Marine Carbohydrates at High Altitudes of the Arctic Atmosphere

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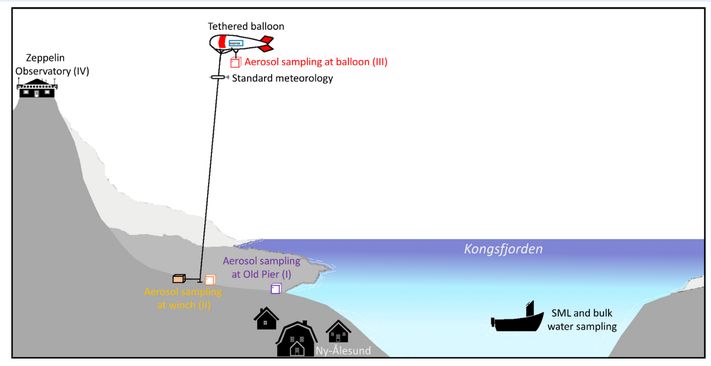
Scientific background

Do sea spray aerosol compounds reach the altitudes relevant for cloud formation?

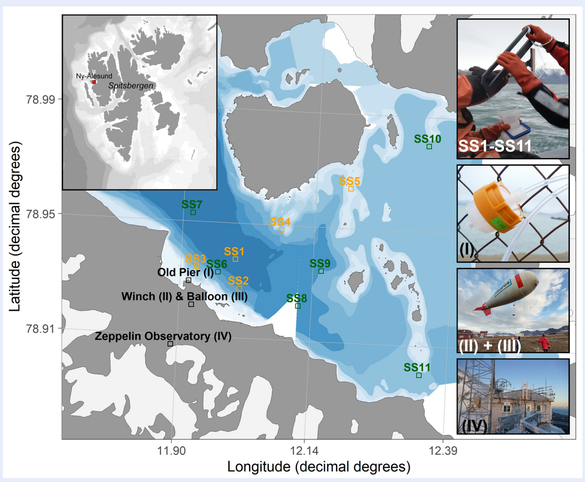
The Arctic atmosphere is a complex system shaped by interactions between oceanic and atmospheric processes, which are particularly impactful for regional climate.

Sea spray aerosol (SSA) particles, generated by wind-driven waves, introduce a mixture of salts and organic materials from the ocean surface into the atmosphere. These aerosol particles often contain marine carbohydrates (CHO), which have been shown to have the potential to influence cloud formation by acting as cloud condensation nuclei and ice nucleating particles (e.g. Orellana et al., 2011; Galgani et al. 2016, Hartmann et al. under review). The sea surface microlayer (SML), enriched with organic materials, is a significant source of these carbohydrates, transferring them into SSA upon wave action. While SSA is transported upward through the atmosphere, few studies have confirmed that its chemical components reach higher altitudes where they can actually influence cloud formation and properties (Köllner et al., 2021).

Detailed measurements are limited, leaving a gap in understanding the full extent of SSA's impact on cloud microphysics. In this study, we used a tethered balloon system to analyze SSA up to 1200 m altitude, providing new insights into how ocean-derived compounds (CHO, sea salts) interact with Arctic atmospheric processes and cloud formation.



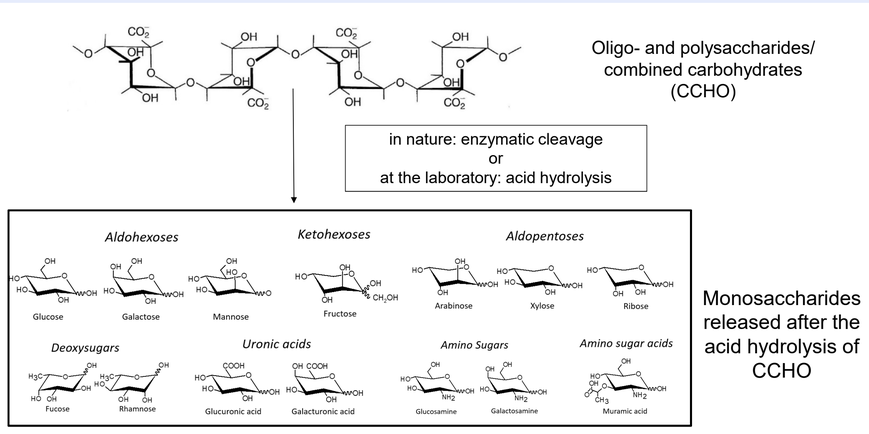
Sampling in Ny-Alesund, Svalbard, Arctic



BELUGA Campaign in Ny-Ålesund (Autumn 2021 & Spring 2022):

- **Surface Microlayer (SML) and Bulk Water Sampling:** Conducted in the Kongsfjorden (Stations SS1-SS11)
- **Continuous Aerosol Sampling:** Performed directly at the shore of the Kongsfjorden next to the Old Pier (I)
- **Aerosol Sampling at Various Altitudes:** Conducted using a tethered balloon (III) with ground-level measurements taken simultaneously at the winch (II)
- **Comparative Aerosol Measurements:** Benchmarked against routine data from the Zeppelin Observatory (IV)

Chemical structure of marine carbohydrates (CHO)

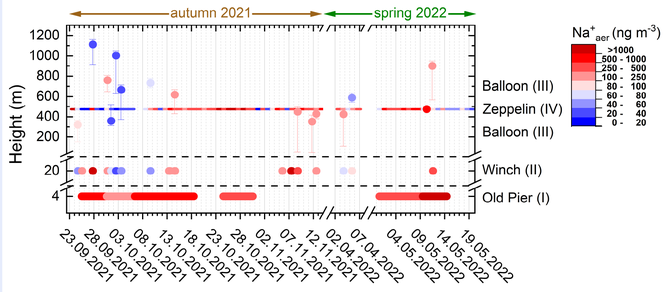


Marine carbohydrates (CHO) exist as either Dissolved Free Carbohydrates (dFCHO) or Combined Carbohydrates (CCHO).

Through acid hydrolysis, CCHO are broken down into individual monosaccharides, enabling further analysis via High-Performance Anion Exchange Chromatography coupled with Pulsed Amperometric Detection (HPAEC-PAD).

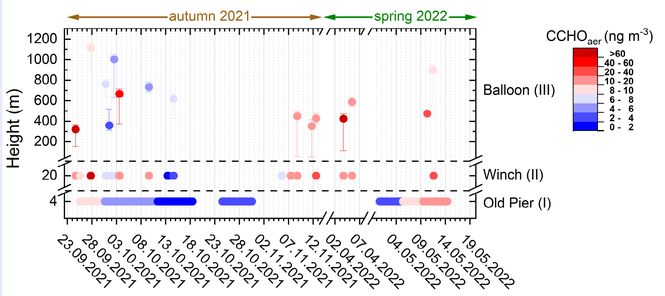
SSA particles at high altitude and ground

Sodium ions (Na^+):



- **Na^+ as Tracer:** Serves as conservative tracer for Sea Spray Aerosol (SSA) emissions
- **Concentration Levels:** Highest concentrations observed at the Old Pier at the Kongsfjorden (emission source)
- **Altitude Variation:** Lower and variable concentrations at the winch and at higher altitudes
- **Data Consistency:** Strong alignment between Zeppelin Observatory monitoring and balloon measurements at comparable altitudes

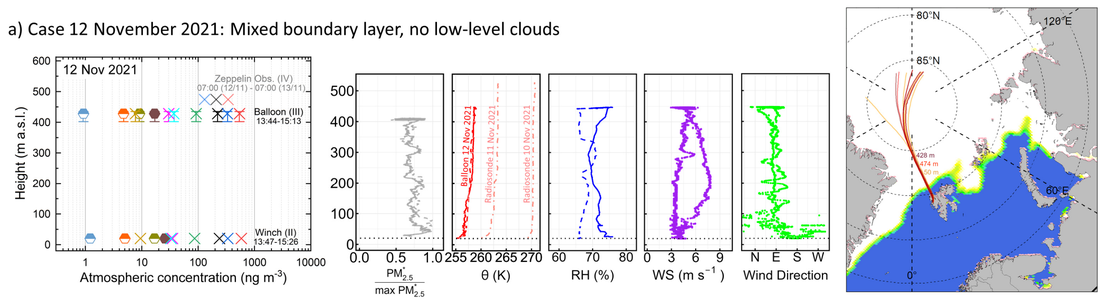
Combined Carbohydrates (CCHO):



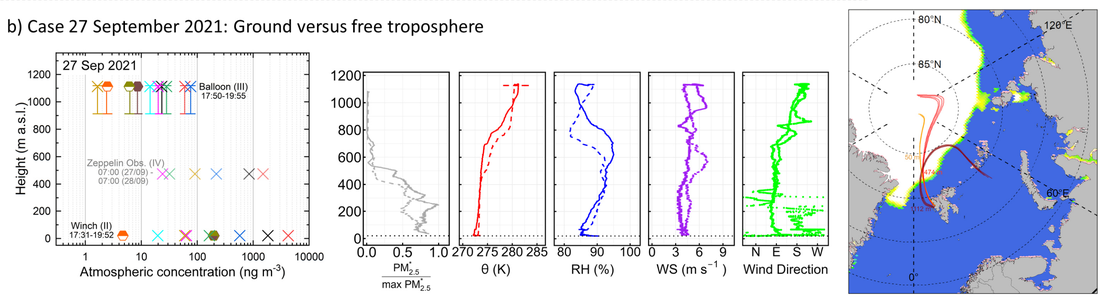
- **Seasonal Variation at the Old Pier:** Lower concentrations observed during the dark season and higher concentrations during the light seasons
- **CCHO Presence Across Altitudes:** CCHO detected at all altitudes, sometimes even in higher concentrations than at ground level (!)

Three different atmospheric cases

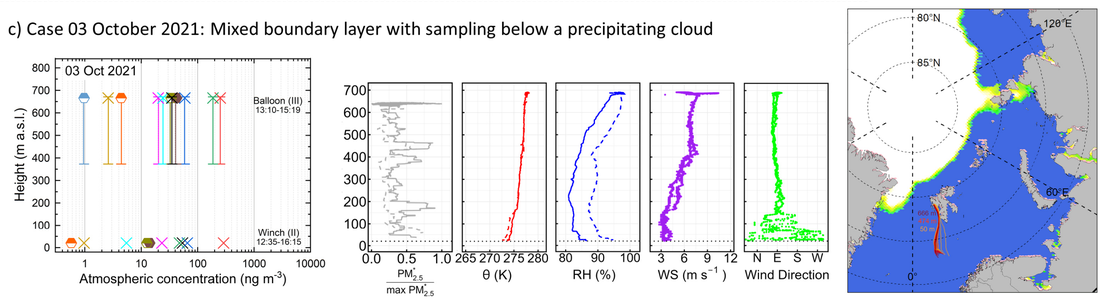
a) Case 12 November 2021: Mixed boundary layer, no low-level clouds



b) Case 27 September 2021: Ground versus free troposphere

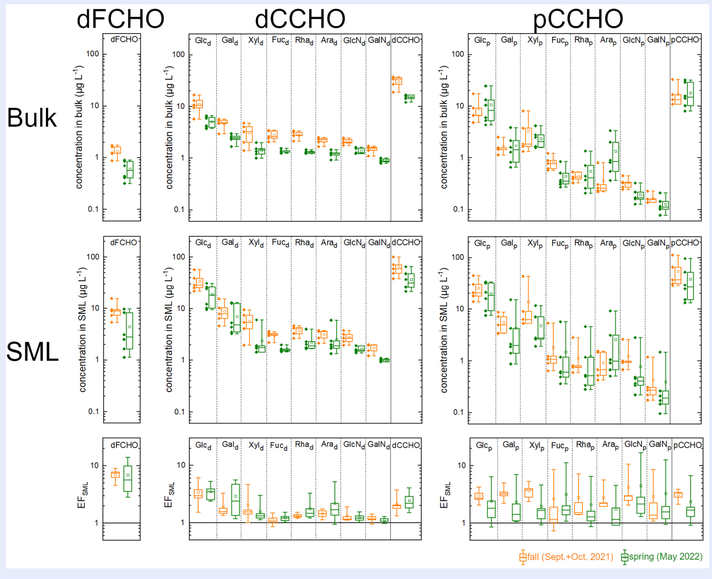


c) Case 03 October 2021: Mixed boundary layer with sampling below a precipitating cloud



Comparison of Atmospheric Cases: Three distinct atmospheric cases examined major inorganic ion and CCHO concentrations based on offline analysis at ground level (winch) and at various balloon altitudes. These chemical concentrations were compared with meteorological profiles (potential temperature, relative humidity, wind speed, wind direction) from standard meteorological sensors on the balloon, as well as aerosol profiles from the Cubic Aerosol Measurement Platform (CAMP, Pilz et al., 2022). Additionally, HYSPLIT back-trajectories (Stein et al., 2015) were calculated at various arrival heights and presented alongside daily prevailing sea ice concentrations (SIC).
Case I: In a mixed boundary layer, similar concentrations of ions and CCHO were observed at ground level and at higher altitudes.
Case II: Measurements in the free troposphere, indicated by a strong inversion layer, showed significantly lower concentrations of major ions and CCHO. However, all constituents were still detectable. HYSPLIT back-trajectory analysis indicated distinct air mass histories at different arrival heights.
Case III: In a mixed boundary layer below a precipitating cloud, inorganic ion concentrations remained consistent, while CCHO concentrations were higher near the cloud at higher altitudes, suggesting possible in-situ formation in the presence of a cloud.

CHO in the surface water of the Kongsfjorden



- **CCHO in seawater (SML and Bulk):** Differentiation between dissolved CCHO (dCCHO, <0.2 µm) and particulate CCHO (pCCHO, >0.2 µm)
- **dCCHO:** Shows low variability within seasons but strong seasonality, with mid-autumn levels generally higher than mid-spring. This pattern suggests that dCCHO results from a series of mid-term production and degradation processes
- **pCCHO:** Highly variable, reflecting recent production, intermediate reactions to environmental stress, or photosynthesis, and masks seasonality
- **Mechanistic Link to Aerosols:** The connection between CCHO in seawater and CCHO in aerosol particles is still under investigation

References and Acknowledgments

Galgani et al. *Scientific Reports* (2016), Hartmann et al. under revision for ES&T, Köllner et al. *ACP* (2021), Orellana et al. *PNAS* (2011), Pilz et al. *AMT* (2022), Stein et al. *B. Am. Meteorol. Soc.* (2015)

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Summary

→ Major sea spray aerosol (SSA) constituents, including sodium, chloride, and CCHO, are present in aerosol particles at all altitudes in the Arctic
→ Their concentrations vary based on meteorological conditions, such as mixing state, air mass history, and cloud presence.
→ CCHO exhibits clear seasonal patterns in both seawater and aerosol particles
→ CCHO can also form next to clouds

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