

Answers to the Referees' comments regarding the manuscript:

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

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Preprint egusphere-2025-4336

We thank the reviewers for their positive evaluation of our revised manuscript and for their detailed and constructive comments. In the following document, all remarks have been addressed carefully. [The referees' comments are marked blue](#), our replies black, and [changed text in the manuscript green](#). The given line numbers of changed sentences are referring to the new lines in the revised manuscript.

Reviewer 1 (Report #1):

General comments:

The manuscript increased in clarity and profits from its condensed form. Significance and gain for the scientific community are better outlined, while the discussion is appropriately balanced. Some details can be still further adjusted.

Authors: We thank the reviewer for this positive reassessment of our revised manuscript. We greatly appreciate the recognition that the revised version has improved in clarity, conciseness, and that the discussion is now better balanced. All remaining comments and suggested adjustments have been carefully considered and addressed below.

L111 'have been unavailable' or rather not been used for this purpose until to date?

Authors: Our intention was to emphasize that such vertically resolved datasets for marine CCHO_{aer} have not previously existed, mainly because their generation requires the combination of highly specific analytical techniques and demanding sampling methodology. To clarify this point, we revised the wording accordingly: 'Vertically resolved field data comparing ground-level and elevated altitudes using mobile platforms for marine CCHO_{aer} have, however, been lacking to date.' (Lines 109-111)

L114 First two reasons are in their current state somehow redundant: offline measurements (upconcentrated sample) reach their detection limit, no online-techniques (highly diluted sample) available. Maybe better combine to one argument?

Authors: We agree that, in the previous version, the first two arguments could appear redundant when viewed solely from the perspective of concentration and detection limits. However, our intention was to distinguish between two separate methodological limitations: a) the current absence of suitable high-resolution online techniques for CCHO_{aer} (regardless of concentration), which reflects the inherent analytical difficulty of detecting and characterizing this compound class, and b) the additional challenge that offline approaches are further constrained by the very low atmospheric concentrations encountered in the field. To make this distinction clearer, we have revised the text accordingly. It now reads: 'This is due to several methodological challenges, most fundamentally the absence of suitable high-resolution online detection techniques for CCHO_{aer}, reflecting the inherent analytical difficulty of this compound class. As a consequence, current approaches rely on offline analyses, which are further constrained by low atmospheric concentrations that approach their detection limits. In addition, lightweight yet powerful high-flow pumps remain technically challenging to realize for mobile airborne platforms (e.g., drones or balloon-based systems), where payload and power constraints limit the collection of sufficient aerosol mass during short sampling periods.' (Lines 111-118)

L328 Modelling of acidic polysaccharides mentioned here, however, only results for polysaccharide-rich regions reported (L707). Only a very small fraction of marine polysaccharides is acidic, while neutral sugars prevail and are discussed in this study. Clarify.

Authors: In this study, we used dissolved acidic polysaccharides as a proxy for dCCHO. We agree that only a fraction of marine polysaccharides is explicitly acidic. However, while neutral monosaccharides often dominate the dCCHO pool after hydrolysis compared to acidic uronic acids, acidic properties related to sulfate and phosphate side groups are not resolved by standard dCCHO measurements. This suggests that the actual contribution of acidic polysaccharides may be substantially higher than inferred from dCCHO analyses alone. As shown by Zeising et al. (2026), although the modelled dissolved acidic polysaccharides represent only part of the total dCCHO pool, their concentrations are within the same order of magnitude as field observations. We have added this clarification to the main text. The added sentences read 'The modelled dissolved acidic polysaccharides were used as a proxy for dCCHO. Although they represent only

a fraction of dCCHO, their concentrations were shown to be within the same order of magnitude as field observations (Zeising et al., 2026).’ (Lines 336-339)

L424 I am not sure if dynamics are similar as you state at the Old Pier CCHO_{aer} were highest at the beginning and end (Sept and May) while dCCHO peaked in late Sept while being much lower in May. Clarify what 44-67% refers to...

Authors: We agree that the originally implied similarity between CCHO_{aer} and seawater dCCHO dynamics was too strong, as both datasets exhibit clear seasonal variability but do not covary directly, particularly due to differences in spring (May) conditions. We have therefore revised and restructured the section to more clearly distinguish the seasonal behaviour of seawater dCCHO, freshly emitted CCHO_{aer}, and their partial linkage.

The previously reported range of 44–67% is no longer used in the revised manuscript. Instead, the seasonal contrast in dCCHO as a whole is now described more consistently, with spring (May) concentrations being on average about 50% of the corresponding autumn (September/October) values.

Overall, the revised text now more clearly reflects that both seawater and aerosol CCHO exhibit seasonality, but that their temporal dynamics are only partially aligned and cannot be directly interpreted as covarying in detail.

The changed section now reads: ‘A seasonal trend was also observed for dCCHO in Kongsfjorden seawater. Concentrations peaked in late September/early October and were substantially lower in early to mid-May, averaging only about 50% of the autumn values (**Figure 4**). Most monosaccharides in bulk dCCHO showed a strong co-variation with SST ($R^2 = 0.79-0.93$; $n = 11$). This relationship was weaker for xylose ($R^2_{\text{XYL-SST}} = 0.62$) and glucose ($R^2_{\text{GLC-SST}} = 0.53$) in bulk dCCHO, and generally more moderate in the SML for most monosaccharides (**Figure 4**).

In contrast, pCCHO as a whole showed no clear seasonal trend in seawater (**Figure S3**). However, a few monosaccharides within bulk water pCCHO, in particular fucose, glucosamine, and galactosamine, displayed a moderate correlation with SST ($R^2_{\text{FUC-SST}} = 0.73$; $R^2_{\text{GALN-SST}} = 0.39$; $R^2_{\text{GLCN-SST}} = 0.69$; $n=11$). While dCCHO in bulk water exhibited relatively low spatial and intra-seasonal variability, pCCHO and SML samples were considerably more variable, even among samples from the same season (**Figure S4**). This likely reflects the rapid dynamics of pCCHO in relation to phytoplankton blooms (Becker et al., 2020; Engel et al., 2012; Fabiano et al., 1993), as well as the formation of transparent exopolymer particles (TEP) from dCCHO in turbulent waters and vertical transport of pCCHO via sedimentation (e.g., as marine snow) or its accumulation in the SML depending on buoyancy (Burns et al., 2019; Engel, 2004; Robinson et al., 2019a, b; Wurl and Holmes, 2008). The SML, in particular, may be more sensitive to these dynamics than the bulk water, potentially explaining its greater fluctuations.

By comparison, dCCHO in bulk water, like dissolved organic carbon (Hansell, 2013; Keene et al., 2017), is likely dominated by recalcitrant and semi-recalcitrant compounds, while the labile fraction is rapidly consumed by heterotrophic bacteria (Goldberg et al., 2011). Notably, combined glucose showed high variability in both dCCHO and pCCHO, likely due to being the main constituent of abundant storage macromolecules such as laminarin (Becker et al., 2020), whose production and turnover may be enhanced during periods of photosynthetic overflow (Barthelmeß et al., 2025), as well as its relatively rapid microbial utilization (Kharbush et al., 2020).

The seasonal variation of CCHO_{aer} at the Old Pier may be linked to the seasonal dynamics of marine CCHO in the surface waters of Kongsfjorden, the only local emission source of SSA. These dynamics are likely driven by seasonal shifts in primary production and phytoplankton composition (Assmy et al., 2023; Mayot et al., 2018, van de Poll et al., 2021).

Overall, CCHO_{aer} and selected seawater dCCHO monosaccharides showed a broadly consistent seasonal tendency, with elevated values in late summer/early autumn. However, the spring conditions (May) deviate from this pattern, with CCHO_{aer} showing a secondary maximum that is not reflected in seawater

dCCHO, where concentrations remain substantially lower. This indicates that the seasonal coupling between (bulk) seawater CCHO and CCHO_{aer} is strongest in late summer/early autumn, while additional processes such as SML enrichment or atmospheric processing may also contribute to the spring aerosol signal.

In conclusion, the seasonal variation of CCHO_{aer} measured at the Old Pier is partly consistent with marine carbohydrates in Kongsfjorden seawater, suggesting that surface CCHO is an important but not exclusive source of freshly emitted CCHO_{aer}.’ (Lines 427-476)

L453 incomplete sentence: ...likely due to it being...?

Authors: We corrected the sentence by adding the missing “to”.

L454 Reformulate, might be misleading to some. Photosynthetic overflow dynamics could contribute to the variability observed in dissolved glucose, but glucose serves as storage molecule in cells no matter whether photosynthetic overflow occurs...

Authors: We agree that our previous wording could be interpreted as implying that combined glucose serves as a storage molecule only during periods of photosynthetic overflow. This was not our intention. Rather, glucose is a general constituent of cellular storage compounds such as laminarin, while photosynthetic overflow may further enhance their production and contribute to the observed variability. We have revised the sentence accordingly. It now reads: “Notably, combined glucose showed high variability in both dCCHO and pCCHO, likely due to being the main constituent of abundant storage macromolecules such as laminarin (Becker et al., 2020), whose production and turnover may be enhanced during periods of photosynthetic overflow (Barthelmeß et al., 2025), as well as its relatively rapid microbial utilization (Kharbush et al., 2020).” (Lines 457-462)

L457 in the whole chapter, only Old Pier and seawater samples are discussed, correct? I would suggest to state this in the concluding sentence once again.

Authors: The reviewer is correct, and we agree that this should be clarified in the concluding sentence. We have therefore revised the sentence as follows: ‘In conclusion, the seasonal variation of CCHO_{aer} measured at the Old Pier is partly consistent with marine carbohydrates in Kongsfjorden seawater, suggesting that surface CCHO is an important but not exclusive source of freshly emitted CCHO_{aer}.’ (Lines 474-476)

L462 Not sure if this is the only implication... the Old Pier could be sheltered, which would lead to less wind, limited fetch, reduced wave breaking and as a consequence to lower SSA formation than at open ocean sides. Consider including this aspect into your discussion.

Authors: We understand the reviewer’s concern regarding potential sheltering effects at the Old Pier. However, the Old Pier sampling site is located in an open coastal setting and is directly exposed to marine conditions, making strong sheltering effects such as reduced wind, limited fetch, or reduced wave breaking unlikely. This is further supported by the consistently high Na⁺ concentrations observed at this site, which indicate frequent influence of sea spray aerosol. In contrast, the balloon and winch measurements were conducted further inland and are therefore more affected by terrestrial shielding. We thus consider it unlikely that limited fetch or reduced wave breaking at the Old Pier is a major driver of the observed variability in CCHO_{aer}.

L511 'PASCAL', missing reference.

Authors: We have clarified the reference to the PASCAL cruise by adding the appropriate citations (Macke and Flores, 2018; Wendisch et al., 2018).

L519 Reference missing or is this referring to the following and not yet introduced results?

Authors: We clarify that the sentence was intended as a transition to the following section and does not refer to missing references or additional results. The revised text now reads: 'Overall, it can be concluded that both Na_{aer}^+ and CCHO_{aer} are transported from the marine emission source to elevated heights within the lower troposphere. In the following section, we discuss the role of meteorological conditions and atmospheric mixing in linking ground-based and balloon-based samples.' (Lines 536-539)

L688 You are discussing three samples with high CCHO_{aer} concentration originating from high altitudes, potentially indicating atmospheric transformation, processing, secondary production pathways. Two of these samples belong to Case III in which you also highlight their trajectories. Only roughly within the last eight hours, these airmasses seem to have separated from mid and low altitude samples. I would consider this a rather short time frame in terms of atmospheric metabolic/ enzymatic and chemical alterations and at temperatures close to zero degree... Maybe you could include relevant examples and/or potentially previously determined rates for atmospheric aging etc?

Authors: We agree with the reviewer that an 8-hour timescale appears short for substantial metabolic or chemical transformation, particularly under temperatures close to 0°C. However, quantitative approximations on atmospheric carbohydrate transformation rates remain scarce. Zeppenfeld et al. (2021) performed some estimations based on typical bacterial abundances in remote marine and ice-covered regions and biodegradation rates of the cloud-borne bacterium *Bacillus* sp. 3B6 (Matulová et al., 2014), which currently represents one of the few laboratory-based references for microbial carbohydrate processing in atmospheric environments. Their results suggest that biologically driven carbohydrate transformation in sea spray aerosol may occur on timescales ranging from ~20 minutes to several hundred hours, depending on parameter choice. Despite considerable uncertainty, these estimates indicate that measurable transformation within an 8-hour transport period is plausible. We have added this aspect to the main text: 'Although substantial uncertainties remain, Zeppenfeld et al. (2021) estimated, depending on parameter choice, residence times between 20 minutes and several hundred hours during which measurable microbial transformation of carbohydrates in the atmosphere may occur.' (Lines 754-757)

Reviewer 4 (Report #2):

Overall impression

This research provides a comprehensive overview of vertical profile of marine carbohydrates including SSA tracers in aerosol particles and relate it well with the seawater concentration (both bulk surface and SML). I particularly liked the investigation of meteorological conditions and atmospheric mixing state to explain the observed vertical profiles for the three case studies. I recommend acceptance of this manuscript subject to some minor revisions as outlined below.

Authors: We sincerely thank the reviewer for the positive and encouraging assessment of our manuscript. We are grateful for the recommendation for acceptance and have carefully addressed all minor revisions raised below.

Minor Comments

Line 71: I recommend dropping the abbreviation – DFCHO – as it hasn't been used enough.

Authors: We removed the abbreviation DFCHO and rephrased the corresponding sentences to improve clarity and maintain consistency within the section. The revised text now reads: 'They also exist as monosaccharides, known as dissolved free carbohydrates. Both fractions are consumed or transformed by heterotrophic organisms, with turnover rates largely determined by their molecular structure and composition (Arnosti et al., 2021; Engel and Händel, 2011; Ittekkot et al., 1981; Kirchman et al., 2001).'

(Lines 70-74) and 'Dissolved free carbohydrates are measured without hydrolysis, whereas CCHO include those monosaccharides released by acid hydrolysis (0.8 M HCl, 100°C, 20 h).'

(Lines 257-259)

Lines 111-116: I understand that the detection limit and resolving power may explain the lower combined carbohydrate measurements. However, how do the limitations of lightweight, powerful pumps present a challenge? Are you specifically referring to vertical profiles that are measured using a sampler loaded onto a balloon or a drone? If so, I recommend clarifying this point to ensure that all readers, especially those who are not directly measuring vertical gradients, understand your intention.

Authors: We thank the reviewer for this helpful comment and agree that the relevance of lightweight, high-performance pumps was not sufficiently clear in the previous version. Our intention was to refer specifically to vertically resolved measurements using mobile airborne platforms (e.g., drones or balloon-based systems), where payload and power constraints strongly limit the use of conventional high-flow sampling systems. At the same time, sufficiently high flow rates are required to collect enough aerosol mass during the short sampling periods typically available on such platforms. To make this point clearer for all readers, we have revised the sentence accordingly. It now reads: "In addition, lightweight yet powerful high-flow pumps remain technically challenging to realize for mobile airborne platforms (e.g., drones or balloon-based systems), where payload and power constraints limit the collection of sufficient aerosol mass during short sampling periods." (Lines 115-118)

Line 139: Please rename Section 2, as the word "Experimental" is an adjective and cannot stand alone. I suggest using "Methods" instead. Additionally, it would be helpful to include two sentences explaining what this section includes. It feels a bit abrupt to have a main heading followed immediately by a table and then sub-sections.

Authors: The section heading has been changed from "Experimental" to "Methods". In addition, we have added a brief introductory paragraph to guide the reader through the structure of the section and to avoid an abrupt transition from the heading to Table 1 and the subsequent subsections. The revised text now reads: "This section summarizes the observational and modelling approaches used in this work. It covers the study area, field sampling, offline and online measurements, supporting datasets, model calculations,

and statistical and visualization methods for data interpretation. An overview of all relevant parameters and methods is provided in Table 1.” (Lines 141-144)

Lines 163-165: Consider rephrasing this sentence to below to be grammatically correct. ‘Furthermore, boundary layer mixing can occur even when a positive gradient in potential temperature exists, suggesting more stable stratification.’

Authors: Thanks for the suggestion. We changed it accordingly.

Line 233: What does gently thawed mean here? Did you use a controlled warming rate? Or the samples were thawed at room temperature?

Authors: We clarified the sentence to specify that samples were thawed at 4 °C in a refrigerator. The revised sentence now reads: ‘Frozen seawater samples were thawed at 4°C in a refrigerator one day before analysis.’ (Line 236)

Figure 3: Explain the meaning of all marker types. For example, what do hexagonal and pentagonal markers mean here? and what does half shading of markers mean? A figure should be fully understandable on its own

Authors: We agree that the use of different marker shapes and shading may have been unclear without explicit explanation. In fact, the different marker types (e.g., hexagonal, pentagonal, full or half-shaded symbols) do not carry specific meaning. They are used solely as visual identifiers to link sodium and corresponding CCHO values originating from the same sample and sampling date within this plot. To avoid any potential misunderstanding, we have clarified this in the caption of Figure 3. The caption now includes the following statement: “Marker shapes and fill styles serve as identifiers linking corresponding Na^+_{aer} and CCHO_{aer} values from the same sample.”

Line 414: I would recommend changing the subheading to ‘Combined carbohydrates in fresh SSA and seawater’ to keep it coherent with the rest of text (eg line 425) and make it easier to follow for the reader.

Authors: We agree. We changed the subheading to ‘Combined carbohydrates in fresh SSA and surface seawater’.

Line 424: I would recommend adding correlation coefficient here to quantify the similarity in seasonality of monosaccharides and SST as well as monosaccharides (dissolved) and CCHO_{aer} to further support the conclusion of this section, as stated in lines 457-458

Authors: We have added correlation coefficients to the revised text to better quantify the seasonal relationship between seawater monosaccharides and SST. The revised text now reads: “A seasonal trend was also observed for dCCHO in Kongsfjorden seawater. Concentrations peaked in late September/early October and were substantially lower in early to mid-May, averaging only about 50% of the autumn values (Figure 4). Most monosaccharides in bulk dCCHO showed a strong co-variation with SST ($R^2 = 0.79-0.93$; $n = 11$). This relationship was weaker for xylose ($R^2_{\text{Xyl-SST}} = 0.62$) and glucose ($R^2_{\text{Glc-SST}} = 0.53$) in bulk dCCHO, and generally more moderate in the SML for most monosaccharides (Figure 4).

In contrast, pCCHO as a whole showed no clear seasonal trend in seawater (Figure S3). However, a few monosaccharides within bulk water pCCHO, in particular fucose, glucosamine, and galactosamine, displayed a moderate correlation with SST ($R^2_{\text{Fuc-SST}} = 0.73$; $R^2_{\text{GalN-SST}} = 0.39$; $R^2_{\text{GlcN-SST}} = 0.69$; $n=11$)....” (lines 427-436)

Regarding the comparison between dissolved monosaccharides and CCHO_{aer} , we have moderated our original conclusion following Reviewer 1’s comment and restructured the section to better acknowledge the differing seasonal behaviour of the individual components.

Line 435: Please add full form for TEP.

Authors: We introduced the full term for TEP and slightly rephrased the sentence, which now reads: ‘This likely reflects the rapid dynamics of pCCHO in relation to phytoplankton blooms (Becker et al., 2020; Engel et al., 2012; Fabiano et al., 1993), as well as the formation of transparent exopolymer particles (TEP) from dCCHO in turbulent waters and vertical transport of pCCHO via sedimentation (e.g., as marine snow) or its accumulation in the SML depending on buoyancy (Burns et al., 2019; Engel, 2004; Robinson et al., 2019a, b; Wurl and Holmes, 2008).’ (Lines 438-443)

Figure 4: I recommend combining all of these subpanels into one plot. This will make it easier to follow the trends of different monosaccharide units with SST and to compare the concentrations of the monosaccharides with one another. You can use the same marker shapes and change the colors for the different monosaccharides. Also, add unit for concentration on y-axis.

Authors: We added the concentration unit to the y-axis. Regarding the recommendation to combine all subpanels into a single plot, we appreciate the reviewer’s idea and agree that, in principle, a combined figure could facilitate direct comparison among compounds. However, after careful consideration, we decided to retain the multi-panel format because the monosaccharides span substantially different concentration ranges (for example, glucose occurs at markedly higher concentrations than several of the other compounds). Displaying all variables in one panel would therefore require either multiple y-axes, logarithmic scaling, or normalization of the data, each of which would reduce the direct readability of the original concentration values. In addition, several monosaccharides show similar temporal patterns and partially overlapping concentration ranges, so combining all series into one plot would lead to strong visual overlap and reduced clarity. We therefore consider the current panel structure to provide the clearest presentation of both temporal trends and differences between bulk and SML samples while preserving the original concentration scale for each compound.

Line 453: Missing preposition – should be ‘likely due to being...’

Authors: We corrected the sentence by adding the missing “to”.

Line 525-527: The sentence needs to be rephrased. You may consider the following modification: ‘To examine how meteorological conditions and atmospheric mixing influence Na_{aer} and CCHO_{aer} at high altitudes, three distinct cases with constant weather conditions were selected (Figure 5).’

Authors: Thanks for this suggestion. We changed the sentence accordingly.

Figure 5: Need a colon after the word ‘Case’ in the panel headings. It could also be read as Case 12, November 2021, for example.

Authors: We agree that the previous panel headings could lead to this confusion. We have revised them to follow the format “Case I (12 November 2021): Mixed boundary layer, no low-level clouds.”

Line 772: This sentence needs to be rephrased. The word ‘precursors’ is used twice. Or did you mean precursors of oxalic acid precursors?

Authors: We actually meant the precursors of the precursors. However, to avoid confusion, we have rephrased the sentence. ‘But what are the precursors of glyoxal and glycolaldehyde, the precursors of oxalic acid?’ (Line 791)

Figure S3: Is the space for first sub-panel left blank intentionally? It looks incomplete

Authors: This space was intentionally left blank in an earlier version of the manuscript, where we presented a direct comparison between dCCHO and pCCHO. At that time, the blank area was reserved for displaying sea surface temperature (SST) in the dCCHO panel. However, after moving the dCCHO plot to

the main results (now Figure 4) and keeping the pCCHO plot in the supplement (Figure S3), this rationale no longer applies. We have now filled the previously blank space with SST as well. In addition, we have added concentration values to the y-axes, which were missing before.

References:

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