used to assess the source of the INPs at a freezing temperature of -25 °C.”

[11] Methods: p3, line 17: It is not clear to me why there are three circular glass slides on the second stage? It sounds as if they are there simultaneously? Or did you only use three during the whole time? Please clarify!

[A11] Three hydrophobic glass slides were located on the second stage simultaneously. As a result, each collected sample consisted of three hydrophobic glass slides. This has been clarified in Sect. 2.2.1 in the revised manuscript.

[12] p4, line 20: It is crucial what happens to the RH during cooling, in terms of the effect of RH: did the droplets grow or shrink during this phase, if yes, to which extent? (They might get in touch with each other, or they might dry out, as the two extreme ends of what could happen.) Is that prevented somehow? How? A bit of text discussing this should be added here.

[A12] If no gas is passed through the flow cell during cooling, the droplets tend to grow by condensation. To prevent the growth, a small flow of dry helium ( $\sim 0.2 \text{ L min}^{-1}$ ) was passed through the flow cell during cooling. To address the referee's comments, this information has been added to Sect. 2.2.2 in the revised manuscript.

[13] p4, line 28 ff: Do really several separate droplets form on one spot? How large is such a spot? You may want to give this number here.

[A13] The diameter of each spot was approximately 110  $\mu\text{m}$ . This information has been added to Sect. 2.2.2 in the revised manuscript.

[14] p5, line 5 ff: Freezing of “non-spot droplets” may come from impurities on or in the glass, particularly as particles as small as 180 nm may not be very ice active. And if this is the case, your way of analyzing the data leads to an overestimation. Did you try the experiment on a “field blank” glass (glass slides treated as the ones used for sampling, including to put them into the sampler, just without turning on the air flow)? Are those “blanks” you mention on p5, line 20 such “field blanks”? How do the data from your “blanks” compare to the number of “non-spot droplets”? And should not the counts obtained from the “blanks” be subtracted from the counts of the samples, instead of adding “non-spot droplets” to the counts of the samples?

[A14] The blanks mentioned on P5, line 20 and shown in Fig. 2 refer to field blanks (treated exactly as the referee described) and lab blanks (glass slides that were cleaned in the lab the same way as the ones used for sampling in the field). Lab and field blanks shown in Fig. 2 are consistent with lab and field blanks determined in another study using exactly the same techniques as described in the current study (Irish et al., 2018). In short, rarely do we see freezing at -25 °C or above in the field or lab blanks. In the current study, we report INP concentrations at  $\geq -25 \text{ °C}$ . Thus, any freezing events of non-spot droplets that happened at  $\geq -25 \text{ °C}$  were most likely

not from impurities on or in the glass substrates. We assume that non-spot droplet freezing may have been due to INPs  $< 0.18 \mu\text{m}$  in diameter not focused into the spots or a small fraction of INPs  $\geq 0.18 \mu\text{m}$  not concentrated into the spots due to rebound from the hydrophobic glass slides. To address the referee's comments, this information has been added to Sect. 2.2.2 and Sect. 3.1 in the revised manuscript.

[15] p5, line 14 ff: The number 300 confused me. Is that the number of impactor spots that were sampled on that stage? If yes, please clarify! Also, above you discuss a number of sources for uncertainty (several droplet on one spot, merging droplets, . . .), and here now you say that the uncertainties were derived based on Koop et al. (1997). So were these issues (several droplet on one spot, merging droplets, . . .) not considered for the uncertainty?

[A15] 300 is the number of nozzles in the nozzle plate of the impactor, and consequently, the number of spots generated on the second impactor stage below the nozzle plate. The uncertainties based on Koop et al. (1997) account for the uncertainty associated with the limited number of freezing events detected. The overall uncertainty in the concentrations of INPs reported in the manuscript includes this uncertainty as well as the uncertainties discussed earlier. This has been clarified in Sect. 2.2.2 in the revised manuscript.

[16] p5, line 19: More information on the background measurements is needed. Right now, you basically only show this background data in Fig. 2, but it is not said if it had been subtracted, and depending on if you did or did not subtract it, why it was done how it was done. This should be added here.

[A16] To address the referee's comments, more information about the background measurements (i.e. blanks) has been added to the revised manuscript. Specifically, the following sentences have been added to Sect. 2.2.2:

“One field blank was collected by treating the slides in the same manner as the sample hydrophobic slides including locating the hydrophobic slides in the impactor, except that the pump was not turned on. Lab blanks refer to slides cleaned in the same manner as the ones used for sampling in the field but not sent to the field.”

And to Sect. 3.1:

“For the remainder of this document, we focus on INP concentrations at -15, -20, and -25 °C. ... Since freezing was rarely observed at  $\geq -25$  °C in the blank experiments, the INP concentrations determined from the blank experiments were not subtracted from the INP concentrations reported in this study”

[17] p6, line 13 ff: I now know that it is discussed later that the daily filter samples described here have a different time resolution than the samples taken for INP analysis (p-8, line 31). This confused me when reading it for the first time, and it would be good to mention already here, that this is the case and that it is discussed later. At the later discussion of this issue, it would be good to mention if air mass trajectories were at least

similar throughout the whole days when you sampled. For days when that was not the case, less correlation between INP and any information on chemical composition should be expected.

[A17] Thank you for pointing this out. The following statement has been added to Sect. 2.4 where the filter sampling time was first mentioned:

“Hence, collection times for tracer measurements (~ 24h) were different than the collection times for INP measurements (~ 2h). The implications of the different collection times are discussed in Sect. 3.3.”

In addition, air mass trajectories initiated every 2h to illustrate the variability in air mass trajectories over the 24h quartz filter sampling periods have been added to the Supplement (Fig. S1).

[I18] Results and discussion: p9, line 27: “The North Pacific Ocean is not likely the source of mineral dust either.” The high signal in Panel D of Fig. 9 confused me in the beginning. But after looking at the plots a bit, I now assume that it originates in the fact that this Pacific region gets its high signals as it has small signals in both, plot B and C. So it would be the result of bad statistics (dividing a small signal by yet smaller signals).

But still, would not all regions in B have to have higher concentrations as those in C? C, as I understand it, is a subset of B, and yet, there are regions in C that indicate larger concentrations, which cause the high values in the Pacific in D. - How can this be? Please explain the latter, and also mention that high values in the Pacific in “D” come from the division of small signals by small signals, if that is the case.

[A18] The statement that “The North Pacific Ocean is not likely the source of mineral dust” is because that the Pacific Ocean is not considered as a traditional source of mineral dust. This information has been added to the revised manuscript. Besides, the description of Fig. 9 has been revised for clarity. Specifically, the following has been added to the revised manuscript:

“...Figure 9(B) shows the average footprint PES plot for all samples, and Fig. 9(C) shows the average footprint PES plot for the four samples with the highest mineral dust concentrations, which were collected on March 21, 22, 27, and 28. Note, since average footprint PES plots are shown in Fig. 9(B) and 9(C), Fig. 9(C) is not a subset of Fig. 9(B). Shown in Fig. 9(D) is a ratio plot of the sum of footprint PES plots of the four samples with the highest mineral dust concentrations to the sum of footprint PES plots of all samples. These types of ratio plots are often used as a sensitive method to identify the source regions of a component under investigation (Hirdman et al., 2010). ...”

[I19] p10, line 12-16: I agree that you show that mineral dust contributes INP that are ice active at -25°C. You also show that INP that are ice active at higher temperatures have other sources (as they do not correlate with mineral dust). So the statement you make here is a bit twisted, as you do not show what contributed the INP that are ice active at

the higher temperatures, which is the temperature range for which INP from sea spray (generally assumed to be biogenic in origin) may be ice active. And for mixed phase clouds INP active at higher temperatures might be even more important than INP active at  $-25^{\circ}\text{C}$ . Please correct your statement here accordingly, particularly the last sentence.

*[A19]* To address the referee's comments, we have specified the temperature to make the statement more accurate. Specifically, the statement has been modified to:

“Our results suggest that mineral dust is a more important source of INPs at  $-25^{\circ}\text{C}$  than sea spray aerosol for the time and location studied.”

*[20]* p10, line 21 ff: It is VERY well known that salt in general causes a freezing point depression (that's why salt is put on icy roads in winter time). That is why I was somewhat surprised by that part in general, as I would not consider this a new result. There is also other older work on that, not for NaCl but for salts and other soluble material in general, on the temperature depression of ice nucleation for minerals (Koop & Zobrist, 2009, Wex et al., 2014), in which even more general ways of describing this suppression are given. Although this originates from laboratory studies, it may merit including that and mentioning that your results is not such an astonishing finding. Also, as you know the salt concentrations on the filters, it would be interesting to see how much freezing point depression could be expected from that. This is easy to do, and, albeit only being an estimate, it could show if an effect may be expected at all.

*[A20]* To address the referee's comments, we have estimated how much the NaCl can decrease the freezing temperatures of mineral dust by the traditional freezing point depression mechanism, which involves the salt changing the water activity in solution. The maximum concentration of NaCl in our freezing experiments was  $\sim 0.03 \text{ mol L}^{-1}$ , which would only cause a freezing point depression of  $\sim 0.1^{\circ}\text{C}$ . Since this number is within the uncertainty of our measured freezing temperatures, it is too small to explain the negative correlations shown in Fig. 11. On the other hand, very recent studies have illustrated that trace amounts of NaCl can lower the freezing temperature of feldspar (a type of mineral dust) by surface specific interactions between the solute (NaCl) and the mineral dust surface, and the decrease in freezing temperature by this mechanism is more than expected based on traditional freezing point depression mechanism (Whale et al., 2018). Additional discussion has been added to Sect. 3.4 in the revised manuscript to make it clear that our results cannot be explained by the traditional freezing point depression mechanism.

*[21]* p11, line 17: Do you mean “at any of the three temperatures”?

*[A21]* This statement has been revised to improve clarity.

*[22]* p11, line 19ff: As you discuss other non-Arctic studies here, you may want to include a recent study on INP in Beijing, where it is also said that: "All these results indicate that Beijing air pollution did not increase or decrease INP concentrations in the examined temperature range (down to  $-25^{\circ}\text{C}$ ).” (Chen et al., 2018).

*[A22]* Thank you for the suggestion. This reference has been added to the revised manuscript.

*[23]* Figures Caption of Fig. 9: What do you mean by “for all mineral dust sampling periods”? The time when samples were analyzed wrt. INP concentrations? Please clarify, here or in the respective text.

*[A23]* This figure caption has been revised to “panel (B) is the average footprint PES plot from a 20 day FLEXPART analysis for all samples” to improve clarity.

*[24]* Supplement There is a supplement with nothing else than one table that is not even large. This should be included in the main manuscript. It makes no sense to have a separate document for so little of information.

*[A24]* Additional plot and text have been added to the Supplement. Hence, a Supplement is necessary.

Literature Chen, J., Z. Wu, S. Augustin-Bauditz, S. Grawe, M. Hartmann, X. Pei, Z. Liu, D. Ji, and H. Wex (2018), Ice nucleating particle concentrations unaffected by urban air pollution in Beijing, China, *Atmos. Chem. Phys.*, 18, 3523–3539, doi:10.5194/acp-18-3523-2018.

Koop, T., and B. Zobrist (2009), Parameterizations for ice nucleation in biological and atmospheric systems, *Phys. Chem. Chem. Phys.*, 11(46), 10839-10850, doi:10.1039/b914289d.

Wex, H., P. J. DeMott, Y. Tobo, S. Hartmann, M. Rösch, T. Clauss, L. Tomsche, D. Niedermeier, and F. Stratmann (2014), Kaolinite particles as ice nuclei: learning from the use of different kaolinite samples and different coatings, *Atmos. Chem. Phys.*, 14, 5529-5546, doi:10.5194/acp-14-5529-2014.

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# Concentrations, composition, and sources of ice-nucleating particles in the Canadian High Arctic during spring 2016

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**Abstract.** Modelling studies suggest that the climate and the hydrological cycle are sensitive to the concentrations of ice-nucleating particles (INPs). However, the concentrations, composition, and sources of INPs in the atmosphere remain uncertain. Here we report daily concentrations of INPs in the immersion freezing mode and tracers of mineral dust (Al, Fe, Ti, and Mn), sea spray aerosol ( $\text{Na}^+$  and  $\text{Cl}^-$ ), and anthropogenic aerosol ( $\text{Zn}$ ,  $\text{Pb}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and non-sea-salt  $\text{SO}_4^{2-}$ ) at Alert, Canada during a three-week campaign in March 2016. In total, 16 daily measurements of INPs are reported. The average INP concentrations measured in the immersion freezing mode were approximately  $0.005 \pm 0.002 \text{ L}^{-1}$ ,  $0.020 \pm 0.004 \text{ L}^{-1}$ , and  $0.186 \pm 0.040 \text{ L}^{-1}$  at  $-15^\circ\text{C}$ ,  $-20^\circ\text{C}$ , and  $-25^\circ\text{C}$ , respectively. These concentrations are within the range of concentrations measured previously in the Arctic at ground level or sea level. Mineral dust tracers all correlated with INPs at  $-25^\circ\text{C}$  (correlation coefficient,  $R$ , ranged from 0.70 to 0.76), suggesting that mineral dust was a major contributor to the INP population at  $-25^\circ\text{C}$ . Particle dispersion modelling suggests that the source of the mineral dust may have been long-range transport from the Gobi desert. Sea spray tracers were anti-correlated with INPs at  $-25^\circ\text{C}$  ( $R = -0.56$ ). In addition, INP concentrations at  $-25^\circ\text{C}$  divided by mass concentrations of aluminum were anti-correlated with sea spray tracers ( $R = -0.51$  and  $-0.55$  for  $\text{Na}^+$  and  $\text{Cl}^-$ , respectively), suggesting that the components of sea spray aerosol suppressed the ice-nucleating ability of mineral dust in the immersion freezing mode. Correlations between INPs and anthropogenic aerosol tracers were not statistically significant. These results will improve our understanding of INPs in the Arctic during spring.

## 1 Introduction

The formation of ice in clouds can be initiated by homogeneous or heterogeneous nucleation. Heterogeneous nucleation of ice in clouds occurs on only a small subset of atmospheric particles, referred to as ice-nucleating particles (INPs) (Vali et al., 2015). A variety of aerosol particle types have been identified as possible INPs, including, but not limited to, mineral dust, sea spray aerosol containing biological material, and primary biological particles from terrestrial sources (Coluzza et al., 2017; Cziczo et al., 2017; Hoose and Möhler, 2012; Kanji et al., 2017; Murray et al., 2012). INPs can change the frequency and properties of clouds in the atmosphere and influence climate and precipitation (Andreae and Rosenfeld, 2008; Curry,

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1995; DeMott et al., 2010; Du et al., 2011; Lohmann and Feichter, 2005; Prenni et al., 2007; Xie et al., 2013). As a result, to predict precipitation and Earth's climate, an understanding of the concentrations, composition, and sources of INPs in the atmosphere is required.

INPs have been measured in the Arctic since as early as the 1970s (Bigg, 1996; Bigg and Leck, 2001; Borys, 1989; Conen et al., 2016; Creamean et al., 2018; DeMott et al., 2016; Flyger et al., 1973, 1976; Flyger and Heidam, 1978; Fountain and Ohtake, 1985; Mason et al., 2016; McFarquhar et al., 2011; Prenni et al., 2007, 2009; Radke et al., 1976; Rogers et al., 2001). Nevertheless, our understanding of the concentrations, composition, and sources of INPs in this region is incomplete. In the following, we focus on INPs in the Arctic during the spring. In the Arctic winter-spring, land is mostly covered by snow and hence local emissions of mineral dust and primary terrestrial biological particles are expected to be small. In addition, a large fraction of the Arctic Ocean is covered by ice, limiting emissions of sea spray aerosol. On the other hand, during the winter-spring, long-range transport of particles from mid-latitudes is important, and removal processes of aerosols are reduced, leading to elevated aerosol particle concentrations in the region, referred to as Arctic haze (Barrie, 1986; Barrie et al., 1981; Norman et al., 1999; Pacyna, 1995; Quinn et al., 2007; Shaw, 1995).

There have only been a small number of measurements of INPs in the Arctic during the spring (Borys, 1989; Creamean et al., 2018; Fountain and Ohtake, 1985; Mason et al., 2016; McFarquhar et al., 2011; Radke et al., 1976; Rogers et al., 2001). Of note, Borys (1989) measured INP concentrations in April 1986 and found that low concentrations of INPs were associated with tracers of pollution, while high concentrations of INPs were associated with a tracer of mineral dust. Fountain and Ohtake (1985) measured surface INP concentrations in Alaska from August 1978 to March 1979 and observed that high INP concentrations in March were correlated with air masses transported from Eurasia. Rogers et al. (2001) measured concentrations and the chemical composition of INPs during aircraft studies in May 1998. Chemical analysis of the INPs indicated that a significant fraction of the INPs (approximately 37 %) contained mineral dust. Mason et al. (2016) carried out size-resolved measurements of INPs from the end of March to late July 2014. They found that a large fraction (> 60% at -20 °C) of the INPs were larger than 1 µm, indicating supermicron particles such as mineral dust or sea spray aerosol containing biological material contributed to a majority of the INPs.

In the following, we investigate the composition and sources of INPs at Alert, Nunavut in the Canadian High Arctic from 11 to 29 March 2016. This study was carried out as part of the Network on Climate and Aerosols: Addressing Key Uncertainties in Remote Canadian Environments (NETCARE). Specifically, we measured daily the concentrations of INPs in the immersion freezing mode, and concentrations of tracers of mineral dust (Al, Fe, Ti, and Mn), sea spray aerosol (Na<sup>+</sup> and Cl<sup>-</sup>), and anthropogenic aerosol (Zn, Pb, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, and non-sea-salt SO<sub>4</sub><sup>2-</sup>). These data were used to determine if mineral dust, sea spray aerosol, and anthropogenic aerosol are a major contributor to the INP population at -15, -20, and -25 °C in the Canadian High Arctic during spring. Studies as a function of temperature are necessary since different types of aerosols are ice-active at different temperatures. Although a few studies have identified mineral dust particles as an important contributor to the INP population in the Arctic during spring, additional studies are needed to determine how often mineral dust is an

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important contributor. The measurements [reported here](#) together with particle dispersion modelling [were also used to assess the source of the INPs at a freezing temperature of -25 °C](#).

## 2 Methods

### 2.1 Sampling location

- 5 Sampling was conducted at the Dr. Neil Trivett Global Atmosphere Watch Observatory (82.5° N, 62.5° W) at Alert, Nunavut (Fig. 1), a research laboratory operated by Environment and Climate Change Canada. The research laboratory is on a plateau that is approximately 185 m above sea level and is powered by electricity generated by the Canadian Forces Station Alert, which is 6 km to the north of the research laboratory. The population at Alert is about 75 on a regular basis. The closest community is Grise Fiord (population 129) located approximately 800 km to the south of Alert.

### 10 2.2 INP measurements

Concentrations of INPs were determined by first collecting particles on hydrophobic glass slides with an inertia impactor followed by determining the freezing temperatures of the collected aerosol particles with the droplet freezing technique (DeMott et al., 2016; Irish et al., 2018). Details are given below.

#### 2.2.1 Inertia impactor

- 15 The inertia impactor (model 100-180nm-10lpm; MSP Corp., Shoreview, MN, USA) consisted of two impactor stages. The first impactor stage collected particles with aerodynamic diameters  $> 10 \mu\text{m}$ , and the second impactor stage collected particles with aerodynamic diameters between 0.18 and  $10 \mu\text{m}$ . [For each collection period, three circular hydrophobic glass slides \(HR3-277; Hampton Research, USA\) were placed on the second impactor stage simultaneously to collect particles for INP analysis. As a result, each collected sample consisted of three hydrophobic glass slides.](#) Due to the design of the impactor, the collected particles were concentrated into spots on the hydrophobic glass slides. Particles collected on the first stage were not analyzed. The hydrophobic glass slides were cleaned with Millipore water and dried with ultrapure nitrogen gas before being sent to the field for particle collection. After collection, all slides were placed in petri dishes, wrapped in aluminum foil, and stored at 4 °C before analysis. The inertia impactor was operated at a flow rate of  $10 \text{ L min}^{-1}$  and the average collection time for INP samples was approximately 2h.
- 20
- 25 The inertia impactor was located within the Dr. Neil Trivett Global Atmosphere Watch Observatory. Aerosol particles were first sampled through a louvered total suspended particulate (TSP) inlet (Mesa Labs Inc., Butler, NJ, USA) that was approximately 10 m above ground level. Next, the aerosol particles were passed through a humidifier (model FC125-240; Perma Pure LLC, Lakewood, NJ, USA) to condition the aerosol particles to an average relative humidity (RH) of 84 % at room temperature. Finally, the aerosol particles were passed through the inertia impactor to collect the particles on the hydrophobic glass slides for INP analysis. [In total, 16 daily measurements of INPs are reported in this study.](#)
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When sampling aerosol particles with an inertia impactor, a possible issue is particle rebound from the collection substrate. Rebound occurs when the kinetic energy of the particles striking the impactor substrate exceeds the adhesion and dissipation energies at impact (Bateman et al., 2014). If rebound is a factor, the measured INP concentrations will be lower than the actual INP concentrations in the atmosphere. Previous work has shown that particle rebound can be reduced when RH is above 70 % (Bateman et al., 2014; Chen et al., 2011; Fang et al., 1991), although this will depend on the chemical composition of the particles. In addition, field measurements of INP concentrations using a similar inertia impactor have shown good agreement with INP concentrations measured by a continuous flow diffusion chamber (a technique that is not susceptible to rebound) when the RH of the sampled aerosol stream was as low as 40 % (DeMott et al., 2017; Mason et al., 2015). In our experiments, the RH was increased to an average value of 84 % at room temperature with a humidifier to reduce rebound. However, rebound cannot be completely ruled out.

### 2.2.2 Droplet freezing technique

The INP number concentrations in the immersion freezing mode were determined with the droplet freezing technique (Iannone et al., 2011; Mason et al., 2015; Wheeler et al., 2015). Briefly, the hydrophobic glass slides used to collect aerosol particles with the inertia impactor were placed in a temperature- and humidity-controlled flow cell coupled to an optical microscope (Axiolab; Zeiss, Oberkochen, Germany) with a CCD camera. Typically between 15-25 spots of particles (out of 300 spots generated by the inertia impactor) were visible in the microscopic field of view. The diameter of each spot was approximately 110  $\mu\text{m}$ . After locating the hydrophobic glass slides in the flow cell, the temperature in the flow cell was decreased to approximately 0 °C and a humidified flow of helium with a dew point of approximately 3 °C was passed over the hydrophobic glass slide, resulting in water droplets with diameters of approximately 100-500  $\mu\text{m}$  condensing on the spots of particles as well as other areas on the glass slides. Droplets that condensed on the spots of particles are referred to as spot droplets, while droplets that condensed on other areas of the slide are referred to as non-spot droplets. After condensation of the water droplets, the flow cell was cooled down to -40 °C at a rate of -10 °C  $\text{min}^{-1}$  while images of the droplets were recorded. A small flow of dry helium ( $\sim 0.2 \text{ L min}^{-1}$ ) was passed through the flow cell during the cooling process to prevent further condensation of water vapour. The freezing temperature of each droplet was then determined from the images. The droplets that overlapped with spots of particles were also identified from these images. The number of INPs within the microscopic field of view was calculated as a function of temperature using the following equation:

$$\#INP(T) = \left( -\ln \left( \frac{N_{us}(T)}{N_s} \right) \right) N_s, \quad (1)$$

where  $N_{us}(T)$  is the number of unfrozen spot droplets at temperature  $T$ , and  $N_s$  is the total number of spots analyzed in the microscopic field of view. Equation (1) represents the cumulative nucleus spectrum or the concentrations of INPs active at all temperatures  $\geq T$ . The use of Eq. (1) to quantify the concentrations of INPs active at all temperatures  $\geq T$  from droplet freezing experiments has been justified using Poisson's law and Monte Carlo simulations (Vali, 1971).

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Equation (1) assumes that each droplet covered only one spot. However, sometimes more than one droplet formed on one spot. In these cases, the first droplet to freeze was considered in Eq. (1), which should give the equivalent result to one droplet condensing on one spot. Another situation is when one droplet covered two spots (this occurred for less than 5 % of the total analyzed spot droplets). For these cases, an upper limit of the number of INPs was calculated by assuming two droplets covered the two spots and both droplets froze at the observed freezing temperature. A lower limit was calculated by assuming two droplets covered the two spots with one droplet freezing at the observed freezing temperature, and the other freezing at  $-37^{\circ}\text{C}$  (approximately the homogeneous freezing temperature of the droplets). A similar approach was applied to cases where one droplet covered three or more spots.

Freezing of non-spot droplets was less frequent than freezing of spot droplets at temperatures  $\geq -25^{\circ}\text{C}$ . For example, the ratio of frozen non-spot droplets to frozen spot droplets was 0.2 at  $-25^{\circ}\text{C}$ . Freezing of non-spot droplets may have been due to INPs  $< 0.18\text{ }\mu\text{m}$  in diameter not focused into the spots or a small fraction of INPs  $\geq 0.18\text{ }\mu\text{m}$  not concentrated into the spots due to rebound from the hydrophobic glass slides. To take into account the INPs in the non-spot droplets, we assumed each frozen non-spot droplet contained one INP, and the number of frozen non-spot droplets,  $N_{ns2}$ , was added to Eq. (1), resulting in the following equation:

$$\#INP(T) = \left( -\ln\left(\frac{N_{us}(T)}{N_s}\right) \right) N_s + N_{ns2} \quad (2)$$

During the freezing experiments, most freezing events occurred by immersion freezing, while some occurred by contact freezing, which refers to the freezing of liquid droplets coming into contact with neighboring frozen droplets. Contact freezing only accounted for approximately 2 % of the total freezing events, and droplets that froze by contact freezing were not considered when determining the INP concentrations.

The INP concentration as a function of temperature in the atmosphere,  $[INPs(T)]$ , was calculated using the following equation:

$$[INPs(T)] = \#INP(T) \left( \frac{300}{N_s V} \right) f_{ne} \quad (3)$$

where 300 is the number of nozzles in the nozzle plate of the impactor, and consequently, the number of spots of particles generated on the second stage of the impactor,  $V$  is the total volume of air sampled by the impactor, and  $f_{ne}$  represents the uncertainty associated with the limited number of nucleation events detected and is based on nucleation statistics (Koop et al., 1997). In Eq. (3), the ratio of  $300/N_s$  accounts for the fact that only a fraction of total spots was analyzed with the droplet freezing technique. The overall uncertainty in the concentrations of INPs reported here includes the uncertainty from the limited number of nucleation events, as well as the other uncertainties discussed above.

Droplet freezing experiments were also performed on glass slides that were not exposed to any particles. These slides are referred to as blanks. One field blank was collected by treating the slides in the same manner as the sample hydrophobic slides including locating the hydrophobic slides in the impactor, except that the pump was not turned on. Lab blanks refer to slides cleaned in the same manner as the ones used for sampling in the field but not sent to the field. For the blanks, we

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assumed the  $\#INP(T)$  was equal to the number of observed freezing events since multiple INPs within the same droplet was unlikely at temperatures  $\geq -25$  °C.  $[INPs(T)]$  was then calculated using Eq. (3) with the assumptions that  $N_s$  is 21 (average number of spots analyzed in one experiment) and  $V$  is 1208 L (average air volume sampled).

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### 2.3 Meteorological parameters

- 5 Local ground-level meteorological conditions were monitored at the site by Environment and Climate Change Canada. The March 2016 data was retrieved from climate.weather.gc.ca (climate IDs 2400305 and 2400306). The ambient temperature, ambient RH, wind speed, and wind direction were measured hourly. The sum of the total rainfall and the water equivalent of the total snowfall in millimeters were measured daily as total precipitation.

### 2.4 Tracers of mineral dust, sea spray, and anthropogenic aerosol

- 10 The elements Al, Fe, Ti, and Mn were used as tracers of mineral dust, as done previously (Balasubramanian et al., 2003; Barrie and Barrie, 1990; Formenti et al., 2003; Malm et al., 1994; Quinn et al., 2004). These elements are components of the Earth's crust (Usher et al., 2003; Wedepohl, 1995). The species  $\text{Na}^+$  and  $\text{Cl}^-$ , which are the major inorganic components of seawater (Holland, 1978), were used as tracers of sea spray, as done previously (Balasubramanian et al., 2003; Malm et al., 1994; Quinn et al., 2002, 2004). For tracers of anthropogenic aerosol, we used Zn, Pb,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and non-sea-salt  $\text{SO}_4^{2-}$  (nss- $\text{SO}_4^{2-}$ ). Pb and Zn are almost exclusively from anthropogenic sources (Macdonald et al., 2000; Nriagu and Pacyna, 1988; Pacyna, 1995). The major anthropogenic sources for Pb are gasoline combustion and, to a lesser extent, non-ferrous metal industry and fossil fuel combustion, and the major anthropogenic sources for Zn are non-ferrous metal industry followed by fossil fuel combustion (Barrie et al., 1992; Nriagu and Pacyna, 1988; Pacyna, 1995).  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and nss- $\text{SO}_4^{2-}$  can come from both anthropogenic and natural sources, but mostly from anthropogenic sources.  $\text{NO}_3^-$  is mainly formed from  $\text{NO}_x$ , which is emitted from combustion processes (Seinfeld and Pandis, 2006).  $\text{NH}_4^+$  originates mainly from agricultural activities (Follett and Hatfield, 2001). The main anthropogenic source of nss- $\text{SO}_4^{2-}$  is fossil fuel combustion (Schwikowski et al., 1999; Ward, 2009).

- To determine the concentrations of the tracers discussed above, aerosol particulate samples were collected on 20 x 25 cm Whatman-41 quartz filters daily using a high-volume sampler (Barrie et al., 1981, 1989), which was located approximately 500 m away from the laboratory on the ground. The face velocity of sampling ( $50 \text{ cm s}^{-1}$ ) and typical filter loadings ensured collection efficiencies better than 95 % (Watts et al., 1987). The average sampling time was 23.5h except for the first sample, which was collected over two days due to a storm making it difficult to change the filter. Hence, collection times for tracer measurements (~ 24h) were different than the collection times for INP measurements (~ 2h). The implications of the different collection times are discussed in Sect. 3.3. For the filter samples, the average total air volume sampled was roughly 2,300  $\text{m}^3$  at standard conditions of 1 atm pressure and 0 °C. The precision of volume sampled was estimated to be  $\pm 5$  % (Sirois and Barrie, 1999). The quartz filter samples were stored at room temperature before analysis.

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The concentrations of Al, Fe, Ti, Mn, Zn, and Pb were determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES). These experiments were carried out at Chester LabNet, Oregon, USA. Punches from the quartz filters were submerged in a solution containing ultrapure HNO<sub>3</sub> and HCl. The solution was then heated and sonicated, and then further diluted and filtered before being nebulized and analyzed by the ICP-AES (Perkin-Elmer Optima 8300). Sample  
5 duplicates were also analyzed to estimate the method precision. The accuracy and precision of the technique were estimated to be ± 10 %.

The concentrations of Cl<sup>-</sup>, Na<sup>+</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and NH<sub>4</sub><sup>+</sup> were determined using ion chromatography (IC) (Macdonald et al., 2017; Toom-Sauntry and Barrie, 2002). These experiments were carried out at Environment and Climate Change Canada in Ontario, Canada. To quantify water-soluble cations and anions, punches taken from each quartz filter were extracted in  
10 deionized water, and the extraction solution was passed through an IC (Dionex IC: DX600). The concentration of nss-SO<sub>4</sub><sup>2-</sup> was calculated using the following equation based on the assumption that the chemical composition of sea salt particles is the same as that of seawater (Millero, 1974):

$$[nss - SO_4^{2-}] = [SO_4^{2-}] - 0.14[Cl^-], \quad (4)$$

To determine whether the calculated concentrations of nss-SO<sub>4</sub><sup>2-</sup> was sensitive to depletion of Cl<sup>-</sup> in the particles, we also  
15 calculated nss-SO<sub>4</sub><sup>2-</sup> using Na<sup>+</sup> rather than Cl<sup>-</sup> (Balasubramanian, 2003). The difference between the concentrations of nss-SO<sub>4</sub><sup>2-</sup> based on Cl<sup>-</sup> and the concentrations based on Na<sup>+</sup> was less than 5 %.

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## 2.5 Particle dispersion modelling

The source regions of measured air masses were investigated using the Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005), which was driven using operational meteorological analyses from the European Center for Medium-  
20 Range Weather Forecasts (ECMWF). FLEXPART was run at hourly intervals in the backward mode for each sample collected for INP analysis. In each run, 40,000 particles were released over 1 minute in a 0.1 deg x 0.1 deg area. The particles were followed backward for 20 days with output generated at 1-hour intervals. In backward mode FLEXPART provides potential emission sensitivities as output. This is the response function of a source-receptor matrix (Seibert and Frank, 2004) and corresponds directly to the residence time of the released particles in a given volume of air. For each run,  
25 the hourly output was integrated over the 20 days to produce a potential emission sensitivity (PES) plot. For a given INP sample, the mean PES plot was generated by averaging all hourly PES plots. Since the focus of this study are INP sources close to the surface, near surface PES plots (from the surface up to 100 m) were plotted as footprint PES plots.

## 2.6 Statistical analysis

To compute a correlation coefficient (*R*), Pearson correlation analysis was applied between INPs and the variables measured  
30 in this study. *P*-values were also calculated using t-test for Student's t-distribution to determine if the correlations were statistically significant at the 95 % confidence level (*P* < 0.05).

### 3 Results and discussion

#### 3.1 Concentrations of INPs

Concentrations of INPs in the immersion mode are plotted as a function of temperature in Fig. 2. Concentrations of INPs measured were higher than concentrations of the blanks. In addition, all of the samples have warmer onset temperatures than the blanks, with onset temperatures of the samples varying from approximately -11 to -22 °C. For the remainder of this document, we focus on INP concentrations at -15, -20, and -25 °C. INP concentrations at temperatures warmer than -15 °C are not discussed because freezing events at these temperatures were rare. INP concentrations at temperatures below -25 °C are not discussed since freezing of the blanks became significant at these temperatures. Since freezing was rarely observed at  $\geq -25$  °C in the blank experiments, the INP concentrations determined in the blank experiments were not subtracted from the INP concentrations reported in this study.

In the current study, the mean number concentration of INPs was  $0.005 \pm 0.002 \text{ L}^{-1}$  at -15 °C,  $0.020 \pm 0.004 \text{ L}^{-1}$  at -20 °C, and  $0.186 \pm 0.040 \text{ L}^{-1}$  at -25 °C (Fig. 3). These concentrations are within the range of INP concentrations measured in previous studies at ground level or sea level in the Arctic (Fig. 3). The sampling platform, location, and dates of previous Arctic INP studies at ground level or sea level that are shown in Fig. 3 are summarized in Table 1 and Fig. 1 for comparison purposes.

The time series of INP concentrations measured in the current study at -15, -20, and -25 °C is plotted in Fig. 4. The difference between the highest INP concentration at -25 °C (measured on March 28) and the lowest INP concentration at -25 °C (measured on March 17) was roughly a factor of 50.

#### 3.2 Meteorological parameters and correlations with INPs

Shown in Fig. 5 is the time series of meteorological parameters measured at Alert during the field campaign. Precipitation was rare throughout the campaign, and the average ambient temperature was approximately -30 °C. The RH was above 70 % for most of the time. The wind speed was below  $10 \text{ km h}^{-1}$  except for two storm events during the first part of the campaign. During the field campaign, the wind came mainly from the SW (more than half of the time) with some contribution from the SE and NW.

In the current study, INP concentrations at freezing temperatures of -15, -20, and -25 °C were not correlated with any meteorological parameters (Table S1). This result is consistent with the results from Fountain and Ohtake (1985), who also did not find any correlations between meteorological variables (including air temperature, precipitation, fog, and wind direction, etc.) and INP concentrations measured at Barrow, Alaska from August 1978 to April 1979. In contrast, measurements by Radke et al. (1976) in Barrow, Alaska during March found that the INP concentrations were affected by local weather conditions.

### 3.3 Tracers of mineral dust: concentrations, correlations with INPs, and sources

The time series of concentrations of mineral dust tracers (Al, Fe, Ti, and Mn) is plotted in Fig. 6. Figure 6 shows that concentrations of different tracers of mineral dust are correlated with each other. Shown in Table 2 is a correlation coefficient matrix between each of the tracers based on a Pearson correlation analysis. Correlations that are both strong ( $R \geq 0.7$ ) and statistically significant ( $P < 0.05$ ) are highlighted in bold and red. The correlation coefficients show that the mineral dust tracers are strongly correlated with each other ( $R \geq 0.85$ ), which is expected since these elements are almost exclusively from the mineral dust sources.

In Fig. 7(a), we compared the mean mass concentration of aluminum we measured at the site during the campaign with the concentrations reported in Sirois and Barrie (1999) for the same site and for time period from 1980 through 1995. The mean mass concentration of aluminum measured during the current study was lower than the previous concentrations measured in March at Alert, indicating that the concentration of mineral dust during the current campaign was lower than many of the previous measurements at Alert during March.

Pearson regression analysis was also done between INP concentrations at -15, -20 and -25 °C, and tracers of mineral dust. The correlation coefficients and  $P$ -values are summarized in Table 3. The linear regression plots between INP concentrations at -25 °C and mineral dust tracers are shown in Fig. 8. When calculating the correlation coefficients, a value of zero was used when tracer concentrations and INP concentrations were below the detection limits. It is important to keep in mind that the collection time was different for INPs (~2h) and the aerosol tracers (~24h), and that the analysis presented here is based on the assumption that the average concentrations of the aerosol tracers during the INP sampling time were the same as the average concentrations determined from the quartz filters. If this assumption is not correct, correlation coefficients between

INPs and aerosol tracers will be smaller than expected. Shown in Fig. S1 are air mass back trajectories initiated every 2h during each 24h quartz filter sampling period. The back trajectories suggest that for a large fraction of the samples, the source of the air masses during INP sampling was similar to the source of the air masses measured during quartz filter sampling.

As shown in Table 3, at -15 °C, the correlations between INP concentrations and tracers of mineral dust were not statistically significant ( $P > 0.05$ ). At -20 °C, the correlations between INPs and Ti and Mn were not statistically significant. Al and Fe were moderately correlated with INPs at -20 °C ( $R = 0.53$  and  $0.60$ , respectively), and the correlations were statistically significant ( $P < 0.05$ ). At -25 °C, all four tracers were strongly correlated with INPs ( $R$  ranged from  $0.70$  to  $0.76$ ), and the correlations were statistically significant. This suggests that mineral dust was a component of the sampled INPs at a freezing temperature of -25 °C. This is consistent with previous field measurements that have identified mineral dust as a major component of the INP population at different locations (Boose et al., 2016; Cziczo et al., 2013; DeMott et al., 2003; Klein et al., 2010; Pratt et al., 2009; Prenni et al., 2009). A previous study of ice nucleation of Arctic aerosol found that the elements of crustal or natural dust were associated with high concentrations of INPs at -15 °C and -25 °C (Borys, 1989). Chemical analysis of the INPs during aircraft studies in the Arctic indicated that a significant fraction of the INPs (approximately 37 %)

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contained mineral dust (Rogers et al., 2001). Another study of snow crystals during summer on the Greenland Ice Cap also suggested that the natural snow crystals mainly formed on clay mineral particles by heterogeneous nucleation (Kumai and Francis, 1962).

Mineral dust at Alert can come from both local sources and long-range transport, but during the spring local sources are not likely important since land is covered with snow during this time of the year. To investigate the origins of mineral dust measured in this study, the FLEXPART model was used to generate the footprint PES plots, which show the residence time of aerosol particles in the layer from 0 to 100 m in altitude during the 20 days prior to sampling. Results are shown in Fig. 9. Figure 9(A) shows the surface coverage type on the first day of sampling (March 12). The sampling location at Alert was surrounded by ice and snow during the campaign. Figure 9(B) shows the average footprint PES plot for all samples, and Fig. 9(C) shows the average footprint PES plot for the four samples with the highest mineral dust concentrations, which were collected on March 21, 22, 27, and 28. Note, since average footprint PES plots are shown in Fig. 9(B) and 9(C), Fig. 9(C) is not a subset of Fig. 9(B). Shown in Fig. 9(D) is a ratio plot of the sum of footprint PES plots of the four samples with the highest mineral dust concentrations to the sum of footprint PES plots of all samples. These types of ratio plots are often used as a sensitive method to identify the source regions of a component under investigation (Hirdman et al., 2010). In these types of plots, a value close to one indicates a more likely source region. Figure 9(D) suggests that the North Pacific Ocean, Alaska, and the Gobi Desert were possible source regions of the mineral dust sampled. Alaska was covered in snow during the sampling period, so this was not likely the source of the mineral dust, although mineral dust re-suspended from blowing snow cannot be ruled out. Mineral dust from shorelines also cannot be ruled out. The North Pacific Ocean is not likely the source of mineral dust either since the Pacific Ocean is not considered as a traditional source of mineral dust. Based on the surface coverage types (Fig. 9(A)) and the ratio plot (Fig. 9(D)), a possible source of the mineral dust was long-range transport of dust from the Gobi desert. This conclusion is consistent with previous studies that have shown that a substantial fraction of the dust reaching Alert in spring months comes from long-range transport of Asian dust (Drab et al., 2002; Franzén et al., 1994; Pacyna and Ottar, 1989; Sirois and Barrie, 1999; Welch et al., 1991).

### 3.4 Tracers of sea spray aerosol: concentrations and correlations with INPs

The time series of sea spray aerosol tracers ( $\text{Na}^+$  and  $\text{Cl}^-$ ) is shown in Fig. 10, and the results of the Pearson correlation analysis between these tracers are listed in Table 2. The correlation coefficients show that sea spray tracers are strongly correlated with each other ( $R = 0.95$ ), which is expected since these species are almost exclusively from sea spray.

A comparison between the mean mass concentrations of  $\text{Na}^+$  and  $\text{Cl}^-$  measured in the current study and the values measured by Sirois and Barrie (1999) from 1980 to 1995 is shown in Fig. 7(b-c). Figure 7(b-c) illustrates that the concentrations of  $\text{Na}^+$  and  $\text{Cl}^-$  measured in the current study were consistent with many of previous measurements at Alert during March.

The correlation coefficients between INP concentrations at -15, -20, and -25 °C, and tracers of sea spray aerosol are listed in Table 3. The liner regression plots between INP concentrations at -25 °C and the sea spray tracers are shown in Fig. 11. At -15 °C and -20 °C, the correlations between INP concentrations and tracers of sea spray aerosol were not statistically

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significant ( $P > 0.05$ ). At  $-25\text{ }^{\circ}\text{C}$ , the sea spray tracers were negatively correlated with INPs with moderate correlation coefficients ( $R = -0.56$ ) and statistical significance ( $P < 0.05$ ). Previous field studies and modelling studies have suggested sea spray aerosol as an important source of ambient INPs in marine environments when other sources of INPs, such as mineral dust, are low (Burrows et al., 2013; DeMott et al., 2016; Rosinski et al., 1986, 1988; Schnell, 1982; Vergara-Temprado et al., 2017; Wilson et al., 2015). Our results suggest that mineral dust is a more important source of INPs at  $-25\text{ }^{\circ}\text{C}$  than sea spray aerosol for the time and location studied.

Due to the method of particle collection and the method of forming liquid droplets in the freezing experiments, soluble material, such as salts including NaCl, were mixed with mineral dust particles within the same droplets during the freezing experiments. Salts decrease the freezing temperature of droplets by decreasing the water activity in the solution. However, the decrease in freezing temperature by this mechanism is not expected to be important in our experiments since the concentration of salts within a droplet is small. For example, the maximum concentration of NaCl in the water droplets formed during the droplet freezing experiments was estimated to be  $\sim 0.03\text{ M}$  based on the mass concentrations of  $\text{Na}^+$  and  $\text{Cl}^-$  sampled. This concentration of NaCl in the droplets would only cause a freezing point depression of  $\sim 0.1\text{ }^{\circ}\text{C}$ , which is within the uncertainty of the measured freezing temperatures and is too small to explain the negative correlation between INP concentrations and tracers of sea spray aerosol shown in Fig. 11.

On the other hand, other studies have illustrated that trace amounts of NaCl can lower the freezing temperature of mineral dust by surface specific interactions between NaCl and the mineral dust surface, and the decrease in freezing temperature by this mechanism is more than expected based on the traditional freezing point depression mechanism (Reischel and Vali, 1975; Whale et al., 2018). For example, Reischel and Vali (1975) studied the effects of 0.01 M, 0.1 M and 1 M solutions of NaCl on the nucleating ability of kaolin, and found that the presence of NaCl led to lower freezing temperatures, by as much as  $4\text{ }^{\circ}\text{C}$ , for kaolin. A recent study by Whale et al. (2018) also found that a 0.015 M NaCl solution caused a decrease in freezing temperature of approximately 2 to  $4\text{ }^{\circ}\text{C}$  for BCS376 microcline, Eifel sanidine, quartz and Arizona test dust, but had no effect on silica. To investigate the influence of NaCl on the ice-nucleating ability of mineral dust at  $-25\text{ }^{\circ}\text{C}$  in the current study, INP concentrations at a freezing temperature of  $-25\text{ }^{\circ}\text{C}$  ( $[\text{INPs}]_{-25^{\circ}\text{C}}$ ) was divided by the mass concentration of aluminum ( $[\text{Al}]$ ), and this ratio was then plotted as a function of  $\text{Cl}^-$  and  $\text{Na}^+$  (Fig. 12). The variable  $[\text{INPs}]_{-25^{\circ}\text{C}}/[\text{Al}]$  is used as an estimation of the ice-nucleating ability of mineral dust, and it was negatively correlated with  $\text{Cl}^-$  and  $\text{Na}^+$  with moderate correlation coefficients ( $R = -0.51$  and  $-0.55$ , respectively) and statistical significance ( $P < 0.05$ ), which suggests that NaCl suppressed the ice-nucleating ability of mineral dust in the current study. This result is consistent with the two previous laboratory studies mentioned above and is the first field study that we are aware of that suggests that NaCl can suppress the ice-nucleating ability of mineral dust particles in the immersion freezing mode even in dilute solution droplets.

Another possible explanation for the negative correlation between INP concentrations at  $-25\text{ }^{\circ}\text{C}$  and sea spray tracers is that the air masses containing sea spray aerosol have relatively few mineral dust particles. In this case, the sea spray aerosol does not influence the ice-nucleating ability of mineral dust, rather appears as a negative correlation by coincidence. However, Table 2 shows that the correlations between tracers of mineral dust and tracers of sea spray aerosol are not statistically

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significant, suggesting that this possible explanation is not the major reason for the negative correlation between INP concentrations at -25 °C and sea spray tracers.

### 3.5 Tracers of anthropogenic aerosol: concentrations and correlations with INPs

The time series of anthropogenic aerosol tracers ( $\text{Zn}$ ,  $\text{Pb}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{nss-SO}_4^{2-}$ ) is plotted in Fig. 13. The results of the Pearson correlation analysis between these different tracers are listed in Table 2. Not all the anthropogenic aerosol tracers are correlated with each other. This is not surprising since these tracers come from different sources. However, there is a correlation between  $\text{Zn}$  and  $\text{Pb}$ , and between  $\text{nss-SO}_4^{2-}$  and  $\text{NH}_4^+$ .  $\text{Zn}$  and  $\text{Pb}$  are strongly correlated with each other ( $R = 0.84$ ,  $P < 0.01$ ), and  $\text{nss-SO}_4^{2-}$  and  $\text{NH}_4^+$  are strongly correlated with each other ( $R = 0.93$ ,  $P < 0.01$ ).

The comparison between the mean mass concentrations of  $\text{Zn}$ ,  $\text{Pb}$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{NH}_4^+$  measured in the current study and the values measured by Sirois and Barrie (1999) from 1980 to 1995 is shown in Fig. 7(d-h). The concentrations of  $\text{Zn}$ ,  $\text{Pb}$ ,  $\text{SO}_4^{2-}$ , and  $\text{NH}_4^+$  were lower than previous concentrations measured in March at Alert, which might be due to the reduced emissions from Eurasia after the dissolution of the former USSR in 1991 (Christensen, 1997). The concentration of  $\text{NO}_3^-$  was higher than previous concentrations measured in March. An increasing trend of annual concentrations of  $\text{NO}_3^-$  has been observed by others in the Arctic (Hole et al., 2006; Neftel et al., 1985).

The correlation coefficients between INP concentrations at -15, -20, and -25 °C, and tracers of anthropogenic aerosol are listed in Table 3. None of the correlations were statistically significant, consistent with a recent study that showed no effect of atmospheric pollution on INP concentrations in Beijing at freezing temperatures down to -25 °C (Chen et al., 2018). In contrast, Borys (1989) found that pollution derived Arctic haze aerosol had lower INP concentrations than unpolluted troposphere aerosol.

There have been a few studies that investigated the effect of sulfate coating on the ice-nucleating properties of mineral dust particles. Most laboratory studies have shown decreased freezing abilities for sulfate coated mineral dust particles compared to uncoated particles (Chernoff and Bertram, 2010; Eastwood et al., 2008; Gallavardin et al., 2008; Sullivan et al., 2010). However, other studies have shown an increase in the freezing temperature of droplets containing mineral dust particles in the presence of ammonium sulfate (Reischel and Vali, 1975; Whale et al., 2018). To investigate the effect of ammonium, sulfate, and the ammonium-to-sulfate ratio on the ice-nucleating ability of mineral dust at -25 °C, the parameter  $[\text{INPs}]_{-25^\circ\text{C}}/[\text{Al}]$  was plotted as a function of  $\text{NH}_4^+$ ,  $\text{nss-SO}_4^{2-}$ , and  $\text{NH}_4^+/\text{nss-SO}_4^{2-}$  ratio as shown in Fig. 14. The  $\text{NH}_4^+/\text{nss-SO}_4^{2-}$  ratio was between 0 and 1. The correlations between the ice-nucleating ability of mineral dust at -25 °C and  $\text{NH}_4^+$ ,  $\text{nss-SO}_4^{2-}$ , and  $\text{NH}_4^+/\text{nss-SO}_4^{2-}$  ratio were not statistically significant.

### 4 Conclusions

The INP concentrations measured at Alert during March 2016 fell into the range of previously reported INP concentrations measured in the Arctic at ground level or sea level. At -25 °C, the INP concentration was strongly correlated with tracers of

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mineral dust (Al, Fe, Ti, and Mn), anti-correlated with tracers of sea spray ( $\text{Cl}^-$ ,  $\text{Na}^+$ ), and not correlated with tracers of anthropogenic aerosol ( $\text{Zn}$ ,  $\text{Pb}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and  $\text{nss-SO}_4^{2-}$ ) or meteorological variables. This suggests that mineral dust was a major contributor to INP populations at  $-25^\circ\text{C}$  at this site during the sampling period. The ice-nucleating ability of mineral dust, represented as the ratio of INP number concentration to the mass concentration of aluminum, was also anti-correlated with the tracers of sea spray at  $-25^\circ\text{C}$ , which suggests that NaCl suppressed the ice-nucleating ability of mineral dust particles in the immersion freezing mode. This is the first field study that we are aware of that showed the suppression effect of NaCl on the ice-nucleating ability of mineral dust. Due to the way particles were collected and droplets were formed in our experiments, soluble material, such as NaCl, was internally mixed with mineral dust within the same droplets. However, in the atmosphere, the soluble material, such as NaCl, may not be internally mixed with mineral dust. Studies of the mixing state of soluble material with mineral dust at Alert during the same time of the year are needed.

The particle dispersion model analysis suggests that a likely source of mineral dust that caused freezing at  $-25^\circ\text{C}$  was long-range transport of dust from the Gobi desert. Additional measurements of the composition of individual INP or ice crystal residuals in the Arctic are needed to confirm the conclusions reached in the current study.

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**Deleted:** The results from this study should be useful for testing and improving models used to predict INPs and climate in the Arctic.

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**Table 1: Measurements of the concentrations of INPs at ground level or sea level in the Arctic. Included is the information on the sampling platform (ground vs. ship based), location, and dates.**

Study	Platform	Location	Dates
Radke et al. (1976)	Ground	Barrow, Alaska, USA	Mar 1970
Flyger & Heidam (1978)	Ground	North Greenland	Jun-Aug 1974
Fountain & Ohtake (1985)	Ground	Barrow, Alaska, USA	Aug 1978-Apr 1979
Bigg (1996)	Ship	Central Arctic Ocean	Aug-Oct 1991
Bigg & Leck (2001)	Ship	Arctic Ocean	Jul-Sep 1996
DeMott et al., (2016)	Ship	Canadian Arctic	Jul 2014
Mason et al. (2016)	Ground	Alert, Nunavut, Canada	Mar-Jul 2014
Conen et al. (2016)	Ground	Northern Norway	Jul 2015
Creamean et al. (2018)	Ground	Alaska oilfields, USA	Mar-May 2017
Current study	Ground	Alert, Nunavut, Canada	Mar 2016

Table 2. The correlation coefficient ( $R$ ) matrix between each of the measured aerosol constituents. Correlations that are statistically significant at the 95 % confidence level ( $P < 0.05$ ) are highlighted in red. Strong correlations ( $R \geq 0.7$ ) are highlighted in bold.

	Al	Fe	Ti	Mn	Cl <sup>-</sup>	Na <sup>+</sup>	Zn	Pb	NO <sub>3</sub> <sup>-</sup>	NH <sub>4</sub> <sup>+</sup>
Fe	<b>0.97</b>									
Ti	<b>0.91</b>	<b>0.85</b>								
Mn	<b>0.90</b>	<b>0.90</b>	<b>0.95</b>							
Cl	-0.23	-0.33	-0.29	-0.42						
Na <sup>+</sup>	-0.15	-0.25	-0.13	-0.25	<b>0.95</b>					
Zn	0.38	0.49	0.23	0.38	-0.18	-0.15				
Pb	0.46	<b>0.59</b>	0.33	<b>0.53</b>	-0.39	-0.32	<b>0.84</b>			
NO <sub>3</sub> <sup>-</sup>	<b>0.54</b>	0.37	<b>0.65</b>	0.47	0.10	0.27	-0.11	-0.18		
NH <sub>4</sub> <sup>+</sup>	<b>0.60</b>	<b>0.56</b>	<b>0.82</b>	<b>0.83</b>	-0.30	-0.04	0.17	0.30	<b>0.59</b>	
nss-SO <sub>4</sub> <sup>2-</sup>	0.41	0.44	<b>0.65</b>	<b>0.74</b>	-0.46	-0.20	0.20	0.39	0.33	<b>0.93</b>

Table 3. Results of Pearson correlation analysis between the INP number concentrations (at freezing temperatures of -15, -20, and -25 °C) and tracers of aerosol components measured in this study.  $R$  is the correlation coefficient,  $P$  is the probability value (two tailed), and the sample number is 16. Correlations that are statistically significant at the 95% confidence interval ( $P < 0.05$ ) are highlighted in red. Strong correlations ( $R \geq 0.7$ ) are highlighted in bold.

	INP number concentrations (L <sup>-1</sup> )					
	-15 °C		-20 °C		-25 °C	
	$\underline{R}$	$\underline{P}$	$\underline{R}$	$\underline{P}$	$\underline{R}$	$\underline{P}$
<b>Mineral dust tracers</b>						
Al (ng/m <sup>3</sup> )	0.29	0.27	<b>0.53</b>	0.03	<b>0.72</b>	<0.01
Fe (ng/m <sup>3</sup> )	0.25	0.36	<b>0.60</b>	0.01	<b>0.76</b>	<0.01
Ti (ng/m <sup>3</sup> )	0.05	0.85	0.29	0.28	<b>0.70</b>	<0.01
Mn (ng/m <sup>3</sup> )	0.06	0.83	0.32	0.22	<b>0.74</b>	<0.01
<b>Sea spray tracers</b>						
Cl <sup>-</sup> (ng/m <sup>3</sup> )	0.48	0.06	-0.01	0.97	<b>-0.56</b>	0.02
Na <sup>+</sup> (ng/m <sup>3</sup> )	0.49	0.06	-0.05	0.86	<b>-0.56</b>	0.02
<b>Anthropogenic aerosol tracers</b>						
Zn (ng/m <sup>3</sup> )	0.07	0.81	0.09	0.75	0.18	0.51
Pb (ng/m <sup>3</sup> )	0.09	0.74	0.24	0.37	0.30	0.26
NH <sub>4</sub> <sup>+</sup> (ng/m <sup>3</sup> )	-0.02	0.93	0.01	0.96	0.42	0.10
nss-SO <sub>4</sub> <sup>2-</sup> (ng/m <sup>3</sup> )	-0.19	0.47	-0.08	0.77	0.38	0.15
NO <sub>3</sub> <sup>-</sup> (ng/m <sup>3</sup> )	0.24	0.37	0.02	0.95	0.22	0.41

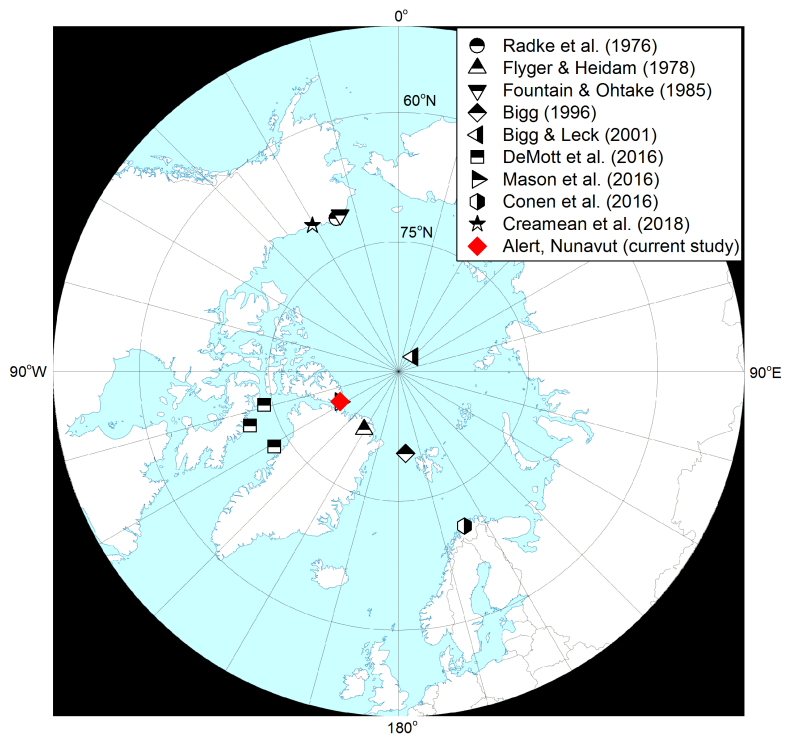


Figure 1. The locations of previous ground-based and ship-based INP studies in the Arctic. For Bigg (1996) and Bigg & Leck (2001), samples were collected along the ship track, but only one location for each study is shown in this map. The red diamond represents the location of current study.

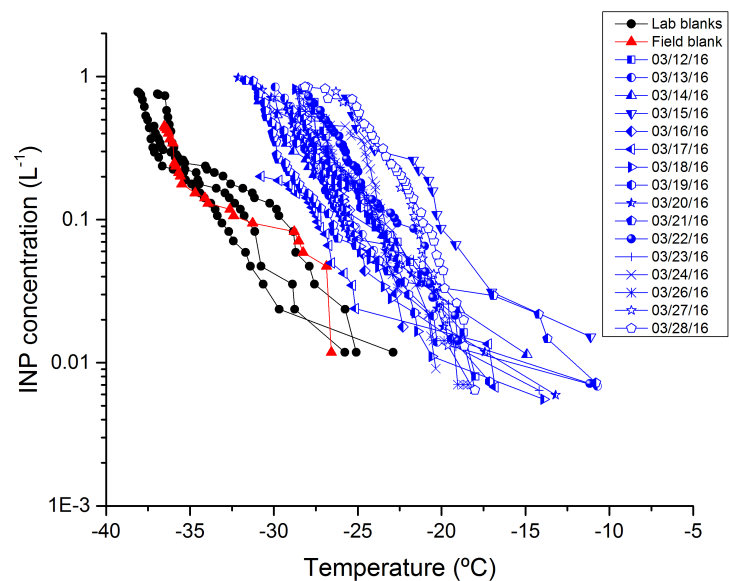


Figure 2. The INP concentrations from each sample plotted as a function of temperature. Black represents lab blanks. Red represents the field blank, which was collected by putting the glass slides briefly in the inertia impactor without turning on the pump. Blue represents samples, and the legend gives the collection date of each sample.

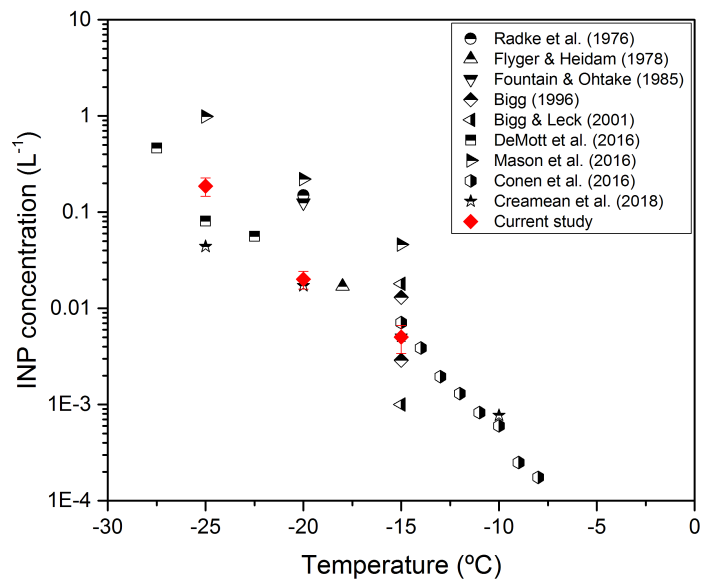


Figure 3. INP concentrations measured at ground level or sea level in the Arctic. Black represents previous ground-based and ship-based studies, and red represents the current study. The error bars on current study data points represent the standard error of the mean.

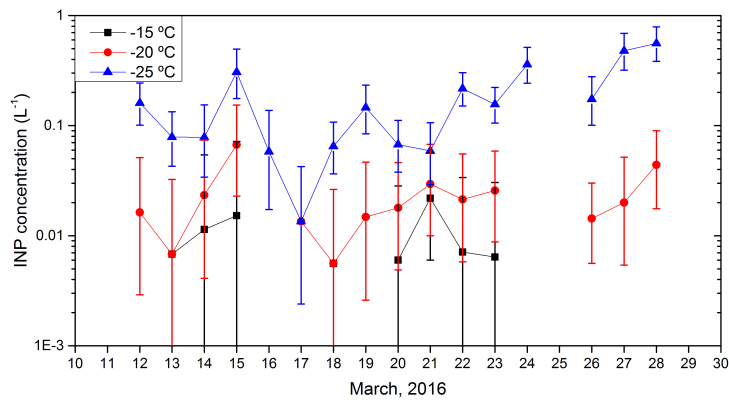


Figure 4. The time series of INP concentrations at -15 °C, -20 °C, and -25 °C. INP samples were not collected on March 25 due to time constraint. On several days, INP concentrations are shown at -25 °C, but not at -15 °C and some cases not at -20 °C. In these cases, INP concentration was below the detection limit, and hence not shown in this figure. The uncertainties in INP concentrations were calculated as described in Sect. 2.2.2.

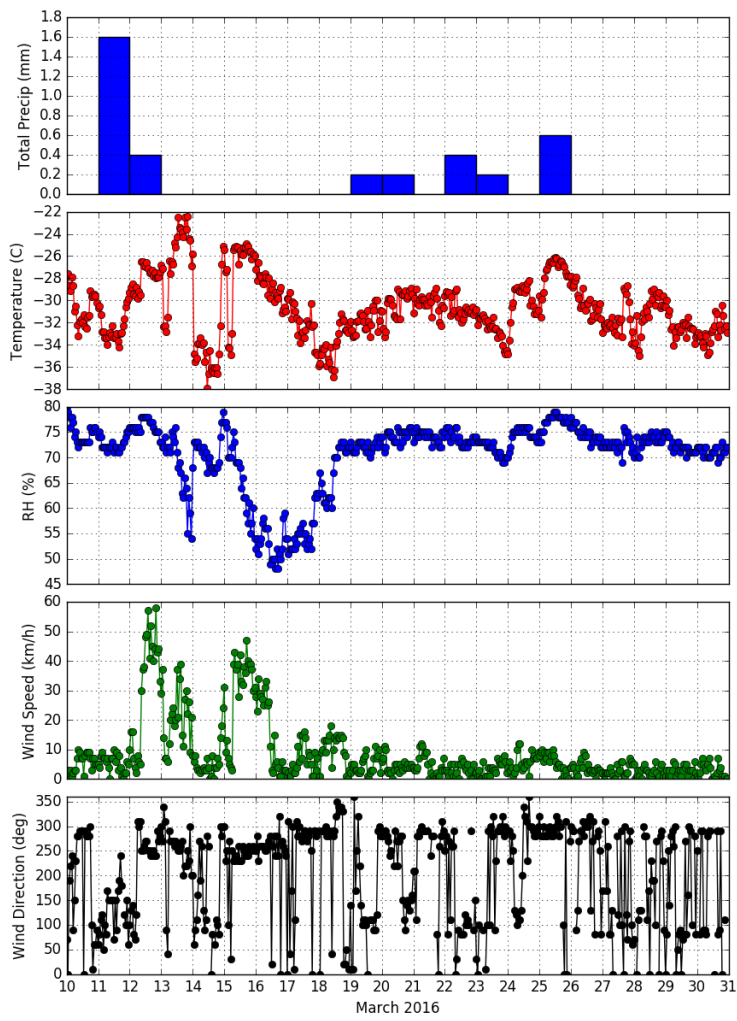


Figure 5. The time series of meteorological parameters, including total precipitation, ambient temperature, ambient relative humidity (RH), wind speed, and wind direction. The meteorological data were retrieved from [climate.weather.gc.ca](http://climate.weather.gc.ca). For wind direction, a value of zero denotes a calm wind, and a value of 360 denotes a wind blowing from the geographic North Pole.

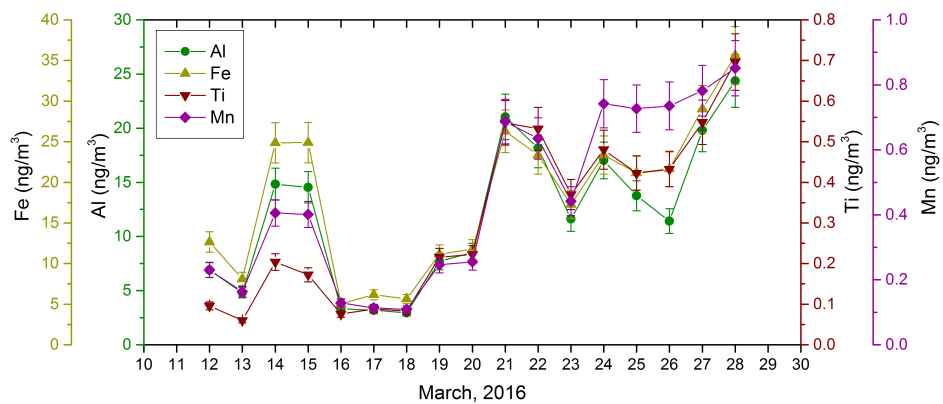


Figure 6. The time series of mineral dust tracers (Al, Fe, Ti, and Mn).

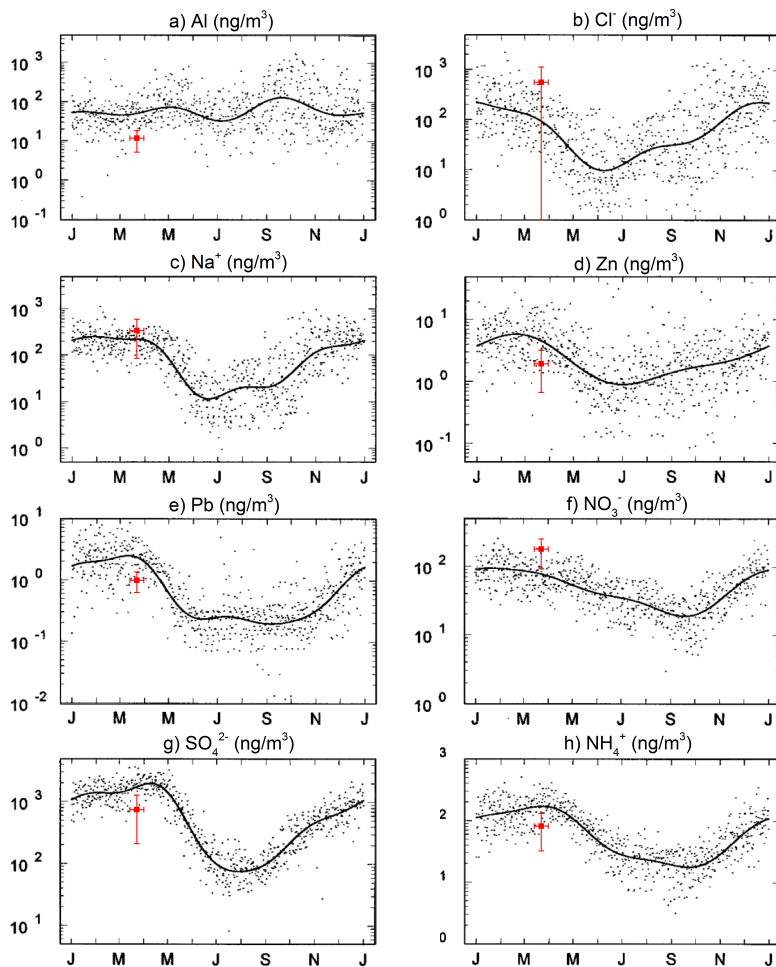
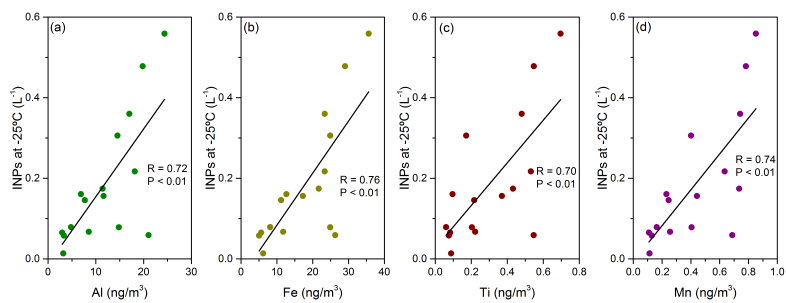


Figure 7. Comparison between the mean mass concentrations of each of the aerosol tracers measured at the Alert site during the current study and the concentrations reported from a 15-year (1980-1995) study at the same site (Sirois and Barrie, 1999). The x-axis represents the months. The black dots represent the weekly concentrations measured by Sirois and Barrie (1999), and the black line represents the estimated seasonal variations. The red symbols represent the mean mass concentrations measured in this study with standard deviation as y-error bars. The x-error bars represent the time period of this study. The mass concentrations of Fe, Ti and Mn were not available from the 15-year study.



**Figure 8.** Correlation plots between INP concentrations at -25 °C and mineral dust tracers (Al, Fe, Ti, and Mn). Included in each panel are the correlation coefficient (*R*) and the probability value (*P*).

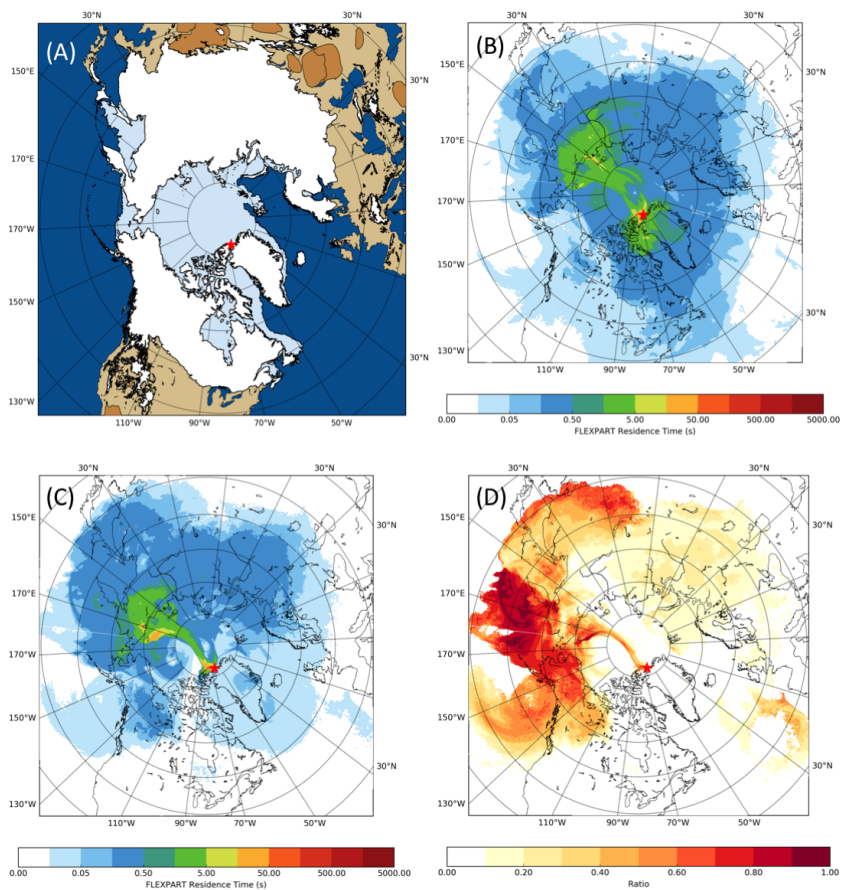


Figure 9. Panel (A) shows the surface coverage types on the first day of sampling (March 12, 2016). White represents snow, light blue represents ice, dark blue represents ocean, light brown represents land, and dark brown represents desert (data from National Snow & Ice Data Center, <https://doi.org/10.7265/NS2R3PMC>); panel (B) is the average footprint PES plot from a 20 day FLEXPART analysis for all samples; panel (C) is the average footprint PES plot for the four samples with the highest mineral dust concentrations, which were collected on March 21, 22, 27, and 28; panel (D) shows the ratio of the sum of footprint PES plots of the four samples with the highest mineral dust concentrations to the sum of footprint PES plots of all samples. Red star indicates the sampling location.

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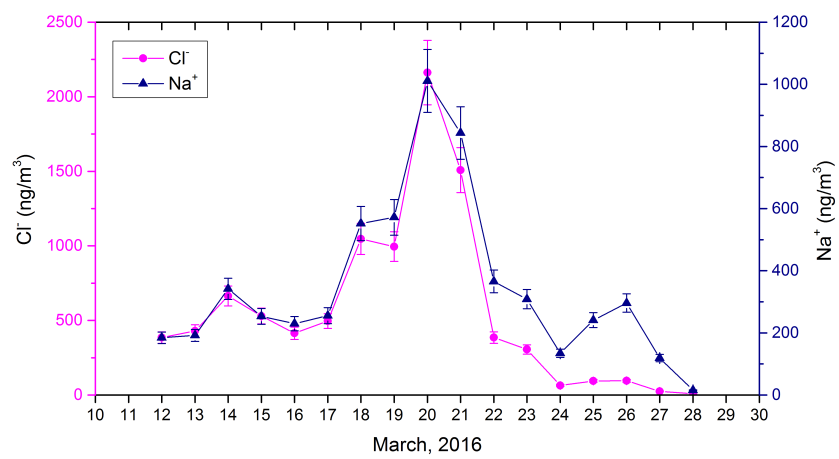
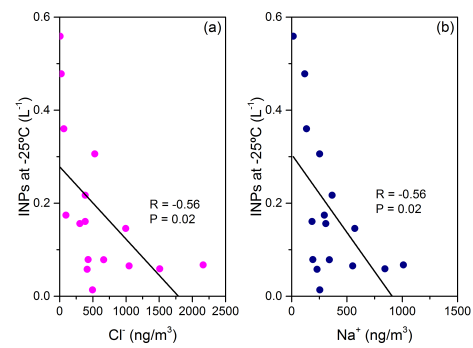
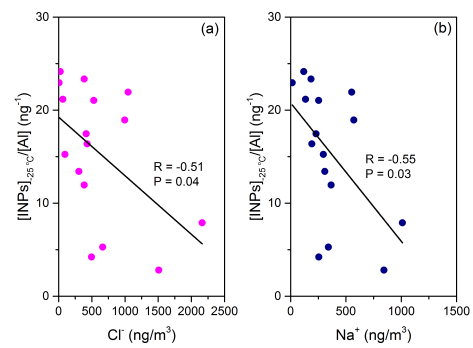


Figure 10. The time series of sea spray tracers ( $\text{Cl}^-$  and  $\text{Na}^+$ ).



**Figure 11.** Correlation plots between INP concentrations at -25 °C and sea spray tracers ( $Cl^{-}$  and  $Na^{+}$ ). Included in each panel are the correlation coefficient ( $R$ ) and the probability value ( $P$ ).



**Figure 12.** Correlation plots between the ratio of INP number concentration at -25 °C to aluminum mass concentration and sea spray tracers (Cl⁻ and Na⁺). Included in each panel are the correlation coefficient ( $R$ ) and the probability value ( $P$ ).

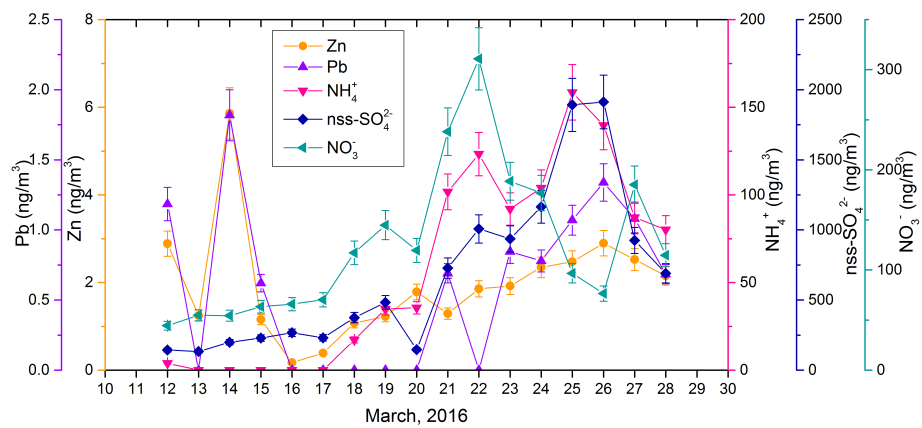
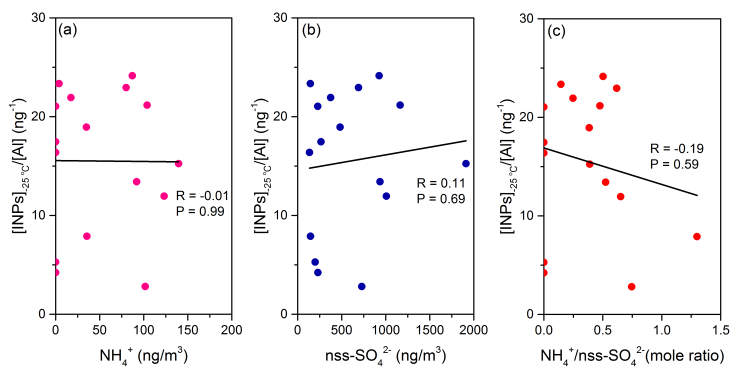


Figure 13. The time series of anthropogenic aerosol tracers (Zn, Pb,  $\text{NH}_4^+$ ,  $\text{nss-SO}_4^{2-}$ , and  $\text{NO}_3^-$ ).



**Figure 14.** Correlation plots between the ratio of INP number concentration at -25 °C to aluminum mass concentration and anthropogenic pollution tracers ( $\text{NH}_4^+$  and  $\text{nss-SO}_4^{2-}$ ) and  $\text{NH}_4^+/\text{nss-SO}_4^{2-}$  mole ratio. Included in each panel are the correlation coefficient ( $R$ ) and the probability value ( $P$ ).

## Supplemental Information

### **S1 Back trajectories**

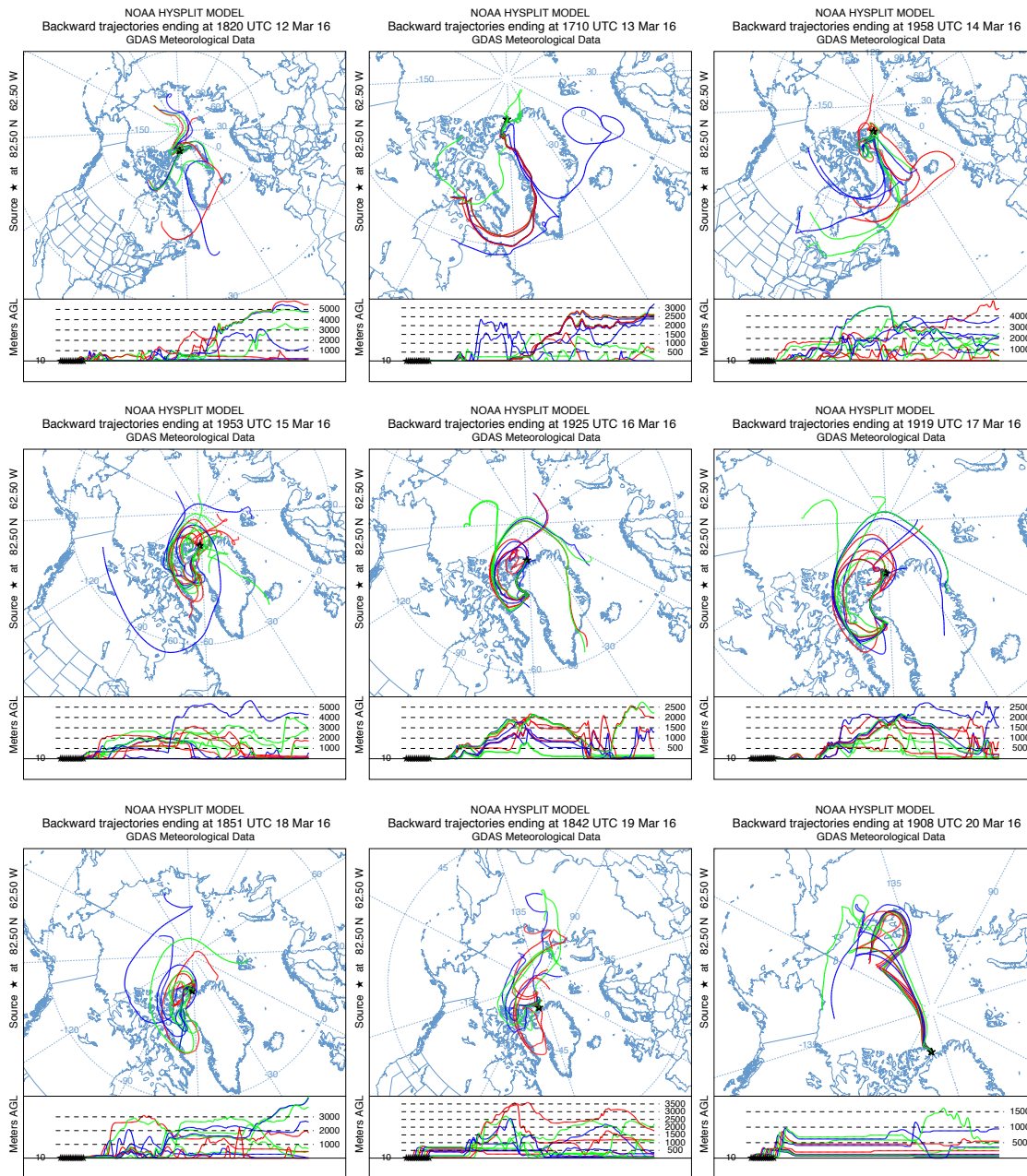
For each high-volume (i.e. quartz filter) sampling period (~ 24h), 10-day back trajectories were calculated using the HYSPLIT4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) model of the NOAA Air resources Laboratory (Stein et al., 2015). The GDAS (Global Data Assimilation System) 1° meteorological data were used as input. Back trajectories were initiated at the beginning of each quartz filter sampling period and at every 2h until the end of the sampling period. The initiation height was 10m a.g.l.. The results are shown in Fig. S1.

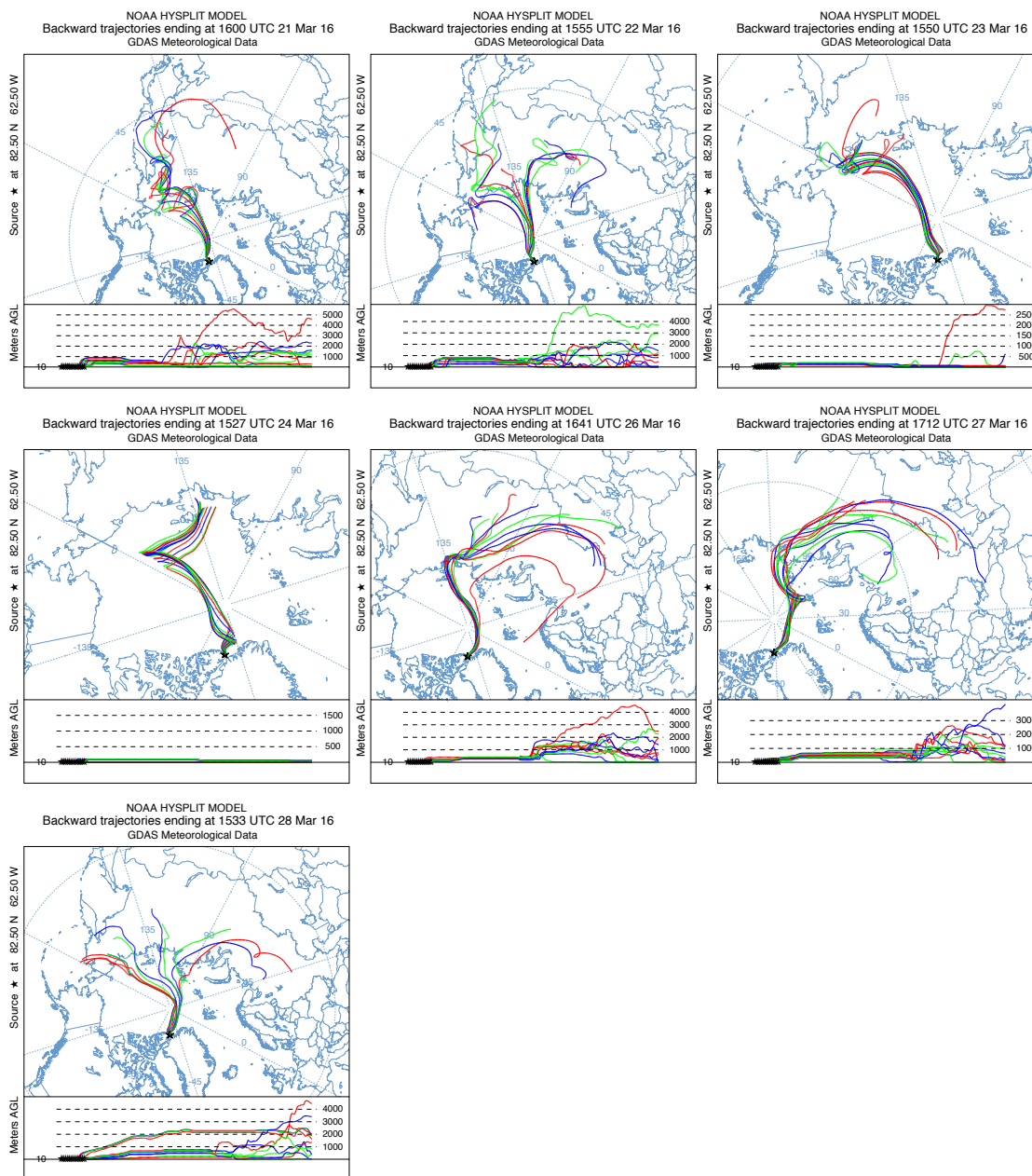
### References:

Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D. and Ngan, F.: NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System, Bull. Am. Meteorol. Soc., 96(12), 2059–2077, doi:10.1175/BAMS-D-14-00110.1, 2015.

**Table S1. Results of linear correlation analysis between the INP number concentrations (at freezing temperatures of -15, -20, and -25 °C) and meteorological parameters measured in this study.  $R$  is the correlation coefficient,  $P$  is the probability value (two tailed), and the sample number is 16.**

Meteorological parameters	INP number concentrations ( $L^{-1}$ )					
	-15 °C		-20 °C		-25 °C	
	$\underline{R}$	$\underline{P}$	$\underline{R}$	$\underline{P}$	$\underline{R}$	$\underline{P}$
Ambient temperature (°C)	<0.01	0.99	0.17	0.52	0.26	0.33
Ambient RH (%)	0.07	0.80	0.15	0.59	0.45	0.08
Wind direction (degree)	-0.04	0.87	-0.16	0.56	-0.10	0.72
Wind speed (km/h)	0.07	0.80	0.24	0.38	-0.07	0.80





**Figure S1.** The 10-day HYSPLIT back trajectories for each quartz filter sample. The back trajectories were calculated at the beginning and for every 2h during each 24h sampling period. Each panel represents one sample, and the black stars represent the sampling site. Global Data Assimilation System (GDAS) meteorological data at  $1^{\circ} \times 1^{\circ}$  spatial resolution were used as input to calculate the back trajectories using HYSPLIT.