

Vertically-resolved source contributions to climate-relevant aerosol properties in Southern Greenlandic fjord systems

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15 **Abstract.**

Greenlandic fjords are rapidly changing environments where ocean, ice, land, and atmosphere interact, yet aerosol sources and their role in cloud formation remain relatively unconstrained. To address this gap, we conducted an intensive field campaign in Narsaq, Southern Greenland, during summer 2023 as part of the *GreenFjord* project, combining ground-based sampling with a tethered balloon (24 flights to ~700 m). Over six weeks, we measured meteorology, aerosol [number](#) size distributions, particle and CCN number concentrations, as well as optical properties. Aerosol characteristics varied markedly in time and altitude, reflecting four main sources: fresh anthropogenic pollution, aged background aerosols from local anthropogenic and marine origins, transported biomass burning, and new particle growth events. Local pollution dominated ~50% of the campaign, while growth events and Canadian wildfire plumes each contributed ~8%; the remainder (~34%) reflected aged marine background aerosols. Number size distributions were typically Aitken-mode dominated, presumably due to the frequently observed growth events. Biomass burning advection, in contrast, was marked by accumulation-mode particles. During plume periods, median CCN concentrations at 0.5% supersaturation increased by a factor of 1.7-3.7 relative to median background concentrations, while median absorption and scattering coefficients increased by factors of 1.8-4.0 and 1.4-4.8, respectively. The enhancement factors are similar to or even exceed the enhancements from local anthropogenic pollution and highlight the substantial role that long-range transported biomass burning may have in modulating aerosol–cloud–radiation interactions over southern Greenland.

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1 Introduction

Greenland hosts the world's second-largest ice sheet and is undergoing rapid transformation due to climate change. The Greenland Ice Sheet currently discharges $\sim 1000 \text{ Gt yr}^{-1}$ of freshwater into coastal oceans primarily via fjord systems (Bamber et al., 2012). In the last two decades alone, freshwater runoff and glacial discharge in Southern Greenland have increased by nearly 50 % (Bamber et al., 2012; Böning et al., 2016; Sejr et al., 2017), significantly impacting the marine (Meire et al., 2017; Oksman et al., 2022), terrestrial (Wrona et al., 2016), and atmospheric components of fjordic ecosystems (Meire et al., 2015). Arctic coastal regions are hotspots of chemical and biological activity (Schmale et al., 2024) with increasing freshwater input from glaciers altering fjord dynamics, stratification, sedimentation, and bio-productivity (Arrigo et al., 2017; Holding et al., 2019; Juul-Pedersen et al., 2015; Meire et al., 2016, 2017, 2023; Mortensen et al., 2013, 2014). These changes also have critical implications on regional greenhouse gas fluxes and potentially influence aerosol formation and cloud properties in the Arctic (Feltracco et al., 2021; Meire et al., 2015; Schmale et al., 2024; Verdugo et al., 2022). For example, Holding et al. (2019) found that fjords with marine-terminating glaciers are more bio-productive than land-terminating glacier fjords where only streams enter the fjord. It is hence conceivable that marine-terminating glacier fjords lead to relatively higher emissions of marine biogenic compounds in two ways: either by emission of volatile organic compounds (VOCs) such as dimethylsulfide (DMS), which act as precursors to aerosol formation (Beck et al., 2020), or as direct, primary emissions of biogenic aerosols through sea spray emissions (Fahlgren et al., 2015). Hence fjord systems can be important local contributors to the aerosol population. Understanding how glacial retreat affects aerosols and clouds is essential for assessing climate feedbacks comprehensively in this rapidly evolving environment, particularly in fjordic ecosystems that sit at the intersection of the ocean, land, cryosphere, atmosphere, and biosphere. These interconnected systems are especially sensitive to climate change, with each component responding acutely to the pressures of rising global temperatures.

While geographically remote, southern Greenland is influenced by a combination of local anthropogenic emissions and long-range transported pollutants. Both source types inject aerosols at different altitudes throughout the atmosphere, and paired with frequent temperature inversions in the boundary layer, add an additional layer of complexity to fjord-atmosphere interactions. Local emissions from urbanised areas may contribute to aerosols in the region, despite the relatively low population density. These emissions most likely arise from combustion processes, such as those associated with transportation and residential activities. A recent study in Qaanaaq, Greenland, demonstrated that open waste burning can significantly elevate local $\text{PM}_{2.5}$ levels, underscoring the potential for measurable local emission contributions even in sparsely populated Greenlandic settlements (Yasunari et al., 2024). Beyond local anthropogenic sources, long-range transport of pollution from more industrialized regions—particularly the East coast of the United States (Quennehen et al., 2011; Schmale et al., 2011), also affects the aerosol composition over Greenland. In addition, it has been shown that also emissions from biomass burning in North America contribute episodically and significantly to specifically absorbing aerosol concentrations in Greenland during the summer months (Dibb et al., 1996; Thomas et al., 2017), affecting the melt of the ice sheet. With increasing incidences of boreal forest fires in Canada due to a warming climate (Kirchmeier-Young et al., 2017; Wang, 2024), emissions

from biomass burning (BB) are expected to have an increasingly important influence (Flannigan et al., 2006; Gramlich et al., 2024; Kommula et al., 2024; Soja et al., 2007).

Understanding of aerosol sources in Southern Greenland is crucial, as they directly affect the availability of cloud condensation nuclei (CCN), which in turn play a key role in cloud formation. CCN influence cloud properties by modulating droplet size distributions, cloud phase, lifetime, and albedo (e.g., Fan et al., 2016), ultimately impacting surface radiative fluxes and the surface energy balance. This is of particular importance during Arctic summer, when surface melt of the Greenland Ice Sheet is at its peak. Low-level mixed-phase and liquid clouds, which are common during summer, have been shown to significantly affect the surface energy budget increasing surface melt (Hofer et al., 2019; Niwano et al., 2019; Ryan et al., 2022; Van Tricht et al., 2016). However, more recent analysis by Ryan (2024) suggests that, over the period of 2002-2023, cloud radiative forcing exerted only a limited influence on ice sheet melt. This contrast highlights the need to better understand the processes governing cloud properties. Since a particle's ability to act as a CCN is dependent on the particle size and composition, it is important to understand aerosol sources and processes relevant for Greenland. This understanding will allow us to better predict Arctic warming consequences such as ice sheet melt and glacial retreat.

In this study, we discuss results from the atmospheric part of the project 'Greenlandic Fjord ecosystems in a changing climate: Socio-cultural and environmental interactions' (*GreenFjord*). *Greenfjord* is a multi-disciplinary program that aims to better understand how climate change is reshaping Southern Greenlandic fjord ecosystems from both an environmental and a social perspective. Here we elucidate processes which influence the aerosol characteristics, specifically CCN and optical properties, in Southern Greenland during summer. We seek to answer how local, remote, natural and anthropogenic aerosol sources contribute, taking into account the vertical structure of the boundary layer.

2 Methods

2.1 Sampling site

The field campaign took place from June 16th to August 5th 2023, in Narsaq, Greenland with remote sensing, and in-situ ground-based and vertical observations via tethered balloon (helikite)-supported measurements. Airborne measurements took place between 25th June and 2nd August 2023. The ground station and helikite base were established at the Narsaq International Research Station, hereafter referred to as NIRS, (lat: 60.92°, lon: -46.05°, 17 m a.s.l), an independent non-profit research platform. A schematic of the region with station and sampling locations is given in Fig. 1.

Narsaq is primarily a fishing town, and agriculture is also practised. It is the ninth largest town of Greenland, and home to an estimated 1,285 people, 2.27 % of Greenland's population. The town holds around 230 motor vehicles, 120 of which are cars, and electricity is provided by a 70 km distant hydropower plant (Statistics Greenland, 2024). The region immediately surrounding Narsaq is only accessible by boat with few land-based transport routes. Narsaq is situated at the intersection

between two fjords: Nordre Sermilik fjord, and Tunulliarfik fjord, and is therefore, surrounded on three sides by water. [Directly](#)
95 [Approximately 2 km](#) east of Narsaq lie the Tasiigaaq and Qaqqarsuaq mountains, rising to 364 m and 685 m, respectively. To
the northeast, the surrounding mountain range includes peaks approaching 1,400 m.



Figure 1: Map of the region around Narsaq with insets highlighting the region's location in Southern Greenland and a zoomed in view on Narsaq. Maps with blue background are from <https://maps.greenmin.gl/geusmap/>, the satellite image is from © Google Earth.

100 2.2 Ground-based measurements

The atmospheric measurement hut was equipped with four inlets: one PM₁ [interstitial](#) inlet with a flow of ~15 L min⁻¹, two total suspended particle inlets with a flow ~15 L min⁻¹, and a third total suspended particle inlet with a flow rate of ~100 L min⁻¹ (all inlet heads by Digital Elektronik AG). [The particle cutoff for the interstitial](#)
105 [interstitial and total inlets were 1 μm and 40 μm respectively.](#) A schematic of the instrumental and inlet set up is provided in Fig. [S24](#). An overview of all ground-based

instruments used in this work can be found in Table 1.

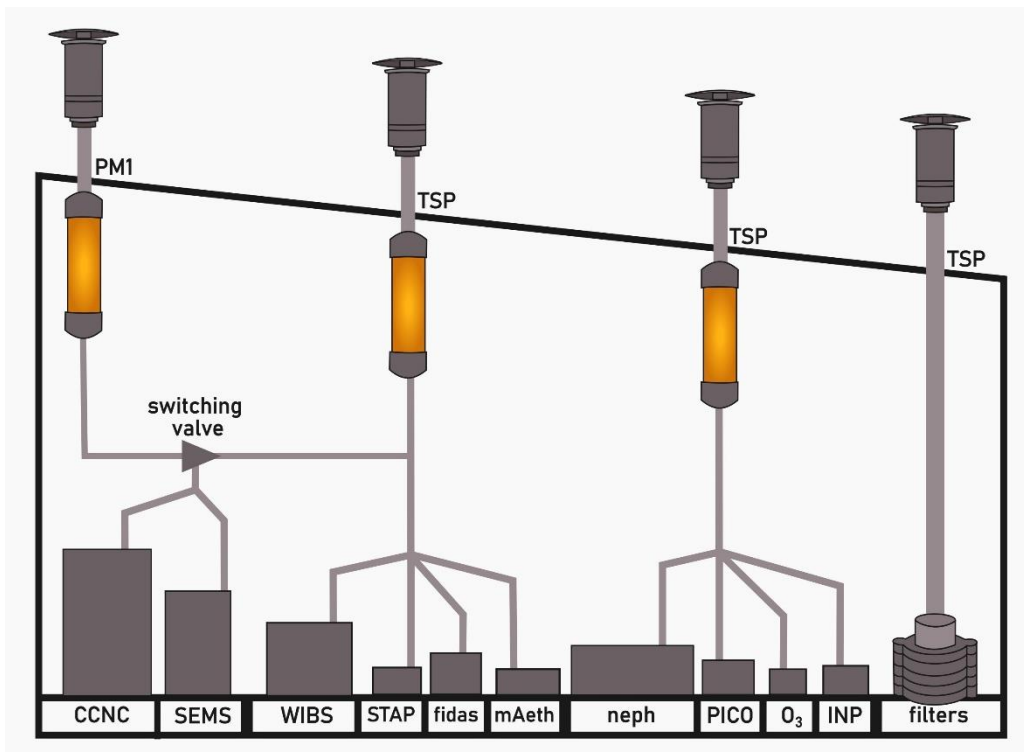


Figure 2. Schematic of the instrumental setup inside the measurement hut at NIRS. All four aerosol inlets are shown and their connections, via Silicagel dryers (orange), to the instruments. CCNC: Cloud Condensation Nuclei Counter; SEMS: Scanning Electrical Mobility Spectrometer; WIBS: Wideband Integrated Bioaerosol Sensors; STAP: Single-channel tricolor absorption photometer; fidas: Fidas Frog fine dust monitor; mAeth: micro aethalometer; neph: nephelometer; PICO: MIRA Pico Nitrous Oxide and CO gas analyser; O₃: ozone monitor; INP: Ice Nucleating Particle filter sampler; filters: cascade impactor.

Inlets 1 to 3 were fitted with hygrometers (model HC2, Rotronic AG, Bassersdorf, Switzerland) in a flow bypass to measure temperature (T) and relative humidity (RH). Silica gel dryers before the hygrometers were intended to reduce the ambient RH sampled by the instrumentation to below 40 %. However, due to the consistently high ambient humidity, it was not always possible to sufficiently reduce the ambient RH within the sample line. Note, inlet air T and RH are given in the data files (see data availability). All inlets were positioned directly upwards through the roof of the hut with a clearance of 1.5 m. Inlets 1 (interstitial) and 2 (total) were connected to a valve that could be switched to allow quantification of both activated and non-activated particles in the case of ground-level fog. Without fog, inlet 2 was used as standard. Ground-level fog events were rare with interstitial inlet data representing less than 1 % of the aerosol data. As such, we only report here on the total inlet data.

Table 1. List of instruments at NIRS ground station and in the helikite payload as well as measured variables.

Instrument	Ground/Helikite	Inlet no.	Manufacturer (Model)	Variable	Reference
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Scanning Electrical Mobility Spectrometer (SEMS)	Ground	1/2	Brechtel Manufacturing, Inc, USA (model 2100)	Particle number size distribution (8 – 1000 nm)	(Brechtel, 2017)
micro Aethalometer (mAeth)	Ground	2	Aethlabs (model MA200)	Equivalent black carbon mass (375, 470, 528, 625 and 880 nm)	(Liu et al., 2021a; Salo et al., 2024)
Fidas Frog fine dust measuring device	Ground	2	Palas GmbH	Particle number concentration and size distribution (0.19 – 93 μm)	(Bächler et al., 2021; Fang et al., 2023).
Cloud Condensation Nuclei Counter (CCNC)	Ground	1/2	Droplet Measurement Technologies, Inc (model CCN-100)	CCN number concentration at the following supersaturations: 0.1, 0.15, 0.2, 0.5 and 1 %	(Roberts and Nenes, 2005)
Nephelometer	Ground	3	Acoem	Light scattering coefficient (σ_{sc}) and backscatter coefficient (σ_{bsc}) at 450, 525 and 625 nm	(Müller et al., 2011)
Lidar Ceilometer (CL61)	Ground	-	Vaisala (model CL61)	Attenuated backscatter (β) and linear depolarisation ratio at 910.55 nm	(Vaisala, 2022)
Atmos 41 Gen 2 Weather Station	NIRS roof	-	METER	T, RH, P, wind/gust speed, wind direction, solar radiation, precipitation, lightning strike count and distance	(Dombrowski et al., 2021)
INP filter sampler	Ground	3	Built in-house	INP concentration (not reported here)	
Wideband Integrated Bioaerosol Sensor (WIBS)	Ground	2	Droplet Measurement Technologies, Inc	Particle number concentration and size distribution of fluorescent bioaerosols (0.5 – 30 μm , not reported here)	(Beck et al., 2024; Gabey et al., 2010; Savage et al., 2017)
Ozone monitor	Ground	3	2B Technologies (model 205, dual beam)	Ozone mixing ratio (not reported here)	(Johnson et al., 2014)
MIRA Pico CO/N ₂ O Gas Analyser	Ground	3	Aeris Technologies, Inc	H ₂ O vapour, CO and N ₂ O mixing ratio (not reported here)	(Scherer et al., 2019)
HFI stage impactor	Ground	4	TSI Inc. (model 131A)	Chemical analysis (IC ^a , ICP-MS ^b)	(Marple et al., 1991)
Portable Optical Particle Sizer (POPS)	Ground and Helikite	2	Handix Scientific LLC	Particle number size distribution (186 – 3370 nm)	(Liu et al., 2021b; Mei et al., 2020; Pilz et al., 2022)

Single-channel tricolour Absorption Photometer (STAP, model 9406)	Ground and Helikite	2	Brechtel Manufacturing, Inc	Aerosol light absorption coefficient (σ_{abs}) at 450, 525 and 624 nm)	(Bates et al., 2013; Düsing et al., 2019; Pikridas et al., 2019; Pilz et al., 2022)
Eight-Channel filter sampler (FILT)	Ground and Helikite	2	Brechtel Manufacturing, Inc	Offline analysis – SEM-EDX ^c / TEM-EDX ^d (not reported here)	(Pohorsky et al., 2024)
CO ₂ sensor	Helikite	-	Vaisala (CARBOCAP GMP343)	CO ₂ mixing ratio	(Brus et al., 2021)
Miniaturised Scanning Electrical Mobility Spectrometer (mSEMS)	Helikite	-	Brechtel Manufacturing, Inc	Particle number size distribution (8 – 240 nm)	(Pohorsky et al., 2024)
SmartTether Weather Sonde	Helikite	-	Anasphere, Inc	Altitude, T, RH, P, wind speed, wind direction, latitude, longitude	(Pohorsky et al., 2024)
RH/T sensors	Helikite	-	Sensirion (SHT85)	RH, T	(Lorek and Garland, 2025)

^aIC: Ion Chromatography, ^bICP-MS: Inductively Coupled Plasma Mass Spectrometry, ^cSEM-EDX: Scanning Electron Microscopy with Energy Dispersive Xray analysis, ^dTEM-EDX: Transmission Electron Microscopy with Energy Dispersive Xray analysis.

120 2.2.1 Particle number concentration and size distributions

A Scanning Electrical Mobility Spectrometer (SEMS) and Fidas Frog (fidas) were used to measure particle number concentration (PNC) and size distributions (PNSD) for both fine and coarse mode aerosols, respectively. The SEMS is a particle size spectrometer which provides high time resolution aerosol PNSD measurements with a diameter range of 8 to 1000 nm. It is made up of a soft X-ray neutraliser, a differential mobility analyser (DMA) column, and a mixing condensation
125 particle counter (mCPC). The SEMS was run in scanning mode with 120 bins, at 1 second per bin, for both up and down scans. The DMA sheath air flow was set at 4 L min⁻¹, and the sample flow at 0.34 L min⁻¹. The effect on particle losses by tubing was considered in the SEMS software and is therefore accounted for (Brechtel Manufacturing Inc., 2021). The fidas is an optical particle counter and was used for the continuous measurements of particle number size distributions between 0.19 and 93 µm, also allowing coarse mode aerosol mass to be derived (PM₁, PM_{2.5}, PM₅, PM₁₀ and PM_{tot} (Bächler et al., 2021; Fang
130 et al., 2023)).

2.2.2 Aerosol absorption and scattering coefficients

Two instruments were implemented for measuring aerosol absorption to provide a continuous timeseries on the ground and during flights: a micro-aethalometer (mAeth) and a Single-channel Tricolour Absorption Photometer (STAP). Both instruments use filter-based absorption techniques, which measure the absorption of light by a sample filter at multiple

135 wavelengths and compare this to a clean reference filter. The STAP uses quartz-fiber glass filters to detect attenuation at 450, 525 and 624 nm, at a resolution of 1 s, with a measurement uncertainty reported by Brechtel Manufacturing Inc of 0.2 Mm^{-1} for a 60 s averaging period. The performance of the STAP has been studied previously (Bates et al., 2013; Düsing et al., 2019; Pikridas et al., 2019; Pilz et al., 2022). Note, Brechtel Inc. includes within the STAP software a correction for scattering incurred in filter-based measurements, based on correction by (Ogren, 2010).

140 The mAeth (MA200) is a portable photometer that uses deposition of aerosols onto a PTFE filter band to measure attenuation at 375, 470, 528, 625 and 880 nm (Aethlabs). Measurements were outputted at a resolution of 60 s for this work. Using SingleSpot™ mode, Aethlabs reports the limit of detection of 30 ng m^{-3} of black carbon (BC) for a 5-minute averaging period at a 150 ml flow rate. The performance of the mAeth has previously been studied (Liu et al., 2021a; Salo et al., 2024). The mAeth gives equivalent BC (eBC) values and the absorption coefficients at each wavelength can be calculated using the mass
145 absorption coefficient (MAC). The MAC value is calculated from the wavelength specific attenuation cross-section, $\sigma_{ATN}(\lambda)$, and the multiple scattering coefficient, C , given by the manufacturer as 1.3, which compensates for the enhanced scattering properties of the filters used. The MAC and absorption coefficients are calculated as follows:

$$MAC = \frac{\sigma_{ATN}(\lambda)}{C} \quad \text{Eq 1}$$

$$\sigma_{abs} = MAC [m^2 g^{-1}] \times BC [ng m^{-3}] \quad \text{Eq 2}$$

The $\sigma_{ATN}(\lambda)$ values are provided by AethLabs and are 10.120, 14.091, 17.028, 19.070 and $24.069 \text{ m}^2 \text{ g}^{-1}$ for 880, 625, 528, 470 and 375 nm, respectively. For comparison of the calculated σ_{abs} from the mAeth measurements with the measured σ_{abs}
150 from the STAP, in addition to discussion on the differences between these values, see Fig. [S2-S1](#) and associated text. Unless otherwise stated, absorption coefficients discussed in this manuscript refer to those measured by the STAP, while eBC concentrations are taken from the mAeth.

The Aurora 3000 nephelometer was used to measure scattering and backscattering coefficients with an LED light source from
155 9-170°. This instrument is a multi-wavelength integrating nephelometer and measures in-situ light scattering of aerosols at 450 nm, 525 nm and 635 nm. The time resolution during the campaign was 60 s, with the lower limit of detection given by the manufacturer as 0.3 Mm^{-1} for a 60 s averaging period. [The Nephelometer was calibrated before the campaign and periodic zero tests were conducted throughout the campaign. A span gas check and calibration were performed on 11 July 2023 but with values that fell outside the recommended range of the manufacturer. Therefore, a flag is applied to data following the](#)
160 [unsuccessful calibration although no discernible shift or step-change in the data was observed.](#)

2.2.3 Cloud condensation nuclei number concentrations

CCN concentrations were measured using a Cloud Condensation Nuclei Counter (CCNC) at a 1 second time resolution, stepping through supersaturations (SS) of 0.1, 0.15, 0.2, 0.5 and 1.0 % at a frequency in minutes of 20, 10, 10, 10 and 10

leading to a 1-hour measurement cycle. The supersaturation of 0.1 % was measured for double the time due to the increased
165 time needed to equilibrate from the drop of 1 % to 0.1 % SS. Data points where the temperature was not stable within the
instrument column were removed in postprocessing as were data which occurred within the cooling cycle at the start of the 1
hour run, and data directly after switching between the later supersaturations. This corresponds to removing the first 10 minutes
of each hour, followed by the first two minutes and last one minute of data at the start of 0.15, 0.2, 0.5 and 1 % supersaturations.
Calibration of the CCNC was performed [following the ACTRIS procedure, detailed in \(Gysel and Stratmann, 2013\) and \(Rose
170 et al., 2008\), with ammonium sulfate particles](#) both before and after the campaign to ensure supersaturation values remained
consistent, with post-campaign results showing good agreement with the pre-campaign calibration. For correlation of 1 % SS
 N_{CCN} with N_{8-1000} across the entire campaign at different activation diameter cut-offs see Fig. S23.

2.2.4 Ceilometer attenuated backscatter and linear depolarisation ratio

Measurements of attenuated back-scatter (β) and linear depolarisation ratio (LDR), providing insight into the presence of
175 hydrometeors and aerosols and the phase of water in clouds, respectively, were made by a light detection and ranging (LiDAR)
instrument with depolarisation measurement capabilities (Vaisala Lidar Ceilometer CL61). For the duration of the campaign,
the ceilometer was mounted outside NIRS and took measurements every 1 minute. The ceilometer operates at a wavelength
of 910.55 nm, sending short laser pulses vertically into the atmosphere at a frequency and duration of 9.5 kHz and 160 ns,
respectively, at a maximum average power of 50 mW. The attenuated backscatter is a measure of the proportion of emitted
180 light from the ceilometer, which is scattered back and measured by a receiver, diminished in strength by atmospheric
absorption, scattering and the natural attenuation of light, which occurs during the path of the light up through the atmosphere
and back down once scattered. The LDR is a measure of the change in polarisation of the emitted light when it is scattered
back to the receiver. Since every particle the laser light interacts with will alter light polarisation, with the exception of
spherical particles such as liquid cloud droplets, the water and ice phase of hydrometeors can be inferred from their measured
185 LDR. Spherical particles are expected to return non-depolarised light whereas non-spherical particles return significantly
depolarised light due to reflections at the solid-air interface. Multiple scattering can also occur within optically thick clouds
leading to depolarisation even if scattering particles in clouds are spherical in nature (Vaisala, 2022).

Raw data from the ceilometer, after pre-processing by Vaisala's internal processing algorithm, was then post-processed to
give a clean dataset. Values, where the attenuated backscatter were negative or where the depolarisation ratio value was outside
190 of the expected range between 0 and 1, were removed within a rolling mean smoothing window of 10 minutes and 50 metres.
Noisy data was observed in the case of low backscatter from clear sky days, or when the range of laser light is almost fully
attenuated. The LDR was found to be more susceptible to noise than β .

2.2.5 Meteorological measurements

Meteorological measurements including T, RH, atmospheric pressure (P), wind/gust speed, wind direction, solar radiation and maximum precipitation rate were made by an ATMOS41 Gen 2 weather station. Solar radiation was measured by a pyranometer, which measures total (direct and diffuse) solar radiation. The wind speed and wind direction were measured with an anemometer. The vapor pressure sensor measures RH and T, then uses these measurements to compute the vapor pressure. The sensor was protected by a Teflon enclosure to keep water and dust away from the sensor whilst measurements were being taken. RH was calculated using the following equation:

$$RH_{air} = \frac{e_{air}}{e_{sat}(T_{air})} \quad \text{Eq 3}$$

where e_{air} is the vapor pressure of the air and $e_{sat}(T_{air})$ is the saturation vapor pressure at a given air temperature.

2.3 Tethered balloon measurements

Vertical measurements were taken using a tethered helium balloon system, i.e., helikite (64 m³) and a payload of up to 25 kg (Pohorsky et al., 2024). An overview of all helikite-based instruments used in this work can be found in Table 1. The helikite payload is designed in a modular style meaning the set of instruments can be customised to suit the science question for each flight. Flights typically lasted between 3 and 5 hours, were carried out sometimes twice a day, and reached altitudes up to 760 meters. Flight patterns were flexible and primarily targeted towards specific observations, such as measurements of new particle formation, local anthropogenic emissions and elevated long-range transported pollution plumes. A list of all flights and their details is provided in Table S1. In total 24 instrumented science flights were performed. In brief, flown instruments included the following. The mSEMS is a compact, miniaturised version of the SEMS, working in the same way. The mSEMS was set to measure particles in the diameter range from 8 to 240 nm, for a sample flow rate of 0.36 L min⁻¹, scanning across 40 bins, giving a total run time of 2 minutes and 4 seconds including scan delay. For full details of characterisation of the mSEMS, see Pohorsky et al. (2024). A comparison between the mSEMS and SEMS was done in the field with good agreement in total PNC and PNSD, shown in Fig S4S3. The deployed POPS is a lightweight optical particle counter, which measures PNSD and total PNC between 186 and 3370 nm in our case (Gao et al., 2016; Liu et al., 2021b; Pilz et al., 2022; Pohorsky et al., 2024). The STAP has been described above. More information on the CO₂ probe and Smart Tether can be found in Pohorsky et al. (2024).

2.4 Data processing, pollution flagging, and environmental condition categorization

Data processing was completed using Python 3.11.7 and we report final data, where times with zero-particle filter tests, flow checks, calibrations or inlet maintenance are removed. Those activities were conducted on a 3-day rotational basis on the ground and at a daily frequency for the flight payload. All ground-based aerosol and trace gas datasets were individually flagged for local pollution. Local pollution in this context refers to sudden and short-term perturbations in the data, i.e., spikes, due to anthropogenic emissions from local activities such as vehicle exhausts or occasional firework displays. Flags were

created by first using a pollution detection algorithm (PDA), described fully in Beck et al. (2022), and then any extraneous pollution spikes missed by the algorithm were flagged manually. All pollution flags were double checked by a co-author. This
225 divides the data into a “clean” and “polluted” category with 50.4% ground SEMS data being flagged as polluted, for example. Unless when focusing on pollution, we report “clean” data. Throughout this work, the following terminology will be used to categorize and discuss different conditions:

- 230 1. *Background (BG)* – This category represents conditions primarily influenced by local marine aerosol and aged anthropogenic pollution. While specific sources cannot be individually identified, persistent local emissions are likely encompassed in this background signal. This category therefore represents a *persistent aerosol background*, against which we compare other data categories. To isolate a stable background signal, short-lived spikes from fresh anthropogenic pollution were excluded. In addition, data affected by new particle growth events, fog, and biomass burning were removed, as each of these processes can strongly perturb the background aerosol population.
- 235 2. *Biomass burning plume events (BB)* – this category contains data only from when the biomass burning plumes were identified by the ceilometer. This is regardless of what the balloon-borne vertical or ground-based aerosol data showed. As with the background category, strong, short-lived spikes in anthropogenic pollution have been removed. The plume periods are hereafter defined as the following times: 27th June 21:20 – 29th June 15:00; the 5th July 20:13 to 8th July 01:42; and the 17th July 11:47 to 18th July 18:17 (identification and description of plumes can be found in
240 Section 3.4). Because these periods are strictly time-constrained, coincident and concurrent new particle growth events have been retained within the plume category to provide a more complete representation.
- 245 3. *New particle growth events (GE)* – this category contains data only from time periods defined as growth events. We define this to include the period from the first appearance of a nucleation mode in the PNSD to 6 hours post this time, when typically the distinction over background was not recognizable anymore. As with the *background* and *BB* categories, strong, short-lived spikes in anthropogenic pollution have been removed. We have defined these events as GE rather than new particle formation (NPF) events since our instruments only observed particles > 8 nm and therefore captured the subsequent growth periods. [Growth rates were calculated for all identified events \(Table S4, Table S8\), providing quantitative confirmation of the coherent size evolution in each case.](#)
- 250 4. *Fresh pollution (pollution)* – this category contains only the short-lived spikes from anthropogenic pollution, i.e. the data removed from the previous 3 categories. [Figure S4](#) gives an example time series over one day showing the short-lived pollution spikes visible on top of the background aerosol concentrations. Examples for other conditions can [also](#) be found in Fig S45.

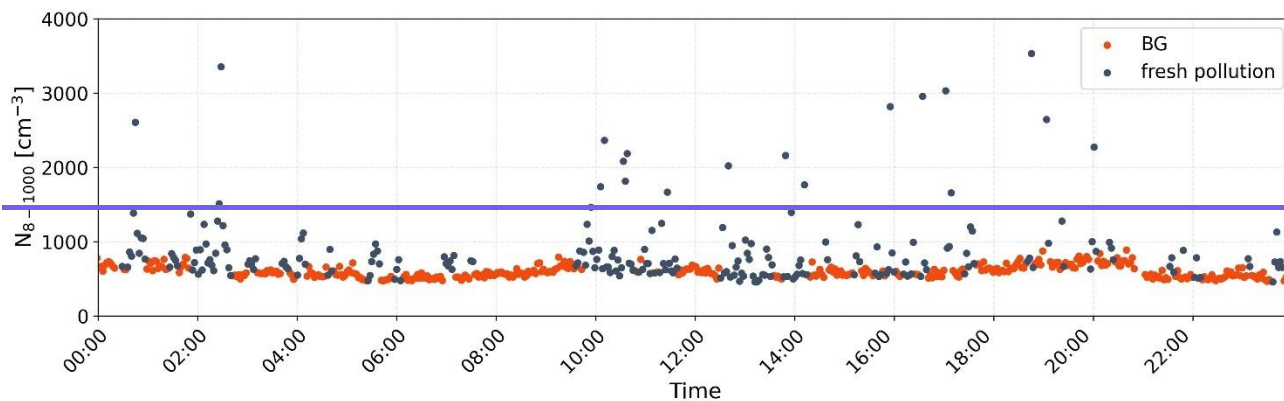


Figure 2. Time series of N_{8-1000} over 24 hours for the 25th of June showing the effect of short lived pollution spikes (navy scatter) on the aerosol background (orange scatter).

2.5 K-means clustering of particle number size distributions

The measured PNSDs from the ground SEMS and the flight mSEMS were categorized using the k-means clustering algorithm, as commonly used for clustering aerosol PNSDs (Beddows et al., 2009; Boyer et al., 2023; Dall’Osto et al., 2018; Heutte et al., 2024; Pernov et al., 2022). Because the PNSD data reflected very large ambient variability, we split the dataset a priori into the four categories - pollution, background, growth events and biomass burning - and kept ground and vertical observations separate. For each data category clustered, the number of clusters was varied between 3 and 10, and it was concluded per dataset what number best described the PNSDs. The optimum cluster number was derived using the Within Cluster Sum of Squares (WCSS) and Silhouette score (Lloyd, 1982; Morissette and Chartier, 2013; Rousseeuw, 1987; Steinley and Brusco, 2011). WCSS is a measure of cluster cohesion made through summing the squared distances between each data point and its assigned clusters centroid. The Silhouette score is another measure of the similarity of the PNSDs within each cluster, compared to other clusters. Larger values therefore indicate a high degree of similarity within a cluster, but a low degree of similarity with PNSDs in other clusters. For PNSDs, the Silhouette score will start initially at higher values and decrease as the separation between clusters decreases. The WCSS will also start high but will decrease rapidly as the addition of clusters provides little additional information describing the system. It is convention to take the point where the graph of WCSS vs cluster number starts to level off, the “elbow point”, as the ideal cluster number. From these two measures and understanding of the data and time series, the ideal cluster number per dataset was judged. WCSS is discussed further in Text S2 and Fig. S6S5. The total number of clusters per data category is detailed in Table S2. To facilitate comparison over the same particle size range for the clusters from the SEMS and the mSEMS instruments, the ground-based data clustering was run on the PNSDs including from 8 to ~240 nm to be in line with the instrumental size range of the mSEMS. [Figure S7 shows a comparison of the ground clustering done for size ranges of 8 to 1000 and 8 to 240 nm and these agree very well. Ground clustering using size ranges of 8–1000 nm and 8–240 nm produced nearly identical particle number size distributions \(PNSDs\), differing on average by less than 5%.](#)

K-means clustering was run again using aerosol PNSDs observed by the mSEMS flown in the helikite payload to compare ground and elevated PNSDs. Data within lowest 100-150 m of each profile were not considered due to the fast take-off phase, where usually only one or very few scans of the mSEMS were available, and hence there were not sufficient data.

3 Main characteristics of aerosols in Narsaq, Southern Greenland

280 3.1 Campaign overview

In 2023, Narsaq experienced an exceptionally sunny and dry summer with average temperatures of 10.4 °C, and daily values ranging between 0 and 21.1 °C (Fig. 3). Throughout the period of 2013-2023, average monthly values of temperature for June, July and August have ranged from 6.2-9.9°C (Statistics Greenland, 2025). Due to the prevalence of clear sky conditions, global solar radiation at the ground was relatively high with average daytime values of 360 W m⁻², ranging between 0 and 1046 W
285 m⁻². Wind speeds were typically low, except for a few days and individual gusts owing to katabatic winds from the ice sheet to the West (Fig. 3c). The dominant wind directions at the ground were westerly (defined here as between 247.5 and 292.5°) for ~51 % of the time, and northerly (defined here as between 337.5 and 22.5°) for 22 % of the time (see Fig. S68). The occurrences of each wind direction category can be found in Table S3. Despite dry conditions, 10 fog events were recorded (Fig. 3b), with 26.4% of their total duration occurring at ground level. Fog periods were identified primarily from ceilometer
290 data using Vaisala's *mid- and high-density liquid* classifications (see Text S3), and were further confirmed by high relative humidity measurements from ground meteorological data, along with direct visual observations. This manuscript does not further address aerosol–fog interactions. In addition, 6 rainy periods were measured. For a full description of precipitation and fog periods see Table S4.

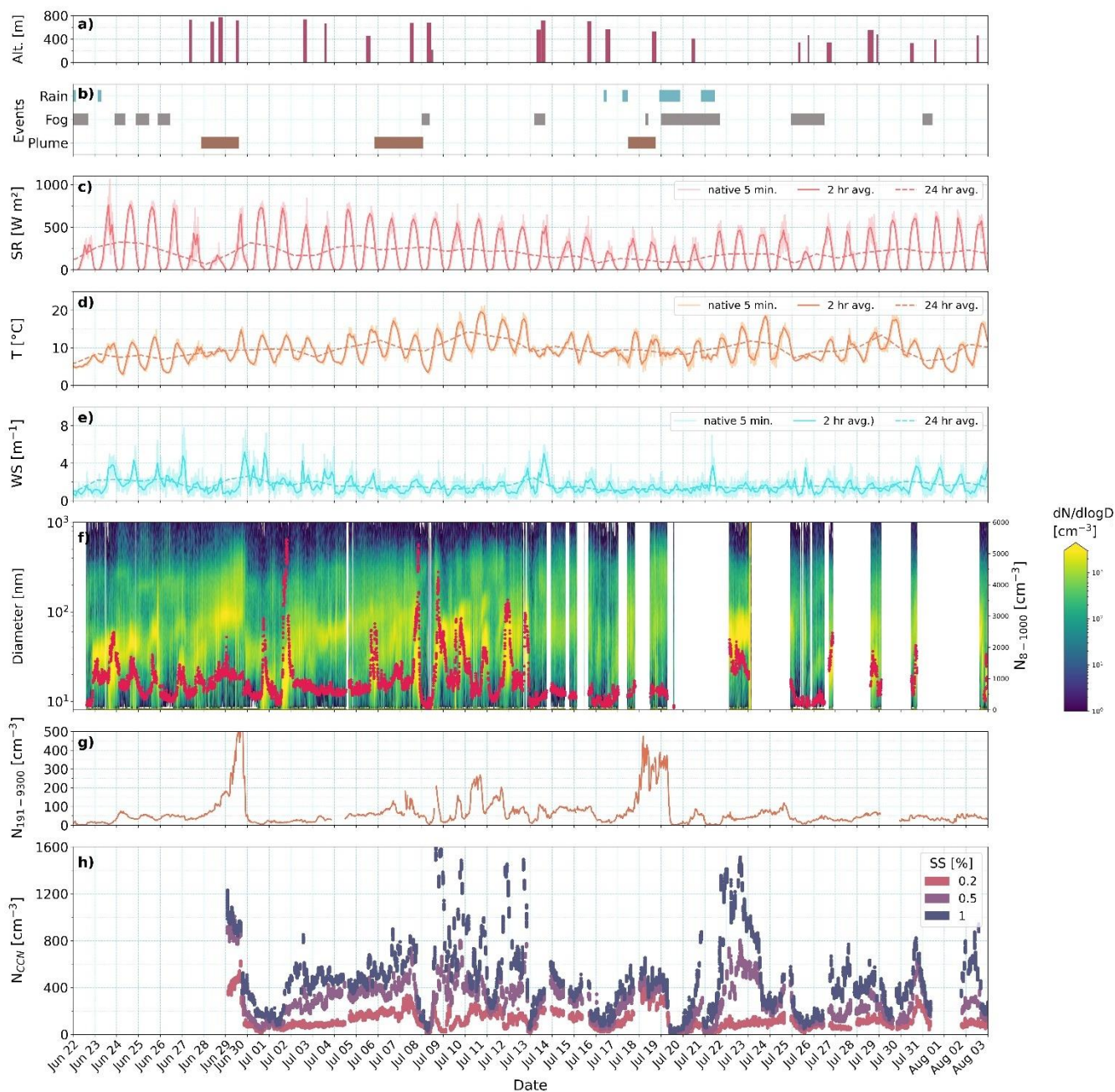


Figure 3: Time series overview of a) flight periods and altitudes above ground level (alt., dark pink bars); b) time periods of rain (mid blue bars), fog (grey) and BB plumes (brown bars); c) solar radiation ($W m^{-2}$, red, median average of 5 min., 2 hrs and 24 hrs); d) air temperature ($^{\circ}C$, orange, median average of 5 min., 2 hrs and 24 hrs); e) wind speed ($m s^{-1}$, light blue, median average of 5 min., 2 hrs and 24 hrs); f) particle number size distribution, $dN/dlogD_p$ (heatmap) between 8 and 1000 nm with total number concentration in the same size range (N_{8-1000} , cm^{-3} , red scatter, based on the SEMs); g) total number concentration between 191 and 9300 nm ($N_{191-9300}$, cm^{-3} , dark orange, from the fidis); h) total number concentration of CCN (N_{CCN} , cm^{-3}) for supersaturations of 0.2% (pink scatter), 0.5% (purple scatter) and 1% (midnight blue scatter). N_{8-1000} , $N_{191-9300}$ and N_{CCN} are without polluted data, see Section 2.4 for details. For the heatmap

of PNSD between 8-1000 nm flagged pollution data have not been removed. Blank spaces in panel f) correspond to time periods when the SEMS instrument was not measuring.

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Aerosol properties exhibited considerable variability over the course of the campaign. After excluding direct perturbations from frequently occurring growth events, long-range transport of biomass burning plumes, and scavenging during ground-level fog periods, the median (interquartile range, IQR) background concentrations of N_{8-1000} and $N_{191-9300}$ were 601 (412 - 941) and 40 (23 - 59) cm^{-3} , respectively. Table 2 summarizes the mean, standard deviation, median, and IQR of several additional aerosol variables. Narsaq is a sub-Arctic coastal site, and particle number concentrations there are considerably lower than at Mace Head, a mid-latitude European coastal site (Dall'Osto et al., 2010), but much higher than at the Zeppelin Mountain Observatory in Svalbard (Tunved et al., 2013) or Villum Research Station in northern Greenland (Nguyen et al., 2016), which are also coastal sites.

300

A notable feature of summer 2023 was the advection of three BB aerosol plumes originating from Canadian forest fires. N_{8-1000} , $N_{191-9300}$ and N_{CCN} all showed enhancements in total number concentrations, though the magnitude varied between the three events (Fig. 3 f-h). Frequent particle growth events were also observed, totalling 17 across the campaign period. After excluding these perturbations, the median background CCN number concentrations were 90, 234, and 375 cm^{-3} at 0.2, 0.5, and 1% SS, respectively (Fig. 3h). In general, high concentrations of coarse-mode aerosols ($N_{191-9300}$) covaried with high CCN number concentrations throughout the time series. However, two periods—early July and 21–24 July—exhibited enhanced CCN number concentrations at higher supersaturations without a corresponding increase in $N_{191-9300}$.

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Table 2: Summary of median, mean, and IQR of N_{8-1000} , $N_{191-9300}$, N_{CCN} for supersaturations of 0.2, 0.5 and 1 %, eBC, $\sigma_{abs,525}$, $\sigma_{sc,525}$ and $\sigma_{bsc,525}$ under BG and polluted BG (BG + pollution events), as well as growth events and plume conditions. Polluted BG conditions have data from particle growth, biomass burning and fog events removed, while keeping short-lived spikes from local anthropogenic emissions. BG also has these events removed, and in addition the anthropogenic emission spikes are removed.

315

Variable	BG			BG + pollution			Growth Events			Plume 1, Plume 2, Plume 3		
	Median	Mean	IQR	Median	Mean	IQR	Median	Mean	IQR	Median	Mean	IQR
N_{8-1000} [cm^{-3}]	600.7	815.1	411.6 – 940.7	928.1	2892.4	619.9 – 1453.8	1366.7	1573.6	1014.0 – 1840.9	1074.7, 924.7, 628.8	1047.1, 1223.4, 638.3	909.5 – 1174.2, 739.3 – 1277.9, 586.2 – 669.0
$N_{191-9300}$ [cm^{-3}]	40.0	50.7	22.8 – 59.2	52.4	78.4	32.2 – 83.9	39.5	54.3	27.8 – 63.1	147.0, 66.9, 235.6	181.0, 75.5, 243.5	78.2 – 189.7, 56.2 – 88.0,

													163.1 – 323.8
$N_{CCN0.2}$ [cm^{-3}]	90.1	100.9	63.4 – 127.9	149.7	183.3	93.7 – 243.5	134.0	128.0	67.3 – 166.1	426.1, 187.9, 294.5	436.4, 183.1, 287.3	398 – 472.3, 157.4 – 210.9, 242.9 – 333.1	
$N_{CCN0.5}$ [cm^{-3}]	234.4	246.6	124.4 – 346.0	312.7	352.4	216.9 – 437.8	328.9	311.5	190.7 – 428.4	858.8, 397.9, 397.9	839.8, 384.5, 400.1	826.7 – 910, 317.7 – 470.2, 375.1 – 426.9	
N_{CCN1} [cm^{-3}]	374.5	411.8	210.1 – 494.4	487.6	582.8	352.7 – 702.2	572.8	600.4	446.5 – 714.5	955.4, 546.5, 455.1	968.7, 503.2, 467.8	912.3 – 1014.4, 401.3 – 655.4, 417.9 – 529.7	
eBC mass [ng m^{-3}]	69.0	85.9	34.0 – 114.0	164.0	441.0	74.0 – 396.0	61.0	73.0	22.0 – 110.0	157.5, 98.0, 191.0	173.3, 133.8, 198.3	109.0 – 223.0, 62 - 151.8, 139.0 – 251.0	
$\sigma_{abs,525}$ [M m^{-1}]	0.5	0.52	0.3 – 0.7	1.4	2.9	0.8 – 2.7	0.4	0.6	0.2 – 0.6	1.3, 0.9, 2.0	4.8, 1.2, 1.8	0.7 – 7.1, 0.7 – 1.4, 13.6 – 2.2	
$\sigma_{sc,525}$ [M m^{-1}]	5.4	6.6	3.0 – 8.2	5.6	7.5	3.4 – 8.8	5.5	7.4	3.1 – 9.0	19.8, 7.7, 26.1	22.7, 8.8, 24.7	11 – 25.1, 6.1 – 10.8, 16.6 – 33.5	
$\sigma_{bsc,525}$ [M m^{-1}]	1.1	1.3	0.7 – 1.6	1.2	1.5	0.8 – 1.7	1.2	1.4	0.9 – 1.8	3.5, 1.7, 3.8	3.6, 1.9, 3.6	2.1 – 4.3, 1.4 – 2.2, 2.6 – 4.8	

320 Figure 4 shows the diurnal variation of solar radiation, T, wind speed, N_{8-1000} , $N_{191-9300}$ and N_{CCN} for supersaturations of 0.2 and 0.5%, as well as optical properties, $\sigma_{abs,525}$, $\sigma_{sc,525}$. The diurnals were plotted taking data from the BG dataset so that the various defined perturbations did not influence the average pattern. Solar radiation peaked on average between 12:00 – 13:00LT (local time), with median peak values of 560 W m^{-2} . Air temperature on the ground peaked between 15:00 and 16:00LT, 3 hours

after solar noon, with a median peak value of 12 °C. The wind speed was generally low throughout the campaign, as is typical
325 for summertime in the region, with wind speed peaking alongside temperature between 15:00 and 16:00 at 2.3 m s⁻¹. The
maximum wind speed reached during the campaign period was between 7 – 8 m s⁻¹. The median N_{8-1000} was at a minimum of
480 cm⁻³ overnight and peaked at 1290 cm⁻³ at ~17:00. N_{8-1000} remained low after sunrise on average, usually until ~10:00LT,
peaking between 16:00-18:00LT, co-varying with diurnal patterns of temperature and wind speed, before falling back to night-
time levels by 22:00LT. In contrast to the finer mode aerosols, $N_{191-9300}$, showed a less distinct, more variable diurnal pattern,
330 while the elevated concentrations over mid-day coincide with wind speed picking up, suggesting potentially wind-induced
particle generation. On average, $N_{191-9300}$ was at a minimum between 22:00 and 24:00LT, with a median value of 40 cm⁻³, while
the concentration peaked on average at 16:00LT at 55 cm⁻³. No strong diurnal pattern was observed in N_{CCN} for lower SS as
expected given the little variation of $N_{191-9300}$. At 0.5 % SS, the N_{CCN} diurnal pattern starts resembling N_{8-1000} , increasing
throughout the morning and peaking in the early afternoon with median peak value of 367 cm⁻³. The optical properties $\sigma_{abs,525}$
335 and $\sigma_{sc,525}$ show little diurnal pattern, except $\sigma_{sc,525}$ features higher values in the 90th percentile between 00:00 and 09:00LT, but
without any clear connection to other aerosol variables.

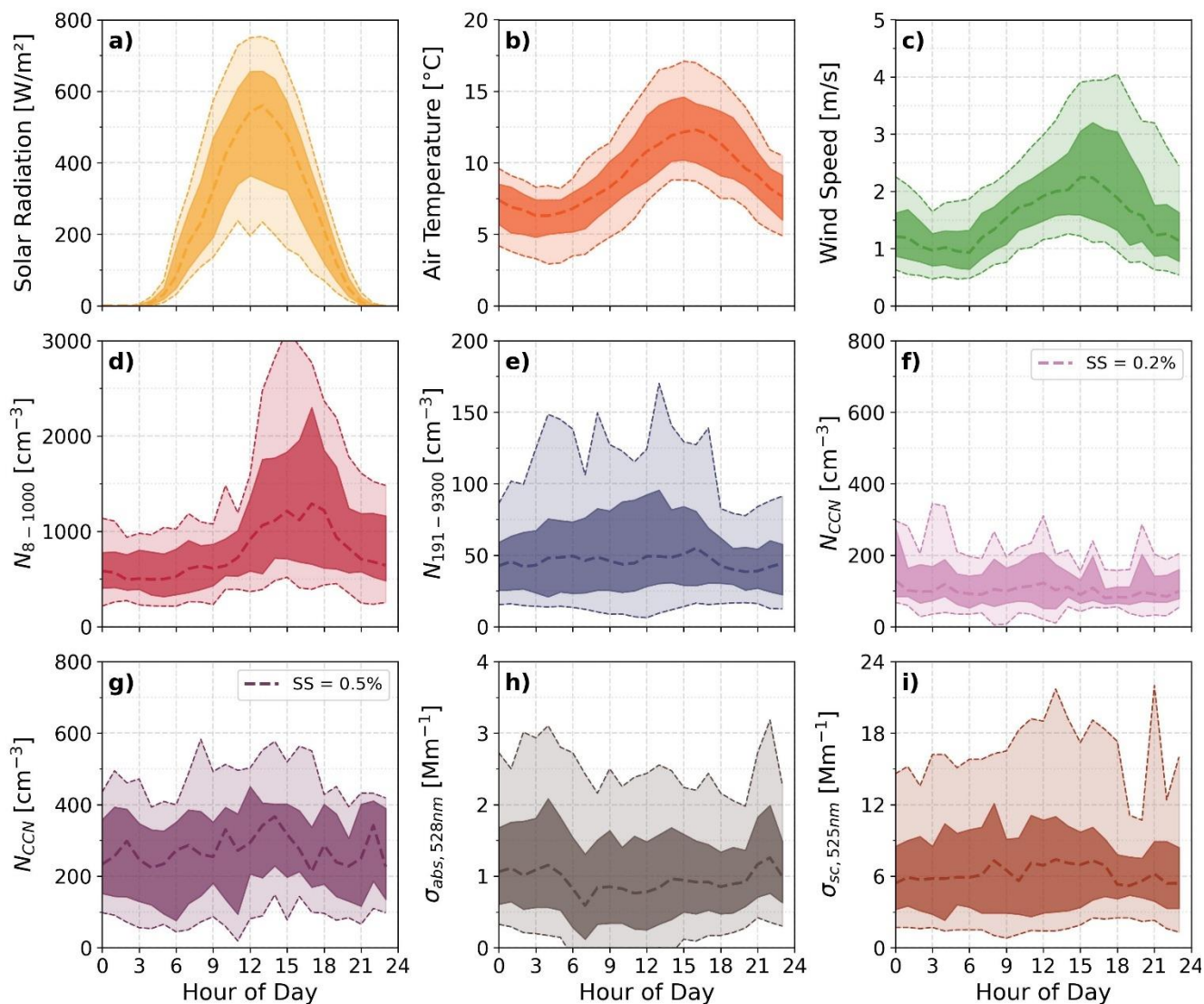


Figure 4. Hourly-resolved diurnal profiles of *BG* data with median (dashed lines), IQR (dark shaded regions) and 10th/90th percentiles (light shaded regions) for measured medians of a) solar radiation, b) T, c) wind speed, d) N_{8-1000} , e) $N_{191-9300}$, f) N_{CCN} with supersaturations of 0.2 and 0.5%, g) $\sigma_{abs,525}$ and h) $\sigma_{sc,525}$.

For deeper analysis we broke the time series into four distinct conditions based on the different proposed sources dominating the aerosol population: 1) a persistent aerosol background influenced primarily by local marine aerosol and aged anthropogenic pollution occurred 33.5 % of the time (Table S2); 2) local short-lived anthropogenic pollution (50.4 % of the time); 3) particle growth events (8.0 % of the time); and 4) biomass burning plume events (8.1 % of the time). We clustered the ground-based and vertical PNSD for each condition separately and discuss results throughout the remainder of section 3. The discussion aims to answer the following:

- Section 3.2: Can local pollution be distinguished from background aerosol?

- Section 3.3: Does particle growth occur on the ground or aloft?
- 345 • Section 3.4: To what extent do biomass burning plumes perturb the PNSD observed on the ground in periods where they are identified above the boundary layer?
- Section 3.5: What effect do pollution, biomass burning plumes and growth events have on climate relevant aerosol properties?

3.2 Background versus fresh pollution regimes

350 As summarised in Table 2, *BG* conditions are characterised by low aerosol and CCN number concentrations, and low eBC mass concentrations and absorption coefficients compared to *fresh pollution* and *BB* plume event periods. Based on the SEMS ground measurements, *BG* conditions accounted for ~33.5% of the campaign period. *Fresh pollution* events consist solely of the short-lived anthropogenic pollution spikes excluded from *BG*, *BB*, and *GE* categories. These spikes represent sharp perturbations to ambient aerosol conditions. They provide an insight into the impact of fresh local anthropogenic emissions on

355 the aerosol population. In contrast to the *BG* conditions, *fresh pollution* periods occurred 50.4 % of the time in the SEMS measurements.

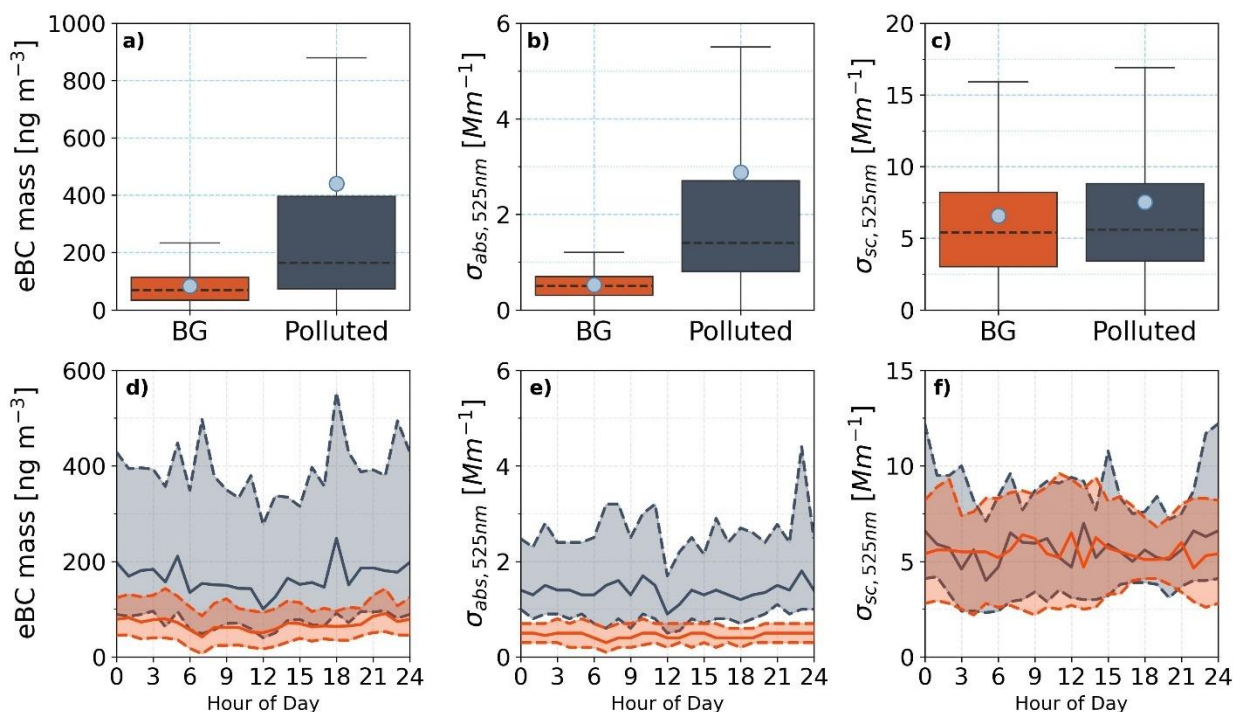


Figure 5. Comparison of BG (33.5 % of time) and fresh pollution (50.4 % of time) source contributions to the aerosol population in Narsaq. The top panel shows boxplots comparing a) aerosol eBC mass (ng m^{-3}), b) $\sigma_{\text{abs},525\text{nm}}$ (Mm^{-1}) and c) $\sigma_{\text{sc},525\text{nm}}$ (Mm^{-1}) for both BG and freshly polluted conditions. Bottom panel shows median average diurnals with interquartile ranges in shaded regions for d) eBC

mass, e) $\sigma_{abs,525nm}$ and c) $\sigma_{sc,525nm}$. It is worth noting that eBC values are given by a mAeth, while $\sigma_{abs,525nm}$ values are measured by the STAP. Due to the reduced data coverage on the ground by the STAP as a consequence of its usage on the tethered balloon, the clean dataset from the mAeth contains 9705 more 1-minute data points than the STAP dataset (26,924 vs 36,629 data points). Due to the increased sensitivity and temporal resolution of the STAP compared to the mAeth, 46 % of the dataset is flagged as polluted while only 40 % of the mAeth data is flagged. This also contributes to the difference in data coverage for both categories. Note: the y axis for panels a), b) and c) differ from that of d), e) and f).

There is a clear statistical difference between the *BG* and *fresh pollution* conditions including diurnal patterns for eBC mass, $\sigma_{abs,525nm}$ and $\sigma_{sc,525nm}$ (Fig. 5). Specifically, eBC mass and $\sigma_{abs,525nm}$ are enhanced in *fresh pollution* conditions with median values increasing by a factor of 2.2 and 2.7, respectively. Median values of $\sigma_{sc,525nm}$ however, show only minor enhancement suggesting that aerosol scattering is largely controlled by the persistent, presumably marine, aerosol background. While the diurnal patterns in all three variables are relatively weak, eBC under *fresh pollution* conditions shows an enhancement over the night compared to daytime, potentially related to domestic activities and a shallower boundary layer. Despite the frequent occurrence of *fresh pollution* spikes, the underlying *BG* conditions exhibit low and stable eBC mass and $\sigma_{abs,525nm}$. These pollution events do not appear to significantly modify the background state.

This interpretation is further supported by the clustering of PNSDs. The result provides additional insight into how transient emissions alter, or do not alter, the prevailing aerosol characteristics (Fig. 6). K-means clustering on the *BG* data yielded one bi-modal and two uni-modal clusters: 1) *BG c₁* peaking broadly in the lower Aitken mode at 30 nm and showing a shoulder in the nucleation mode at ~22 nm; 2) *BG c₂* peaking in the Aitken mode at 51 nm occurring the highest percentage of the time (39%) and at the highest concentration compared to the other clusters; and 3) *BG c₃* peaking in the Aitken mode at the highest diameter of 68 nm. The relatively high prevalence of *BG c₁* (30%) is likely due to the frequent particle growth events observed in the region, indicating that any meaningful representation of background conditions must inherently account for the influence of these growth events (section 3.3). Ground-based cluster classification is summarised with the relative abundance of each cluster per condition in Table S5. Given the generally low wind speeds across the campaign period, with median values of $2.4 \pm 1.3 \text{ m s}^{-1}$, and the dominance of the wind direction from the west and north-west, i.e. from down/across the fjord, ice sheet, and open ocean, it is reasonable to assume background clusters are highly likely to be marine in origin, and potentially mixed with fresh or aged local anthropogenic emissions.

Results from clustering of the pollution events show also three clusters (Fig. 6): 1) *pollution c₁* peaking at 27 nm; 2) *pollution c₂* peaking in the Aitken mode at 52 nm and; 3) *pollution c₃* peaking in the Aitken mode at the highest diameter of 77 nm. Despite the similarity of the main modes of the clusters in the background and polluted conditions, there are clear differences in the magnitude and variability, with pollution being more variable. The internal variability within the background clusters is consistent with the background category definition, which retains aged and processed anthropogenic aerosol alongside the marine signal, with only the short-lived fresh pollution spikes that are excluded, and it reflects natural day-to-day variability in the relative contributions of these sources under changing meteorological conditions. Primary exhaust particles, while their characteristics are strongly affected by vehicle type and fuel, have been shown to have PNSDs dominated by sub-50 nm

particles, and even in some cases sub-10 nm (Alanen et al., 2015; Rönkkö et al., 2006; Rönkkö and Timonen, 2019; Weimer et al., 2009). Due to the large-scale and permanent influence of marine air masses on the aerosol populations, it is conceivable that the *fresh pollution* category still contains a marine aerosol signature and hence it is not surprising that the clustered PNSDs for both conditions are similar. We hypothesize that the local pollution spikes do not modify the *BG* conditions significantly and in the long-term, and instead are short-lived perturbations on top of a dominant marine-influenced background.

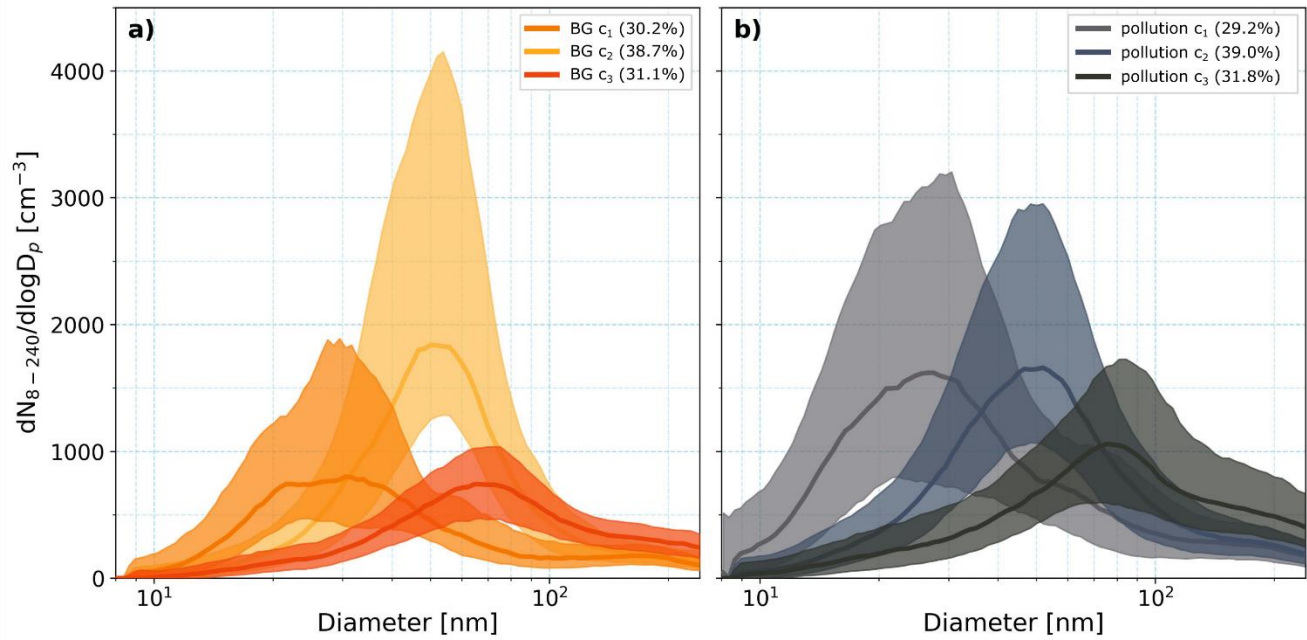


Figure 6: Clustered PNSD for a) BG and b) fresh pollution conditions. Legend shows percentage occurrence of each cluster within each dataset. Median values and IQR are shown for each clustered PNSD.

The vertical structure of specific aerosols properties under background conditions was assessed using tethered balloon and ceilometer data (see Text S3, Tables S6-7 and Fig. S879-9810). These data suggest that the background aerosol regime as observed at the surface generally extends up to ~200–300 m, where a significant decrease in the particle number concentration towards higher altitudes was observed, though the limited number of flights (8 flights) prevents robust conclusions. The ceilometer data covers much more time, and the clustering algorithm suggests that the “smoke/pollution/marine” category extended up to 500 -700 m, depending on the hour of the day. Since helikite flights did not reach higher than 700 m, it cannot be assessed with the in-situ observations whether another sharp decline in concentrations occurred further aloft. It seems justified to assume that the ground-based background observations are representative throughout the boundary layer in absence of specific other events.

3.3 Particle growth events

Throughout the campaign period, 17 growth events were identified. A 6-hour time window (section 2.4) was chosen to capture the initial growth phase while distinguishing both the point in time, when grown particles start resembling the background conditions, and overlapping events, where one event subsides as another begins. Figure 7 displays the PNSD for a representative growth event on the 1st July 2023, clearly showing the increase in N_{8-1000} . The growth rate between 8 and 25 nm (GR_{8-25}) for this event was calculated to be 2.7 nm hr⁻¹. During the campaign, the GR_{8-25} ranged from 1.5 to 7.6 nm hr⁻¹, with an average value of 3.9 nm hr⁻¹. Compared to high Arctic sites, these GRs are higher, more comparable with lower latitude remote and rural sites (Kerminen et al., 2018). A comprehensive summary of each GE , including time intervals, growth rates, coagulation and condensation sinks and the associated aerosol and meteorological conditions, is provided in Table S8 with a full description of how particle growth parameters were calculated (Text S4). The full campaign time series (Fig. 32) shows that the frequent occurrence of these events—at times on a daily basis—substantially influenced the average PNSD over the campaign and that they constitute an intrinsic component of the regional background aerosol population. Based on the 6-hour definition, growth events occurred 8 % of the time during the campaign.

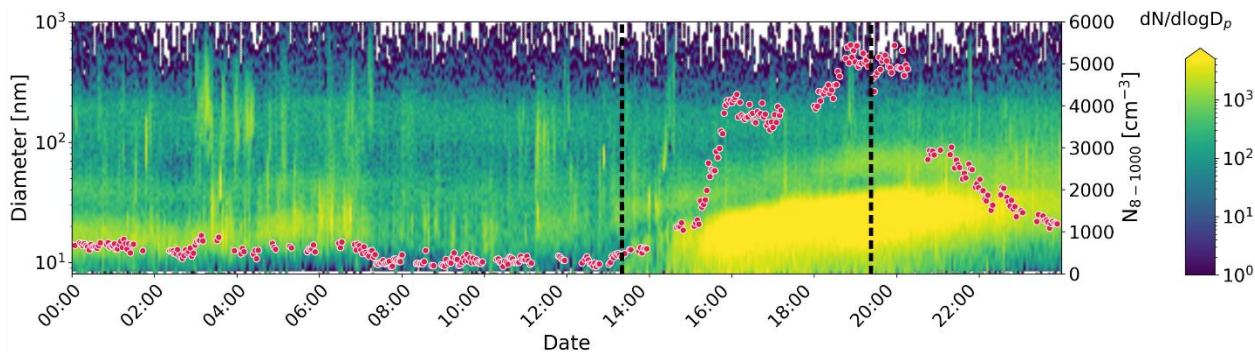


Figure 7. Normalised PNSD, $dN/d\log D_p$ (heatmap), between 8 to 1000 nm with N_{8-1000} (red scatter) overlaid for a growth event on the 1st July 2023. Black dashes represent the defined start and end (start + 6 hours) of this growth event.

Figure 8 shows the clustered PNSDs for the GE periods at the ground. Two bi-modal and one uni-modal clusters were found: 1) $GE c_1$ peaking in the nucleation and Aitken modes at 17 and 40 nm, respectively; 2) $GE c_2$ peaking in the nucleation and Aitken modes at 25 and 73 nm, and 3) $GE c_3$ peaking in the Aitken modes at 53 nm overlapping in the Aitken mode with background cluster $BG c_2$. Notably, 44 % of the GE data reached mode diameters characteristic of $BG c_2$, the most commonly observed BG cluster. The GE clusters likely represent temporal evolution of particle growth, where particles begin at $GE c_1$ at the onset and subsequently growth through $GE c_2$ and c_3 . This interpretation explains why $GE c_1$ is associated with the highest solar radiation – corresponding to the earliest stage of the event and usually corresponding to solar noon when photochemical-induced nucleation is the strongest. Eventually, the PNSD evolves toward that of $BG c_2$, illustrating not only the rapid growth of particles but also that, beyond the 6-hour definition from onset, much of the background aerosol is influenced by growth events. Across clusters, neither wind direction nor speed showed variability. There was no indication of the presence of a clear

mode below 8 nm in any of the three growth-related clusters, although *GE c1* exhibited a tail extending into this range, below the SEMs' lower cut off, with a shoulder around 8 nm, supporting the hypothesis that new particles are mainly transported from their nucleation site to Narsaq. Ground-based *GEs* were consistently associated with westerly winds travelling over the fjord (Fig. 8d). Based on the wind speed and calculated GR_{8-25} between 1.5 to 7.6 nm hr⁻¹, estimated distances from the nucleation source to the measurement site ranged from 5.5 to 31.8 km (see Text S5). Figure S9+ shows a regional map illustrating the estimated distances from nucleation sources as concentric circles around Narsaq. Given that wind is predominantly from the west, NPF appears to happen within the fjord system, close to the termination of the Greenland ice sheet. If biological productivity is active in the marine-terminating glacier fjords, it is expected that there are emissions of gas phase precursors for the nucleation, such as DMS, that can be oxidized and then lead to formation and growth of new particles.

435 It is noteworthy that particle growth events measured on the ground were much less common for periods where the wind direction was not persistently westerly. A full discussion and analysis of the effect of marine and land-terminating glaciers on NPF observed in fjord systems in Southern Greenland will follow in a separate paper based on ship-borne observations.

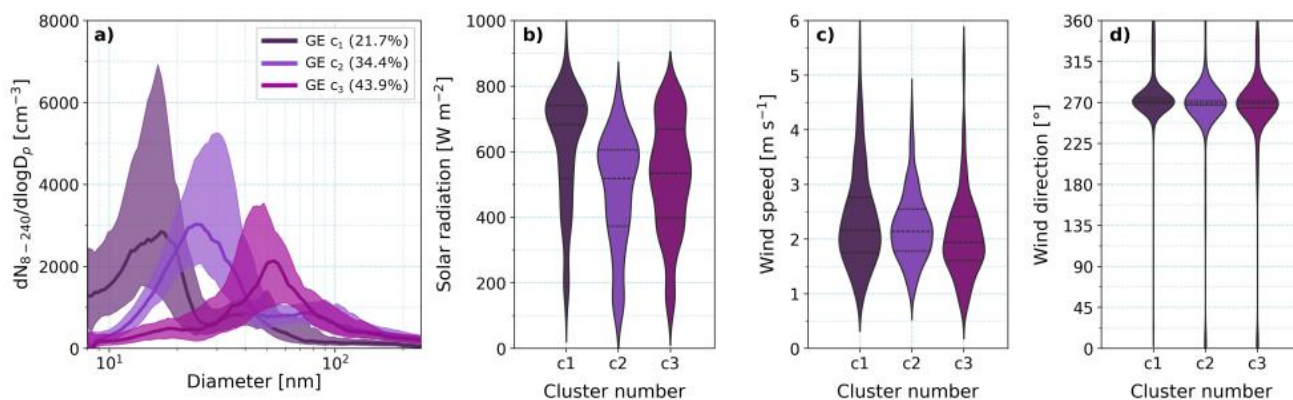


Figure 8: Panel a) shows clustered PNSD for particle *GE*. Anthropogenic pollution spikes have been removed from the data. Median values and IQR are shown. Legend shows percentage occurrence of each cluster within each dataset. Panel b-d) show violin plots of solar radiation, wind speed, and wind direction for each cluster.

440 While ground-based analysis provides a detailed view of *GEs* experienced at the surface, it does not capture how these particles evolve vertically in the lower atmosphere, and whether the source is indeed at the surface. To address this, we examine one vertically resolved case study, for the time period of 12:05 to 18:00 on the 28th July, to explore the influence of atmospheric structure and vertical transport on the dispersion and growth of nucleation-mode particles above Narsaq. Although several flights occurred during particle growth events throughout the campaign, these cases did not allow for vertical clustering due to the helikite's ascent and descent speeds within the lowest 200 m, which were too fast to collect sufficient scans with the mSEMS. Nevertheless, the thermodynamic conditions for the chosen case study were typical of those during growth events

445 across the campaign, featuring an elevated temperature inversion at ~320 m above ground and the maximum measured altitude

of 560 m. Of the 24 flights in total, elevated temperature inversions were observed in 14, and surface-based temperature inversions were observed in seven. The vertical profiles of temperature for the flights during this case study show the thermodynamic structure and the presence of temperature inversions in Fig. S120, while Text S6 describes how inversions were determined.

In this case study, to establish a reference for interpreting the vertical aerosol structure, we first applied k-means clustering to the ground-based PNSDs during the case-study period. Figure 9 shows two distinct surface (s) clusters: 1) $sGE\ c_2$, a tri-modal distribution peaking at 24, 65 and 173 nm, observed after ~14:00 when the wind direction became persistently westerly; and 2) $sGE\ c_1$, a tri-modal distribution peaking at 21, 57 and 172 nm, seen before ~14:00 under mixed westerly and north-westerly influence. The temporal evolution of the two identified clusters (Fig. 9b) suggests they represent different phases of the same growth event: (1) a pre-event stage with lower nucleation mode concentration ($sGE\ c_1$) and (2) a subsequent advected growth event ($sGE\ c_2$). From the time when the westerly airmass became more dominant, the nucleation mode around ~24 nm increasingly dominated over the Aitken mode in the ground distributions.

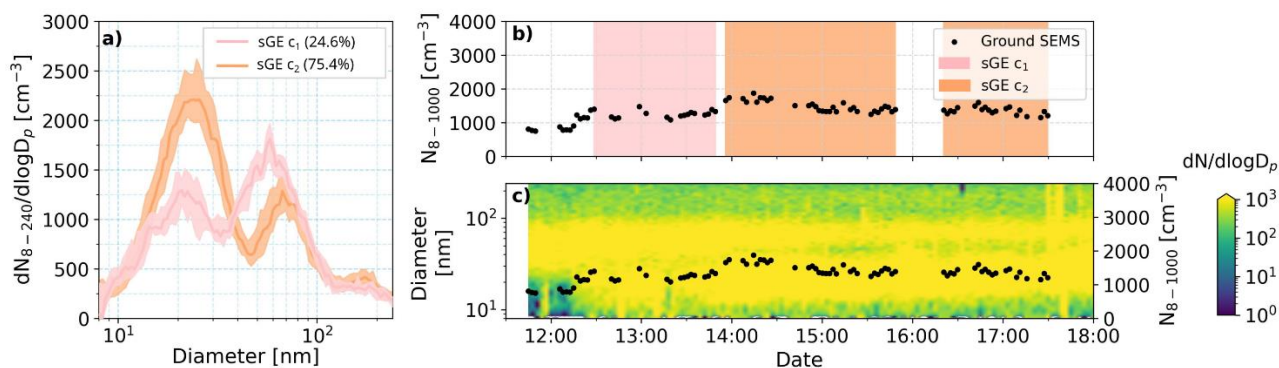
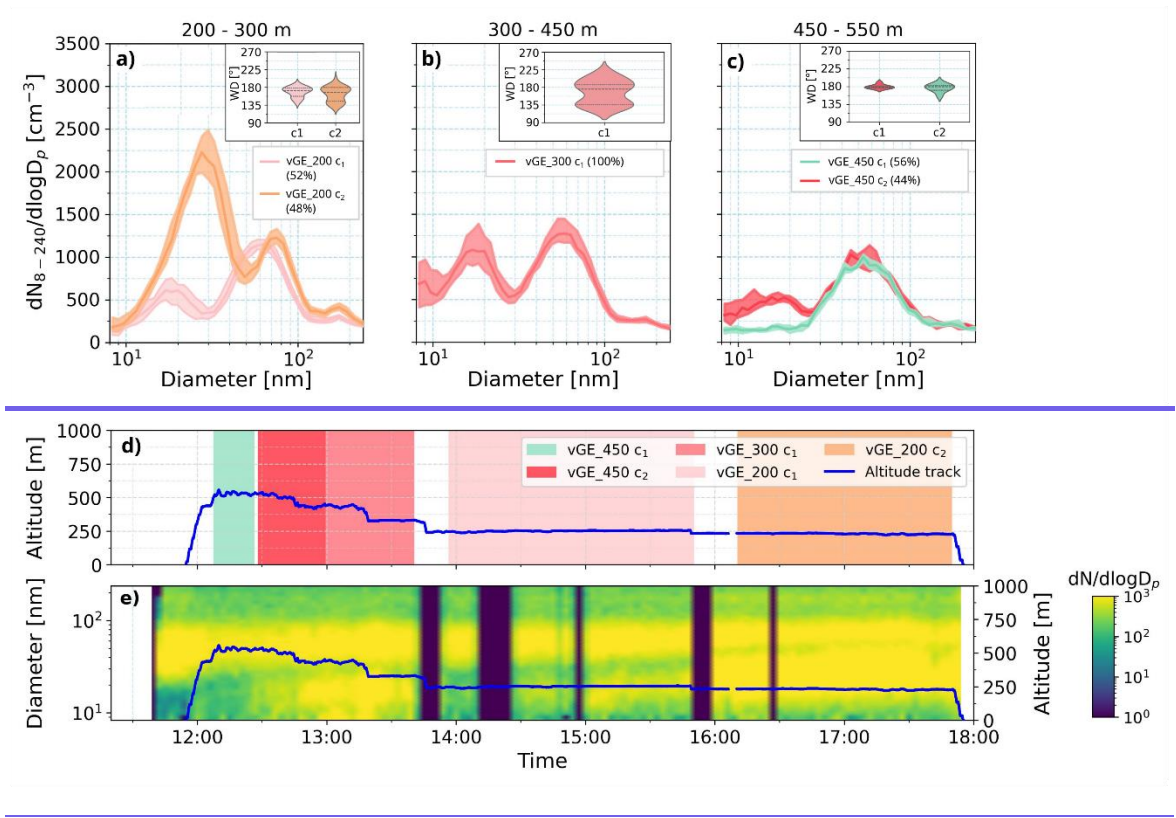


Figure 9. Panel a) shows clustered PNSDs using ground SEMS measurements for the time period of flight 8 on the 28th July, corresponding to vertical measurements shown in Fig. 10. The legend shows of the percentage of SEMS runs for each cluster during the shown period. b) Time series of N_{8-1000} (black scatter, cm^{-3}) with shading corresponding to the occurrence of each cluster. c) Heatmap of particle number size distribution, $dN/d\log D_p$ with same time series of N_{8-1000} as in b).

Clustering of the vertical aerosol profiles revealed varying complexity across altitude layers, with 2, 1, 1, and 2 clusters identified in the ground level, 200–300 m, 300–450 m, and 450–550 m altitude ranges, respectively. Figure 10 a-c shows the clustered PNSDs for the flight, Fig. 10d the time series of the altitude track with the occurrence of each cluster shaded and Fig. 10e shows the heatmap of PNSD over the course of the flight. The slow descent profile from ~550 m to 200 m was used to characterise the vertical aerosol population. Altitude bins for clustering were chosen to represent the distinct vertical aerosol layers observed and the presence of a temperature inversion above ~300 m up to the maximum height of the flight (Fig. S102). Comparing ground and elevated observations at 200-300 m, there is strong similarity in the aerosol populations: the two vertical clusters - $vGE_{200}\ c_1$ and c_2 – closely resemble the ground clusters $sGE\ c_1$ and c_2 , indicating a well-mixed boundary layer up

470 to 300 m (see Fig. S113 for a direct comparison of the clusters). Within the elevated inversion, between 300 and 450 m, only
one cluster was identified, with mode diameters different from the layers below and potentially a mode below 10 nm (Fig.
10b). This suggests that the elevated inversion inhibits mixing of particles from below and hence features a distinct aerosol
population. At the highest elevation range, 450 -550 m, and still above the elevated inversion, two clusters are observed with
475 a similar Aitken mode diameter around 60 nm compared to the layer below, however without or with strongly depleted smaller
modes. Wind direction information (see insets in Fig. 10), indicates different source regions for these layers: in the 450–550
m range, winds came directly from the south, while at 300–450 m they were a mix of southerly and south-easterly, and at the
surface again predominantly southerly with a smaller south-easterly component. This points to potentially different source
regions in these three layers.



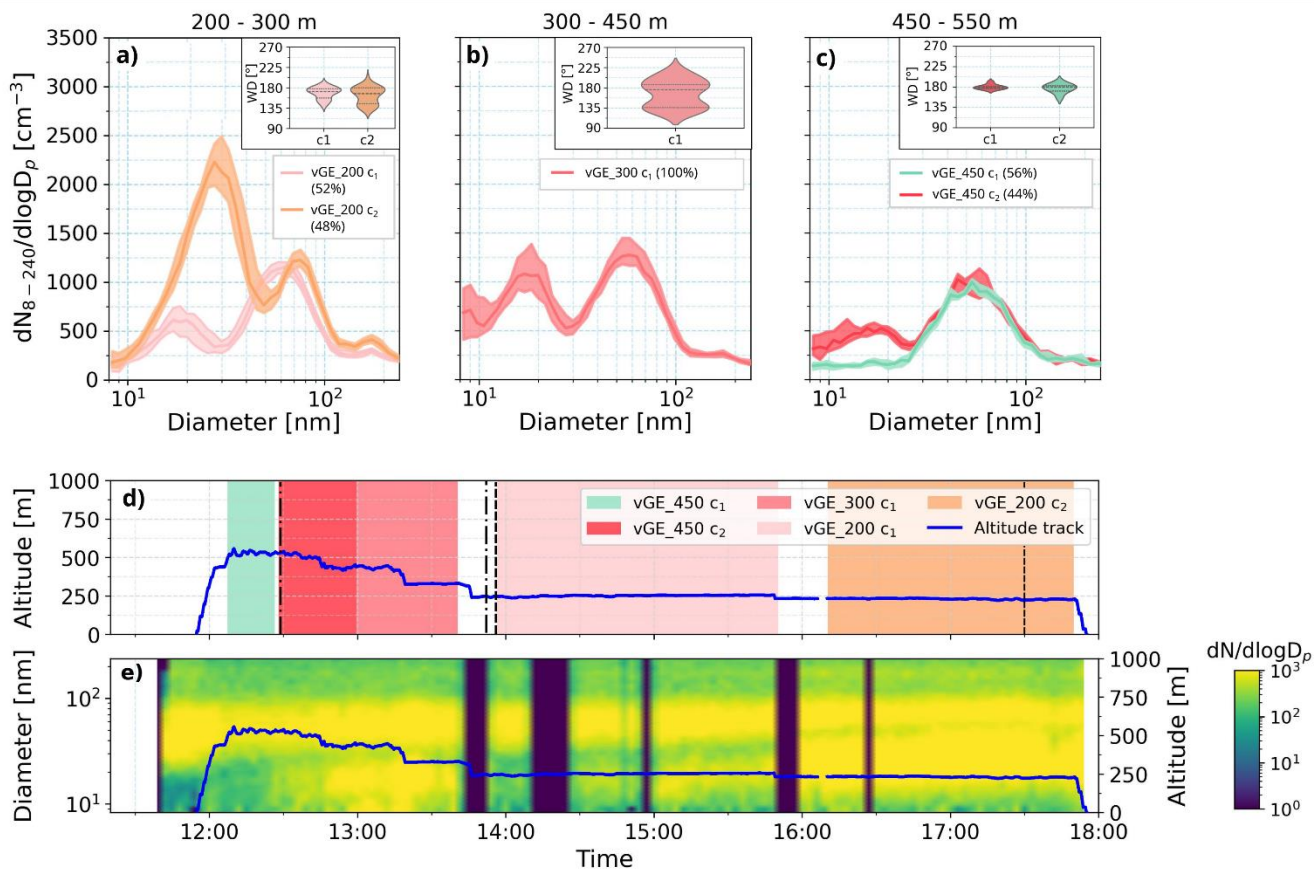


Figure 10: Clustered PNSDs (median and IQR) using mSEMS vertical measurements for altitude bins of a) 200 to 300 m (light pink and orange), b) 300 to 450 m (pink), and c) 450 to 550 m (red and turquoise), above ground level. Legends show percentages of mSEMS scans in each cluster. Insets provide violin plots of wind direction for each cluster. Panel d) displays the altitude of the helikite (blue) over the course of the flight, with shaded regions showing the chronological occurrence of each cluster throughout the flight period, while the heatmap of the particle number size distribution from the flight is shown in panel e). [The region between black dashed lines in panel d\) represents time during which \$sGE_{c1}\$ was present on the ground, while the region between black dash-dotted lines represents time during which \$sGE_{c2}\$ was present on the ground. Direct comparison of ground and vertical cluster distributions is provided in Fig. S11.](#)

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Overall, the analysis suggests that the growth events measured at the surface mainly occur at the ground rather than aloft. The cluster with a smaller nucleation mode (< 25 nm) between 200-300 m, i.e. $vGE_{200_{c1}}$ is diluted compared to its ground analogue sGE_{c1} (Fig. S143), while the cluster with the larger nucleation mode (around 30 nm, $vGE_{200_{c2}}$) is as strong as the ground cluster sGE_{c2} . Given the slightly larger mode diameter aloft, the particles seem to have been around for longer in the region and were able to disperse also in the vertical below the inversion. Moreover, size distributions within and above the

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elevated surface inversion are distinct from the ground. That said, these size distributions contain nucleation mode particles and it can hence not be excluded that NPF and growth also takes place above the well mixed part of the boundary layer, indicating that NPF can occur at the surface as well as aloft. The complex structure of the boundary layer seems to allow multiple particle sources to form distinct vertical layers that will only mix over time scales longer than is possible to observe during helikite flights (i.e. > 5 hours).

3.4 Biomass burning plume advection

Three different *BB* plumes from Canada were transported towards Narsaq, arriving during the following periods: 27th of June 21:20 to 29th of June 15:00; 5th of July 20:13 to 8th of July 01:42; and 17th of July 11:47 to 18th of July 18:17 (all LT). A time series overview including *BB* plume periods can be found in Fig. 3. Figure 11 shows the time series of β observations from the
495 | ceilometer for the three plume periods. Plume 1 arrived about ~~teame~~ within 300 m ~~of~~ above the ground with a thickness of roughly 2 km (Fig. 11a). Throughout the entire plume 1 period, there is pronounced variability in the vertical structure of the plume, including a layer of clouds occasionally capping the plume (light pink color) with several short precipitation events (yellow) reaching the ground. Unlike the first plume, plume 2 maintains an altitude between 1 and 5 km until mid-day on July 6th (Fig. 11b). Thereafter additional plumes occur further aloft. The lower plume 2 gradually thins and increases in altitude
500 | with time and appears to be less affected by vertical mixing. In plume 2, β is lower compared to plume 1, suggesting a less dense aerosol concentration. Plume 3 appears to reach within 1 km of the surface, and is the thinnest plume with regard to vertical extent, with visible layers appearing over the night between 17th and 18th July between 1 and 2 km (Fig. 11c). On the second day of the event, evidence of rising motion can be seen in the afternoon, after a morning ground-level fog event between 07:00 and 10:00.

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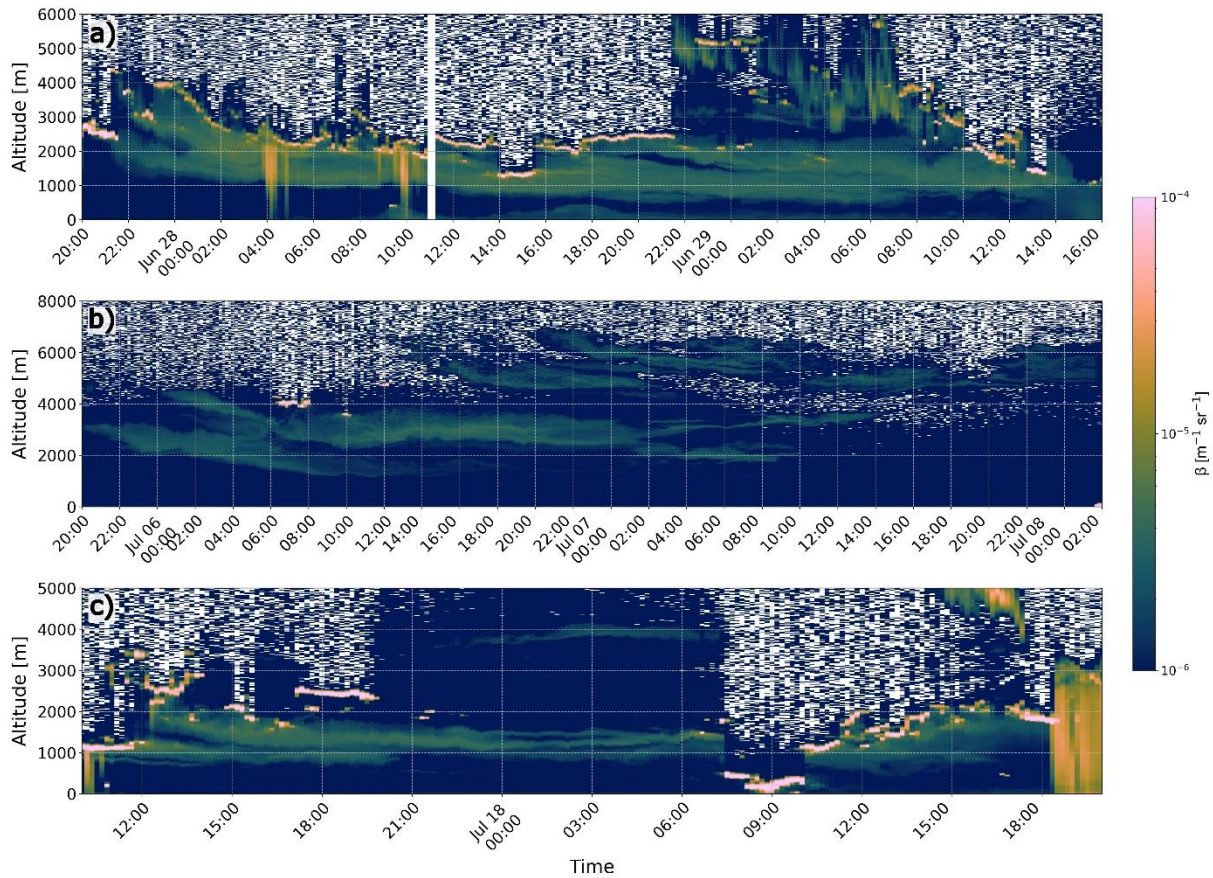


Figure 11. Time series of β ($\text{m}^{-1} \text{sr}^{-1}$) as a function of altitude measured by the ceilometer for periods of a) plume 1, b) plume 2 and c) plume 3. Blue, green, yellow and pink colors signify clear atmosphere, aerosols, mid-density liquid (precipitation) and high-density liquid (clouds), respectively.

While ceilometer observations show the main aerosol layers remained aloft, the instrument's optimal sensitivity to particles over 900 nm means it has limited detection capability for smaller particles (Sundström et al., 2009). Consequently, smaller particle fractions from the plume edges, which likely reached the surface, would not be visible in the ceilometer data. This interpretation is supported by the substantial variability and elevated values observed in ground-based fine and coarse mode aerosol concentrations, which likely reflect the surface arrival of smaller particles not detected by the ceilometer. The variation of N_{8-1000} and $N_{191-9300}$ for each plume is compared to the background conditions (Fig. 12). Across all plume periods, coarse mode particle concentrations were significantly elevated relative to *BG* conditions, with statistical significance confirmed by Mann-Whitney U tests ($p < 0.05$ for all plume–background comparisons for $N_{191-9300}$). The largest enhancement in $N_{191-9300}$ was observed during plume 3, followed by plume 1. In contrast, N_{8-1000} was elevated during plumes 1 and 2, but not plume 3. The relatively stronger enhancement of coarse mode particles during plume 3, compared to plumes 1 and 2, may indicate

difference in emitting fire types, i.e. smouldering vs flaming (Virkkula et al., 2014), or greater atmospheric aging, such as photochemical oxidation and subsequent condensation of low-volatility compounds, promoting particle growth and shifting the aerosol size distribution towards larger diameters. While coagulation dominates in the early hours post-emission of plumes, photochemical aging typically unfolds over longer atmospheric residence times. Some studies report size or mass enhancements linked to secondary organic aerosol formation (Hodshire et al., 2019; Shrivastava et al., 2017), while others note mass loss from evaporation or oxidative fragmentation (Cappa et al., 2020; Reid et al., 2005). Notably, Farley et al., 2022 documented wildfire plumes with transport times of up to 10 days, during which aerosol oxygen-to-carbon (O/C) ratios and modal diameters increased, consistent with progressive aging. The coarse mode enhancement in plume 3 aligns with this aging-related growth, as reported in other multi-day transport studies. This interpretation is further supported by back-trajectory analysis using the Lagranto Lagrangian analysis tool (Sprenger and Wernli, 2015; Wernli and Davies, 1997). Back-trajectories were calculated for 120 hours, using ERA5 3-hourly re-analysis data, with a starting pressure range of 750 – 650 hPa - chosen to represent the upper bound of the observed plume altitudes. Figure S124 shows the trajectories initiated at the plume start time, and 24 and 48 hours later, providing insight into the evolution of the air mass history. For plumes 1 and 2, air masses originated more directly from fire regions, with shorter transport times of 1–2 days. In contrast, air masses associated with plume 3 spent more time over Quebec before crossing the Labrador Sea. Notably, some back-trajectories during plume 3 also crossed fire clusters in Western Canada as early as five days before reaching Narsaq (not shown). The extended transit time may have allowed for enhanced photochemical processing and continued particle growth.

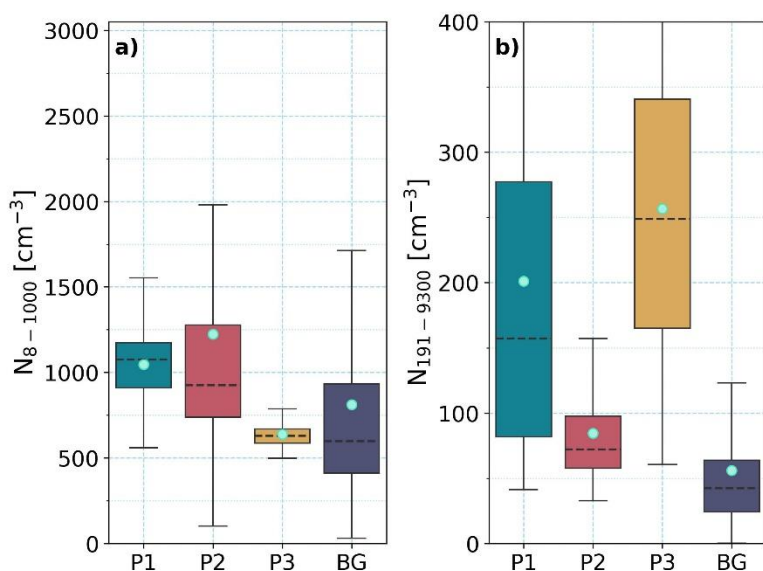


Figure 12: Boxplots with median, IQR and 10/90th percentiles showing variation in a) total number concentration of N_{8-1000} , and b) $N_{191-9300}$, for periods of plume 1 (P1), plume 2 (P2) and plume 3 (P3) compared to the background (BG) observations. Dots represent averages.

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To better understand how these transported plumes impacted the ground-level aerosol population in Narsaq, we applied k-means clustering to the PNSDs measured during each plume period. This allowed us to characterise distinct aerosol modes associated with each plume and assess their evolution over time and with respect to the local boundary layer structure. Figure 13a-d shows the clustered PNSDs for the period of plume 1 from the ground-based SEMs measurements in comparison to the background clusters. For plume 1 four clusters were found to describe this plume on the ground: 1) a tri-modal cluster peaking in the nucleation mode at 18 nm, and twice in the Aitken mode at 48 and 75 nm (P1c₁), 2) a uni-modal cluster peaking at 77 nm (P1c₂), 3) a second uni-modal cluster peaking at 91 nm (P1c₃), and 4) a bi-modal cluster with a peak in the Aitken mode at 91 nm and a broadened peak in the accumulation mode between 100-200 nm (P1c₄). A distinct shift towards higher diameters can be seen for all clusters in comparison to the background clusters (depicted in Fig. 13a-d as the BGc₁ to c₃). Figure 13e shows the time series of N_{8-1000} with the occurrence of each cluster shaded. There is a clear evolution over time as clusters occur in successive order. There is a shift in the Aitken mode peak from 75 nm in P1c₁ to 91 nm in P1c₄, as well as the distinct concentration enhancement in accumulation mode particles in all bins [between](#) >100 nm by up to a factor of ~2.5 from c₁ to c₄. This observed evolution coincides with the dissipation of an elevated temperature inversion (starting at 300 m) over the course of the day (Fig. S135), suggesting that the thermodynamic structure of the atmosphere above Narsaq plays a role in the extent to which the plume can mix towards the surface.

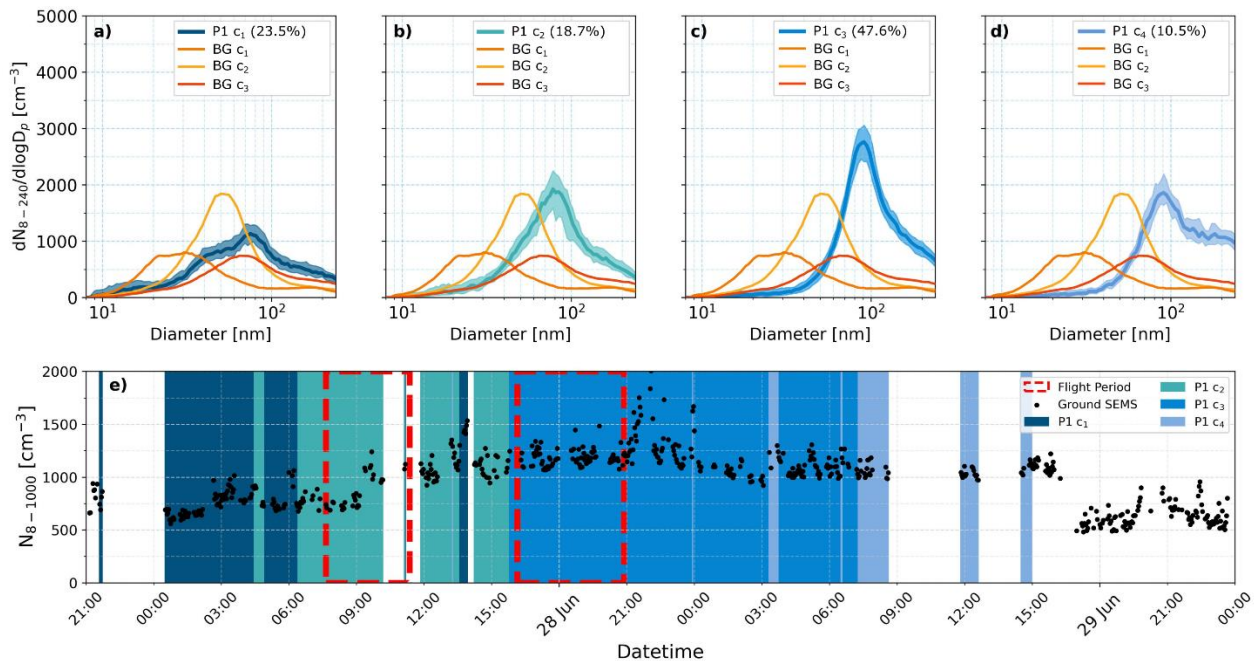


Figure 13: Panels a) to d) show clustered PNSD [at ground level](#) covering the time period of *plume 1* (blue) compared to the clean *background* clusters (yellow, orange and red). Median values and IQR are shown for each cluster. Panel e) shows the time series of N_8 .

1000 (black scatter) with the occurrence of each cluster shaded. Time periods corresponding to helikite flights are boxed in red dashes. Legend shows the percentage occurrence of each cluster in the overall category.

To investigate the vertical structure of the aerosol population per plume event and compare with the ground PNSD, k-means clustering was applied to the mSEMS data for flights occurring within a plume period. For each plume, altitude bins were chosen to best represent a section of the vertical transect, ensuring enough data points per altitude to provide reasonable statistical information. The flights during plume 1, described in this section, are the only flights where we directly measured
555 inside a plume.

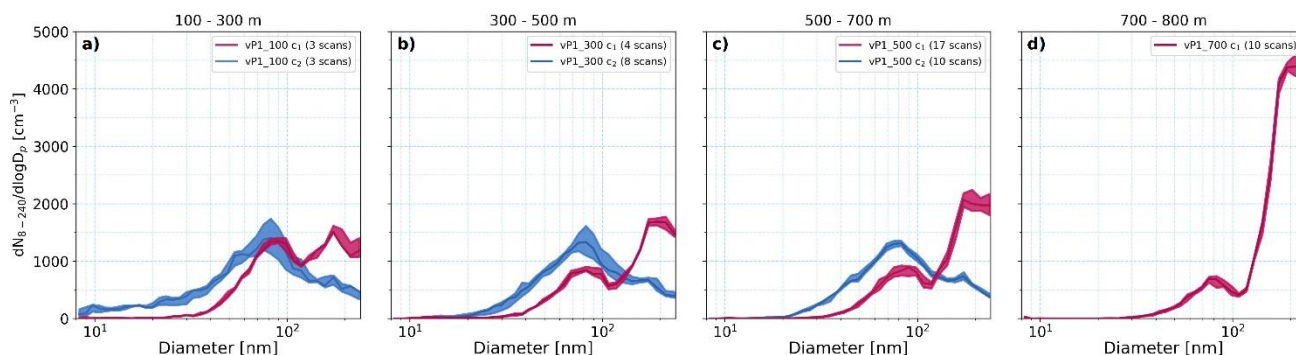


Figure 14: Clustered PNSDs using mSEMS vertical measurements for altitude bins of a) 100 to 300 m, b) 300 to 500 m, c) 500 to 700 m and, d) 700 to 800 m above ground level. Median values and IQR are shown for each clustered PNSD. Legends show number of scans within each cluster.

Figure 14 shows clustered PNSDs for morning and afternoon flights during plume 1, where data from both flights were
560 combined per altitude section. Figure S135 shows the temperature profiles between 07:39 – 11:22LT and 16:08 – 20:52LT. Throughout 100-300 m, a bi-modal cluster (blue, *vP1_100c2*) peaking at ~82 nm in the Aitken mode and weakly at ~170 nm in the accumulation mode, was visible and fading in occurrence up to 700 m (follow the blue cluster in Fig. 14), and was no longer present above 700 m. The altitude range of 700 – 800 m corresponded to when the helikite entered the lowermost boundary of the plume according to the ceilometer observations (Fig. 11). The second cluster (*vP1_100c2*) is relatively similar
565 to *PI c2* observed on the ground (Fig. 13b). This cluster (dark pink) was also bi-modal, peaking at ~65-75 nm and ~190 – 210 nm, and showed increasing dominance of the accumulation mode with larger proximity to the plume going upwards. This shift was accompanied by a gradual increase in Aitken mode diameter while the overall concentration in this mode decreased with each altitude bin. We expect that the vertical distribution of aerosols is affected by the elevated inversion in the morning. Results from the vertical PNSD clusters, observed with the mSEMS at higher altitudes, suggest that upward mixing of the
570 ground-cluster, observed with the SEMS at ground level, higher than 700 m was inhibited, while the inversion did not seem to fully stop the downward mixing of plume cluster. The distinct peak > 100 nm in the accumulation mode, which appears to be an indicative signature of the plume, is however not seen clearly in any of the ground clusters for this period.

This is in line with the vertical observation, where the intensity of the distinct accumulation mode cluster decreases towards the ground. Within the plume itself, the accumulation mode peak is narrower and centred around 213 nm whereas below the plume this mode is closer to 170 nm and spans a wider diameter range. This is likely due to mixing of plume aerosols with other aerosol populations at lower altitudes. Plumes 2 and 3 are discussed in detail in the supplement (Text S7 and S8; Fig S146-1921), and therefore only a summary of these plumes is given here.

Plume 2 showed four ground-level clusters with a shift towards larger Aitken mode diameters and enhanced accumulation mode during the day, while vertical profiles indicated a persistent Aitken accumulation bi-modal structure up to 600 m. Despite ceilometer evidence that the plume remained aloft, partial downmixing is suggested by accumulation mode enhancement near the surface (Fig. S16). Compared to plume 1, its impact was weaker, likely due to multiple inversions confining plume influence (Fig. S18). Plume 3, likely the most aged event, was marked by three ground clusters dominated by broadened Aitken modes and lower overall fine particle concentrations, approaching background levels. Vertical clustering revealed a homogeneous population up to 600 m with consistent Aitken (~60–70 nm) and accumulation (>170 nm) modes, and a nucleation mode below 300 m. Stratification from surface-based temperature inversions separated the near surface air from elevated layers, resulting in weaker ground enhancement while maintaining an accumulation mode signal aloft.

Overall, on the ground, all three plumes show enhancement in accumulation mode concentrations (100–240 nm), with the most pronounced increase observed during plume 1. Plume 1 reached closest to the surface according to the ceilometer observations of β . The fainter signatures on the ground for the other two plumes are likely related to the higher plume altitudes as well as the temperature inversions below the plumes that might have inhibited downmixing. It is noteworthy that higher accumulation mode concentrations were observed on the ground, even though the ceilometer did not indicate a descend of the plume to the surface, meaning the aerosol attenuation might not have been high enough. The clustered size distributions observed on the helikite indicate that the specific biomass burning plume cluster, i.e. accumulation mode more intense than Aitken mode and diameter near 200 nm, is present at all altitudes for plumes 1 and 3 and is decreasing in intensity towards the surface. For plume 2, the accumulation mode is less intense than the Aitken mode, however still slightly enhanced compared to the background conditions.

3.5 Source contributions to climate relevant aerosol properties

To evaluate the influence of distinct aerosol sources on climate-relevant aerosol properties, we assessed changes in N_{CCN} , $\sigma_{obs,525nm}$, and $\sigma_{sc,525nm}$ for the different aerosol source categories relative to background conditions (Fig. 15). We could clearly identify periods of elevated over BG (Fig. 15a–c), with increased droplet activation potential across anthropogenic emissions, particle growth events, and biomass burning sources. The variability in distribution for 0.2 to 1% SS is presented in Fig. S220. Among these different sources, *plume 1* contributed the largest increase in N_{CCN} , followed by *plumes 2* and *3*, highlighting the importance of biomass burning aerosol transport but also aging and transport history in shaping CCN activity with the three plumes showing substantial variability in both N_{CCN} magnitude and distribution shape. This difference is likely influenced not

only by the intrinsic properties of plumes, but also by their vertical position and the degree of dilution before reaching the surface. Plume 1, being closest to the ground, delivered a larger fraction of its BB aerosol directly to the measurement site, resulting in higher N_{CCN} over this period. Plumes 2 and 3, by contrast, occurred at higher altitudes and likely experienced more dilution and mixing before reaching the surface, which could contribute to their lower N_{CCN} despite comparable aerosol loads aloft.

Both local anthropogenic pollution and growth events slightly skew the CCN number concentration to higher values, but much less than the biomass burning plumes. For $\sigma_{abs,525nm}$, polluted conditions display a wide variability (Fig. 15d), driven by fluctuating local emissions. Plumes 2 and 3 exhibit more distinct enhancement in $\sigma_{abs,525nm}$, while plume 1 shows elevated absorption with a broader distribution and higher absolute values, reflecting differences in plume composition, transport history, ageing processes and fire source characteristics. Growth events do not show distinct variation from the BG distribution as expected given the hypothesis that they are a significant source of the background aerosol population and contribute non-absorbing and primarily scattering chemical compounds from low-volatility vapor condensation. [Wavelength dependent absorption differences between plumes are showcased in Fig. S21.](#) ~~Fig. S23 shows the differences in absorption from 450-624 nm for each data category. Plumes 1 and 2 exhibit similar absorption characteristics in terms of mode and wavelength dependence, although plume 2 displays more pronounced tails. In contrast, plume 3 shows stronger overall absorption, with a marked increase at shorter wavelengths. This pattern may suggest a greater contribution from light absorbing organic aerosols, potentially originating from different fuel types and fire conditions, such as increased smouldering or peat combustion, compared to the other plumes.~~ All three plumes show enhanced $\sigma_{sc,525nm}$ relative to BG (Fig. 15i), though plumes 1 and 3 display greater variability, potentially due to differences in particle size distribution or varying proportions of scattering species. In contrast, $\sigma_{sc,525nm}$ remains relatively unchanged under polluted conditions compared to BG (Fig. 15g), indicating a limited influence of short-lived local emissions on aerosol scattering properties. Growth events, however, exhibit a modest shift to larger $\sigma_{sc,525nm}$ (Fig. 15h), consistent with increased particle number concentrations. Overall, local anthropogenic emissions have the largest impact on $\sigma_{abs,525nm}$ compared to their impact on N_{CCN} or $\sigma_{sc,525nm}$. Growth events are similar to background observations as expected, while biomass burning particles strongly enhance all three properties, with their variability driven by differences in plume origin, altitude, transport, dilution, and composition.

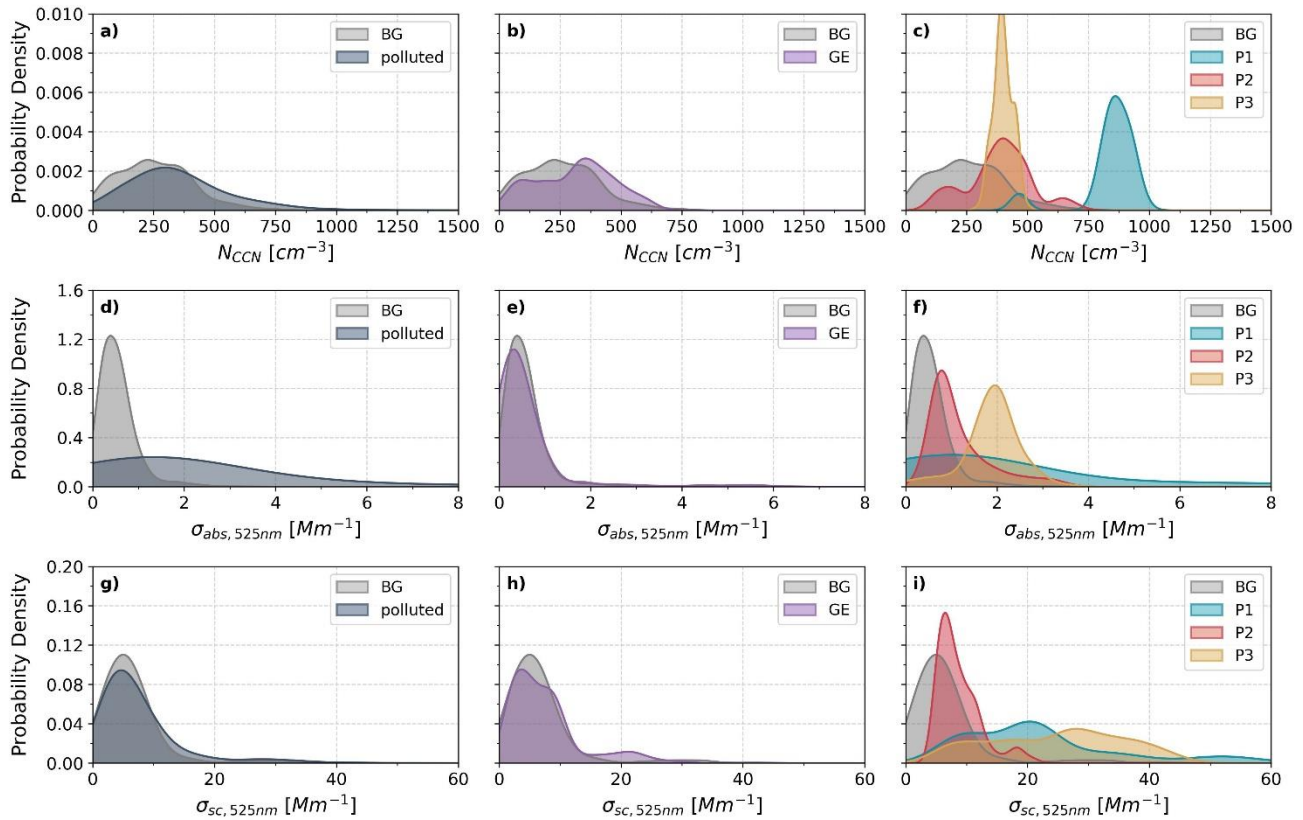


Figure 15: Probability density estimates of a-c) N_{CCN} at 0.5 % SS, d-f) $\sigma_{abs,525nm}$ and g-f) $\sigma_{sc,525nm}$ for polluted (navy), growth event (purple), plume 1 (blue), plume 2 (red) and plume 3 (yellow) conditions relative to BG (grey) conditions.

4 Conclusion

In situ ground-based and tethered-balloon measurements were conducted over a six-week campaign in Narsaq, Southern Greenland, from June to August 2023, as part of the *GreenFjord* project (“Greenlandic Fjord Ecosystems in a Changing Climate: Socio-cultural and Environmental Interactions”). The campaign aimed to quantify the contribution of different aerosol sources to climate-relevant particle properties.

Narsaq, a small town (population $\sim 1,300$) located in a sub-Arctic fjord system, experienced an exceptionally dry summer with high solar radiation and low wind speeds. Based on aerosol characteristics and air mass origins, we classified the observations into four categories: persistent aged aerosol background conditions, local fresh anthropogenic pollution, regional particle growth events, and long-range transported biomass burning (BB) plumes from Canada. Local pollution dominated 50% of the observation period, background conditions accounted for $\sim 34\%$, while growth events and BB plumes each contributed $\sim 8\%$.

Overall, number size distributions were typically Aitken-mode dominated, consistent with marine-influenced environments where new particle formation and growth play a major role (Croft et al., 2019, 2021; Gong et al., 2020; Heintzenberg et al., 2000; O'Dowd et al., 2004; Park et al., 2020). Fresh anthropogenic pollution particle number size distributions were similar to the background conditions and could not be disentangled completely due to their intermittent nature. Local pollution is likely caused by traffic and domestic activities, including residential heating powered by varied fuel sources, and therefore had a strong influence on the aerosol absorption coefficient. Biomass burning plumes, by contrast, contained aged accumulation-mode particles with enhanced N_{CCN} , $\sigma_{abs,525nm}$ and $\sigma_{sc,525nm}$ ~~enhanced optical properties~~. Particle growth events were primarily linked to marine air masses advected from the west, likely associated with biological activity during the summer bloom period. Growth events seem to be an important contributor to the background aerosol population.

Although our observations only extended down to 8 nm particle size, the presence of nucleation modes peaking above this limit indicates that new particle formation occurred upwind of Narsaq, within roughly 30 km. This estimate assumes relatively direct transport, which may not fully apply in the complex fjord terrain, where flow channelling and deflection can occur. Vertical profiles showed that growth events developed near the surface within the lower boundary layer and were partly confined by elevated temperature inversions. On several occasions, nucleation-mode particles were also observed above these inversions, suggesting that new particle formation and growth can occur at multiple altitudes.

Analysis of biomass burning plume periods revealed significant variations in aerosol concentrations, size distributions and optical properties at the ground. A distinct accumulation mode was evident throughout the vertical column, diminishing in intensity toward the surface. Ceilometer and thermodynamic data indicated that mixing of the plumes downward was limited by temperature inversions, yet ground-level perturbations in aerosol properties confirmed that transported BB aerosols influenced the surface even without full plume descent.

Both local pollution and advected BB aerosols substantially modified climate-relevant aerosol properties. Local pollution primarily enhanced the absorption coefficient, though with large temporal variability. The three BB plumes differed markedly in their microphysical and optical properties but consistently produced strong increases in median CCN number concentration (at 0.5% SS), as well as in median absorption and scattering coefficients, by factors of between 1.7-3.7, 1.8-4.0, and 1.4-4.8, respectively. Growth events slightly shifted the probability distributions of the scattering coefficient and CCN number concentration toward higher values, while modestly reducing absorption.

These findings show that different aerosol sources exert distinct influences on particle number, size, and optical characteristics, as well as on CCN activity. Such source-specific characterization is essential for constraining aerosol–cloud–radiation interactions, particularly in rapidly changing sub-Arctic coastal regions where both climate and human activity are evolving. The pronounced impact of biomass burning aerosols is especially noteworthy given the projected increase in wildfire frequency and intensity across the boreal belt.

Data availability

675 Data is publicly available on Zenodo.

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Author Contributions

J.A, N.B, L.F, B.H, J.W, M.S, K.V and J.S took part in field campaign to collect ground-based and vertical measurements. J.S. conceptualized the measurements and acquired funding. J.A processed and analysed ground data with support from N.B and
715 B.H. J.A processed and analysed helikite data. J.A and J.S interpreted the data and wrote the manuscript. J.W, M.A and P.W provided their expertise on biomass burning plume content. K.V, J.W and P.W provided the expertise with offline filter and chemical composition analysis. S.H provided expertise on CCN measurements. L.F provided technical support and expertise during the field campaign with both ground-based instrumentation and the helikite platform. All co-authors contributed to the editing and improvement of the manuscript.

720 **Competing interests**

The authors declare that there are no competing financial or personal interests affecting the research presented in this paper.

Acknowledgements

This work was supported by funding from the Swiss Polar Institute grant no. SPI-FLAG-2021-002 Greenfjord, Swiss National Science Foundation grant no. 200021_212101, and the ENAC Flagship 2022 ECO-Plains. This project has received funding
725 from the European Union's Horizon 2020 research and innovation program under grant agreement no. 101003826 via project CRiceS (Climate Relevant interactions and feedbacks: the key role of sea ice and Snow in the polar and global climate system). J.S holds the Ingvar Kamprad chair for Extreme Environments Research sponsored by Ferring Pharmaceuticals. We thank the Narsaq International Research Station team, especially Lise Autogena, and the residents of Narsaq for their generosity, curiosity, and close collaboration throughout our work. We are grateful to the Kujalleq commune for their support and for
730 granting the land-use permissions that made this research possible. We recognize that our study took place on the traditional lands of the Greenlandic people and deeply appreciate the opportunity to carry out this research in a spirit of respect and cooperation with the community. We would like to thank Jakob Pernov, Michael Lonardi, Yolanda Temel and Kathleen Alden

for their contribution to multiple scientific discussions and their support of J.A for the preparation of this manuscript and the science within.

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1025