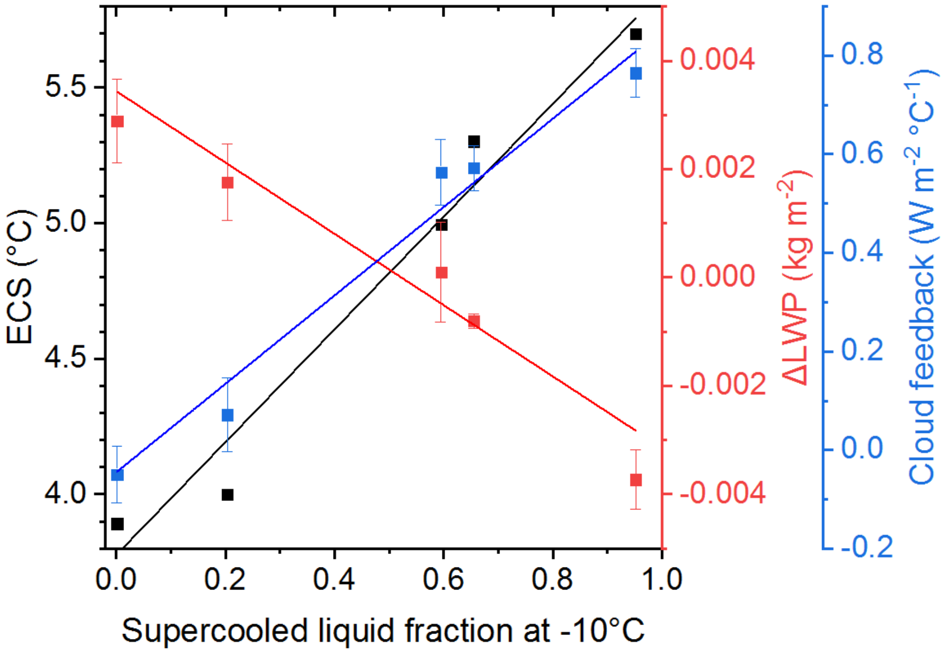
Supporting online material

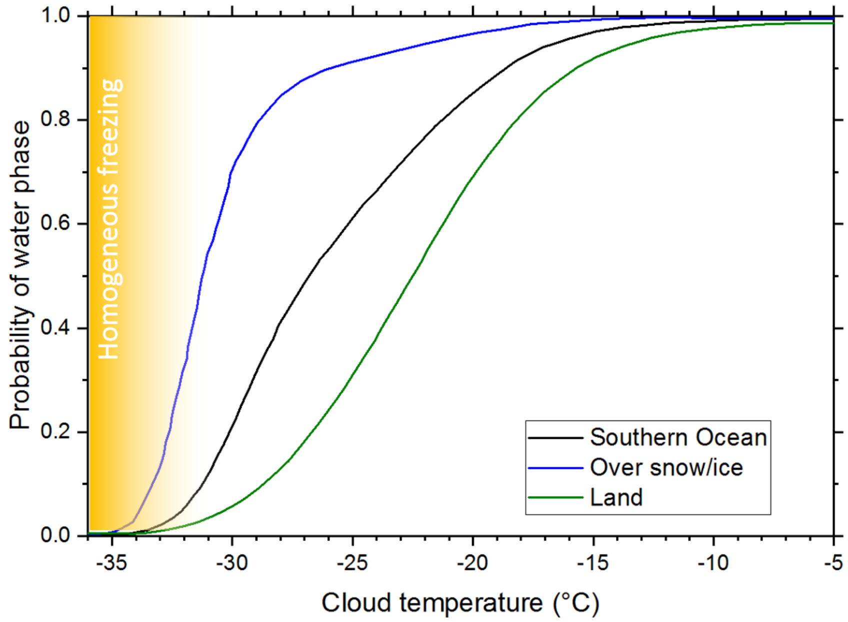
Opinion: Cloud-phase climate feedback and the importance of ice-nucleating particles

Benjamin J. Murray, Kenneth S. Carslaw, Paul R. Field

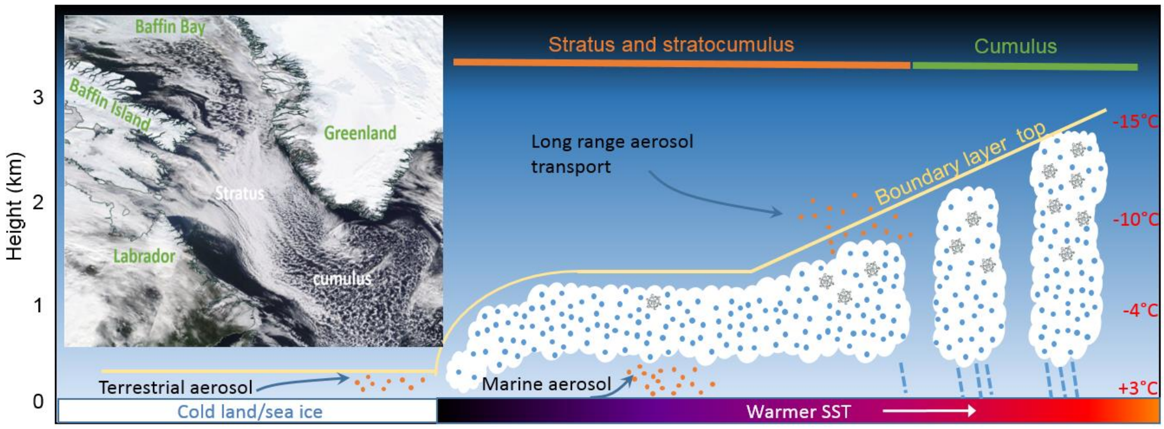
1. Supplementary Figures:



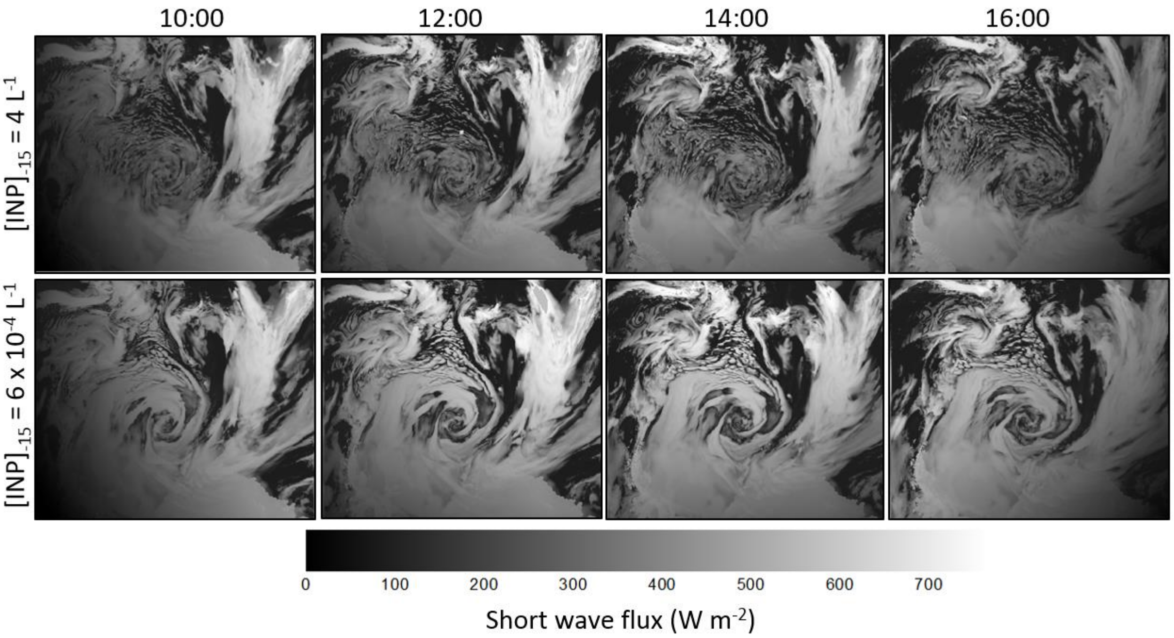
Supplementary Figure 1. Response of feedback and equilibrium climate sensitivity to supercooled liquid fraction in one model**.** Data are taken from Tan et al. (2016). The supercooled liquid fraction is for the -10°C isotherm in the present day. The model used was version 5.1 of the National Center for Atmospheric Research’s Community Atmosphere Model (CAM5.1). The points at 0.6 and 0.65 supercooled liquid fraction were constrained by space-borne lidar measurements, whereas the point at 0.2 was the model run in its standard configuration.



Supplementary Figure 2. Satellite based lidar (CALIPSO) measurements of the probability of finding the water phase at cloud top as a function of cloud top temperature (Hu et al., 2010). The range over which homogeneous freezing up water becomes dominant is indicated in yellow shading and becomes increasingly important below -33°C (Herbert et al., 2015).



Supplementary Figure 3. An illustration of a cold-air outbreak. As cold air streams off the sea ice or cold land masses at higher latitudes, stratus clouds will initially form in the deepening boundary layer (on the order of 1 km deep), but then as the CAO extends over warmer ocean, the clouds become more broken, first forming stratocumulus and then shallow cumulus which can become around 2-3 km deep. As they become deeper, the cloud top temperatures decrease and precipitation becomes more likely. This progression from stratus to cumulus cloud can occur over thousands of kilometres and therefore exert a substantial impact on radiation and global climate. The inset satellite image showing an example of a cold-air outbreak is from the NASA Worldview application for the 13th October 2017 (https://go.nasa.gov/34e8yfp).



Supplementary Figure 4. The effect of INP concentration on model clouds in the cold air sector of a cyclone system over the Southern Ocean with a cloud top temperature of around -15°C over the course of a simulation. This is an expanded version of Figure 3. The top row is for a relatively high INP case, whereas the bottom row is for a relatively low, more realistic, case. This model data is from Vergara-Temprado et al. (2018).

Supplementary Table 1. Details of INP measurements at the mid- to high-latitudes

Study	Platform	Instrument	Location	Dates	Notes
Southern Hemisphere					
Schmale et al. (2019)	Ship	Filter-DFA	Southern Ocean, Antarctica circumnavigation	December 2016 – March 2017	These are averages from each of three legs. More detailed data is yet to be published.
McCluskey et al. (2018)	Ship	CFDC and filter-DFA	Southern Ocean, South of Australia	March–April 2016	Measurements collected during the CAPRICORN cruise on the RV Investigator. Used open faced filter holders 23 m above ocean surface. Used a sector specific sampler to minimise ship stack emission sampling. Blanks regularly collected. 19 to 55 m^3 of air sampled per filter.
Bigg (1973)	Ship	Filter-diffusion chamber	Southern Ocean. Half the SH cantered around Australia	1969-72	In Figure 4 we have included the average of the latitude dependent data from Fig 2 for -10, -15 and -20°C. We have only included the data from 50 to 74°S. There is data down to 30°, which becomes affected by the continent of Australia. These are averages themselves. There is a breakdown of data at -15°C, which shows a greater spread in values than indicated by the data in the plot. The more detailed data for temperatures other than -10 or -15°C is not reported. Bigg (personal communication) states that the original data is now lost. A sample volume dependence is reported, with larger concentrations

					reported for lower volumes. Bigg (1973) concludes that this is related to mixing with sea salt aerosol on the filter reducing the activity of the INP; this is supported by Mossop and Thorndike (1966).
Bigg and Hopwood (1963)	Land	Mixing cold chamber	McMurdo base, 78 S.	Dec 1961 – Feb 1962	Bigg (personal communication): Potential local contamination sources were the McMurdo camp, construction work on a proposed nuclear power plant, New Zealand's Scott Base and Mt. Erebus that emitted a constant smoke plume. A site was chosen a site on a hill about 200m above McMurdo and Scott and about 150m above the construction work. The winds didn't blow from McMurdo or the construction site and particle collections at Scott Base didn't suggest any obvious pollutants. The plume from Erebus emitted at above 3000m in a region of strong winds was unlikely to be a problem. In addition, Bigg noted that there were problems with mixing chambers of this type that had to be avoided: (1) frost on the chamber walls leading to over-counts, (2) insufficient cloud density to capture potential IN and (3), matching sugar solution concentration to the temperature of measurement. If this is not done, or the solutions not replaced frequently, some ice crystals unrelated to atmospheric INP may form. Precautions to counter these problems were strictly observed at McMurdo.

Bird et al. (1961)	Ship	Expansion cold box	Southern Ocean, 42 to 67°S (we included measurements above 66° only.	January and February, 1960	Data in sea ice, on return leg are included only. Bigg (personal communication) noted that the conditions in open water on the outward leg were ‘almost impossible for the operators until they were within the pack ice region’.
Belosi et al. (2014)	Land	Filter and dynamic processing chamber	Zucchelli Station (74°41′ S–164°07′ E), Terra Nova Bay, Antarctica.	November, 2001	Seven filters collected, from a range of locations around the base. No discussion of handling blank in paper.
Kikuchi (1971)	Land	Mixing chamber	Syowa station 69*00'S, 39*35'E), Antarctica.	February 1968 to January 1969	Several protocals were employed. Lower apparent INP concentrations were reported when the air was heated prior to being admitted to the instrument. This may indicate a population of microscopic ice or preactivated aerosol. We only plot data where air was heated prior to analysis. There was a strong peak in INP during winter. This may be related to preactivated aerosol despite the heating step.
Saxena and Weintraub (1988)	Land	Filter and DFA	Palmer station (64°46′27″S 64°03′10″W), Antarctic peninsula	February to December, 1983	Very high INP concentrations. No discussion of handling blanks, therefore we did not include this data in the paper.

Northern Hemisphere					
Sanchez-Marroquin et al. (2020)	Aircraft	Filter and DFA (NIPI)	Around Iceland	October 2017	Boundary layer flights in air with wide range of Icelandic dust concentrations.
Wex et al. (2019)	Land	Filter and DFA	Alert (Nunavut, northern Canadian archipelago on Ellesmere Island). Utqiagvik, (N. Alaska). Ny-Ålesund (Svalbard). Villum Research Station (northern Greenland)	2012-2016, intermittent and different periods for each station	Strong indication of seasonal cycles. Generally lower INP concentrations in winter. Generally higher activity in summer and autumn (and early winter). Substantial variability in all seasons and all locations.
Irish et al. (2019)	Ship	Single stage impactor and DFA	Canadian Arctic	July and August 2014	Conclude that mineral dust from local sources is important

Creamean et al. (2019)	Ship	Rotating-drum impactor and DFA	Bering and Chukchi Seas	August and September 2017	Size resolved into 4 bins. Only show total here, however bin B was missing, hence the total INP will be biased low.
Creamean et al. (2018)	Land	Rotating-drum impactor and DFA	N. Alaska, Oliktok Point	March to May 2017	Size resolved into 4 bins. Only show total here.
Si et al. (2018)	Ship	Impactor and DFA	Lancaster Sound Labrador Sea	July 2014	Size resolved measurements.
Si et al. (2019)	Land	Impactor and DFA	Alert	March 2016	
Bigg (1996)	Ship	Filter and diffusion chamber	High Arctic	August to October 1991	The time series data has been digitised from Fig 1, but there is some ambiguity as to the interpretation of the plot. The range of concentrations is taken from their Figure 2, where discrete values are given.
Bigg and Leck (2001)	Ship	Filter and diffusion chamber	High Arctic	July to September 1996	The time series has been digitised from their Figure 4a.
Bigg (Personal communication)	Ship	Filter and diffusion chamber	High Arctic	July to August, 2001	Third in a series of cruises on the Oden
Fountain and Ohtake (1985)	Ground	Filter and diffusion	Alaska:		Daily filters in each location.

		chamber similar to Bigg's	Barrow, Fairbanks and Homer	Barrow: August 78 to April 79. Fairbanks: June 78 to April 79 Homer: Oct 78 to April 79	Averages quotes as 0.125, 0.14 and 0.167 L^{-1} in Barrow, Fairbanks and Homer at -20°C. There was very large variability. The time series are not plotted in a way amenable to digitisation, hence data is not included in Figure 4.
Borys (1989)	Aircraft	Filters and dynamic developing chamber	Alaska, northern Canada and Greenland. Surface to tropopause	April 1986	We split the data into regions, excluding measurements which crossed multiple regions. Higher INP in between Thule and Alert than Alaska. Several results in baseline in Alaska.
Rogers et al. (2001)	Aircraft	CFDC	Arctic Ocean north of Alaska, associated with SHEBA	May 1998	Data is show for supersaturations below and above (or equal to) water saturation. We show the latter binned into broad altitude ranges.
Prenni et al. (2007)	Aircraft	CDFC	N. Alaska	September-October 2004	Data include frequent periods where the INP concnentration was below detection limit.
DeMott et al. (2016)	Ship	Various. Filter/impactor and DFA	Bering Sea Baffin Bay	Bering Sea: summer 2012	Paper presents range of marine INP measurements across all latitudes. We have shown data relevant for this work.

				Baffin Bay: summer 2014	
Flyger and Heidam (1978)	Land	Filter, but few details on analysis technique	Kap Harald Moltke, (82° 09'N, 29° 53'W) N. Greenland	June to August 1974	One event where they observed relatively high INP for several days. Satellite images indicate it has the potential to be a good dust source.
Radke et al. (1976)	Land	Mixing cloud chamber with acoustic detector	Utqiagvik, N. Alaska	March 1970	Authors note very large, but poorly quantified uncertainty in measurements, so results not shown.

References

Belosi, F., Santachiara, G., and Prodi, F.: Ice-forming nuclei in Antarctica: New and past measurements, Atmos. Res., 145-146, 105-111, https://doi.org/10.1016/j.atmosres.2014.03.030, 2014.

Bigg, E. K., and Hopwood, S. C.: Ice Nuclei in the Antarctic, J. Atmos. Sci., 20, 185-188, 10.1175/1520-0469(1963)020<0185:INITA>2.0.CO;2, 1963.

Bigg, E. K.: Ice Nucleus Concentrations in Remote Areas, J. Atmos. Sci., 30, 1153-1157, 10.1175/1520-0469(1973)030<1153:incira>2.0.co;2, 1973.

Bigg, E. K.: Ice forming nuclei in the high Arctic, Tellus B, 48, 223-233, 10.1034/j.1600-0889.1996.t01-1-00007.x, 1996.

Bigg, E. K., and Leck, C.: Properties of the aerosol over the central Arctic Ocean, J. Geophys. Res., 106, 32101-32109, 10.1029/1999jd901136, 2001.

Bird, I., Cresswell, G., Humble, J., Norris, D., and Bigg, E. K.: ATMOSPHERIC ICE NUCLEI IN HIGH SOUTHERN LATITUDES, Journal of Meteorology, 18, 563-564, 10.1175/1520-0469(1961)018<0563:ainihs>2.0.co;2, 1961.

Borys, R. D.: Studies of ice nucleation by Arctic aerosol on AGASP-II, Journal of Atmospheric Chemistry, 9, 169-185, 10.1007/BF00052831, 1989.

Creamean, J. M., Kirpes, R. M., Pratt, K. A., Spada, N. J., Maahn, M., de Boer, G., Schnell, R. C., and China, S.: Marine and terrestrial influences on ice nucleating particles during continuous springtime measurements in an Arctic oilfield location, Atmos. Chem. Phys., 18, 18023-18042, 10.5194/acp-18-18023-2018, 2018.

Creamean, J. M., Cross, J. N., Pickart, R., McRaven, L., Lin, P., Pacini, A., Hanlon, R., Schmale, D. G., Ceniceros, J., Aydell, T., Colombi, N., Bolger, E., and DeMott, P. J.: Ice Nucleating Particles Carried From Below a Phytoplankton Bloom to the Arctic Atmosphere, Geophys. Res. Lett., 46, 8572-8581, 10.1029/2019GL083039, 2019.

DeMott, P. J., Hill, T. C. J., McCluskey, C. S., Prather, K. A., Collins, D. B., Sullivan, R. C., Ruppel, M. J., Mason, R. H., Irish, V. E., Lee, T., Hwang, C. Y., Rhee, T. S., Snider, J. R., McMeeking, G. R., Dhaniyala, S., Lewis, E. R., Wentzell, J. J. B., Abbatt, J., Lee, C., Sultana, C. M., Ault, A. P., Axson, J. L., Diaz Martinez, M., Venero, I., Santos-Figueroa, G., Stokes, M. D., Deane, G. B., Mayol-Bracero, O. L., Grassian, V. H., Bertram, T. H., Bertram, A. K., Moffett, B. F., and Franc, G. D.: Sea spray aerosol as a unique source of ice nucleating particles, P. Natl. Acad. Sci. USA, 113, 5797-5803, 10.1073/pnas.1514034112, 2016.

Flyger, H., and Heidam, N. Z.: Ground level measurements of the summer tropospheric aerosol in Northern Greenland, J. Aerosol Sci., 9, 157-168, https://doi.org/10.1016/0021-8502(78)90075-7, 1978.

Fountain, A. G., and Ohtake, T.: Concentrations and Source Areas of Ice Nuclei in the Alaskan Atmosphere, Journal of Climate and Applied Meteorology, 24, 377-382, 10.1175/1520-0450(1985)024<0377:casaoi>2.0.co;2, 1985.

Herbert, R. J., Murray, B. J., Dobbie, S. J., and Koop, T.: Sensitivity of liquid clouds to homogenous freezing parameterizations, Geophys. Res. Lett., 42, 1599-1605, 10.1002/2014GL062729, 2015.

Hu, Y., Rodier, S., Xu, K.-m., Sun, W., Huang, J., Lin, B., Zhai, P., and Josset, D.: Occurrence, liquid water content, and fraction of supercooled water clouds from combined CALIOP/IIR/MODIS measurements, J. Geophys. Res., 115, 10.1029/2009JD012384, 2010.

Irish, V. E., Hanna, S. J., Willis, M. D., China, S., Thomas, J. L., Wentzell, J. J. B., Cirisan, A., Si, M., Leaitch, W. R., Murphy, J. G., Abbatt, J. P. D., Laskin, A., Girard, E., and Bertram, A. K.: Ice nucleating particles in the marine boundary layer in the Canadian Arctic during summer 2014, Atmos. Chem. Phys., 19, 1027-1039, 10.5194/acp-19-1027-2019, 2019.

Kikuchi, K.: Observation of concentration of ice nuclei at Syowa station, Antarctica, J. Meteor. Soc. Japan, 49, 20-31, 1971.

McCluskey, C. S., Hill, T. C. J., Humphries, R. S., Rauker, A. M., Moreau, S., Strutton, P. G., Chambers, S. D., Williams, A. G., McRobert, I., Ward, J., Keywood, M. D., Harnwell, J., Ponsonby, W., Loh, Z. M., Krummel, P. B., Protat, A., Kreidenweis, S. M., and DeMott, P. J.: Observations of Ice Nucleating Particles Over Southern Ocean Waters, Geophys. Res. Lett., 45, 11,989-911,997, 10.1029/2018gl079981, 2018.

Mossop, S. C., and Thorndike, N. S. C.: The Use of Membrane Filters in Measurements of Ice Nucleus Concentration. I. Effect of Sampled Air Volume, J. App. Meteorol., 5, 474-480, 10.1175/1520-0450(1966)005<0474:TUOMFI>2.0.CO;2, 1966.

Prenni, A. J., Harrington, J. Y., Tjernstrom, M., DeMott, P. J., Avramov, A., Long, C. N., Kreidenweis, S. M., Olsson, P. Q., and Verlinde, J.: Can ice-nucleating aerosols affect arctic seasonal climate?, B. Am. Meteorol. Soc., 88, 541-+, 2007.

Radke, L. F., Hobbs, P. V., and Pinnons, J. E.: Observations of Cloud Condensation Nuclei, Sodium-Containing Particles, Ice Nuclei and the Light-Scattering Coefficient Near Barrow, Alaska, J. App. Meteorol., 15, 982-995, 10.1175/1520-0450(1976)015<0982:OOCCNS>2.0.CO;2, 1976.

Rogers, D. C., DeMott, P. J., and Kreidenweis, S. M.: Airborne measurements of tropospheric ice-nucleating aerosol particles in the Arctic spring, J. Geophys. Res., 106, 15053-15063, 2001.

Sanchez-Marroquin, A., Arnalds, O., Baustian-Dorsi, K. J., Browse, J., Dagsson-Waldhauserova, P., Harrison, A. D., Maters, E. C., Pringle, K. J., Vergara-Temprado, J., Burke, I. T., McQuaid, J. B., Carslaw, K. S., and Murray, B. J.: Iceland is an episodic source of atmospheric ice-nucleating particles relevant for mixed-phase clouds, Science Advances, 6, eaba8137, 10.1126/sciadv.aba8137, 2020.

Saxena, V. K., and Weintraub, D. C.: Ice forming nuclei concentrations at Palmer Station, Antarctica, Atmospheric Aerosols and Nucleation, Berlin, Heidelberg, 1988, 679-682,

Schmale, J., Baccarini, A., Thurnherr, I., Henning, S., Efraim, A., Regayre, L., Bolas, C., Hartmann, M., Welti, A., Lehtipalo, K., Aemisegger, F., Tatzelt, C., Landwehr, S., Modini, R. L., Tummon, F., Johnson, J. S., Harris, N., Schnaiter, M., Toffoli, A., Derkani, M., Bukowiecki, N., Stratmann, F., Dommen, J., Baltensperger, U., Wernli, H., Rosenfeld, D., Gysel-Beer, M., and Carslaw, K. S.: Overview of the Antarctic Circumnavigation Expedition: Study of Preindustrial-like Aerosols and Their Climate Effects (ACE-SPACE), B. Am. Meteorol. Soc., 100, 2260-2283, 10.1175/BAMS-D-18-0187.1, 2019.

Si, M., Irish, V. E., Mason, R. H., Vergara-Temprado, J., Hanna, S. J., Ladino, L. A., Yakobi-Hancock, J. D., Schiller, C. L., Wentzell, J. J. B., Abbatt, J. P. D., Carslaw, K. S., Murray, B. J., and Bertram, A. K.: Ice-nucleating ability of aerosol particles and possible sources at three coastal marine sites, Atmos. Chem. Phys., 18, 15669-15685, 10.5194/acp-18-15669-2018, 2018.

Si, M., Evoy, E., Yun, J., Xi, Y., Hanna, S. J., Chivulescu, A., Rawlings, K., Veber, D., Platt, A., Kunkel, D., Hoor, P., Sharma, S., Leaitch, W. R., and Bertram, A. K.: Concentrations, composition, and sources of ice-nucleating particles in the Canadian High Arctic during spring 2016, Atmos. Chem. Phys., 19, 3007-3024, 10.5194/acp-19-3007-2019, 2019.

Tan, I., Storelvmo, T., and Zelinka, M. D.: Observational constraints on mixed-phase clouds imply higher climate sensitivity, Science, 352, 224-227, 10.1126/science.aad5300, 2016.

Vergara-Temprado, J., Miltenberger, A. K., Furtado, K., Grosvenor, D. P., Shipway, B. J., Hill, A. A., Wilkinson, J. M., Field, P. R., Murray, B. J., and Carslaw, K. S.: Strong control of Southern Ocean cloud reflectivity by ice-nucleating particles, P. Natl. Acad. Sci. USA, 10.1073/pnas.1721627115, 2018.

Wex, H., Huang, L., Zhang, W., Hung, H., Traversi, R., Becagli, S., Sheesley, R. J., Moffett, C. E., Barrett, T. E., Bossi, R., Skov, H., Hünerbein, A., Lubitz, J., Löffler, M., Linke, O., Hartmann, M., Herenz, P., and Stratmann, F.: Annual variability of ice-nucleating particle concentrations at different Arctic locations, Atmos. Chem. Phys., 19, 5293-5311, 10.5194/acp-19-5293-2019, 2019.