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Marine Carbohydrates and Other Sea Spray Aerosol Constituents Across Altitudes in the Lower Troposphere of Ny-Ålesund, Svalbard

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Abstract

Marine combined carbohydrates in aerosol particles ($CCHO_{aer}$) have the potential to influence cloud formation and properties, but it remains unclear to what extent they reach altitudes relevant for cloud processes. Balloon-borne measurements of major sea spray aerosol (SSA) constituents, including sodium (Na^+_{aer}) and $CCHO_{aer}$, were conducted in autumn 2021 and spring 2022 in Ny-Ålesund (Svalbard). Total suspended particles were collected at 321-1112 m, covering both the marine boundary layer and the free troposphere, with Na^+_{aer} ranging 23–850 ng m^{-3} and $CCHO_{aer}$ 3.8–274 ng m^{-3}. The chemical composition of balloon-borne aerosol samples was compared with synchronized ground level measurements at the balloon's winch (Na^+_{aer}: 35–3710 ng m^{-3}; $CCHO_{aer}$: 1.9–194 ng m^{-3}), and at the Old Pier (Na^+_{aer}: 140–1470 ng m^{-3}; $CCHO_{aer}$: 1.6–10.0 ng m^{-3}), where freshly emitted SSA particles were sampled. Surface seawater from the Kongsfjorden was analyzed to evaluate the sea-air transfer of marine CCHO. Air mass histories, atmospheric mixing, and cloud conditions were evaluated for three selected cases to explain vertical concentration patterns. A strong correlation (R=0.78,

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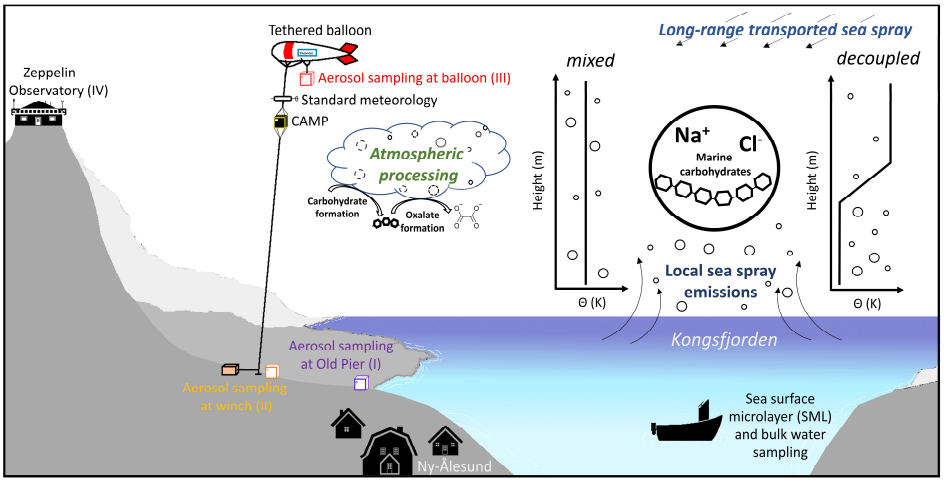
p<0.001) between combined xylose (<0.2–14.1 ng m^{-3}) in $CCHO_{aer}$ and $oxalate_{aer}$ (<1–67 ng m^{-3}) across all altitudes, suggests either coproduction or a connection through atmospheric processing. These results provide a first comprehensive picture of local primary sea-air transfer of marine combined carbohydrates and highlight the roles of long-range transport, in-situ formation, and chemical aging in shaping their atmospheric distribution.



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1. Introduction

Aerosol particles in the High Arctic atmosphere originate from a complex interplay of primary and secondary emissions from oceanic, terrestrial, cryospheric, and anthropogenic sources, followed by diverse atmospheric processes (Schmale et al., 2021). They play a crucial role in the radiation balance, both directly by scattering and absorbing sunlight and indirectly by influencing cloud formation and phase state through functioning as cloud condensation nuclei (CCNs) and ice-nucleating particles (INPs) (Lohmann and Feichter, 2005; Penner et al., 2001; Quinn et al., 2015; Yu et al., 2006). These effects are strongly governed by the particles' size distribution and chemical composition (Dusek et al., 2006; Farmer et al., 2015; Kanji et al., 2017; Pilinis et al., 1995).

The High Arctic predominantly consists of marine areas, characterized by a seasonally variable extent of sea ice cover and open waters. Consequently, sea spray aerosol (SSA) particles represent a key group of primary aerosol particles in this region (Heutte et al., 2025; Kang et al., 2025; Schmale et al., 2022). As Arctic sea ice coverage continues to decline due to global warming, enhanced by Arctic amplification (Cai et al., 2021; Francis and Wu, 2020; Wendisch et al., 2017, 2023), larger expanses of open ocean are anticipated to become significant sources of SSA emissions (Browse et al., 2014; Struthers et al., 2011). Although direct measurements remain sparse, Sharma et al. (2019) readily observed increasing sea salt aerosol production from sea spray over 34 years at the Arctic air chemistry observatory in Alert, Canada.

SSA particles are generated through wind-driven wave action, which causes bubbles at the sea surface to burst, ejecting film and jet droplets into the atmosphere (Veron, 2015). SSA particles primarily consist of inorganic sea salt ions, mainly sodium and chloride, along with organic matter (OM), including significant amounts of marine carbohydrates originating from the sea surface microlayer (SML) and the underlying bulk seawater (Müller et al., 2010; van Pinxteren et al., 2023; Quinn et al., 2015; Russell et al., 2010). In seawater, carbohydrates are produced by both unicellular and multicellular organisms, predominantly as linear or branched oligo- and polysaccharides (Aluwihare et al., 1997; Borch and Kirchman, 1997; Engel and Händel, 2011; Khadem, 2012), collectively referred to as combined carbohydrates (CCHO). They also exist as dissolved free carbohydrates (DFCHO), monosaccharides often rapidly

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consumed by heterotrophic bacteria (Arnosti et al., 2021; Engel and Händel, 2011; Ittekkot et al., 1981; Kirchman et al., 2001).

Sodium in aerosol particles (Na^{+}_{aer}) is highly abundant in the marine boundary layer with only minor terrestrial sources and greater atmospheric stability compared to chloride (Cl^{-}_{aer}) (Chi et al., 2015; Keene et al., 1986; Manders et al., 2010; Sander et al., 2003). This makes it a valuable conservative tracer for studying the sea-to-air transfer and atmospheric transformation of organic compounds, including marine carbohydrates, as well as other inorganic SSA constituents. Notably, the ratio of OM to Na^{+} is significantly higher in SSA particles than in seawater, reflecting not only the preferential enrichment of surface-active substances at the interface but also a more complex interplay of factors such as water solubility, biological activity within the ocean surface, and co-adsorption processes involving matrix constituents (Burrows et al., 2014; Gantt et al., 2011; Hasenecz et al., 2020, 2019; Hoffman and Duce, 1976; Jayarathne et al., 2016; van Pinxteren et al., 2017; Quinn et al., 2015; Russell et al., 2010; Schill et al., 2018). This enrichment is particularly pronounced in submicron particles compared to supermicron particles. Furthermore, following the sea-to-air transfer of OM and CCHO, recent laboratory (Hasenecz et al., 2020; Malfatti et al., 2019) and field (Zeppenfeld et al., 2021, 2023) observations suggest their molecular transformation or additional in-situ formation, driven by abiotic, microbial or enzymatic activities in the atmosphere.

SSA particles are known to function as both CCNs (Orellana et al., 2011; Xu et al., 2022) and INPs (Alpert et al., 2022; DeMott et al., 2016; Hill et al., 2023; Mirrielees et al., 2024), underscoring their important role in cloud microphysics, cloud formation, and precipitation processes. Recently, Hartmann et al. (2025) demonstrated, through a combination of lab and field data, that SSA particles' ice-nucleating activity is likely attributable to the polysaccharides they contain. Model simulations further indicated that the ice-nucleating activity of marine polysaccharides is particularly significant within the temperature range between -20 and -15°C in remote oceanic regions, where contributions from terrestrial INP sources are minimal or absent. Furthermore, Rocchi et al., (2024) demonstrated that the presence of glucose-rich CCHO, in combination with sea salt, significantly enhances SSA production in eastern Arctic waters. This finding may improve the predictability of SSA emissions in marine models.

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In the field, marine combined carbohydrates in aerosol particles ($CCHO_{aer}$) have been predominantly measured at ship-based or coastal locations, which are in close proximity to local marine emission sources both horizontally and vertically (Leck et al., 2013; van Pinxteren et al., 2023; Zeppenfeld et al., 2021, 2023). In contrast, only a few studies have investigated $CCHO_{aer}$ (Karl et al., 2019; Yttri et al., 2024) at an elevated mountain site in a marine-influenced setting, aiming to assess atmospheric concentrations at higher altitudes. Vertically resolved field data comparing ground-level and elevated altitudes using mobile platforms, however, have been unavailable for marine $CCHO_{aer}$ in the past. As a result, it remains unclear to what extent and under which conditions $CCHO_{aer}$ reach the upper marine boundary layer and the free troposphere. This is due to several challenges, including low atmospheric concentrations pushing the instruments' detection capabilities for offline analyses to their limits, the lack of highly resolving online detection techniques for $CCHO_{aer}$, and in particular the absence of lightweight yet powerful pumps with high flow rates. Additionally, the very short sampling times typically available on mobile airborne measurement platforms pose a further obstacle for measuring marine $CCHO_{aer}$ in aerosol particles across altitudes within the troposphere. This lack of vertical field data leaves high uncertainty about the broader relevance of these biomolecules in cloud formation and glaciation beyond a controlled laboratory setup.

Previous airborne measurements around Svalbard (Hara et al., 2003) and the Canadian Arctic (Köllner et al., 2017) demonstrated that SSA particles—identified by Na^+ and Cl^-—are present in higher altitudes of the lower troposphere, and, to a lesser extent, reach the middle free troposphere (3–6 km a.s.l.). Some of these aerosol particles showed signs of atmospheric aging, such as the replacement of chloride with nitrate and sulfate in the SSA particles. While vertically resolved data exists for major inorganic SSA constituents, such extended information is lacking for marine $CCHO_{aer}$.

Recent methodological advances now allow for a more detailed investigation of the transport mechanisms and atmospheric chemical fate of marine carbohydrates. In this study, we present atmospheric concentrations of these biomolecules alongside common inorganic SSA constituents. Measurements were conducted from ground level up to various altitudes within the boundary layer and lower free troposphere using a tethered helium balloon in Ny-Ålesund on Svalbard during autumn 2021 and spring 2022. For selected cases, we examined the influence of mixing state, meteorological conditions, and air mass history on the observed aerosol composition. Finally, this study addresses the potential atmospheric processing and

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transformation of marine carbohydrates, with a focus on their possible contribution to secondary aerosol formation and their implications for atmospheric chemistry and cloud-relevant processes.

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2. Experimental

2.1 Study area: Ny-Ålesund as an atmospheric observation site

Ny-Ålesund, located at 78.9°N at the Kongsfjorden in Svalbard (Norway), belongs to the world's northernmost permanently inhabited settlements with a year-round accessibility. It serves as a key research site for studying Arctic climate change and Arctic amplification. Ny-Ålesund hosts long-term monitoring sites for aerosols and meteorology, such as the Zeppelin Observatory (Platt et al., 2022), Gruvebadet (Amore et al., 2022), and the AWIPEV Observatory (Maturilli et al., 2013, 2015). These, along with additional research stations operated by various international institutions, provide valuable data for both long-term atmospheric studies and short-term investigations like the present one.

However, Ny-Ålesund is not representative of the entire High Arctic. Its distinct topography, situated within a fjord and surrounded by high mountains up to 800 m, leads to complex atmospheric dynamics, including foehn-like effects (Shestakova et al., 2021). The local boundary layer is relatively shallow characterized by an average mixing layer height below 700 m and a strong influence by orographic effects (Chang et al., 2017; Dekhtyareva et al., 2018; Gierens et al., 2020). While free-tropospheric winds are predominantly westerly, surface winds result from an interplay of land-sea breeze circulations, southeasterly channeled winds along the fjord axis, and katabatic flows from the Zeppelin mountain range, the Broeggerbreen glacier, or the Kongsvegen glacier (Esau and Repina, 2012; Gierens et al., 2020). Additionally, large wind shear has been observed to generate turbulence, leading to frequent neutral stratification (Gierens et al., 2020). Furthermore, boundary layer mixing can occur even when a positive gradient in potential temperature suggests a more stable stratification. During the present field campaign, we observed that near-surface winds often shift unpredictably, changing direction without a clear pattern, making airflow dynamics challenging to interpret.

From an oceanographic perspective, Svalbard is similarly exceptional. The region is influenced by the cold Arctic waters of the Spitsbergen Polar Current and the warm waters of the West Spitsbergen Current (Feltracco et al., 2021). Kongsfjorden, located on the western coast of Spitsbergen, lies at the interface of High Arctic and Atlantic influences, making it a dynamic and variable environment (Bischof et al., 2019).

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168 Therefore, findings from Ny-Ålesund may not be fully transferable to atmospheric processes
169 over sea ice or the open ocean in the High Arctic. However, in general, the representativeness
170 of any single Arctic site is highly questionable, as Freud et al., (2017) found significant
171 heterogeneity in aerosol particle size distribution across all Arctic sites in their study.

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173 2.2 Field sampling

174 The field samples (aerosol particles, bulk seawater and SML) for this study were collected near
175 Ny-Ålesund and from the adjacent Kongsfjorden during autumn 2021 and spring 2022.

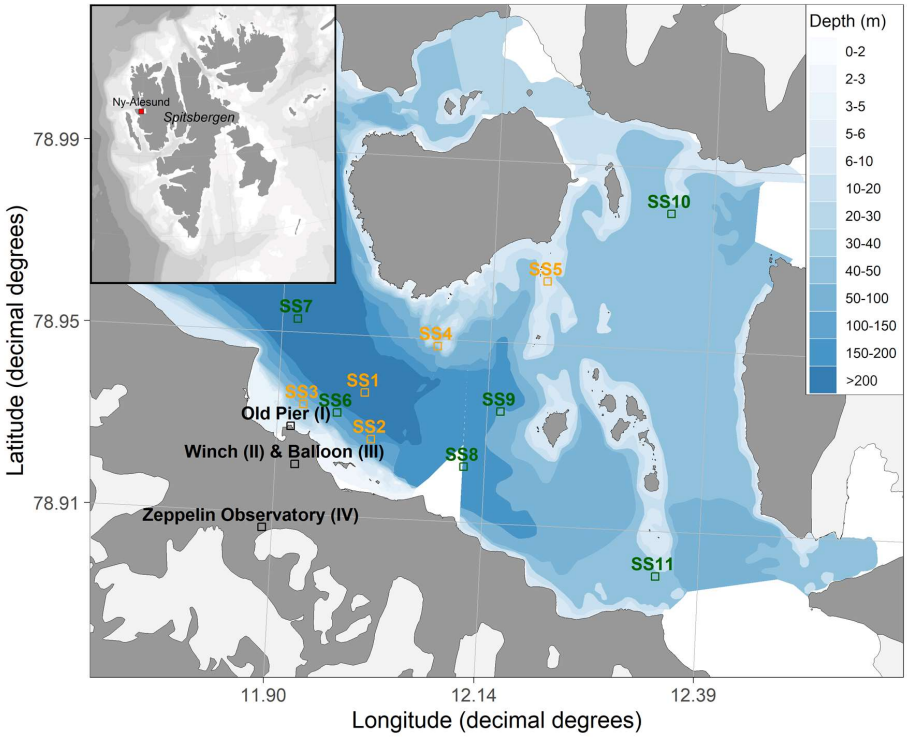

Figure 1. Map of the sampling locations. Aerosol particles were collected at: (I) the Old Pier, representing fresh SSA emissions; (II) the winch, representing ground measurements; (III) the tethered balloon at various altitudes; and (IV) the Zeppelin Observatory, serving as a reference for comparison. Bulk and SML samples were collected from different locations within Kongsfjorden. Orange squares (SS1-SS5) indicate autumn 2021 samples, while green squares (SS6-SS11) represent spring 2022 samples. Blue shading indicates water depth.

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a) Bulk seawater and SML sampling

In total, 11 bulk surface seawater and 11 SML samples were taken from a small boat at various dates and locations across the Kongsfjorden (**Figure 1**, **Table S1**). Bulk water samples were obtained from a depth of 1 m using low-density polyethylene (LDPE) bottles secured to a telescopic rod. The corresponding SML samples were collected using the glass plate technique (Cunliffe and Wurl, 2014; van Pinxteren et al., 2012). A glass plate measuring 50 cm × 20 cm × 0.5 cm, with an oval sampling area of 2000 cm^2, was immersed vertically into the surface of the fjord seawater and withdrawn at a steady rate of 15 cm s^{-1}. The SML film attached to the glass surface was drained into a precleaned wide-neck plastic bottle using a funnel and a framed Teflon wiper. Water samples were filtered through 0.2 µm polycarbonate filters (Whatman® Nuclepore™, 47 mm diameter) to separate dissolved and particulate fractions. The filtrate, filters and field blanks were preserved at −20°C until chemical analyses (inorganic ions, carbohydrates). Sea surface temperature (SST) was measured directly from the boat at a depth of approximately 10 cm using a digital thermometer.

b) Aerosol particle sampling in the surroundings of Ny-Ålesund

Total suspended aerosol particles (TSP) were captured on polycarbonate filters (0.8 µm, Whatman® Nuclepore™, 47 mm diameter) at four locations (**Figure 1**): (I) Near the Old Pier next to Kongsfjorden (8 samples), representing fresh SSA emissions; (II) near the balloon winch close to the AWIPEV Observatory (17 samples), representing ground measurements; (III) at high altitudes at the tethered balloon (14 samples); and (IV) at the Zeppelin Observatory (1 sample), serving as a reference for comparison. **Table S2** provides details of individual aerosol particle samplings near the Old Pier (I), while **Table S3** presents the sampling times, locations and heights of all the individual high-altitude aerosol samples (III & IV), along with the corresponding simultaneous ground-level samples (II) taken near the winch.

For sampling aerosol particles at the Old Pier (4 m above sea level), a filter holder with a polycarbonate filter attached to a pump was used. Sampling lasted between 4 and 7 days. Flow rates, measured at the beginning and the ending of the sampling with a flowmeter, ranged from 5 to 10 L min^{-1}, with total air volumes between 44 and 82 m^3. The estimated diameter-dependent collection efficiency of this TSP sampling setup, assuming a 90° aspiration angle, is shown in **Figure S1**. To reduce the risk of pump failure due to cold temperatures or snow, the pumps were housed in a Zarges box for protection.

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High-altitude TSP samples were collected using the helium-filled tethered balloon BELUGA, as described in detail by Pilz et al. (2023). The balloon's altitude was controlled using an electric winch located near the AWIPEV Observatory, with ascent and descent rates from 1 to 3 m s^{-1}. The tethered balloon operated under various meteorological conditions, including both clear and cloudy skies. At a specified altitude, a HALFBAC (High-volume And Light-weight Filter sampler for BAlloon-borne appliCation) (Grawe et al., 2023) collected aerosol particles 2-3 m below the balloon. The HALFBAC is a custom-designed, lightweight aerosol particle sampler operating at a pump flow between 25 and 35 L min^{-1}. It is capable of collecting sufficient aerosol mass on filters at high altitudes for subsequent offline chemical and microphysical analyses. Simultaneously, another HALFBAC collected ground-level aerosol particles near the electric winch (20 m above sea level). Additionally, one aerosol sample (Filter ID 62, sampling date: 10/05/2022) was collected at the Zeppelin Observatory, a permanent monitoring station located at 474 m a.s.l. on Zeppelinfjellet, using the HALFBAC. Synchronized aerosol particle sampling at the winch and the balloon typically lasted around two hours, as detailed in **Table S3**. The collection efficiency for TSP sampling using HALFBAC is discussed in the supplement (A1) and **Figure S1**.

2.3 Chemical analyses from offline aerosol particle filters and seawater

For the analysis of major cations, anions and marine carbohydrates in aerosol particles, the complete polycarbonate filters were extracted in 6-7 mL of ultrapure water (resistivity > 18.2 MΩ) for two hours followed by a filtration through a 0.45 µm syringe filter. Frozen seawater samples were gently thawed at 4°C one day before analysis.

a) Major cations and anions

Major inorganic ions, including sodium (Na^+), potassium (K^+), magnesium (Mg^{2+}), calcium (Ca^{2+}), chloride (Cl^-), sulfate (SO_4^{2-}), and oxalate, were quantified in 0.45 µm filtered aqueous aerosol extracts, bulk seawater and SML samples using ion chromatography (Dionex ICS-6000, Thermo Scientific) as described by Zeppenfeld et al. (2021). Cations were separated isocratically with a 36 mM methanesulfonic acid eluent on a Dionex IonPac CS16−4 µm column (2 mm × 250 mm), paired with a Dionex IonPac CG16−4 µm guard column (2 mm × 50 mm). For anion separation, a gradient from 4 to 40 mM KOH was applied on a Dionex IonPac AS18

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column (2 mm × 250 mm), along with a Dionex IonPac AG18 guard column (2 mm × 50 mm). The analytical uncertainty for each ion was below 5%. Aerosol extracts were measured undiluted, while bulk seawater and SML samples were analyzed at a 1:15,000 dilution.

b) Dissolved free and combined carbohydrates

Carbohydrates in seawater and aerosol particle extracts were measured according to the protocols outlined by Zeppenfeld et al. (2020, 2021), utilizing high-performance anion-exchange chromatography with pulsed amperometric detection. The system was equipped with a Dionex CarboPac PA20 analytical column (3 mm × 150 mm) and a Dionex CarboPac PA20 guard column (3 mm × 30 mm). The applied eluent gradient separated the following monosaccharide units: fucose (Fuc), rhamnose (Rha), arabinose (Ara), galactose (Gal), glucose (Glc), xylose (Xyl), mannose (Man), fructose (Fru), galactosamine (GalN), glucosamine (GlcN), muramic acid (MurAc), galacturonic acid (GalAc), and glucuronic acid (GlcAc). The analytical uncertainty for each monosaccharide was below 10%. dFCHO represents the total of identifiable free monosaccharides, whereas CCHO include only those monosaccharides released through acid hydrolysis (0.8 M HCl, 100°C, 20 h). For seawater samples, particulate combined carbohydrates (pCCHO, >0.2 µm) were measured from 0.2 µm polycarbonate filters, while dissolved combined carbohydrates (dCCHO, <0.2 µm) were measured from the filtrate after desalination via electrodialysis. Both fractions were later summed to represent the total CCHO.

2.4 Vertical profiles from online measurements

a) Size-resolved aerosol particles number concentrations

An optical particles size spectrometer (POPS, Handix), integrated into the Cubic Aerosol Measurement Platform (CAMP) as described by Pilz et al. (2022), provided the integrated total number concentrations (N_{150}) for aerosol particles between 150 and 2900 nm at a temporal resolution of 1 second. On selected dates of HALFBAC sampling, CAMP was operated simultaneously 25 m below the balloon providing insight into the vertical profile of N_{150} during specific events. Vertical profiles are presented as rolling averages over 30 seconds.

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b) Meteorological observations and calculations

Standard meteorological parameters—including altitude, ambient temperature (T), wind speed (U), wind direction, air pressure (p), and relative humidity (RH)—were measured for the elevated-altitude samples using a standard meteorology package positioned approximately 20 m below the balloon (Pilz et al., 2023). The potential temperature (θ) within the atmospheric column - as a measure of the static stability of the unsaturated atmosphere - was calculated using Eq. I, where T is the ambient temperature (K), p is the atmospheric pressure (hPa), p_0 is the reference pressure (1000 hPa), R is the specific gas constant (287 J kg^{-1} K^{-1}) and c_p is the specific heat capacity of dry air at constant pressure (1004 J kg^{-1} K^{-1}).

$$\theta = T\left(\frac{p_0}{p}\right)^{\frac{R}{c_p}} \quad \text{(Eq. I)}$$

Specific humidity (q)–remaining constant during adiabatic ascent or descent as long as no phase changes occur–was calculated using Eq. II from Egerer et al. (2021), where R_d/R_v (the ratio of specific gas constants for dry air and water vapor) is approximately 0.622, and $e_s(T)$ represents the temperature-dependent saturation vapor pressure.

$$q = \frac{R_d/R_v \cdot e_S(T) \cdot RH}{p - (1 - R_d/R_v) \cdot e_S(T) \cdot RH} \quad \text{(Eq. II)}$$

Meteorological data measured 2 m above the ground (13 m above sea level) at the AWIPEV Atmospheric Observatory (Maturilli, 2020), represented the weather conditions during aerosol sampling at the winch.

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2.5 Supporting observations and model calculations

Major inorganic ions measured at the Zeppelin Observatory with a 24-hour resolution using a statically installed aerosol sampler (Filter_3pack) as part of the European Monitoring and Evaluation Programme (Tørseth et al., 2012) by the Norwegian Polar Institute (NPI) and the Norwegian Institute for Air Research (NILU) were obtained from the EBAS database for the study duration (Aas et al., 2022, 2023). The Filter_3pack data were utilized in two ways:

1. **Comparing sampling techniques:** Data from the Filter_3pack were compared with one HALFBAC aerosol particle sample collected directly at the Zeppelin Observatory (Filter ID 62, 10/05/2022) to evaluate potential artifacts arising from differences in sampling techniques and filter media. Despite variations in time resolution and methods, sodium, potassium, chloride, and sulfate concentrations showed strong agreement (detailed in the supplement A2 and **Figure S2**).
2. **Comparison with balloon data:** Sodium concentrations measured at the Zeppelin Observatory were directly compared with those obtained from the tethered balloon sampling.

Information on the occurrence of clouds and hydrometeor types at Ny-Ålesund were taken from the Cloudnet classification product (Illingworth et al., 2007; Nomokonova et al., 2019), which is based on a combination of ground-based cloud radar, ceilometer, and numerical weather prediction output. Vertically integrated ice water content (IWC), i.e. ice water path (IWP) has been calculated from the Cloudnet IWC product following Hogan et al. (2006). Vertically integrated cloud liquid water (liquid water path; LWP) and water vapor (IWV) were taken from zenith HATPRO microwave radiometer measurements (Nomokonova et al., 2019).

The 48-hour back-trajectories for the aerosol sampling periods were generated using the NOAA HYSPLIT model (Stein et al., 2015). Trajectories were calculated hourly based on GDAS1 meteorological data (Global Data Assimilation System; 1° spatial resolution; 3-hour intervals) for various arrival heights: 50 m (ground level), 474 m (Zeppelin Observatory), and the specific balloon sampling altitudes. Sea ice concentration data were obtained from the NOAA-maintained ERDDAP server (Environmental Research Division's Data Access Program). The back-trajectories were used to assess the relative influence of distant sources, such as the marginal ice zone, versus local ice-free oceanic emissions on the aerosol chemical composition. Given the rather short atmospheric residence times of SSA particles—typically

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less than two days for supermicron particles (Madry et al., 2011; Veron, 2015), which account for most of the SSA mass —and the increasing uncertainties associated with longer back-trajectory periods, we consider 48 hours back-trajectory length appropriate for this analysis.

Ocean surface concentrations for total chlorophyll *a* (TChl-a) and dissolved acidic polysaccharides were obtained by a coupled setup of the ocean sea ice biogeochemistry model FESOM2.1-REcoM3 (Gürses et al., 2023), to which additional state equations have been added to simulate dissolved and particulate organic carbon following Engel et al. (2004) and Schartau et al. (2007). The simulation was set up following Gürses et al. (2023) and using the Arctic-specific tuning of Oziel et al. (2022). Monthly model output was obtained on an irregular grid with approximately 4.5 km resolution in the Arctic Ocean. This configuration has already been applied successfully in Leon-Marcos et al. (2025).

2.6 Statistics, data processing, visualization and text optimization

Statistical analyses, calculations and visualization were conducted using OriginPro 2024, Microsoft Excel, IDL, python3 and R version 4.2.1 with the ncdf4 (Pierce, 2023), openair (Carslaw and Ropkins, 2012), reshape2 (Wickham, 2007), scales (Wickham et al., 2023b), lubridate (Grolemund and Wickham, 2011), cmocean (Thyng et al., 2016), maps (Brownrigg, 2023), mapdata (Brownrigg, 2013), rgdal (Bivand et al., 2022), raster (Hijmans, 2023), RColorBrewer (Neuwirth, 2022), sp (Bivand et al., 2013), dplyr (Wickham et al., 2023a), ggplot2 (Wickham, 2016), and PlotSvalbard (Vihtakari, 2020) packages. Box-and-whisker plots illustrate the interquartile range (box), the median (horizontal line inside the box), the mean (open square), the minimum and maximum values (whiskers). Text and language were optimized using Open AI's ChatGPT-4 Turbo.

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3. Results and Discussion

3.1 Chemical constituents in marine aerosol particles from their oceanic source to elevated altitudes

Sodium in aerosol particles (Na^+_{aer})

Sodium, a dominant and chemically stable component of SSA, is commonly used as a tracer for tracking ocean-derived emissions in atmospheric studies (Manders et al., 2010; van Pinxteren et al., 2017; White, 2008). In this study, consistently high Na^+_{aer} concentrations were observed on the TSP filters at the Old Pier next to Kongsfjorden in both autumn 2021 and spring 2022 (**Figure 2a**), ranging from 140 to 1470 ng m^{-3} (median: 495 ng m^{-3}; n=8). During the campaign, sea ice coverage in Kongsfjorden varied; however, significant areas around Ny-Ålesund—particularly near the Old Pier—remained ice-free, suggesting that local sea ice likely had only little influence on sea spray emissions.

Na^+_{aer} concentrations measured at the winch site, located further inland but still at ground level (35–3710 ng m^{-3}; median: 155 ng m^{-3}; n=17), and in elevated HALFBAC samples between 321 and 1112 m (23–850 ng m^{-3}; median: 124 ng m^{-3}; n=15) were generally lower in both minimum and median values compared to those at the Old Pier. Nevertheless, occasional high concentration events were observed across all three sampling locations. The large variability in Na^+_{aer} concentrations, ranging from the lower ng m^{-3} to a few µg m^{-3}, aligns with other marine sampling locations and altitudes (Fomba et al., 2014; Li et al., 2024; Ooki et al., 2002; Theodosi et al., 2010; Triesch et al., 2021; Zeppenfeld et al., 2021, 2023).

Since aerosol particle sampling at the winch and the balloon was always synchronized, direct comparisons between the two sites were possible. Several events showed nearly identical sodium concentrations at both locations (e.g., 30 September: 191 ng m^{-3} at the winch and 207 ng m^{-3} at the balloon; 2 October: 35 ng m^{-3} at the winch and 36 ng m^{-3} at the balloon; 9 October: 59 ng m^{-3} at the winch and 60 ng m^{-3} at the balloon; 12 November: 240 ng m^{-3} at the winch and 223 ng m^{-3} at the balloon). In contrast, other dates revealed significant differences, with much lower concentrations at higher altitudes than at the ground (e.g., 27 September: 1840 ng m^{-3} at the winch and 23 ng m^{-3} at the balloon; 5 April: 84 ng m^{-3} at the winch and 54 ng m^{-3} at the balloon; 11 May: 496 ng m^{-3} at the winch and 125 ng m^{-3} at the balloon), but also two instances of even higher concentrations recorded at the balloon (24

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September: 47 ng m^{-3} at the winch and 99 ng m^{-3} at the balloon; 3 April: 77 ng m^{-3} at the winch and 194 ng m^{-3} at the balloon). These variations can likely be attributed to atmospheric depletion processes, including dry and wet deposition (Farmer et al., 2021), atmospheric dilution during vertical and horizontal transport from the emission region (Wong et al., 2019), vertical mixing conditions (Pilz et al., 2024) and differences in air mass histories (Willis et al., 2018). The influence of atmospheric meteorological conditions on the vertical distribution of chemical compounds will be analyzed and discussed in detail for three selected cases later in this study.

Concentrations measured at the Zeppelin Observatory, where Na^{+}_{aer} is routinely monitored, were largely consistent with those observed in the balloon samples, showing an overall agreement ranging between 56 and 213%, with five events demonstrating excellent agreement of 92–107% (**Table S6**). This similarity is remarkable considering the differences in sampling time resolution (24 h at Zeppelin vs. 1–2 h for the balloon), sampling altitudes, the horizontal distance between the sites, the complex topography of Svalbard (Gierens et al., 2020; Shestakova et al., 2021), and the fact that meteorological conditions and atmospheric mixing states have not yet been considered. Up to 1100 m altitude, encompassing both the boundary layer and the free troposphere, sodium was detectable and quantifiable, sometimes at concentrations comparable to those near the emission source. This indicates that SSA compounds are either effectively mixed throughout the vertical atmospheric column, or reach heights relevant for cloud formation through advection. Such observations are consistent with aircraft-based sodium measurements reported by Hara et al. (2003) and Köllner et al. (2017).

The longer SSA particles remain in the atmosphere, the greater their exposure to atmospheric aging processes, which can alter their impact on cloud formation. While Na^{+}_{aer} in SSA is considered chemically stable, co-emitted OM including carbohydrates may undergo physical, chemical and microbial transformations, as suggested by Zeppenfeld et al. (2021, 2023). This aspect will be explored further in the following sections.

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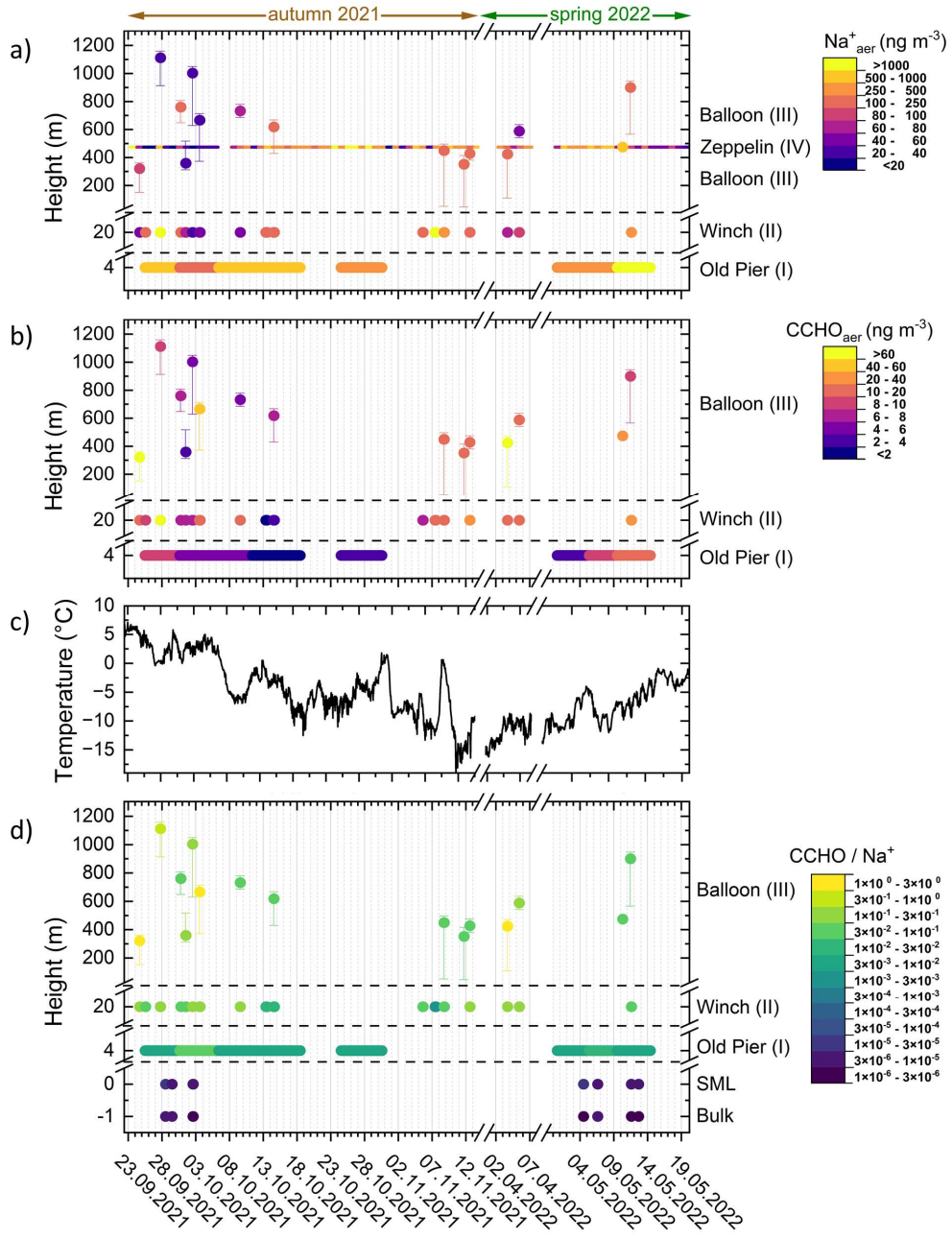

Figure 2. Time-resolved atmospheric concentrations of a) sodium and b) $CCHO_{aer}$ in aerosol particles (TSP) collected in autumn 2021 and spring 2022 in Ny-Ålesund at several heights (m a.s.l.) from four sites: Old Pier, winch near the AWIPEV Observatory, balloon and the Zeppelin Observatory. Dots represent the median height during the total sampling time and vertical error bars represent maximum and minimum height of the sampler during the active sampling. The x-axis ticks represent the start of each date at midnight. c) Air temperature (2 m above ground) measured at the AWIPEV Observatory. d) CCHO/Na^+ ratios within the bulk seawater, the SML and in the aerosol particles at several heights.

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Combined carbohydrates in fresh SSA and their oceanic origin

Similar to sodium, $CCHO_{aer}$ were detected in all atmospheric samples across all sites and altitudes (**Figure 2b**). At the Old Pier, $CCHO_{aer}$ concentrations ranged from 1.6 to 10.0 ng m^{-3} (median: 5.0 ng m^{-3}; n=8). A seasonal pattern emerged, with the highest values observed at the beginning (September 2021) and end (May 2022) of the study period, while lower concentrations were recorded during the colder, darker months in between (**Figure 2c**). However, it should be noted that no aerosol samples were collected during the coldest winter months.

The seasonal variation of $CCHO_{aer}$ at the Old Pier may be linked to the seasonal dynamics of marine CCHO in the surface water of Kongsfjorden, their most probable local emission source for SSA. These dynamics are likely driven by seasonal shifts in phytoplankton composition as well as overall primary production (Assmy et al., 2023; Mayot et al., 2018). Reduced or absent production in Kongsfjorden during the winter and early spring, as indicated by low phytoplankton and TChl-a concentrations, contrasts with significantly higher values from mid-spring to mid-autumn (van de Poll et al., 2021).

Similar seasonality was observed for selected monosaccharides among the dissolved combined carbohydrates (dCCHO) in Kongsfjorden seawater, the primary source of atmospheric $CCHO_{aer}$. In particular, fucose, galactosamine, and rhamnose in bulk water dCCHO exhibited a distinct pattern, closely following SST. Their concentrations peaked in late September/early October, while being much lower (44-67%) in early to mid-May (**Figure S3**). However, this trend was less pronounced for glucose in bulk water dCCHO and even weaker in the SML across most monosaccharide units (**Figure S3**).

In contrast, particulate combined carbohydrates (pCCHO)—including contributions from transparent exopolymer particles (TEPs), detritus, and dead or living cells—showed no clear seasonal trend in seawater (**Figure S4**). While dCCHO in bulk water exhibited relatively low spatial and intra-seasonal variability, pCCHO and SML samples were considerably more variable among all samples, even among samples from the same season (**Figure S5**). This variability likely reflects pCCHO's rapid dynamics in relation to phytoplankton blooms (Becker et al., 2020; Engel et al., 2012; Fabiano et al., 1993). Additional drivers include the spontaneous formation of TEPs from dCCHO precursors in turbulent waters, as well as the

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vertical transport of pCCHO through sedimentation (e.g., as marine snow) or its accumulation in the SML depending on its buoyancy (Burns et al., 2019; Engel, 2004; Robinson et al., 2019b, a; Wurl and Holmes, 2008). The SML, in particular, may be more sensitive to these dynamics than the more stable bulk water, potentially explaining the greater fluctuations.

On the other hand, dCCHO measured in bulk water—similar to dissolved organic carbon (Hansell, 2013; Keene et al., 2017)—may be generally dominated by recalcitrant and semi-recalcitrant compounds. The labile fraction of dCCHOs is likely more rapidly consumed by heterotrophic bacteria (Goldberg et al., 2011) and the remaining dCCHOs presumably represent substrates that are less available to microbial metabolism. Notably, combined glucose showed high variability in both dCCHO and pCCHO, likely due being the main constituent of abundant storage macromolecules such as laminarin (Becker et al., 2020) during periods of photosynthetic overflow (Barthelmeß et al., 2025), as well as its relatively rapid microbial utilization (Kharbush et al., 2020). In conclusion, the seasonal variation of $CCHO_{aer}$ aligns with the observed seasonality of certain marine carbohydrates in the Kongsfjorden, indicating that dCCHO in surface seawater may be the major origin of freshly emitted $CCHO_{aer}$.

$CCHO_{aer}$ at the winch and higher altitudes

$CCHO_{aer}$ concentrations were also measured at the winch site (1.9–194 ng m^{-3}; median: 10.6 ng m^{-3}; n=17) and at the balloon (3.8–274 ng m^{-3}; median: 10.2 ng m^{-3}; n=15), showing a broader range and significantly higher median and maximum values compared to the Old Pier. The higher concentrations, found more inland and at higher altitudes, compared to the Old Pier were unexpected and suggest sources beyond an exclusive primary sea-air transfer, which will be discussed below. Unlike the Old Pier samples, no clear seasonal pattern was evident in these locations, nor was there any apparent dependence on sampling height. The lacking seasonality at the Winch site, unlike at the Old Pier, may be due to its more inland position, making it more sensitive to wind direction and changing weather. Also, the higher temporal resolution of the samples likely captured short-term fluctuations rather than integrated seasonal trends. In addition, atmospheric processing during transport and the lack of true winter samples may have further obscured any clear seasonal signal.

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Similar to sodium, certain events showed comparable $CCHO_{aer}$ concentrations between the winch and balloon samples (e.g., 30 September: 6.5 ng m^{-3} at the winch and 8.0 ng m^{-3} at the balloon; 2 October: 6.3 ng m^{-3} at the winch and 5.8 ng m^{-3} at the balloon; 8 November: 11.1 ng m^{-3} at the winch and 10.2 ng m^{-3} at the balloon; 12 November: 26 ng m^{-3} at the winch and 17 ng m^{-3} at the balloon). On other dates, however, $CCHO_{aer}$ concentrations were markedly lower at higher altitudes (e.g., 27 September: 194 ng m^{-3} at the winch and 8.6 ng m^{-3} at the balloon; 11 May: 25 ng m^{-3} at the winch and 8.1 ng m^{-3} at the balloon). Interestingly, there were also instances where $CCHO_{aer}$ concentrations were much higher at elevated altitudes than at ground level (e.g., 24 September: 10.2 ng m^{-3} at the winch and 136 ng m^{-3} at the balloon; 3 April: 15.9 ng m^{-3} at the winch and 275 ng m^{-3} at the balloon). In most cases, $CCHO_{aer}$ covaried with sodium. However, a notable exception occurred on 3 October, when Na^{+}_{aer} concentrations were slightly lower at the balloon (35 ng m^{-3}) than at the ground (54 ng m^{-3}), whereas $CCHO_{aer}$ showed the opposite pattern, with higher concentrations at the balloon (42 ng m^{-3}) than at the winch (14 ng m^{-3}).

To investigate the emission processes from the ocean and the atmospheric fate of marine CCHO, CCHO/Na^{+} ratios were calculated for all aerosol particle, bulk seawater and SML samples representing the primary sources of the SSA particle constituents studied here (**Figure 2d**). The lowest ratios, with minimal CCHO/Na^{+} variability, were observed in bulk seawater (2.0×10^{-6} to 6.0×10^{-6}). Slightly higher ratios were found in the SML (3.3×10^{-6} to 2.5×10^{-5}), which can be explained with the known enrichment of CCHO in the SML compared to bulk seawater. Specifically, the enrichment factors (EF_{SML}) ranged between 1.3 and 4.1 for dCCHO and between 0.9 and 6.8 for pCCHO (**Figure S5**), which aligns well with the typical single-digit and occasionally two-digit enrichment factors reported in previous studies (Engel and Galgani, 2016; Gao et al., 2012; Zäncker et al., 2021; Zeppenfeld et al., 2021, 2023).

At the Old Pier, where fresh SSA was sampled, the ratios were significantly higher, ranging from 6.2×10^{-3} to 3.3×10^{-2}. This pronounced enrichment of CCHO relative to sodium in SSA particles compared to seawater has been discussed previously and is attributed to a chemo-selective sea-air transfer (Hasenecz et al., 2020, 2019; Jayarathne et al., 2016; Schill et al., 2018; Zeppenfeld et al., 2021, 2023). This process preferentially transfers surface-active organics during bubble bursting to the atmosphere, while highly water-soluble inorganic ions of the sea salt remain in the seawater. The enrichment effect is typically more pronounced in submicron particles, which have a higher relative contribution of organics than inorganic ions

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(Quinn et al., 2015). In contrast, supermicron particles are predominantly composed of sea salts, although organic substances are still notably enriched compared to the surface seawater. Since this study measured total suspended particles, and the majority of SSA particle mass typically resides in the supermicron size range (Facchini et al., 2008; O'Dowd et al., 1997), our results could be considered more representative of supermicron aerosol particles.

At the winch sampling station, located at ground level but further inland, the CCHO/Na^+ ratios within TSP aerosol particles ranged from 2.9×10^{-3} to 2.6×10^{-1}, being either similar to or slightly higher than those at the Old Pier. In contrast, balloon samples from elevated altitudes showed higher CCHO/Na^+ ratios, ranging from 3.9×10^{-2} to 1.4×10^{0}. This increase possibly results from the depletion of larger, salt-dominated supermicron SSA particles through dry and wet deposition (Croft et al., 2009; Hoppel et al., 2002; O'Dowd and de Leeuw, 2007), leading to a relatively greater contribution of OM-dominated submicron particles during transport and with extended atmospheric residence time. Furthermore, the increasing absolute concentration of CCHO at higher altitudes (**Figure 2b**) suggests an atmospheric formation process contributing to the elevated CCHO/Na^+ ratios, potentially linked to microbial activity in the atmosphere, as discussed in detail in section 3.3.

The CCHO/Na^+ ratios observed at the Old Pier and the Winch closely align with ship-based measurements in the High Arctic during the PASCAL cruise (2×10^{-3} to 2×10^{-1} for PM_{10} from summed Berner impactor stages) conducted in May–July 2017 (Zeppenfeld et al., 2023). In contrast, the very high CCHO/Na^+ values ($>1 \times 10^{0}$) observed at some elevated altitudes in this study were reported only occasionally for submicron particles (0.14–0.42 µm) during PASCAL. This may support the idea that supermicron particle deposition caused the shift in balloon sample ratios, though microbial contributions in the atmosphere are also possible. Moreover, these ratios far exceed those from the Southern Ocean near the western Antarctic Peninsula (8×10^{-4} to 7×10^{-3}) (Zeppenfeld et al., 2021), likely due to differences in surface seawater productivity.

Overall, it can be concluded that both Na^+ and $CCHO_{aer}$ are transported from the marine emission source to elevated heights within the lower troposphere. However, with longer atmospheric residence times, the chemical composition of aerosol particles appears increasingly altered in certain samples. As a key factor influencing these observations, the role

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of meteorological conditions and atmospheric mixing in linking ground and balloon samples will be discussed in the next section.

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3.2 Impact of meteorological conditions on SSA particle constituents in higher altitudes

To examine how meteorological conditions and atmospheric mixing influenced Na^{+}_{aer} and $CCHO_{aer}$ at elevated altitudes, three distinct cases with distinct, unvarying constant weather conditions were selected (**Figure 3**). These conditions allow for a detailed interpretation of the observed chemical values.

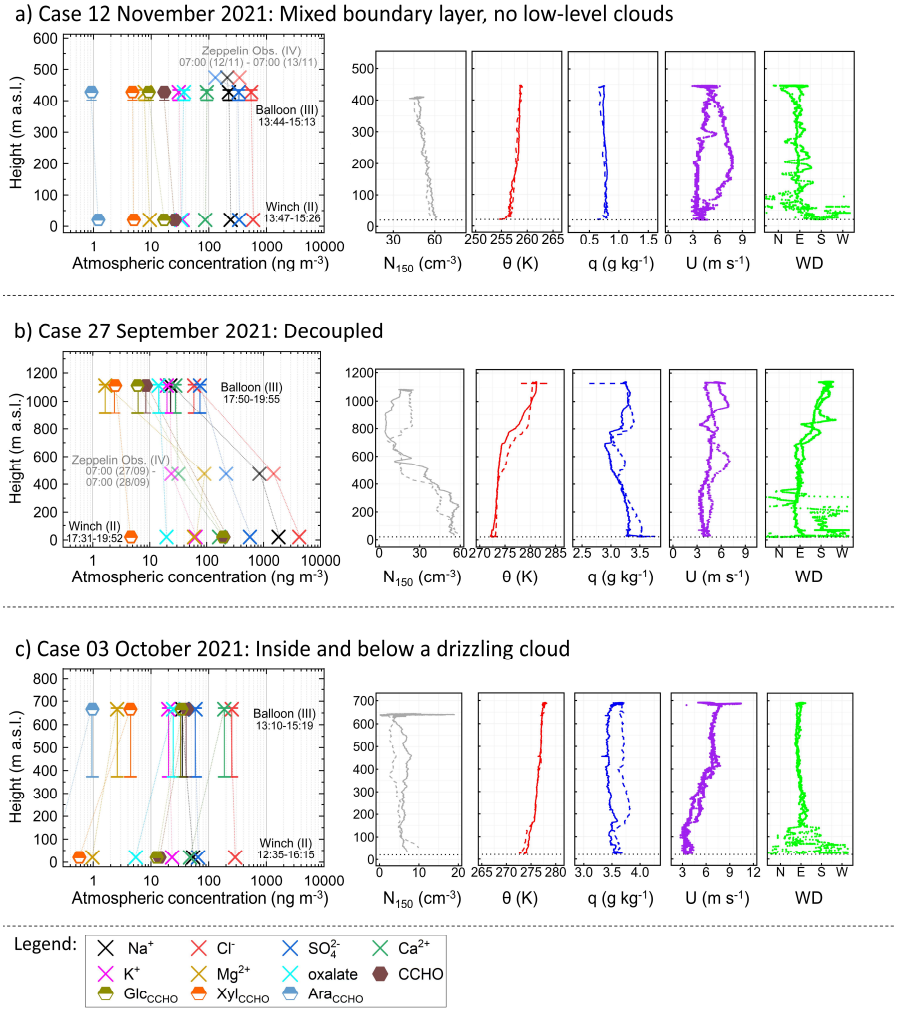

Figure 3. Vertical profiles of three atmospheric cases showing mass concentrations of chemical constituents (inorganic ions, oxalate, total $CCHO_{aer}$, and major monosaccharides within $CCHO_{aer}$) in aerosol particles, measured on the ground (winch) and aloft (balloon) using offline filters. Vertical error bars indicate the range between minimal and maximal heights during active sampling at the balloon, while the symbols denote the median sampling heights. Data from the Zeppelin Observatory are also included when available and above detection limits, albeit with a 24-hour resolution. Dotted lines are included to aid in reading the vertical distribution of individual chemical substances. These profiles are complemented by aerosol particle number concentrations of particles bigger than 150 nm (N_{150}), potential temperature (θ), specific humidity (q), wind speed (U), and wind direction (WD) measured during the ascents (solid lines) and descents (dashed lines) of the balloon.

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To assess atmospheric stability and layering in these cases, vertical profiles of potential temperature were utilized. To further confirm aerosol mixing conditions, additional meteorological parameters (specific humidity, wind speed and direction), vertical aerosol particle number concentrations of particles larger than 150 nm (N_{150}) (**Figure 3**), cloud conditions (**Figure S6**) and back-trajectory analyses (**Figure 4**) were considered. The selected cases include (a) a cloud-free mixed boundary layer (12 November 2021), (b) a free troposphere decoupled from the ground (27 September 2021), and (c) a boundary layer capped by precipitating clouds (03 October 2021).

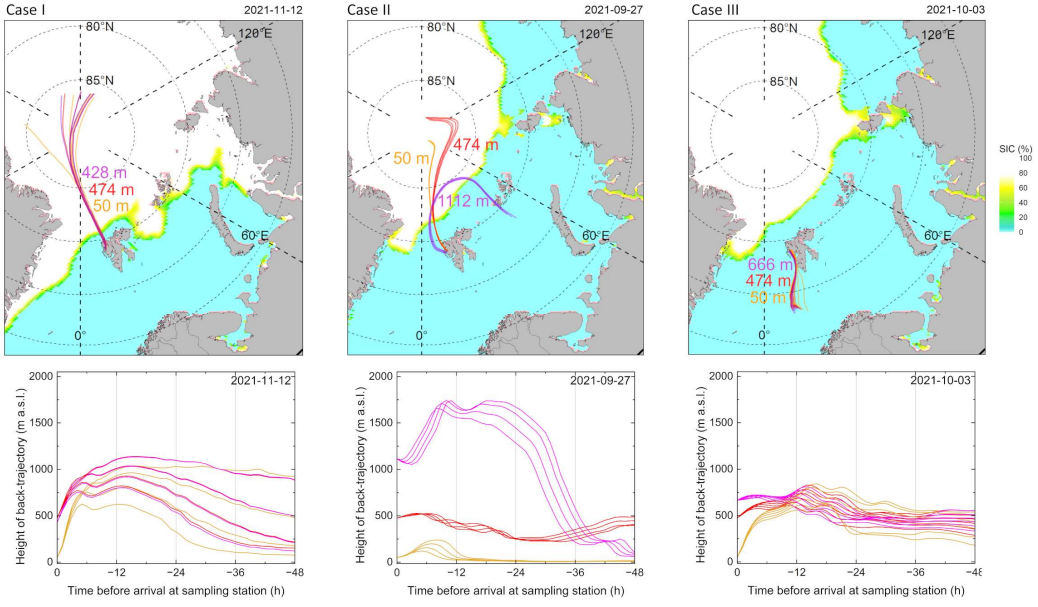

Figure 4. 48-hour back-trajectories calculated on an hourly basis for three arrival heights: orange (50 m, ground-level air masses), red (474 m, height of the Zeppelin Observatory), and purple (variable arrival height, high-altitude air masses sampled at tethered balloon). These are accompanied by daily sea ice concentration (SIC) maps (top) and height profiles (bottom) for three selected aerosol particle sampling cases.

Case I: Mixed boundary layer & no low-level clouds

On 12 November 2021, during the polar night, two HALFBACs were simultaneously operated at the ground and the balloon (median altitude of 428 m) for approximately 90 minutes. During active sampling at the ground, the average temperature was -16.7°C with a relative humidity of 69% and a wind speed of 1.5 m s^{-1} mostly from the southwest. At the balloon, sampling occurred at a similar average temperature of -17.5°C, a relative humidity of 72%, but a higher wind speed of 4.3 m s^{-1} from the northeast and southeast. With an IWV of less than 3 kg m^{-2}, the atmosphere was in general very dry (**Figure S6a**). The sky near the balloon was

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clear of low-level clouds, with only a very thin mixed-phase cloud layer from 4.5 to 5 km altitude with insignificant LWP and IWP values (**Figure S6a**). While these clouds may have had some radiative effects, they were not expected to influence aerosol chemistry measurements within the boundary layer.

During the balloon's ascent to the aerosol sampling height, potential temperature increased from 255 K to 258 K, with the strongest gradient near the surface. Together with the wind speed profile, this allowed to estimate the surface mixing layer height using the Richardson number approach (Akansu et al., 2023), resulting in a very low value of approximately 12 m, likely due to recent surface cooling. This surface inversion, together with the slightly stable to near-neutral part of the boundary layer above, would limit recent vertical mixing of aerosols from the ground into higher layers of the troposphere. However, while surface mixing layer height reflects momentary conditions and can vary within minutes to hours, the aerosol and humidity profiles represent the integrated effects of mixing over longer time scales. We therefore concluded that the current coupling state is not a reliable indicator of the actual mixing state of the atmospheric boundary layer.

Furthermore, as noted in Section 2.1, Ny-Ålesund's complex orography can induce localized turbulent mixing even under stable stratification. During the descent, the wind speed profile clearly revealed the presence of a low-level jet, with a wind speed maximum at least 2 m s^{-1} higher than the minimum values both above and below. This low-level jet is a significant additional source of turbulence and vertical mixing within the boundary layer (Egerer et al., 2023). At the ground, N_{150} was around 60 cm^{-3}, and gradually decreased to 45 cm^{-3} at the balloon's sampling height, indicating a fairly uniform aerosol number distribution and a dominant influence of primary ground-level emissions. Combined with the nearly constant specific humidity (~0.7–0.8 g kg^{-1}), a slight increase of wind speed with altitude measured during the balloon's ascent, the low-level jet observed at the decent and a consistent wind direction on the measurement day (**Figure 3a**), these observations suggest that the boundary layer was largely well-mixed.

For this event, chemical analyses from both HALFBACs (**Figure 3a**) showed similar, sometimes almost identical, concentrations of almost all inorganic ions (sodium: 240 & 223 ng m^{-3}, chloride: 586 & 543 ng m^{-3}, sulfate: 336 & 330 ng m^{-3}, calcium: 87 & 92 ng m^{-3}, magnesium: 9.5 & 7.8 ng m^{-3}, potassium: 34 & 30 ng m^{-3}), oxalate (34 & 37 ng m^{-3}) and major CCHO-bound

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monosaccharide compounds (Glc_{CCHO}: 17 & 9.1 ng m^{-3}, Xyl_{CCHO}: 5.0 & 4.7 ng m^{-3}, Ara_{CCHO}: 1.2 & 0.9 ng m^{-3}) at the ground and at the balloon. As outlined above, there are clear indications for a well-mixed boundary layer, as evidenced by the similar concentrations of all these compounds, including marine $CCHO_{aer}$, at both the ground and balloon. Despite their potentially diverse origins—such as SSA, terrestrial dust, anthropogenic emissions, and secondary formation processes—the vertical distribution of aerosol particle constituents remained uniform. The major inorganic compounds sodium, chloride, and sulfate were also measured at the Zeppelin Observatory with a 24-hour time resolution, showing only slightly lower concentrations and good agreement with the chemical results from the balloon.

Back-trajectory analysis for three arrival heights—ground, balloon, and Zeppelin Observatory (**Figure 4, Case I**)—revealed that air masses at all levels followed the same path within the 48 hours before sampling. Originating from the Arctic pack ice, they crossed the marginal ice zone with a short residence time before passing over the ice-free ocean and Kongsfjorden, where most SSA compounds were likely taken up. The back-trajectory heights indicate a vertical connection between the three air masses, confirming a similar transport history, influenced by the same emission sources.

This case demonstrates that major SSA particle constituents, including Na^{+}_{aer}, Cl^{-}_{aer}, and $CCHO_{aer}$, can mix effectively within the boundary layer. Under favorable meteorological conditions, this mixing allows these compounds to reach elevated altitudes relevant to cloud formation, maintaining concentrations nearly identical to those at the ground. Moreover, such a mixed state can persist even during temporarily decoupled conditions, provided there is no additional aerosol particle source at the ground or aloft.

Case II: Free troposphere decoupled from the ground

On 27 September 2021, balloon measurements were conducted at a median altitude of 1112 m—significantly higher than both the Zeppelin Observatory and the balloon in Case I. At this altitude, the balloon had ascended beyond the boundary layer and into the free troposphere. This observation is supported by the strong increase of potential temperature between 700 m (θ≈274 K) and 900 m (θ≈280 K) in the vertical profile (**Figure 3b**), indicating a significant temperature inversion. N_{150} concentrations were highest near the ground and remained stable within the lower 200 m. Above this layer, values gradually decreased up to around 700 m. From there, they increased slightly toward 1112 m, likely indicating influence

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from sources other than the ground. Specific humidity also showed substantial variability, fluctuating between 2.6 and 4 g kg^{-1}, further confirming the decoupled state of the sampled layer.

During active aerosol particle sampling at the ground, the average temperature was 3°C, with a relative humidity of 89% and a mean wind speed of 0.7 m s^{-1}, predominantly from the southwest (**Table S5**). In contrast, at balloon altitude, the air was slightly colder, with a mean temperature of -1.9°C, a relative humidity of 87%, and a significantly higher average wind speed of 5.5 m s^{-1}, primarily from the south and southwest (**Table S4**). On that day, IWV was high increasing from 13 kg m^{-2} to about 15 kg m^{-2} during the balloon sampling (**Figure S6b**). A dense layer of warm front clouds was present between 2 and 8 km, mainly consisting of cloud ice with IWP values up to 1.4 kg m^{-2} (**Figure S6b**). No precipitation reached the balloon or the ground up to the last 15–30 minutes of sampling, when snowfall reached the balloon and sampling was stopped.

On this date, we observed a strong vertical gradient in both Na^{+}_{aer} and $CCHO_{aer}$ concentrations (**Figure 3b**), starting from the winch (Na^{+}_{aer}: 1840 ng m^{-3}, $CCHO_{aer}$: 199 ng m^{-3}), decreasing at the Zeppelin Observatory (Na^{+}_{aer}: 850 ng m^{-3}), and dropping sharply at the balloon's altitude (Na^{+}_{aer}: 23 ng m^{-3}, $CCHO_{aer}$: 8.6 ng m^{-3}). Similarly, decreasing trends with altitude were observed for other constituents, including sulfate (580; 220; 76 ng m^{-3}), chloride (4230; 1500; 60 ng m^{-3}), and calcium (165; 32; 28 ng m^{-3}). This pronounced decline in major inorganic ions and $CCHO_{aer}$ concentrations with increasing altitude suggests a separation of ground-level air masses and those at higher elevations. It is unlikely that the substances detected at 1112 m originated primarily from fresh local sea spray emissions from Kongsfjorden or the west coast of Svalbard.

This assumption is supported by back-trajectory analysis (**Figure 4b**). While air masses arriving at 50 m and 474 m originated from the pack ice region in the central Arctic Ocean with a subsequent residence time over the ice-free Fram Strait, the air mass arriving at 1112 m followed a different pathway, passing over the Barents Sea near Franz Josef Land. 48 hours before sampling, this air mass had come into contact with the ground and the marine boundary layer in that region. However, after leaving the vicinity of Franz Josef Land, it remained predominantly at altitudes between 1000 and 1800 m. This suggests that SSA

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emissions contributing to the measured concentrations at 1112 m in Ny-Ålesund likely originated from that distant region.

In summary, Case II demonstrates that in the presence of an atmospheric inversion, major SSA constituents, including sodium, calcium, chloride, sulfate and CCHO, can be present at higher altitudes. However, in contrast to the mixed boundary layer (Case I), they occur at concentrations different from those at the ground and likely originate from more distant emission sources through long-range transport.

Case III: Inside and below a drizzling cloud

On 03 October 2021, the ground temperature was 3°C with a high relative humidity of 89%. Winds were light, shifting between east, south, and west at 0.7 m s^{-1} during sampling. At the balloon's altitude of 666 m, the average temperature was -1.3°C, the relative humidity 96% and the wind speed 6.8 m s^{-1} from the east and northeast. The day was overcast, with continuous drizzle from a 2 km deep mixed-phase cloud layer with LWP values of up to 300 g m^{-2} and IWV of around 13 to 14 kg m^{-2}. The balloon's altitude was close to the melting layer.

During the balloon's ascent and descent to the aerosol sampling height of 666 m, a positive gradient in potential temperature was observed (272 K at the ground vs. 278 K at the balloon, **Figure 3c**) indicating a stably stratified boundary layer. Similar to Case I, the specific humidity remained relatively uniform throughout the vertical column, with values between 3.2 and 3.8 g kg^{-1}. The vertical N_{150} distribution showed generally lower aerosol number concentrations compared to Case I, ranging between 3 and 10 cm^{-3}, and exhibited greater relative fluctuation. In summary, the boundary layer mixing conditions on 03 October 2021 (Case III) appeared similar to those in Case I. The key difference, however, was that in this case, aerosol sampling occurred partially inside or below a drizzling low-level cloud.

Back-trajectory analysis for ground, balloon, and Zeppelin Observatory (**Figure 4, Case III**) showed that air masses at all arrival heights followed the same path within the 48 hours before sampling, originating from the ice-free ocean south of Svalbard. As observed in Case I, trajectory heights indicate a vertical movement during this period, suggesting that air masses at all three levels shared a similar transport history and were influenced by the same emission sources.

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In line with the lower aerosol number concentrations, offline measurements of chemical constituents were also generally lower than in the previous cases. Furthermore, major inorganic ions (**Figure 3c**) were similar or slightly higher at the ground than at the balloon (chloride: 289 & 252 ng m^{-3}; sulfate: 66 & 59 ng m^{-3}; Na^+: 53 & 35 ng m^{-3}; K^+: 23 & 20 ng m^{-3}). At the Zeppelin Observatory, only Na^+ was measured above the detection limit, which was quantified at 38 ng m^{-3}—almost identical to the concentration measured at the balloon. This consistency indicates a rather mixed boundary layer. Creamean et al. (2021) analyzed three years of Arctic aerosol vertical distributions using a tethered balloon in Alaska and found that, when a uniform aerosol distribution below clouds was observed, it primarily occurred in autumn, aligning well with Case III.

Interestingly, despite the same levels of major inorganic ions, some chemical constituents exhibited increased concentrations at higher altitudes. These included major monosaccharides bound within CCHO (ground & balloon: Glc_{CCHO}: 12.6 & 34 ng m^{-3}; Xyl_{CCHO}: 0.57 & 4.4 ng m^{-3}; Ara_{CCHO}: below detection limit & 0.97 ng m^{-3}), as well as oxalate (5.5 & 24 ng m^{-3}), Ca^{2+} (47 & 187 ng m^{-3}), and Mg^{2+} (0.97 & 2.6 ng m^{-3}). The origin of these elevated concentrations remains unclear, as neither local sea spray emissions nor remote sources appeared suitable to account for the observed patterns.

While organics like $CCHO_{aer}$ and $oxalate_{aer}$ might originate through secondary in-situ atmospheric chemical or microbial processes —particularly in the aqueous phase, as discussed more in detail in the following section—this does not apply for inorganic elements such as calcium and magnesium. Instead, their ionic forms were possibly released from preexisting complex organic structures in SSA particles, becoming soluble and thus detectable by our analytical methods. OM-bound Ca^{2+}, as already found in Antarctic SSA (Su et al., 2023), may originate from SML-derived polysaccharide gels and airborne algal cells or fragments, which can release Ca^{2+} and Mg^{2+} through gel dispersion or cell dissolution under the acidic conditions of chemically aged SSA aerosol particles (Aller et al., 2017; Angle et al., 2021; Orellana and Leck, 2015; van Pinxteren et al., 2022; Trainic et al., 2018; Zhu et al., 2014).

In summary, Case III, demonstrates that certain organic SSA constituents can change within the vertical column due to atmospheric aging after primary emissions and vertical transport.

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Overall, the three cases presented highlight distinct meteorological scenarios that can lead to similar, lower, or even higher concentrations of chemical constituents at different altitudes. Interestingly, comparable patterns, with higher, lower, or similar levels at ground and balloon altitude, were also observed at the North Pole by Porter et al. (2022), who measured INP concentrations and combined these with trajectory analyses and heat sensitivity tests to conclude on their sources. While this effect-based approach provides valuable insights into aerosol particle properties, direct chemical analyses, as performed in this study, can further enhance certainty regarding the origin and composition of particles relevant for cloud droplet and cloud ice formation.

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3.3 Factors affecting SSA constituents beyond local sea-air transfer

Long-range transport and size-dependent deposition

SSA particles originate from both local and remote marine regions. However, our sampling methods make it challenging to determine the relative contribution of long-range transported SSA constituents, particularly when a local marine source, such as the Kongsfjorden is adjacent to the sampling site and may dominate other marine emissions.

As demonstrated in Case II, long-range transport of SSA can become dominant when air masses at elevated altitudes are decoupled from those at the ground. In this case, vertical and horizontal trajectory analysis suggests that the measured SSA constituents may have been emitted and incorporated into the atmosphere approximately 48 hours earlier over the Barents Sea, near Franz Josef Land. Atmospheric processes, such as the removal of larger supermicron particles, cloud activation, and precipitation, likely reduced their absolute mass concentration by one to two orders of magnitude before the arrival of the air masses in Ny-Ålesund.

In several balloon-borne TSP filter samples, however, an elevated CCHO/Na^+ ratio was observed, most notably on 24 September 2021; 03 October 2021 (Case III); 03 April 2022 (see **Figure 2d**). These values far exceeded both ground-based aerosol measurements from this study and previously reported values (Zeppenfeld et al., 2021, 2023), particularly for supermicron SSA particles that dominate the TSP mass. A slight increase of this ratio may be explained by a longer atmospheric residence time of these particles, leading to a relative reduction of supermicron aerosol particles through deposition (Croft et al., 2009; Hoppel et al., 2002), which are typically dominated by sea salt (O'Dowd and de Leeuw, 2007), while submicron aerosol particles–rich in surface-active CCHO–remain. This could lead to a shift of the CCHO/Na^+ ratios more similar to submicron than supermicron particles in the TSP samples of this study, as seen in Case II.

However, in the three cases with the most pronounced increase of the $CCHO_{aer}/Na^+_{aer}$ ratios in TSP at higher altitudes (24 September 2021; 03 October 2021; 03 April 2022), absolute $CCHO_{aer}$ concentrations were elevated as well (compare **Figures 2b and 2d**). Such increases of absolute concentrations cannot be explained by selective removal processes. This raises the question of whether the observed $CCHO_{aer}$ concentrations could result from the long-range

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transport of SSA compounds from a distant marine source with significantly higher CCHO levels in the seawater than the local Kongsfjorden.

Model simulations using FESOM2.1-REcoM3 (Gürses et al., 2023) (**Figure S7**) and field data (Assmy et al., 2023; Feltracco et al., 2021; Grosse et al., 2021; Wietz et al., 2024) confirm that the eastern Fram Strait as well as coastal Svalbard waters are productive and polysaccharide-rich regions. While the FESOM2.1-REcoM3 model does not resolve the SML separately, previous studies have shown significant CCHO enrichment in this layer (Compiano et al., 1993; Engel and Galgani, 2016; Gao et al., 2012; Zäncker et al., 2021), particularly in the productive marginal ice zone (Zeppenfeld et al., 2023). However, in the cases of high $CCHO_{aer}$ at elevated altitudes in the current study, air mass trajectories did not cross the marginal ice zone within 48 hours before reaching Svalbard (**Figure S8**). These findings suggest that long-range transport of SSA from more productive marine sources is unlikely to explain elevated $CCHO_{aer}$ concentrations at elevated altitudes within the lower troposphere, further supporting a rather local source or atmospheric in-situ formation.

In summary, while long-range transport of SSA constituents at elevated altitudes appears relevant in cases of decoupled atmospheric layers such as in Case II, it may not explain the significantly higher $CCHO_{aer}$ concentrations at high altitudes compared to ground levels. Instead, in-situ formation of $CCHO_{aer}$ could be a more plausible explanation for these observations.

Atmospheric in-situ formation of marine $CCHO_{aer}$

Bacteria can be transported into and persist in the Arctic atmosphere (Jensen et al., 2022; Šantl-Temkiv et al., 2018), with sources including terrestrial environments and surface seawater, particularly the SML (Aller et al., 2005). Our field study confirmed such dynamics by finding diverse marine and terrestrial bacteria in aerosol particles collected at the Old Pier (Wietz et al., to be submitted). Some aerosolized taxa, for instance *Polaribacter*, encode multiple genes for CCHO metabolism (Avcı et al., 2020) and consistently occur in the Kongsfjorden atmosphere during the spring bloom (Feltracco et al., 2021). These observations might underpin microbial CCHO transformations in the atmosphere, for instance the production of sticky, polysaccharide-based gels as protection against temperature fluctuations, salinity changes, and desiccation (Aller et al., 2005; Ramasamy et al., 2023; Šantl-

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Temkiv et al., 2022). Under highly humid conditions—especially in the presence of liquid water (such as in Case III)—airborne bacteria can become metabolically active (Ervens and Amato, 2020; Haddrell and Thomas, 2017). Atmospheric OM formation, including CCHO, through microbial activity has been documented for cloud water and aerosol particles (Bianco et al., 2019; Klein et al., 2016; Matulová et al., 2014). Consequently, metabolically active bacteria in the atmosphere could explain the increased $CCHO_{aer}$ concentrations observed within or near drizzling clouds in Case III of this study.

$CCHO_{aer}$ versus $oxalate_{aer}$: Co-production or atmospheric processing?

Since both combined glucose and combined xylose were consistently detected in $CCHO_{aer}$ of nearly all aerosol samples, we examined their correlation with other atmospheric chemical parameters. We observed a strong correlation between atmospheric xylose in $CCHO_{aer}$ and $oxalate_{aer}$ with an $R=0.78$ ($p<0.001$) across all sampling locations and heights (**Figure 5**). Oxalate, the ionic form of oxalic acid, is the most abundant dicarboxylic acid in aerosol particles (Kerminen et al., 1999; Rinaldi et al., 2011), with atmospheric concentrations in this study between <1 and 67 ng m^{-3}. The strong correlation raised the question of whether oxalic acid could be chemically linked to combined carbohydrates in aerosol particles.

$Oxalate_{aer}$ is known to originate from several primary sources and secondary formation pathways in both terrestrial and anthropogenic environments (Kawamura and Bikkina, 2016; Yang et al., 2022). In remote marine environments, the atmospheric formation of oxalic acid was proposed by Warneck (2003) through the aqueous-phase oxidation of glyoxal and glycolaldehyde, a process also investigated by field measurements inside and above marine clouds (Crahan et al., 2004; Sorooshian et al., 2007) and modeling (Herrmann et al., 2005; Tilgner and Herrmann, 2010). The possible aqueous-phase formation was supported by Case III of this study, where higher $oxalate_{aer}$ concentrations were observed within and in vicinity of clouds compared to ground level. In contrast, in the drier conditions of Cases I and II, $oxalate_{aer}$ levels remained vertically uniform. Additionally, since overall $oxalate_{aer}$ levels at the Old Pier (1.1–10.1 ng m^{-3}; mean=4.3±3.5 ng m^{-3}) were relatively low compared to the more inland Winch (<1–58 ng m^{-3}; mean=19.8±16.2 ng m^{-3}) and elevated altitudes samples (4.6–67 ng m^{-3}; mean=29.6±17.8 ng m^{-3}), direct primary oceanic emission was likely not its dominant source.

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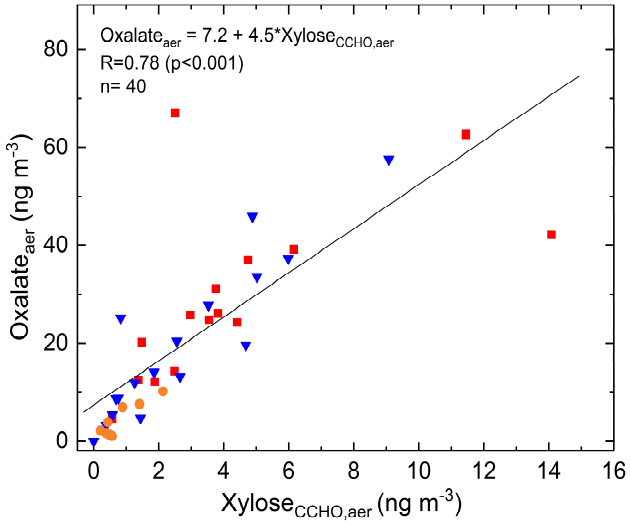

Figure 5. Atmospheric oxalate as a function of xylose in $CCHO_{aer}$ (R=0.78; p<0.001) measured in TSP from the Old Pier (orange circles), the winch site (blue triangles) and at elevated altitudes (red squares).

But what are the precursors of oxalic acid's precursors? While Warneck (2003) suggested that the anthropogenic volatile organic compounds acetylene and ethene can be transformed to atmospheric glyoxal, other studies suggest the photochemical degradation of marine OM (McNeill, 2015; Sinreich et al., 2010; Turekian et al., 2003; Zhou et al., 2014), with oligo- and polysaccharides representing a known subclass. Although not explicitly measured in this study, previous findings have shown that both $CCHO_{aer}$ (Leck et al., 2013; Zeppenfeld et al., 2021, 2023) and $oxalate_{aer}$ (Guo et al., 2016; Rinaldi et al., 2011; Turekian et al., 2003) are present across both the accumulation and coarse size modes. However, no consistently dominant size mode has been identified, which may support a common mechanism of formation or similar atmospheric processing pathways.

Here, based on known chemical reactions, we propose possible atmospheric pathways linking xylose-containing oligo-and polysaccharides as the precursors to oxalate as the final product (**Figure 6**). The initial depolymerization of CCHO presumably occurs either via enzymatic degradation, e.g. by glycoside hydrolases, or acid hydrolysis (Panagiotopoulos and Sempéré, 2005), both of which are plausible in the atmospheric context. Active microbial enzymes have been detected in SSA, often exhibiting activities 1–2 orders of magnitude higher than in bulk seawater (Malfatti et al., 2019). Additionally, SSA particles are known for reaching very low pH

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levels within minutes after their emissions due to the uptake and reactions with acidic gases, as well as water loss (Angle et al., 2022, 2021). Furthermore, although not explicitly investigated in an atmospheric context, Zhu et al. (2023) observed rapid depolymerization of xylose-containing oligosaccharides into the monosaccharide xylose within minutes in a UV/H_2O_2 system, generating hydroxyl radicals.

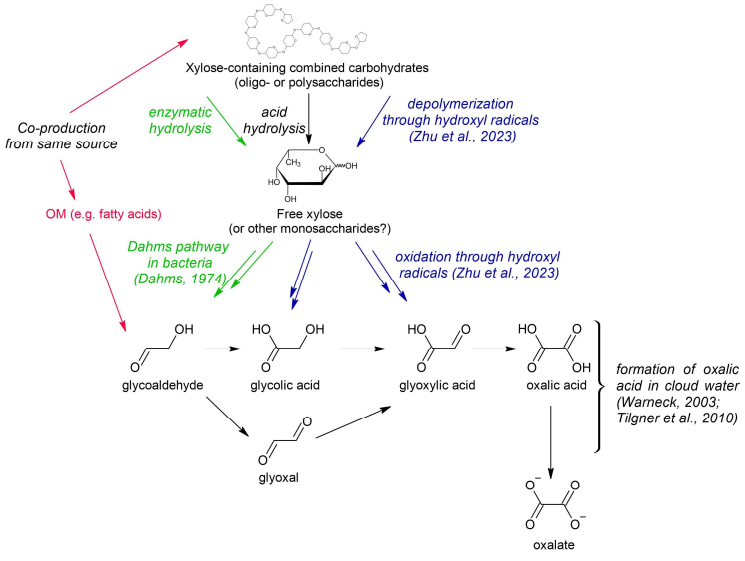

Figure 6. Possible pathways for the formation of atmospheric oxalate from xylose in combined carbohydrates in marine aerosol particles.

Free xylose was—with one exception—never detected in any aerosol sample of this study, suggesting that it is rapidly processed in the atmosphere. Two potential pathways may link monomeric xylose to precursors of Warneck's oxalate formation: (1) a follow-up reaction with hydroxyl radicals, where the pyranose ring of xylose is cleaved after the more susceptible glycosidic bonds have been readily broken. Zhu et al. (2023) observed glycolic acid and glyoxylic acid among other products following the UV/H_2O_2 treatment of xylooligosaccharides. (2) Bacterial metabolism via the Dahms pathway converting free xylose into pyruvate and glycolaldehyde (Dahms, 1974). However, only few bacteria encode this pathway; and it is highly questionable whether these occur in sufficient atmospheric concentrations for a measurable effect.

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One indication that direct formation from xylose-containing oligo- and polysaccharides cannot be the sole source of atmospheric oxalate in the marine environment is the discrepancy in concentrations: atmospheric oxalate levels were seven times higher than those of combined xylose. This confirms the involvement of additional precursors or a co-production/co-emission of combined xylose with gaseous precursors, such as isoprene (Carlton et al., 2009; Kawamura and Bikkina, 2016), or other primary marine organic matter, such as phytoplankton-derived fatty acids (Kawamura et al., 1996b, a) undergoing photo-oxidation. Further targeted laboratory and modeling studies are needed for clarity.

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4. Summary and Atmospheric Implications

In autumn 2021 and spring 2022, we performed balloon-borne measurements of major SSA constituents at Ny-Ålesund (Svalbard). Our evidence demonstrated that both sodium and marine CCHO reach elevated altitudes within the boundary layer, and even the free troposphere as part of aerosol particles. The relationship between ground-level and high-altitude measurements was strongly influenced by meteorological conditions and the mixing state of the lower atmosphere, as discussed in three representative cases. Long-range transport of Na^{+}_{aer} and $CCHO_{aer}$ from remote marine sources is presumably relevant for high-altitude measurements, especially when the upper air masses were decoupled from the ground. However, in cases of a well-mixed lower atmosphere, the local marine source (here, the Kongsfjorden) was the dominant contributor for atmospheric Na^{+}_{aer} and $CCHO_{aer}$. Under very humid conditions particularly in the presence of liquid precipitating clouds, in-situ formation of $CCHO_{aer}$ was observed, possibly linked to microbial metabolism. To establish more generalizable patterns, we recommend further field studies using airborne platforms.

The significant correlation between combined xylose within $CCHO_{aer}$, and $oxalate_{aer}$ suggests underlying pathways for oxalic acid formation from combined xylose and other monosaccharide units within $CCHO_{aer}$; alternatively, a co-production of xylose-containing oligo- and polysaccharides alongside oxalate precursors.

CCNs and INPs are key drivers in cloud formation, influencing radiative and precipitation properties and, consequently, climate processes. Considerable uncertainties remain regarding the origin and chemical composition of these particles, particularly in remote Arctic regions, which affects the accuracy of climate models. Since marine polysaccharides have been identified as relevant ice-nucleating molecules in the remote marine atmosphere (Hartmann et al., 2025), our findings have implications for cloud microphysics, especially given that these carbohydrates are transported to altitudes relevant for cloud formation. Furthermore, atmospheric processing, as observed here, may alter the ice-nucleating properties of these macromolecules, potentially creating new INPs in-situ or deactivating existing ones.

As the Arctic continues to change, expanding ice-free ocean areas will serve as emission sources for SSA particles, influencing cloud properties, and finally the radiative budget.

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Consequently, our findings contribute to an improved understanding of the complex interplay of environmental processes resulting in Arctic amplification (Wendisch et al., 2017, 2023).

Author contributions

SZ wrote the manuscript with input from all co-authors. SZ, JS, CP, HS, BW, MW, and MvP collected field samples in Ny-Ålesund. HS and BW served as principal investigators for balloon operations during the field campaign. SZ conducted the laboratory carbohydrate analyses and data processing. MZ and AB carried out the FESOM2.1-REcoM3 simulations. KE assessed cloud conditions for the case studies using remote sensing data. All co-authors reviewed and commented on the manuscript.

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Competing interests

All authors declare no financial or non-financial competing interests. Some authors are members of the editorial board of ACP.

Data availability

Chemical data from offline TSP filters are publicly available in PANGAEA for seawater (Zeppenfeld and Schmidt, 2025) and aerosol particles (Zeppenfeld et al., 2025). The microwave radiometer LWP and IWV data are available in PANGAEA (Ebell and Ritter, 2022). The Cloudnet classification and ice water content products (Ebell et al., 2025) can be downloaded via the ACTRIS Cloudnet data portal (https://cloudnet.fmi.fi).

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