

Supporting Information

Jingwei Yun¹, Erin Evoy¹, Soleil Worthy¹, Melody Fraser², Daniel Veber², Andrew Platt², Kevin Rawlings², Sangeeta Sharma², Richard Leaitch², Allan Bertram¹

¹Department of Chemistry, University of British Columbia, Vancouver, BC, V6T1Z1, Canada

²Climate Research Division, Environment and Climate Change Canada, Toronto, ON, M3H 5T4, Canada

The supporting information file includes 12 pages, 5 tables, and 4 figures.

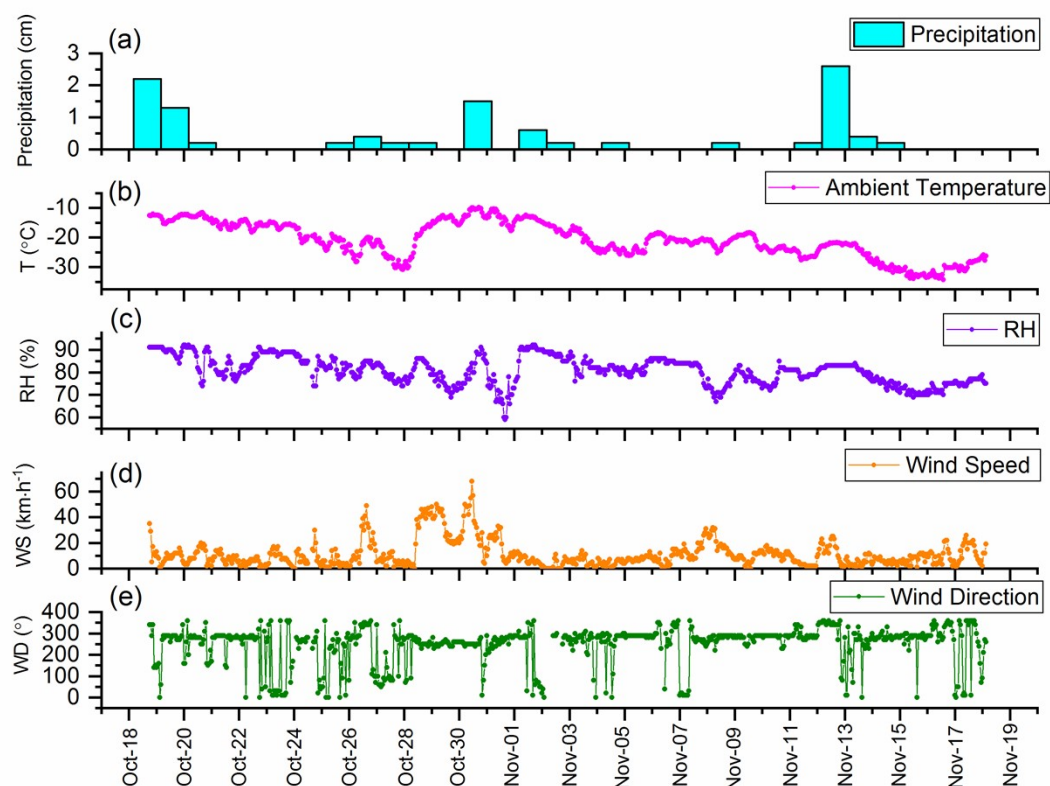


Figure S1. The time series of meteorology data at Alert during the campaign. Panel (a): Precipitation. Panel (b): Ambient temperature. Panel (c): Relative humidity. Panel (d): wind speed. Panel (e): wind direction. The meteorology data were retrieved from the Environment Canada website, <http://climate.weather.gc.ca/>.

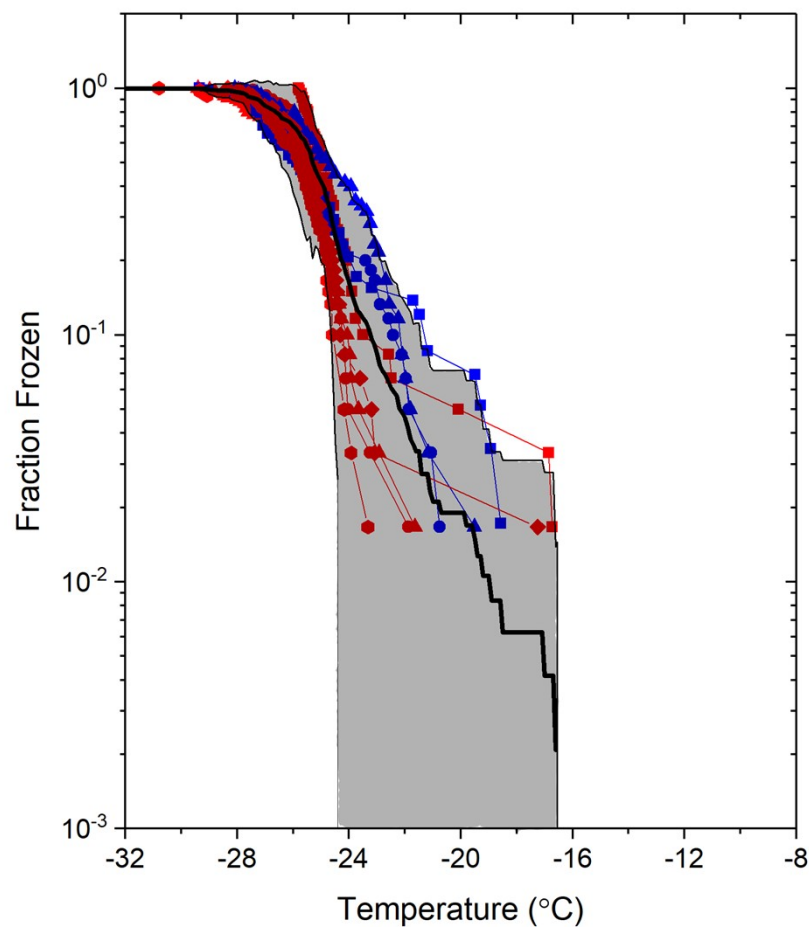


Figure S2. The fraction of frozen droplets as a function of temperature for the blanks (8 trials). Red symbols (5 trials) correspond to extracted solutions from the clean filters using Milli-Q water and blue symbols (3 trials) correspond to 0.05 M $(\text{NH}_4)_2\text{SO}_4$ solutions prepared with Milli-Q water. The freezing data for the 0.05 M $(\text{NH}_4)_2\text{SO}_4$ trials was corrected for freezing point depression caused by the salt using the method described in Section 2.3 of the main text. The black solid line is the average of the 8 trials and the shaded region corresponds to two times the standard deviations of the blanks from the 8 trials.

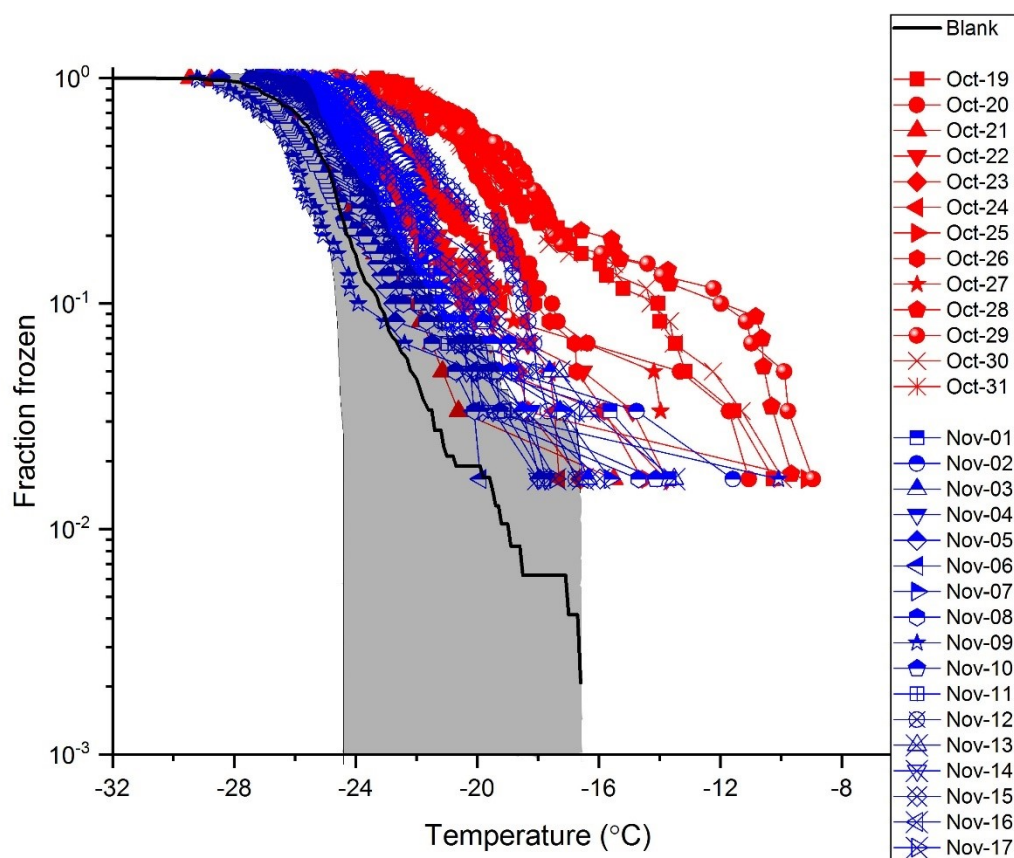


Figure S3. The fraction of frozen droplets for the 30 samples collected in the Arctic during October and November. Also included is the average of the blanks and two times the standard deviations of the blanks from the 8 trials. The black solid line is the average of the 8 trials and the shaded region corresponds to two times the standard deviations of the blanks from the 8 trials.

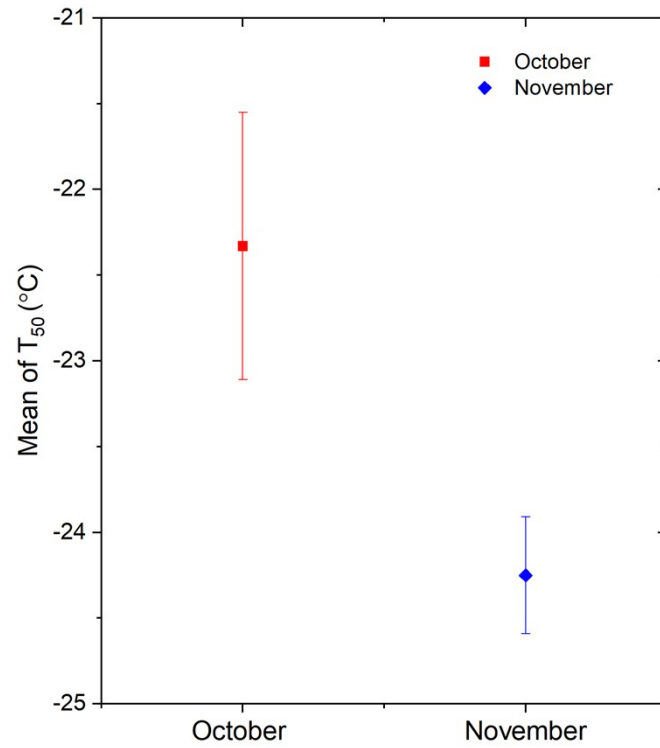


Figure S4. The mean of T_{50} , the freezing temperature at which 50% of droplets froze, for October samples and November samples. The error bars are the 83% confidence intervals. Two datasets are statistically different the 95% confidence level if their 83% confidence intervals do not overlap (Goldstein and Healy, 2009).

Table S1. Compilation of previous studies of the effect of NH_4^+ at dilute concentrations (≤ 0.1 M) on the freezing properties of different types of mineral dust INPs. In this table, ΔT_{50} is the median freezing temperature, ΔT_{onset} is the onset freezing temperature, n_m is the number of ice nucleating active sites per gram of materials, and n_s is the number of ice nucleating active sites per surface area of materials.

INP type	Freezing temperature range	Ammonium salt concentrations	Observations	References
Amorphous silica	~ -35 °C	0.004 M and 0.075 M	No significant change	Kumar et al. (2019a)
Arizona Test Dust	-20 °C to -10 °C	0.05 M	$\Delta T_{50} \cong +6$ °C n_m increased by a factor of ~ 20	Worthy et al. (2021)
Kaolinite	-25 °C to -10 °C	0.05 M	$\Delta T_{50} \cong +8$ °C n_m increased by a factor of ~ 30	Worthy et al. (2021)
K-rich feldspar	-15 °C to -5 °C	0.05 M	$\Delta T_{50} \cong +3$ °C n_m increased by a factor of ~ 10	Worthy et al. (2021)
Montmorillonite	-15 °C to -5 °C	0.05 M	$\Delta T_{50} \cong +6$ °C n_m increased by a factor of ~ 10	Worthy et al. (2021)
Feldspars	-35 °C to -20 °C	7.6×10^{-6} M to 0.1 M	$\Delta T_{onset} = +1$ °C to $+6$ °C	Kumar et al. (2018, 2019b)
Gibbsite	^a N/A	0.004 M and 0.04 M	No significant change	Kumar et al., (2019b)
Kaolinite	-35 °C to -25 °C	0.00076 M to 0.1 M	$\Delta T_{onset} = +1$ °C to $+3$ °C	Kumar et al. (2019b)
Micas	~ -32 °C	0.004 M to 0.1 M	Ratio of heterogeneous freezing signal to homogeneous freezing signal increased;	Kumar et al. (2019b)
Quartz	~ -25 °C	0.004 M to 0.1 M	Ratio of heterogeneous freezing signal to homogeneous freezing signal increased; No significant change in ΔT_{onset}	Kumar et al. (2019a)
Amorphous silica	-20 °C to -10 °C	0.015 M	No significant change	Whale et al. (2018)
Arizona Test Dust	-20 °C to -5 °C	0.015 M	No significant change	Whale et al. (2018)
Feldspars	-20 °C to -5 °C	0.00015 M to 0.015 M	$\Delta T_{50} = +1$ °C to $+4$ °C; n_s increased by a factor of ~ 10	Whale et al. (2018)
Quartz	-20 °C to -15 °C	0.015 M	$\Delta T_{50} \cong +1.5$ °C n_s increased by a factor of ~ 5	Whale et al. (2018)

^aNot Applicable

Table S2. Compilation of previous studies of the effect of NH_4^+ at dilute concentrations (< 0.1 M) on the freezing properties of different types of non-mineral dust INPs. ΔT_{50} is the median freezing temperature.

INP type	Freezing temperature range	Ammonium salt concentration	Observations	References
Fungi	-20 °C to -10 °C	0.05 M	No significant change	Worthy et al. (2021)
Humic Substances	-25 °C to -10 °C	0.015 M to 0.05 M	No significant change	Whale et al. (2018); Worthy et al., (2021)
Leaf-derived INPs	-10 °C to -5 °C	0.01 M to 0.1 M	No significant change	Reischel and Vali (1975)
<i>P. syringae</i>	-10 °C to 0 °C	0.05 M	No significant change	Worthy et al. (2021)
Sea-ice diatom exudates	-20 °C to -15 °C	0.05 M	No significant change	Worthy et al. (2021)
Sea-surface microlayer samples	-25 °C to -5 °C	0.05 M	No significant change	Worthy et al. (2021)
Snowmax	-15 °C to -5 °C	≤ 0.1 M	No significant change	Koop and Zobrist (2009); Worthy et al. (2021)
<i>X. campestris</i>	-15 °C to -5 °C	0.05 M	A small decrease in ΔT_{50} ($\cong -0.43 \pm 0.19$ °C)	Worthy et al. (2021)

Table S3. Relevant information on the previous measurements of INP concentrations in the Arctic from ground-based or ship-based platforms shown in Fig.2.

Study	Platform	Location	Sampling time
Radke et al. (1976)	Ground	Utqiagvik, Alaska, USA	March, 1970
Fountain and Ohtake (1985)	Ground	Utqiagvik, Alaska, USA	August, 1978 – April, 1979
Bigg (1996)	Ship	Arctic Ocean	August – October, 1991
Bigg and Leck (2001)	Ship	Arctic Ocean	July – September, 1996
Conen et al. (2016)	Ground	Finnmark, Norway	July, 2015
Mason et al. (2016)	Ground	Alert, Nunavut, Canada	March – July, 2014
Creamean et al. (2018)	Ground	Oliktok Point, Alaska, USA	March – May, 2017
Si et al. (2019)	Ground	Alert, Nunavut, Canada	March, 2016
Irish et al. (2019)	Ship	Arctic Ocean	July – August, 2014
Šantl-Temkiv et al. (2019)	Ground	Villum, Greenland	August, 2016
Wex et al. (2019)	Ground	Alert, Nunavut, Canada	April, 2015 – April, 2016
Wex et al. (2019)	Ground	Utqiagvik, Alaska, USA	June, 2012 – June, 2013
Wex et al. (2019)	Ground	Villum, Greenland	January, 2015 – November, 2015
Wex et al. (2019)	Ground	Ny-Ålesund, Norway	March, 2012, - September, 2012
Creamean et al. (2019)	Ship	Bering Strait, Arctic Ocean	August – September, 2017
Welti et al. (2020)	Ship	Arctic Ocean	July – August, 2001
Hartmann et al. (2020)	Ship	Arctic Ocean	May – July, 2017
Tobo et al. (2019)	Ground	Ny-Ålesund, Norway	July, 2016
Tobo et al. (2019)	Ground	Ny-Ålesund, Norway	March, 2017
Rinaldi et al. (2021)	Ground	Ny-Ålesund, Norway	April – August, 2018

Table S4. The residence time that each air mass spent in the 7 days prior to sampling in the footprint layer over a specific surface type (land, sea ice, sea, and snow) for each sample.

Collection date of the samples	Residence over land (s)	Residence over sea ice (s)	Residence over Sea (s)	Snow (s)
October 19 th	513.33	15078.3	7204.04	14894.8
October 20 th	1133.96	46781.1	4055.8	24232.5
October 21 st	154.79	76579.4	5886.24	29009.1
October 22 nd	326.99	40152.7	7783.69	16190.6
October 23 rd	31.59	77622.3	4748.68	8489.87
October 24 th	20.55	24062.2	2390.79	11421.8
October 25 th	65.59	21270.2	3353.69	35879
October 26 th	71.94	36990.9	2438.77	32094.6
October 27 th	16.85	30637.1	600.07	39952.4
October 28 th	3.05	15228.9	385.48	37134.8
October 29 th	726.07	22659	577.20	61923.9
October 30 th	1065.72	27905.5	2208.78	69060.7
October 31 st	645.80	43054.8	6167.5	46937.3
November 1 st	193.51	23231.4	16735.1	20140.4
November 2 nd	374.33	49282.1	12775.7	13058.1
November 3 rd	628.16	16966	12181.8	29180.7
November 4 th	155.14	31995.4	2944.26	19794.5
November 5 th	8.88	73468.8	1865.23	37721
November 6 th	0.0087	120622	27.62	58710.9
November 7 th	0.3243052	94172.4	107.14	28177.2
November 8 th	0.005026	128938	6.39	95118.9
November 9 th	0	154951	1.27	97707.6
November 10 th	5.72×10^{-5}	122226	1.16	145639
November 11 st	7.57×10^{-5}	60350.2	133.21	37868.3
November 12 nd	0.018	75497.6	58.15	10154.7
November 13 rd	8.8×10^{-5}	54308.4	1.73	31823.2
November 14 th	8.7×10^{-5}	19470.1	3.41	58576.7
November 15 th	0.0015	47464.7	11.77	63784.9
November 16 th	0.28	71579.1	10.07	41065.1
November 17 th	4.11	99929.7	247.25	21651

Table S5. Starting time and ending time of collection for each sample and total volume of air sampled for each sample.

Collection date of the samples	Starting time	Ending time	Total volume (L)
October 19 th , 2018	2:18pm	2:18pm	17280
October 20 th , 2018	2:26pm	2:26pm	17280
October 21 st , 2018	2:56pm	2:56pm	17280
October 22 nd , 2018	3:00pm	3:00pm	17280
October 23 rd , 2018	3:10pm	3:10pm	17280
October 24 th , 2018	3:20pm	3:20pm	17280
October 25 th , 2018	3:27pm	3:27pm	17280
October 26 th , 2018	3:38pm	3:38pm	17280
October 27 th , 2018	3:44pm	3:44pm	17280
October 28 th , 2018	3:51pm	3:51pm	17280
October 29 th , 2018	3:57pm	3:57pm	17280
October 30 th , 2018	4:03pm	4:03pm	17280
October 31 st , 2018	4:09pm	4:09pm	17280
November 1 st , 2018	4:16pm	4:16pm	17280
November 2 nd , 2018	4:22pm	4:03pm	17052
November 3 rd , 2018	4:16pm	3:16pm	16560
November 4 th , 2018	3:27pm	3:27pm	17280
November 5 th , 2018	3:34pm	3:34pm	17280
November 6 th , 2018	3:42pm	3:42pm	17280
November 7 th , 2018	3:48pm	3:48pm	17280
November 8 th , 2018	3:54pm	2:54pm	16560
November 9 th , 2018	3:05pm	3:05pm	17280
November 10 th , 2018	3:14pm	3:14pm	17280
November 11 st , 2018	3:20pm	3:20pm	17280
November 12 nd , 2018	3:25pm	3:25pm	17280
November 13 rd , 2018	3:37pm	3:37pm	17280
November 14 th , 2018	3:42pm	3:42pm	17280
November 15 th , 2018	3:49pm	3:49pm	17280
November 16 th , 2018	3:58pm	3:58pm	17280
November 17 th , 2018	4:03pm	9:41am	12696

Reference

- Bigg, E. K. (1996) 'Ice forming nuclei in the high Arctic', *Tellus B: Chemical and Physical Meteorology*, 48(2), pp. 223–233.
- Bigg, E. K. and Leck, C. (2001) 'Cloud-active particles over the central Arctic Ocean', *Journal of Geophysical Research Atmospheres*, 106(D23), pp. 32155–32166. doi: 10.1029/1999JD901152.
- Conen, F., Stopelli, E. and Zimmermann, L. (2016) 'Clues that decaying leaves enrich Arctic air with ice nucleating particles', *Atmospheric Environment*. Elsevier Ltd, 129, pp. 91–94. doi: 10.1016/j.atmosenv.2016.01.027.
- Creamean, J. M. et al. (2018) 'Marine and terrestrial influences on ice nucleating particles during continuous springtime measurements in an Arctic oilfield location', *Atmospheric Chemistry and Physics*, 18(24), pp. 18023–18042. doi: 10.5194/acp-18-18023-2018.
- Creamean, J. M. et al. (2019) 'Ice Nucleating Particles Carried From Below a Phytoplankton Bloom to the Arctic Atmosphere', *Geophysical Research Letters*, 46(14), pp. 8572–8581. doi: 10.1029/2019GL083039.
- Fountain, A. G. and Ohtake, T. (1985) 'Concentrations and Source Areas of Ice Nuclei in the Alaskan Atmosphere', *Journal of Climate and Applied Meteorology*, pp. 377–382. doi: 10.1175/1520-0450(1985)024<0377:casaoi>2.0.co;2.
- Goldstein, H. and Healy, M. J. R. (2009) 'The Graphical Presentation of a Collection of Means', *Royal Statistical Society*, 158(1), pp. 175–177. doi: 10.2307/2983411.
- Hartmann, M. et al. (2020) 'Terrestrial or marine? – Indications towards the origin of Ice Nucleating Particles during melt season in the European Arctic up to 83.7° N', *Atmospheric Chemistry and Physics Discussions*, (November), pp. 1–35. doi: 10.5194/acp-2020-1211.
- Irish, V. E. et al. (2019) 'Ice nucleating particles in the marine boundary layer in the Canadian Arctic during summer 2014', *Atmospheric Chemistry and Physics*, 19(2), pp. 1027–1039. doi: 10.5194/acp-19-1027-2019.
- Koop, T. and Zobrist, B. (2009) 'Parameterizations for ice nucleation in biological and atmospheric systems', *Physical Chemistry Chemical Physics*, 11(46), pp. 10741–11064. doi: 10.1039/b914289d.
- Kumar, A. et al. (2018) 'Ice nucleation activity of silicates and aluminosilicates in pure water and aqueous solutions - Part 1: The K-feldspar microcline', *Atmospheric Chemistry and Physics*, 18(10), pp. 7057–7079. doi: 10.5194/acp-18-7057-2018.
- Kumar, A., Marcolli, C. and Peter, T. (2019a) 'Ice nucleation activity of silicates and aluminosilicates in pure water and aqueous solutions-Part 2: Quartz and amorphous silica', *Atmospheric Chemistry and Physics*, 19(9), pp. 6035–6058. doi: 10.5194/acp-19-6035-2019.

- Kumar, A., Marcolli, C. and Peter, T. (2019b) ‘Ice nucleation activity of silicates and aluminosilicates in pure water and aqueous solutions-Part 3: Aluminosilicates’, *Atmospheric Chemistry and Physics*, 19(9), pp. 6059–6084. doi: 10.5194/acp-19-6059-2019.
- Mason, R. H. et al. (2016) ‘Size-resolved measurements of ice-nucleating particles at six locations in North America and one in Europe’, *Atmospheric Chemistry and Physics*, 16(3), pp. 1637–1651. doi: 10.5194/acp-16-1637-2016.
- Radke, L. F., Hobbs, P. V. and Pinnons, J. E. (1976) ‘Observations of Cloud Condensation Nuclei, Sodium-Containing Particles, Ice Nuclei and the Light-Scattering Coefficient Near Barrow, Alaska’, *Journal of Applied Meteorology*, pp. 982–995. doi: 10.1175/1520-0450(1976)015<0982:ooccns>2.0.co;2.
- Reischel, M. T. and Vali, G. (1975) ‘Freezing Nucleation in Aqueous Electrolytes’, *Tellus*, 27(4), pp. 414–427. doi: 10.1111/j.2153-3490.1975.tb01692.x.
- Rinaldi, M. et al. (2021) ‘Ice-nucleating particle concentration measurements from Ny-Ålesund during the Arctic spring-summer in 2018’, *Atmospheric Chemistry and Physics*, 21(19), pp. 14725–14748. doi: 10.5194/acp-21-14725-2021.
- Šantl-Temkiv, T. et al. (2019) ‘Biogenic Sources of Ice Nucleating Particles at the High Arctic Site Villum Research Station’, *Environmental Science & Technology*, 53(18), pp. 10580–10590. doi: 10.1021/acs.est.9b00991.
- Si, M. et al. (2019) ‘Concentrations, composition, and sources of ice-nucleating particles in the Canadian High Arctic during spring 2016’, *Atmospheric Chemistry and Physics*, 19(5), pp. 3007–3024. doi: 10.5194/acp-2018-950.
- Tobo, Y. et al. (2019) ‘Glacially sourced dust as a potentially significant source of ice nucleating particles’, *Nature Geoscience*. Springer US, 12(4), pp. 253–258. doi: 10.1038/s41561-019-0314-x.
- Welti, A. et al. (2020) ‘Ship-based measurements of ice nuclei concentrations over the Arctic, Atlantic, Pacific and Southern oceans’, *Atmospheric Chemistry and Physics*, 20(23), pp. 15191–15206. doi: 10.5194/acp-20-15191-2020.
- Wex, H. et al. (2019) ‘Annual variability of ice-nucleating particle concentrations at different Arctic locations’, *Atmospheric Chemistry and Physics*, 19(7), pp. 5293–5311. doi: 10.5194/acp-19-5293-2019.
- Whale, T. . et al. (2018) ‘The enhancement and suppression of immersion mode heterogeneous ice-nucleation by solutes’, *Chemical Science*. Royal Society of Chemistry, 9, pp. 4142–4151. doi: 10.1039/C7SC05421A.
- Worthy, S. E. et al. (2021) ‘The effect of (NH₄)₂SO₄ on the freezing properties of non-mineral dust ice nucleating substances of atmospheric relevance’, (March), pp. 1–30.