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On coarse patterns in the atmospheric concentration of ice nucleating particles

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ABSTRACT

The atmospheric concentration of ice nucleating particles active at around -10 °C (INP₋₁₀) is very low. Nevertheless, these particles play a role in the development of cloud systems, so their spatial and temporal patterns merit attention. We collated available datasets on INP₋₁₀ to identify such patterns. Among the five low altitude observatories in northern Eurasia, median values throughout May to October were lowest in Scandinavia (4 and 6 m⁻³), somewhat higher in central Europe (11 m⁻³), substantially higher in the West Siberian Plain (69 m⁻³) and highest in the Central Yakutian Lowland (204 m⁻³), suggesting that the abundance of INP₋₁₀ in northern Eurasia may increase with continentality and from West to East. The range of values at the same observatories was narrower throughout November to April (2 to 27 m⁻³). On average, by an order of magnitude smaller values were reported for the four Arctic observatories. Consequently, increasing poleward transport of air masses from the midlatitudes likely raises the concentration of INP₋₁₀ in the Arctic, particularly when air masses had surface contact in eastern parts of northern Eurasia.

1. Introduction

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The transient existence of clouds is subject to meteorological and microphysical conditions (Findeisen, 1938). Ice nucleating particles (INPs) modify microphysical conditions in cloud droplets by lowering the energy barrier to be overcome in the transition from supercooled liquid to ice (Kanji et al., 2017). Certain bacteria, fungal spores and macromolecules aerosolised from vegetation and plant litter act as INPs at a few degrees below 0 °C (Sands et al., 1982; Lindemann et al., 1982; Vasebi et al., 2019). Yet, their concentration in clouds is very low (Joly et al., 2014). To quantify these INPs in the atmosphere a sufficient number of them needs to be collected, typically from several cubic metres of air, and analysed off-line in a drop freeze assay. Demands on time and manual labour limit the data made available through such observations. When temperature drops below about -15 °C, increasingly other kinds of aerosol particles cause freezing of cloud droplets (Murray et al., 2012; Kanji et al., 2017; Huang et al., 2021). At -30 °C the vast majority of INPs above central Europe is contributed by desert dust (Brunner et al., 2021), in which feldspar may be the most active ingredient (Atkinson et al., 2013). The greater abundance of INPs active at -30 °C allows reliable quantification in several litres air through realtime, automated measurement (Brunner and Kanji, 2021).

When there is enough moisture available for the activation of INPs, the number concentration of activated INPs increases with decreasing temperature of an air mass. Hawker et al. (2021) have demonstrated how the slope of this relation determines the development of a convective cloud system. A shallow slope, which compared to a steep slope has a larger number of INPs activated above -17 °C and a smaller number below -17 °C, led their regional model to predict ice formation at lower altitudes and resultingly reduced anvil development, with an overall reduced outgoing radiation from cloudy regions. Also, shallow cumulus clouds can be affected by INPs active at little supercooling as shown by modelling (Mason, 1996) and observation (Crosier et al., 2011; Crawford et al., 2012). Such clouds form precipitation when initial ice formation by INPs is amplified through secondary ice production (Field et al., 2017; Phillips et al., 2017; Sullivan et al., 2018). Increasing concentration of INP may promote precipitation and thereby reduce cloud lifetime, resulting in a positive climate feedback (Murray et al., 2021). Low-level mixed-phase clouds in the Arctic are a further example, where the concentration of INPs active at little supercooling

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probably makes a difference to cloud lifetime and radiation properties (Morrison et al., 2012; Pasquier et al., 2022).

While the number concentration of INPs active below -15 °C can be estimated from the more easily measured number concentration of aerosol particles >0.5 µm (DeMott et al., 2015) combined with the history of an air mass (Mignani et al., 2021), the same does not necessarily hold true for INPs active at around -10 °C (Conen et al., 2015). Although, the number concentration of INPs active at -16 °C tends to increase with increasing PM₁₀ mass concentration, INPs active at -12or -8 °C are completely unrelated to this parameter (Welti et al., 2018a, 2018b). The lack of correlation probably results from INP₋₁₀ having different sources from the majority of similar-sized aerosol particles and the minute share of INP₋₁₀ among them. Aerosol properties more specifically related to biological INPs, such as particle fluorescence, are available only for a very limited number of short studies in combination with measurements of INPs active well below -10 °C (Huffman et al., 2013; Prenni et al., 2013).

Perhaps, the distribution of vegetation and climatic zones is a more applicable predictor of INP_{-10} . Schnell and Vali (1973) discovered that the number concentration of INP_{-10} produced by decaying leaves increases from the tropics to temperate rainy environments and is highest in zones with a cold winter. In addition, a corresponding trend in the atmospheric concentration of INP active at -15 °C was identified by Schnell and Vali (1976) for measurements available at that time and from which they had excluded those made under snowbound conditions. Frequent freeze-thaw cycles excert selective pressure on diverse

populations of microorganisms to the advantage of those that are icenucleation active (Wilson et al., 2012). In addition, microorganisms can substantially increase the production of ice-nucleation active molecules when growing at cold temperature (Yang et al., 2022). Therefore, colder environments may harbour a larger fraction of ice-nucleation active microorganisms.

Geographical boundaries of climatic zones are shifting (Beck et al., 2018). The same probably applies to vegetation as the source of INP_{-10} . A higher plant productivity and expanding forest cover is already observed in the far North of Russia (Schaphoff et al., 2016). However, aerosolisation of INP-10 from standing or decaying vegetation is also modulated by other drivers that are subject to climate change. A major driver of INP aerosolisation is rainfall (Bigg and Miles, 1964; Huffman et al., 2013; Mignani et al., 2021; Gong et al., 2022), but not snowfall (Hara et al., 2016; Conen et al., 2017a); a major suppressor is snow cover (Conen et al., 2017a; Creamean et al., 2018). Large-scale continuous observations are necessary to reveal likely changes in the atmospheric concentration of INP-10 concurrent with shifts in climate zones and changing weather pattern. Such observations should be designed to address specific needs of climate models, which are indispensable to unravel the sign and magnitude of complex INP-cloud-climate interactions (Burrows et al., 2009). Here, we summarise available data on INP₋₁₀ with the objective to identify large scale and coarse temporal patterns that may inform the planning of future observations.



Fig. 1. Location of observatories at which INP concentrations were measured that are summarised in this study. The dotted line indicates the Arctic Circle.

2. Material and methods

We collated datasets from 11 observatories (Fig. 1) in the Northern Hemisphere reporting on INPs active between -8 and -12 °C for both the colder and the warmer half of the year. They include datasets available under a Creative Commons License (CC BY 4.0 or CC BY 3.0) plus own datasets acquired during earlier studies. All field studies during which these data were acquired, except the one in Yakutsk, are described and discussed in the publications referred to in Table 1.

At nine observatories procedures were very similar. Whereever vegetation was present, air was sampled from above the canopy, including trees. Particulate matter $<10 \ \mu m$ (PM₁₀) was collected on Quartz fibre filters and millimetre-size pieces of these filters were analysed for INPs in immersion freezing assays. Size cut-off is not mentioned in the studies reporting data for Ny-Alesund and Alert (Wex et al., 2019a), but data at Ny-Alesund was definitively derived from PM₁₀ samples as reported later by Rinaldi et al. (2021). In contrast, sampling at Alert was probably different. Rodriguez et al. (2020) report to have sampled total suspended particles at the same observatory in Alert as Wex et al. (2019a), with an instrument that shares characteristics (custom-built, flow rate, filter-size, location) with the instrument mentioned in Wex et al. (2019a). Therefore, the concentration of INP_{-10} could be somewhat larger at Alert than might have been, if PM₁₀ was sampled. Also, the data from Hyytiälä (Schneider et al., 2021) is potenitally higher than it might have been, if the same procedures had been followed as at the nine observatories mentioned before. The use of membrane filters at Hyytiälä is unlikely to have made a difference. Yet, sampling below the canopy may have resulted in a higher concentration of INP_{-10} as compared to sampling above the canopy (Seifried et al., 2021).

In Yakutsk, aerosol particles were collected on Quartz fibre filters (Pallflex® Tissuequartz, effective diameter 38 mm, Pall Corporation, Port Washington, NY, USA) using a low-volume sampler (PQ100, BGI, Mesa Labs Inc., Butler, NJ, USA) with a PM₁₀ inlet operating at a flow rate of 16.7 L min⁻¹. The sampler was situated with its inlet about 5 m above ground level on the roof of a house in the southern outskirts of the city of Yakutsk. Eventual air pollution is unlikely to affect INP concentration (Chen et al., 2018). Twenty aerosol particle samples with an average sampling time of 10 h were collected throughout the year 2018 (more details in Supplement A) and analysed for INPs following the method described in Conen et al. (2012). Briefly, 72 filter pieces with 1 mm diameter were cut from each filter sample and each piece was immersed in high-purity water (100 µL) in an Eppendorf safe-lock tube (0.5 mL) and exposed to decreasing temperature $(0.33 \text{ C min}^{-1})$ in a cold bath. The number of frozen tubes was counted by eye after each 1 °C step. Cumulative number concentration of INPs were calculated as proposed by Vali (1971). Blank (background) values were determined for five filters by analysing filter pieces with 1 mm diameter from their 5 mm wide fringe, which had been clamped into the filter holder during sampling and was not exposed to the airstream. We subtracted the median of blank values from the median of sample values.

The coarse pattern we intended to identify concern differences in the median INP concentration between observatories. Hereby, we distinguished between the warmer (01. May to 31. October) and the colder (01. November to 30. April) part of the year. A median does justice to the log-normal distribution of INPs (Welti et al., 2018a), where it is equal to the geometric mean (x^{-*}). Further, the median can accomodate values below and above detection limit as long as each category constitutes <50% of the total number of values. The log-normal distribution of INPs is best characterised by the multiplicative standard deviation (s^{*}), estimated robustly as: [upper quartile/lower quartile]^{0.741} (Limpert et al., 2001). Approximately 68% of all values are between x^{-*} times s^{*} and x^{-*} divided by s^{*} , and around 95% of all values are between x^{-*} times (s^{*})² and x^{-*} divided by (s^{*})² (Limpert et al., 2001).

3. Results and discussion

There is a large variation between observatories and seasons in terms of the number of samples available (Table 1). However, the multiplicative standard deviation (s^*) of the median did not systematically change with sample number. Values of s^* for INP₋₁₀ ranged from 2.2 (Utqiagvik, May–October, n = 20) to 9.3 (Chaumont, November–April, n = 36). In 10 of 22 cases s^* had a value between 2.5 and 3.6, and in 5 cases it could not be estimated because the lower or upper quartile of the samples had values outside the detection limits (May–October: Villum and Alert; November–April: Villum, Ny-Alesund, and Hyytiälä) (Fig. 2).

The median concentration of INP₋₁₀ differed substantially between observatories and at half of the observatories also between the warmer and the colder half of the year. Earlier, the work of Schrod et al. (2020) had revealed "*fairly similar*" average concentration of INPs at four observatories, including two in the tropics, one in the midlatitudes, and one in the Arctic. Annual cycles were not observed in any of these two-year time series (Schrod et al., 2020). These different findings could be because Schrod et al. (2020) investigated INPs across a lower temperature range (-20 to -30 °C) where potential sources might be more evenly distributed. Also, aerosolisation, activation and deposition might vary less between seasons than seems to be the case with INP₋₁₀.

3.1. May to October

Most measurements made in northern Eurasia during the warmer half of the year had values of $INP_{-10} > 10 \text{ m}^{-3}$ (Table 1), except in Scandinavia (Birkenes, Hyytiälä), where such concentrations occurred only in around 30% of the samples. Median values at the five observatories in northern Eurasia ranged from 4 to 204 INP_{-10} m⁻³, with higher values at more continental sites. At three observatories within the same climatic zone (Novosibirsk, Chaumont, Birkenes) median values increased by an order of magnitude from the coastal (Birkenes) to the extremely continental site (Novosibirsk), with the central European site in between. Highest concentrations were found at the easternmost observatory, in the Central Yakutian Lowland (Yakutsk), a permafrost region experiencing very cold winters, but nevertheless vegetated with larch, birch and meadows. This apparent West-East gradient in INP₋₁₀ across northern Eurasia (Fig. 2, left-hand panel) resembles that of bacteria in near-surface air (Burrows et al., 2009), where values also increase from western Europe through Siberia to the Central Yakutian Lowland. In contrast, modeled surface concentration of INPs active at -15 °C associated with K-feldspar (Vergara-Temprado et al., 2017) shows a two orders of magnitude lower value in the Central Yakutian Lowland, as compared with central Europe. Feldspar and other mineral particles are the predominant form of atmospheric INPs active in the colder half of the heterogeneous freezing range (Suski et al., 2018; Huang et al., 2021; Brunner et al., 2021). Because of the opposite West-East trends in INPs active at warmer and at colder temperatures the full INP spectrum (-3 to -38 °C) is probably steep in the western and shallow in the eastern part of northern Eurasia. Yet, little of that is visible in the narrow temperature window from -8 to -12 °C (Table 1). Exponential regressions fitted to INP as a function of temperature indicate an increase in INPs by a factor of only 1.5 and 1.6 per 1 degree of cooling (° C^{-1}) at Chaumont and Birkenes and a larger factor at Yakutsk $(1.9 \ ^{\circ}C^{-1})$ and Novosibirsk $(1.9 \ ^{\circ}C^{-1})$, but the factor at Hyytiälä was similarly large (2.0 $^{\circ}C^{-1}$). Still, these trends can not be extrapolated to much colder temperatures because an INP spectrum does not necessarily follow a simple exponential function from -3 to 38 °C (Petters and Wright, 2015; Creamean et al., 2018; Testa et al., 2021).

Overall, median concentration of INP_{-10} between May and October was up to two orders of magnitude higher in northern Eurasia as compared to the Arctic, where values of $INP_{-10} \geq 10 \ m^{-3}$ occurred in only 3 of 47 measurements (Table 1). Nevertheless, the smallest median observed in northern Eurasia (3.9 m^{-3}), at a coastal site (Birkenes), was very close to the largest median found in the Arctic region, also at a

Table 1

4

Summarised observations of INPs active between -8 and -12 °C. Köppen-Geiger climate class is taken from a map with 0.0083° resolution produced by Beck et al. (2018).

Site description							Observations of ice nucleating particles (INPs)								
Observatory	Year	Latitude	Longitude	Altitude (m a.s.l.)	Köppen-Geiger climate class	Months	n	Median concentration (m ⁻³)					Fraction of observations with INPs at -10 °C		[reference of dataset]
								−8 °C	−9 °C	−10 °C	−11 °C	−12 °C	$\geq 1m^{-3}$	$_{m^{-3}}^{\geq 10}$	
Yakutsk	2018	61.97° N	129.67° E	100	Cold, no dry season, very cold	May - Oct	13	37.1	109.1	204.3	324.6	541.2	1.00	1.00	This study
					winter	Nov - Apr	7	7.6	27.9	27.1	63.2	87.2	0.86	0.86	[This study, Supplement
						ratio		4.9	3.9	7.5	5.1	6.2			A]
Novosibirsk	2016-2017	54.85° N	83.10° E	150	Cold, no dry season, warm summer	May - Oct	30	15.1	38.9	68.6	115.8	n.a.	1.00	0.97	Conen et al., 2017b
						Nov - Apr	26	1.6	4.5	7.5	13.0	18.6	1.00	0.34	[This study, Supplement
						ratio		9.4	8.7	9.1	8.9	n.a.			B]
Chaumont	2012-2013	47.05° N	06.98° E	1136	Cold, no dry season, warm summer	May - Oct	36	5.9	9.0	11.2	16.0	28.7	1.00	0.58	Conen et al., 2015
						Nov - Apr	36	1.5	3.0	8.5	12.8	19.8	0.72	0.44	[This study, Supplement
						ratio		3.9	3.0	1.3	1.3	1.4			C]
Hyytiälä	2018-2019	61.85° N	24.29° E	181	Cold, no dry season, cold summer	May - Oct	99	1.3	3.2	5.7	11.7	21.9	0.96	0.28	Schneider et al., 2021
						Nov - Apr	117	n.a.	0.8	2.2	5.1	7.8	0.66	0.06	[Schneider et al., 2020]
						ratio		n.a.	3.9	2.6	2.3	2.8			
Birkenes	2013-2014	58.38° N	08.25° E	219	Cold, no dry season, warm summer	May - Oct	31	1.5	3.0	3.9	6.5	10.2	1.00	0.29	Conen et al., 2017a
						Nov - Apr	35	2.6	4.3	6.9	8.8	12.0	0.94	0.43	[This study, Supplement
						ratio		0.6	0.7	0.6	0.7	0.9			D]
Ny-Ålesund	2012	78.91° N	11.88° E	40*	Polar, tundra	May - Oct	10	0.46	1.11	2.27	3.76	5.61	0.80	0.00	Wex et al., 2019a
						Nov - Apr	3	0.12	0.12	0.23	0.36	0.60	0.00	0.00	[Wex et al., 2019b]
						ratio		4.0	9.5	9.7	10.6	9.3			
Villum	2013, 2015	81.60° N	16.67° W	24	Polar, tundra	May - Oct	4	0.34	2.59	n.a.	n.a.	n.a.	0.75	0.50	Wex et al., 2019a
						Nov - Apr	7	0.07	0.27	0.64	1.04	1.79	0.29	0.14	[Wex et al., 2019c]
						ratio		5.2	9.6	n.a.	n.a.	n.a.			
Alert	2015-2016	82.50° N	62.37° W	200	Polar, tundra	May - Oct	13	0.77	1.85	3.47	6.69	8.09	0.62	0.00	Wex et al., 2019a
						Nov - Apr	25	0.37	0.79	1.52	2.28	2.78	0.56	0.04	[Wex et al., 2019d]
						ratio		2.1	2.4	2.3	2.9	2.9			
Utqiagvik	2012-2013	71.30° N	156.77° W	60	Polar, tundra	May - Oct	20	0.36	0.83	1.63	2.61	3.64	0.70	0.05	Wex et al., 2019a
						Nov - Apr	21	0.02	0.05	0.15	0.41	0.32	0.19	0.00	[Wex et al., 2019e]
						ratio		18.1	16.3	10.6	6.4	11.4			
Jungfraujoch	2012-2013	46.55° N	07.98° E	3580	Polar, tundra	May - Oct	47	1.14	2.50	3.95	5.43	7.73	0.85	0.15	Conen et al., 2015
						Nov - Apr	36	0.03	0.19	0.47	0.65	0.91	0.22	0.03	[This study, Supplement
						ratio		38.0	13.2	8.4	8.4	8.5			E]
Cape Verde	2008-2013	16.85° N	24.87° W	30	Arid, desert, hot	May - Oct	210	0.27	0.55	0.96	1.57	2.44	0.50	0.05	Welti et al., 2018a
						Nov - Apr	284	0.27	0.64	1.36	2.20	3.38	0.58	0.02	[Welti et al., 2018b]
						ratio		1.0	0.9	0.7	0.7	0.7			

* From Rinaldi et al. (2021).



Fig. 2. Median atmospheric INP_{-10} concentration at 11 observatories alligned by longitude. Observatories are colour-coded according to the Köppen-Geiger climate class that corresponds with their location (Beck et al., 2018). Error bars (multiplicative standard deviation) indicate the range including approximately 68% of the observed values. For the observatory Villum available measurements did not provide for determining a median value for May to October, and for some observatories it was not possible to estimate the multiplicative standard deviation in both seasons because more than a quarter of samples were either above or below detection limit.

coastal site (Alert, 3.5 m^{-3}). Values of INP₋₁₀ differed little between Arctic sites and were like those at the high altitude observatory Jung-fraujoch (3580 m a.s.l.) in central Europe and at the Cape Verde Atmospheric Observatory (30 m a.s.l.) in the subtropical marine environment of Cabo Verde.

3.2. November to April

During the colder half of the year, four of five low-altitude observatories in northern Eurasia reported values $\geq 10~\mathrm{INP}_{-10}~\mathrm{m}^{-3}$ for more than one-third of all measurements and all observatories had at least two-thirds of all measured values $\geq 1~\mathrm{INP}_{-10}~\mathrm{m}^{-3}$. Difference within northern Eurasia was within one order of magnitude in the colder half of the year, except for the high-altitude observatory Jungfraujoch with values similar to those at Arctic observatories. At the three observatories within the same climatic zone (Novosibirsk, Chaumont, Birkenes), median values were almost identical (6.9 to 8.5 $\mathrm{INP}_{-10}~\mathrm{m}^{-3}$) (Fig. 2b, right-hand panel). Overall, median values in northern Eurasia still tended to be one order of magnitude larger than those observed in the Arctic during the same time of the year, where most measured values were $< 1~\mathrm{INP}_{-10}~\mathrm{m}^{-3}$.

3.3. Seasonality

Substantially smaller (> factor of 7) median values of INP_{-10} during the colder half as compared to the warmer half of the year were found at the two most continental observatories (Yakutsk, Novosibirsk), two (perhaps three) Arctic observatories (Ny-Ålesund, Utqiagvik, [Villum]) and at the high altitude observatory (Jungfraujoch) (Table 1). Warming probably results in an increased INP-10 median concentration at these sites during the colder half of the year, partly because INP emissions are strengthened when bare land is exposed for longer due to reduced snow cover (Irish et al., 2019), and partly because the potential sink of INP_{-10} is weakened. Under warmer conditions INP_{-10} are less likely to be activated and deposited with the ice crystals they have assisted to form (Stopelli et al., 2015), as indicated in an earlier study at the high altitude observatory Jungfraujoch (Conen et al., 2022). Minor seasonality was observed at the marine (Cape Verde) and at the less continental observatories (Chaumont, Hyytiälä). There, warming alone may not affect INP_{-10} concentration as much as it might do at the other observatories.

The same probably applies to the coastal observatory (Birkenes), where the median value of INP_{-10} was slightly larger in the colder as compared to the warmer part of the year. Rainfall is the main driver of INPs at Birkenes and was more abundant during the colder part of the measurement period (Conen et al., 2017). Interestingly, the same observation, a rain-driven abundance of INP_{-10} and a higher median value during the colder part of the year, was made by Gong et al. (2022) at Punta Arenas, a coastal site in the very South of Chile.

3.4. Synthesis and implications

Coarse patterns in the atmospheric concentration of INP₋₁₀ are apparent in space and time. During the warmer half of the year INP_{-10} likely start to accumulate in air masses once they have passed the coast and move into northern Eurasia, possibly tilting INP spectra to increasingly shallower slopes from West to East. Already a one-day passage above land can markedly increase atmospheric concentration of biological INPs (Conen et al., 2016). Further, increasingly colder climates from West to East (Beck et al., 2018) most likely provide for stronger sources of INP-10 in eastern as compared to western parts of northern Eurasia (Schnell and Vali, 1973, 1976; Yang et al., 2022). Although INP₋₁₀ are efficiently removed from clouds with precipitation (Stopelli et al., 2015), they are at the same time aerosolised when rainfall impacts on land (Bigg and Miles, 1964; Huffman et al., 2013; Mignani et al., 2021; Gong et al., 2022). The overall effect can be a net increase in the atmospheric concentration of biological INPs, while simultaneously that of mineral INPs active at cold temperatures is decreased (Testa et al., 2021). The net accumulation of INP-10 appears to be limited during the colder half of the year, when precipitation is more likely to reach land surface in form of snowfall and large areas in the interior of the continent are snow-covered. Nevertheless, the concentration of INP_{-10} remains much higher in the eastern part of northern Eurasia as compared to the Arctic.

Storm tracks entering the Arctic from the midlatitudes increase in number and intensity because of climate change (Zhang et al., 2004; Bintanja et al., 2020). Midlatitude air masses advected by such storms have the potential to raise the concentration of INP_{-10} at least temporarily. Based on the patterns observed in this study, we expect storms to have a large impact on Arctic INP_{-10} concentration when air masses have been in contact with eastern parts of northern Eurasia. A smaller impact is expected when air masses have entered from western or northern parts of Europe, particularly so during the warmer half of the year, when the relative difference in INP_{-10} concentration between Europe and the Arctic seems to be comparatively small. However, as isotherms shift upwards also at the height of mixed-phase Arctic clouds (typically 0.5 to 2 km; Morrison et al., 2012), the likelihood declines that INP_{-10} are activated under ambient conditions. Changes in storms and isotherms combined, the frequency of primary ice formation at around -10 °C may change little in Arctic clouds. In contrast, where the concentration of INP_{-10} is already high and not expected to change, such as in the Central Yakutian Lowland, the sole upward shift of isotherms may reduce cloud ice formation and result in a negative cloud feedback (Murray et al., 2021).

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All INP data used in this study is either available from data repositories or as supplementary material to this study.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosres.2023.106645.

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