Answers to the Referees' comments regarding the manuscript:

Marine Carbohydrates and Other Sea Spray Aerosol Constituents Across Altitudes in the Lower Arctic Troposphere

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We thank the reviewers for the evaluation of our manuscript. In this document, all of their constructive comments have been answered thoroughly. 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. Please note that, following reviewer 1's request to move a figure from the supplement to the main manuscript and add another one, the figure numbering has been adjusted.

Reviewer 3:

The Arctic continues to change due to a warmer climate. The expanding ice-free ocean areas emerge as emission sources of sea spray aerosol (SSA) particles. Zeppenfeld et al performed balloon-borne and ground-based measurements of major species in SSA particles in autumn 2021 and spring 2022 at Ny-Ålesund. They claim that the similarities or differences in SSA species between ground level and high altitude are strongly influenced by meteorological conditions and atmospheric mixing. The microbial activity might be an important source of carbohydrates in SSA particles. The language is good, but the current manuscript is descriptive and too long to read. It is easy to get lost when reading the manuscript. Major revisions are required before the manuscript can be considered for publication. Here are the comments that needs to be addressed:

Authors: We thank the reviewer for the careful reading of the manuscript and the constructive comments. We appreciate the overall positive assessment of the language and the relevance of the topic.

We agree that the previous version of the manuscript was too descriptive and, in parts, too long and difficult to follow. In response, we have substantially revised both the structure and content of the paper, as detailed in the answers on the reviewer's comments below. The manuscript has been shortened and redundant descriptions were removed. We believe that these changes significantly improve the clarity, and readability of the paper, and we hope that the revised version addresses the reviewer's concerns.

Major Comments:

1. **Section 3.1 is too long to read. While the authors provide great details and discussion in light of the literature, I find it very difficult to catch the take-home message. Please make it compact and concise so that it will be readable.**

Authors: We thank the reviewer for appreciating the level of detail and discussion in this section. We agree that the readability benefits from a more compact presentation. We have therefore carefully revised the text to reduce redundancy, improve clarity, and make the take-home messages easier to follow. Despite the addition of content in response to other comments by reviewers, the number of lines has been substantially reduced in the following subsections:

- ***Sodium in aerosol particles (Na^+_{aer})***: old version: 53 lines; revised version: 39 lines (lines 351–381 and 402-409)
- ***Combined carbohydrates in fresh SSA and their oceanic origin***: old version: 46 lines; revised version: 35 lines (lines 414–439 and 450-458)
- ***$CCHO_{aer}$ at the winch and higher altitudes***: old version: 78 lines; revised version: 64 lines (lines 459-522)

2. **Section 3.2 is very long and descriptive. The discussion should not only analyze each of the three different cases individually but also compare them to one another. Again make it compact and concise.**

Authors:

- shorter and more concise.

We have reformulated and compacted Section 3.2, shortening the descriptions of each case while improving clarity and maintaining the depth of the information. In particular, chemical species are now consistently referred to by their chemical symbols (e.g. $\mathrm{Na}^+_{\mathrm{aer}}$) instead of their full names (e.g., sodium,

chloride) to enhance readability. The number of lines has been substantially reduced in the following subsections:

- **Case I:** old version: 61 lines; revised version: 45 lines (lines 537–581)
- **Case II:** old version: 46 lines; revised version: 32 lines (lines 582–613)
- **Case III:** old version: 62 lines; revised version: 58 lines (lines 614–671)

– more connection between the three different cases

To address the reviewer's comment regarding comparisons across cases, we have highlighted connections and differences between the three cases. Specific examples include:

- "On 27 September 2021, balloon measurements were conducted at a median altitude of 1112 m, above both the Zeppelin Observatory and the altitude range of Case I, i.e. in the free troposphere above the boundary layer." (lines 583-585)
- "In summary, Case II demonstrates that major SSA constituents (Na^{+}_{aer}, Ca^{2+}_{aer}, Cl^{-}_{aer}, $SO^{2-}_{4\,aer}$ and $CCHO_{aer}$) can be present in the free troposphere and likely originate from a distant source. However, they appear at different concentrations above the temperature inversion than in the mixed boundary layer below, where concentrations, like in Case I, are similar." (lines 610-613)
- "During the balloon's ascent and descent to 666 m, a positive gradient in potential temperature (272 K at the ground vs. 278 K at the balloon, **Figure 5c**) indicated a stably stratified boundary layer. Specific humidity was uniform (3.2–3.8 g kg^{-1}), while N_{150} was lower than in Case I (3–10 cm^{-3}) with higher relative variability, likely influenced by low counting statistics at these low concentrations. Overall, mixing conditions in Case III were similar to Case I, but sampling occurred partly within or below a drizzling low-level cloud." (lines 622-627)
- "Back-trajectory analysis (**Figure 6, Case III**) showed that air masses at the altitudes of ground, balloon, and Zeppelin Observatory followed the same 48-h path from the ice-free ocean south of Svalbard. Vertical trajectory heights indicate shared transport history and influence by the same emission sources, consistent with Case I." (lines 628-631)

Minor Comments:

1. **Section 2: I appreciate the levels of detail provided for measurement. To make it reader-friendly, I would suggest including a table to summarize what parameters have been measured and used.**

Authors: We have added a reader-friendly table at the beginning of Section 2 ("Experimental") to provide a clear overview of all parameters, methods, and sample/media types used in this study. The added text now reads:

Table 1. Overview of parameters, methods and sample/media types used in this study.

Category	Parameters	Method/Instrument	Sample/Medium
Major inorganic ions	Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, oxalate	Ion chromatography	Bulk seawater, SML, aerosol particles (filter)
Free and combined carbohydrates	Fuc, Rha, Ara, Gal, Glc, Xyl, Man, Fru, GalN, GlcN, MurAc, GalAc, GlcAc	HPAEC-PAD	Bulk seawater, SML, aerosol particles (filter)

Sea surface temperature	SST	Digital Thermometer	Ocean surface
Aerosol number concentration	N_{150} (150-2900 nm)	POPS (CAMP)	Atmospheric column
Meteorology	T, U, WD, RH, p, wind, θ, q	Standard meteorology package + thermodynamic equations	Atmosphere at ground (AWIPEV), atmospheric column
Cloud properties	Clouds and hydrometer types, IWP, LWP, IWV	Cloudnet + HATPRO	Atmospheric column
Biogeochemistry (model)	TChl-a, dissolved acidic polysaccharides	FESOM2.1–REcoM3	Ocean surface
Air mass origin	48-h back-trajectories	NOAA HYSPLIT	Several altitudes of atmosphere

2. **Figure 1 is a busy plot. I do appreciate the effort in data visualization. However, I feel it is hard to get the key information about Na^+, CCHO, and CCHO/Na^+ without looking at the legend back and forth. I would suggest having the concentrations of Na^+, CCHO, and their ratios in bulk water, SML, and aerosol particles as the y axes and color code the height.**

Authors: We believe the referee is referring to Figure 2, rather than Figure 1, based on the context of the comments. We acknowledge that Figure 2 may appear busy, as it integrates several layers of information. However, it seems difficult to enhance readability by swapping the visual encodings (using color to represent height instead of concentration, and placing concentration on the y-axis instead of height).

In the current design, the combination of sampling height and date functions as a unique identifier for each sample. This allows readers to track and compare individual samples consistently across panels 2a, 2b, and 2d. Changing the plotting scheme as suggested would introduce several issues:

a) Atmospheric mixing state and vertical structure would become less clear.

b) The vertical error bar associated with sampling height could no longer be displayed.

c) It would no longer be visually obvious where each sample was taken (Old Pier, Winch, Balloon, Zeppelin Observatory).

d) A direct comparison of Na^+, CCHO, and the CCHO/Na^+ ratio within induvial samples across several plots would no longer be possible, since the identification through date and height gets lost.

According to the reviewer's suggestion, we experimented with such an alternative visualization for Figure 2a, as shown below, but found that it reduced clarity for the reasons stated above.

For these reasons, we believe that, even though Figure 2 is visually dense, the current layout offers the most balanced and informative representation of this complex dataset. We therefore prefer to keep Figure 2 in its present form. In addition, we have added a new plot (Figure 3) in response to a comment by Reviewer 1, which we expect will also address the concern raised by Reviewer 3.

Figure 2a (Original version as submitted by the authors):

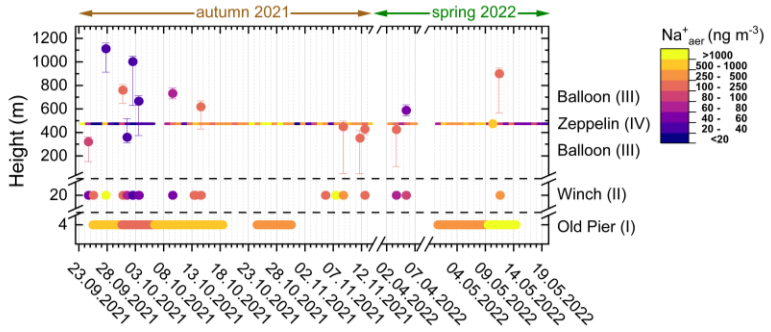
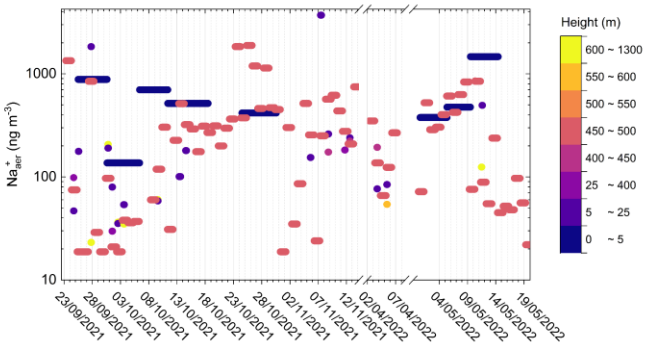


Figure 2a (Version as suggested by the referee):



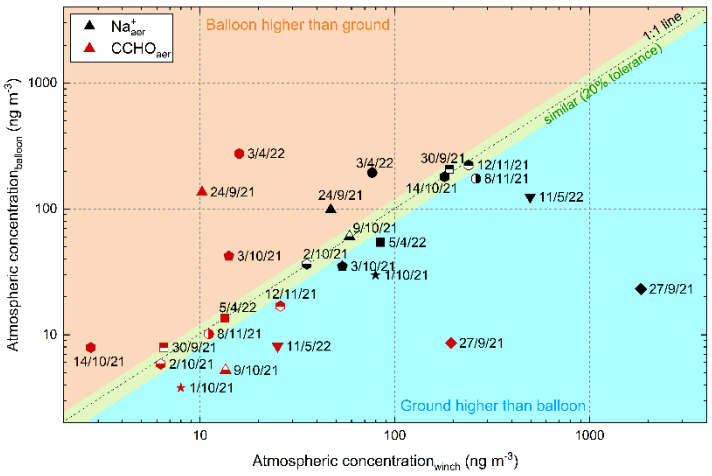


Figure 3. Scatter plot showing Na^{+}_{aer} (black symbols) and $CCHO_{aer}$ (red symbols) concentrations in TSP measured at the winch, and balloon levels. Data points are categorized to indicate whether values were similar, higher at the balloon, or higher at the ground.

3. **Line 394: Please elaborate more on the atmospheric ageing processes.**

Authors: As referee 1 suggested, it is preferable to use the term "atmospheric processing" instead of "ageing" in this manuscript. Since we already dedicate substantial text to atmospheric processing in section 3.3, which addresses aerosol deposition, in-situ formation, and degradation of marine $CCHO_{aer}$, and considering the comment on manuscript length by referee 3, we decided to keep this section brief here and refer the reader to section 3.3 for a more detailed discussion. The revised text now reads: "Longer atmospheric residence increases the exposure of SSA particles to processing, which can alter their impact on cloud formation. While Na^{+}_{aer} is considered chemically stable, co-emitted OM including carbohydrates may undergo physical, chemical and microbial changes (Zeppenfeld et al., 2021, 2023). This aspect will be explored further in section 3.3." (Lines 405-409)

4. **Line 435: What is the rationale behind the claim "most probable local emission source for SSA"?**

Authors: We agree that the original wording was not clear. We meant that Kongsfjorden is the only local source of SSA, because all other ocean regions are much farther away and rather act as long-range sources, as discussed later in the manuscript. We have changed the sentence to: "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, the only local emission source of SSA." (Lines 420-421)

5. **Lines 442-443: Again what is the rationale behind the claim "the primary source of atmospheric $CCHO_{aer}$"? I guess this statement only applies to the study here.**

Authors: We explained the rationale for this claim in the comment above and changed the text accordingly. However, we realized that the statement (the primary source of atmospheric $CCHO_{aer}$) is now redundant in this context, so we have preferred to remove it. The revised text now reads: "Similar seasonality was observed for selected monosaccharides among dissolved combined carbohydrates (dCCHO) in Kongsfjorden seawater." (Lines 424-425)

6. **Liens 743 – 746: How did the authors come up with a one to two order magnitude reduction in absolute particle masses?**

Authors: We acknowledge that our original phrasing was imprecise. We have now clarified that the reduction refers specifically to the atmospheric concentrations of major inorganic ions and $CCHO_{aer}$, and we have referenced the observations shown in Figure 5b. The revised text now reads: "Typical removal processes of supermicron particles, such as dry and wet deposition or cloud droplet activation, likely reduced the atmospheric concentrations of major inorganic ions and $CCHO_{aer}$ by one to two orders of magnitude before the arrival of the air masses in Ny-Ålesund (**Figure 5b**)." (Lines 683-686)

Technical Comments:

1. **CCNs and INPs are only used a few times. It is redundant to use the abbreviations, since they are not the focus of the work.**

Authors: We have removed the abbreviations and replaced them with the full terms, "cloud condensation nuclei" and "ice-nucleating particles" throughout the manuscript.

2. **Lines 363-367: The sentence can be shortened by saying "… at both locations, winch and balloon (e.g., 30 September: 191 vs 207 ng m^{-3};…)" Repeating "at the winch" and "at the balloon" is not necessary. The same applies to Lines 367-373.**

Authors: We appreciate the reviewer's suggestion and have adopted a shorter and more elegant way to indicate whether the values refer to the winch or the balloon. The revised text now reads: "Several events showed nearly identical Na^+_{aer} concentrations (winch vs. balloon), e.g., 30 Sep: 191 vs. 207 ng m^{-3}; 2 Oct: 35 vs. 36 ng m^{-3}; 9 Oct: 59 vs. 60 ng m^{-3}; 12 Nov: 240 vs. 223 ng m^{-3}. In contrast, other periods exhibited strong vertical gradients with higher ground-level concentrations (e.g., 27 Sep: 1840 vs. 23 ng m^{-3}; 5 Apr: 84 vs. 54 ng m^{-3}; 11 May: 496 vs. 125 ng m^{-3}), while two cases showed higher values at the balloon (24 Sep: 47 vs. 99 ng m^{-3}; 3 Apr: 77 vs. 194 ng m^{-3})." (Lines 367-372)

3. **Line 374: Just wondering if atmospheric depletion processes is the right term. Can we just use "atmospheric processes"?**

Authors: We agree that using a more general term without "depletion" better matches the examples that follow as well as the preceding sentence. The revised text now reads: "These variations are likely driven by atmospheric processes, including dry and wet deposition (Farmer et al., 2021), dilution during vertical and horizontal transport from the emission region (Wong et al., 2019), vertical mixing (Pilz et al., 2024) and differing air mass histories (Willis et al., 2018), which will be examined in detail for three selected cases later in this study." (Lines 372-376)

4. **Lines 391-392: What was consistent between studies? Na^+ concentration?**

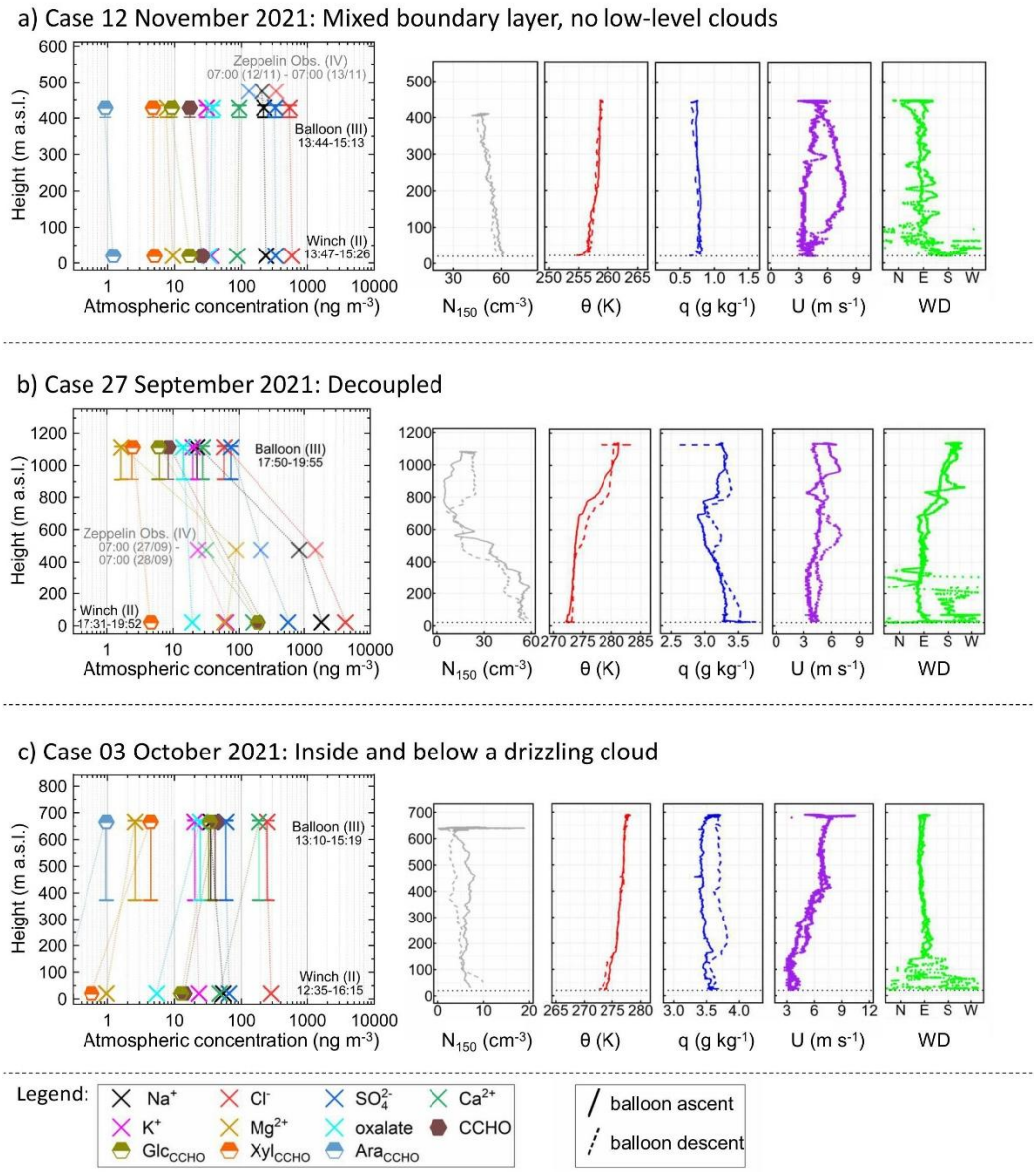
Authors: We clarified that the consistency refers to the observation that SSA compounds reach elevated, cloud-relevant altitudes, in agreement with the cited studies. The revised sentence now reads: "This vertical distribution is consistent with the aircraft-based SSA measurements reported by Hara et al. (2003) and Köllner et al. (2017)." (Lines 404-405)

5. **Line 431: It is unclear when the colder and darker months are.**

Authors: We agree that our original wording was not precise, especially since we only had measurements for one month (October) during the "colder and darker" period. We have rephrased the text to make the timing clearer. The revised text now reads: "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), showing a seasonal pattern with the highest values at the beginning (end of September 2021) and end (mid of May 2022) of the study, and lower values in October 2021 (**Figure 2c**). No samples were collected between November and April, so winter trends remain unknown." (Lines 415-419)

6. **Figure 3: Could the authors provide the legend for solid and dashed lines for the subplots on the right-hand side?**

Authors: We have added a legend indicating that the solid lines represent the balloon ascent and the dashed lines represent the balloon descent in the right-hand subplots of former Figure 3, now Figure 5 (see below).



7. **Some of the abbreviations are not necessary when they are used a few times throughout, e.g., LWP, IWC.**

Authors: We appreciate the reviewer's suggestion regarding the use of abbreviations that appear only a few times, such as LWP, IWP, and IWC. We considered replacing them with the full terms, such as "vertically integrated water vapor" or "cloud liquid water path". However, we found that using the full terms throughout the manuscript considerably reduced readability. To maintain clarity and conciseness, we have therefore retained these abbreviations.

8. Line 761: What do selective removal processes mean?

Authors: In the previous section, we readily discussed the potential deposition of supermicron versus submicron particles ("selective removal processes") over time as a possible explanation for shifts in $CCHO/Na^+$ ratios in TSP. In this section, we clarify that this mechanism (of "selective removal") does not explain the observed pattern regarding absolute $CCHO_{aer}$ concentrations for some atmospheric cases. We have rephrased the text to make this reference clearer to the reader. The revised section now reads: "However, for the three cases with the most pronounced increases in **$CCHO_{aer}/Na^+_{aer}$ ratios** in TSP at higher altitudes (24 Sep 2021; 03 Oct 2021; 03 Apr 2022), absolute $CCHO_{aer}$ concentrations were also elevated (compare **Figures 2b and 2d**). Such increases in absolute concentrations cannot be explained by the selective removal of supermicron particles as hypothesized above." (Lines 698-702)

9. Figure S7: Could you include the measurement location on the plot?

Authors: We added red stars to the plot (former Figure S7, now Figure S6) marking the measurement location.