Reviewer #1

This study presents measurements of trace elements and major ions in surface snow collected during three field campaigns from 2018 to 2021. The authors report higher concentrations of marine-origin species in late spring 2020, likely driven by specific meteorological and oceanic conditions. The results also show a strong correlation of impurities in Ny-Ålesund during cold seasons. The manuscript lacks a clear explanation of how these findings contribute to our understanding of climate change. The introduction sets up climate relevance, but the discussion does not adequately follow through. A more focused and contextualized discussion is necessary to justify the broader implications claimed by the authors. The paper can be published after addressing these comments.

A: We thank the reviewer for their helpful comments. As suggested, we have improved the manuscript by clarifying the discussion of our findings in the context of climate change. Specifically, we now better highlight how the observed seasonal variability and impurity correlations relate to Arctic atmospheric and oceanic changes. We believe these revisions strengthen the manuscript and address the reviewer's concerns.

Major comments.

R-1. Line 89: You mention that this study contributes to understanding trace element and ion interactions in the context of recent climatic changes. However, this connection is not clearly addressed in Section 5, Summary and Conclusion. If the stated goal is "to enhance the understanding of these interactions, particularly in the context of recent climatic changes," then the conclusion should explicitly discuss how your findings support or inform that objective. As it stands, the broader relevance to climate change is implied but not directly explained. This should be clearly articulated before the manuscript can be considered for publication.

A-1. *We see the point raised by the Reviewer 1, and we agree that the relevance to climate change has to be better explained in Summary and Conclusion. For this reason, we modified the text from L620, changing this statement: "*In fact, sea ice has a role in concentrating, storing, and releasing marine species, as well as influencing atmospheric and oceanic processes that affect their production and distribution." *with "*These results provide direct evidence of how sea ice extent modulates the storage, release, and transport of marine-derived impurities, thereby influencing snow-atmosphere chemical exchange processes under varying climatic conditions.". *Furthermore, from L627, the following paragraphs:*

"Therefore, our results highlighted a close dependence of high concentrations of impurities found in the snowpack at Ny-Ålesund on meteorological conditions, especially during cold years, when the production of sea spray related aerosol likely derives by a larger extension of sea ice and stronger local Arctic circulation. The identification of geogenic, marine, and anthropogenic sources in the snowpack was allowed by a chemometric approach (HCA), which clarified the EFs results. The back trajectories analysis revealed distinct seasonal air mass patterns. During fall and winter, air mass predominantly originated from Northern Canada in addition to air masses arriving from Arctic Ocean and Kara seas during spring. On the contrary, no prevalent mid-latitude air currents were revealed in spring as expected, considering the period of the three sampling campaigns (2018-2021). These findings offer new insights into how specific meteorological and oceanic conditions, such as sea ice extent, wind speeds, and Arctic Oscillation phases, influence the chemical composition of the snowpack in Svalbard, particularly within the context of Arctic Amplification."

*have been changed with: "*Such findings illustrate how large-scale atmospheric circulation anomalies associated with Arctic Amplification can significantly alter the deposition patterns of both natural and anthropogenic species in the snowpack. This is particularly evident especially in cold years, when the production of sea spray related aerosol likely derives by a larger extension of sea ice and stronger local Arctic circulation. The identification of geogenic, marine, and anthropogenic sources in the snowpack was allowed by a chemometric approach (HCA), which clarified the EFs results. The use of chemometric techniques (HCA) and back-trajectory analysis enabled a clearer attribution of sources and transport pathways, improving the interpretation of snow composition in relation to meteorological drivers. Specifically, the distinct seasonal air mass patterns revealed, characterised by dominant Arctic-origin air masses in fall and winter and a lack of expected mid-latitude inputs in spring, underscore the changing dynamics of snow-atmosphere interactions in a warming Arctic. Overall, these insights advance our understanding of how recent climatic anomalies, such as altered sea ice extent, shifts in Arctic Oscillation phases, and stronger polar vortices, modulate the chemical composition of the snowpack in Svalbard. Our findings highlight the sensitivity of snow-atmosphere exchanges to both local and large-scale climatic processes, offering important context for interpreting snow chemistry trends in a rapidly changing Arctic environment."

We think that a clearer explanation has been provided to the readers in this way.

R-2. Line 114: Did you collect and analyze background concentrations or include any blank/control samples during sample handling? It is important to clarify how you ensured that the snow samples

were not contaminated during collection, transport, or analysis. Please explain the procedures used to confirm sample integrity and rule out possible contamination.

A2. *We appreciate Reviewer1's comment and agree that the procedures to ensure sample integrity and minimise contamination should be clarified. To assess potential contamination during sampling and transport, we collected field blanks using metal-free vials (Avantor, VWR Centrifuge Tubes, CHN). Some of these vials were opened to ambient air at the sampling site for a few minutes without collecting any snow, then sealed and transported to the Ny-Ålesund laboratory, where they were filled with 2% HNO3.*

In parallel, we prepared analytical blanks by following the same procedure (vials opened to air but without snow sampling), then sealed and transported to Venice, where they were filled with 2% HNO3 and ultrapure water (UPW) from the Venice lab. Both set of blanks were analysed to detect any background contamination and were consistently below LODs or one order of magnitude lower than the lowest concentration detected for all analytes.

*We have integrated this clarification into the main text to better explain the control measures used to ensure sample integrity throughout collection, transport, and analysis: "*To assess potential contamination during sampling, handling, and transport, field blanks were collected during each campaign. Metal-free vials (Avantor, VWR Centrifuge Tubes, CHN) were opened to ambient air at the sampling sites for a few minutes without collecting snow, then sealed and transported to the Ny-Ålesund laboratory. There, they were filled with 2% HNO_3 and stored under the same conditions as the snow samples. In parallel, analytical blanks were prepared by opening vials to air, sealing them, and transporting them directly to Venice, where they were filled with 2% HNO_3 and ultrapure water from the laboratory. Both field and analytical blanks were analyzed alongside the snow samples, confirming that background contamination levels were below detection limits for all target analytes."

R-3. Line 150: It would significantly improve the clarity of the manuscript to include a map showing the sampling locations. Since the sampling was conducted across multiple sites during three separate field campaigns, a visual representation in the main text (not only in the supplementary materials) would help readers better understand the spatial context of the study.

A-3. *We agree with the Reviewer's suggestion, and we included a map showing the sampling locations in the manuscript, moving the modified Fig. S1 from the Supplementary materials.*

R-4. Line 202: The manuscript should clearly explain the rationale for selecting a 6-hour back-trajectory interval with a propagation time of 120 hours.

A-4. *Authors welcome the suggestion of Reviewer1 and implemented this part in the manuscript. L201-210: "Back-trajectories were calculated every 6 h, with a propagation time of 120 h for each sampling period. The choice of a 6-hour interval for the calculation of back-trajectories allows for the capture of temporal variability in air mass origins over the day, which is particularly important in polar regions where atmospheric circulation patterns can change rapidly. This temporal resolution strikes a balance between computational efficiency and the need for sufficient detail to characterise the variability in source regions during each sampling period. The propagation time of 120 hours was selected to provide an adequate temporal window to trace long-range transport pathways that influence air mass composition at Ny-Ålesund. This configuration is consistent with previous studies on atmospheric circulation in the same site (Feltracco et al., 2021)."*

R-5. Line 238-239: You state that "the difference in concentration trends appears very low or negligible, except for sporadic peaks in sea salt and crustal tracers present in the Ny- Ålesund record from November to February." However, the term negligible needs to be supported with quantitative data. Please specify the concentration values and the average concentration differences compared to the November–February period to substantiate this claim.

A.5 *We modify the sentence as suggested by Reviewer1: "the difference in concentration trends appears* ***not statistically significative (p values < 0.05, Wilcoxon test)*** *for all the analysed species, except for sporadic peaks in sea salt and crustal tracers present in the Ny-Ålesund record from November to February".*

R-6. Line 405: Please provide appropriate references to support the statement that enrichment factors (EF) below 10 indicate a crustal origin of the elements.

A-6. *We added the appropriate references here, as suggested: Wedepohl, 1995; Gabrieli et al., 2011*

R-7. Line 520: You refer to correlation results multiple times throughout the manuscript, but you do not indicate which figures or tables support these findings. Please clearly reference the relevant figures or tables in the text. If these results are not currently included, they should be added to the supplementary information to support the discussion.

A-7. *We thank the Reviewer for noting the lack of a supporting figure/table. We added Fig. 5 in the main manuscript.*

R-8. Line 554: As previously mentioned, you need to provide specific values rather than stating that "the errors associated with the EFs are quite high."

A-8. *We see the point of the Reviewer. However, our intention here was not to provide specific values, but to highlight that the EF calculations may reflect larger uncertainties than the HCA method. For this reason, we decided to modify the sentence with the following statement: "This apparent discrepancy may reflect the relatively larger uncertainties typically associated with EF calculations, which can inherit errors from the choice of reference element, assumptions about crustal composition, and variability in background concentrations, compared to the more integrative approach of HCA (e.g., Reimann and de Caritat, 2000)."*

Minor comments.

R-9. Line 210: Provide the full name for the acronym NCEP/NCAR at its first mention in the text to ensure clarity for all readers.

A-9. *We thank the Reviewer1 for raising this point. We clarified the acronym in the text. L217-218: "National Centers for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR)".*

R-10. Line 233: There is a missing period at the end of number 3. Please add it to maintain proper punctuation.

A-10. *We added the missing period to maintain proper punctuation, as kindly suggested.*

R-11. Line 271: In Table 1, "Total" should be capitalized to maintain consistency with proper noun formatting. Change "total" to "Total."

A-11. *We capitalized "Total" in Table 1, as suggested.*

R-12. Line 396: Instead of using vague terms like "slightly above," I recommend providing the exact number to improve clarity and precision in your results.

A-12. *We agree with the point raised by the Reviewer1, and we modified the vague expression with "(i.e., EF = 26)."*

R-13. Figure S2: In the figure, the plots for Pb and Ca^{2+} slightly overlap, particularly where their highest concentrations coincide. To improve clarity, consider adjusting the plot style to avoid confusion in interpreting the peaks.

A13. *We thank the Reviewer for raising up this point and we adjusted the plot style to avoid confusion in interpreting the peaks.*

R-14. Figure S3: You need to explain why the graph includes a gray background for the periods from autumn 2018 to autumn 2020 and from spring 2019 to spring 2021.

A-14. *We added the explanation in the caption: "The grey areas correspond to the distinct snow seasons."*

R-15. Table S2: The "k" in "Km²" should be lowercase. Correct it to "km²" to follow proper SI unit formatting.

A-15. *We put "Km^2" in lowercase to follow proper SI unit formatting.*

R-16. Figure S7: The figure needs to be provided in higher resolution. The text and numbers under the Ice Categories are difficult to read in the current version.

A16. *We agree with the Reviewer1, and we provided a higher resolution figure (ex Fig. S7, now Fig. S8).*

Reviewer #2

Note from the Authors: We would like to kindly inform that Reviewer 2 appears to have based their evaluation on an earlier version of the manuscript (the preprint uploaded before the first round of revisions). As a result, a few discrepancies between the lines and content of the text were identified during the revision process. Additionally, some comments referred to figures and text that had already been modified. Nevertheless, the Authors have made every effort to address all the comments thoroughly and would like to express their sincere thanks to the Reviewer for the valuable and insightful suggestions.

Review of the manuscript entitled "Impact of Arctic Amplification variability 1 on the chemical composition of the snowpack in Svalbard"

This is an interesting paper reporting a lot of data on snow composition from Svalbard. While many papers have already been published about this, the novelty of the present work is the attempt to link compositional data obtained over a 3 year period and meteorological conditions. This is definitely the added value of the manuscript. The idea is interesting and can potentially deepen our understanding of snow chemistry in the Arctic environment, but in my opinion it is not very well developed. **The weakest aspect of the work is the link between snow data and meteorology, which is now only qualitative.** The discussion is made reporting things like: the season of that year was coldest and so sea-ice related elements were higher. Since the authors have in their hand a bunch of detailed meteorological data, **I would really expect to find a correlation study between snow composition and meteorological variables.** Adding this kind of discussion would really improve the novelty of this work and increase its scientific significance.

R1 - In general **I would try to be more quantitative across the entire manuscript**, **applying statistical tests to highlight the similarity of dissimilarity of data** recorded in different seasons or years.

A1. *We really appreciate this valuable suggestion from Reviewer 2, and we try to be more quantitative across the entire manuscript. We applied statistical tests (Kruskal-Wallis, Dunn, Spearmann correlation) to highlight the similarity/dissimilarity of data (see answer to R3).*

R2 - One other thing concerns the comparison between data gathered at GSRS and Ny-Alesund. In the end the authors decide to focus only on GSRS data. I am wondering whether deciding not to include Ny-Alesund data would be a good choice. In the end all the discussion about Ny-Alesund data is rather useless to the aim of the work.

A2. *The comparison between Gruvebadet and Ny-Ålesund concentrations is crucial for assessing the spatial variability of the measured parameters, which is a key aspect of our study. While the Ny-Ålesund data are not used directly in further analyses, they provide important context and serve as a reference point to highlight local-scale differences in concentrations. For this reason, we believe it is important to include the comparison in the main manuscript, as it strengthens the interpretation of our findings. However, since 2019 renovation activities at the Ny-Ålesund site have limited the reliability of data from that station, and therefore a robust comparison is only possible for the first sampling campaign. For this reason, we decided to focus further analyses exclusively on the GSRS data while retaining the comparison with Ny-Ålesund as a meaningful reference point in the discussion of spatial variability.*

R3 - I suggest to follow a more schematic approach to discuss the variability of elements/ions in snow across seasons and years. I would start **distinguishing** (**statistically** of course) **those species which present always the same concentration from those presenting significant differences.** I would apply this approach **considering the whole years and single seasons** (a comparison between the 3 winters considers, 3 springs and so on). In this way it would be possible **to clearly highlight where you should focus to explore a correlation between snow composition and meteo/climatic conditions.** At this point it would be rather simple to apply some statistical tool to assess whether snow data are correlated to meteorological variables.

A3. *The authors appreciate Reviewer 2's valuable suggestion. Considering that the Reviewer 2 has evaluated a previous version of the manuscript, this aspect was already improved in the second version thanks to the previous reviewers. However, to better address this point, we first applied the robust, non-parametric Kruskal-Wallis test to assess whether significant differences existed among seasons. This was followed by the post-hoc Dunn test to identify which specific seasons differed, with significance evaluated using adjusted p-values (< 0.05). The results, presented at the end of # 3.2, are reported in Table S5 and Fig. S3. Finally, we performed a Spearman correlation on the selected variables (the ones that showed a significant variability across seasons and years) to examine potential relationships between snow composition and meteorological/climatic conditions. The correlations between the analysed species (ions and trace elements) and the meteo/climatic variables were generally weak, with all coefficients below 0.4. The weak correlations indicate a complex influence of the considered meteorological variables on the analysed species concentrations, suggesting that the effects of Arctic amplification may involve additional or interacting processes that require additional deep investigations.*

R4 - In general the meteorological description of the three years considered in the study is rather qualitative and not very well supported by data. You report many passages like "Lower surface air temperatures, reduced precipitations, higher wind speed (m sec-1), and minor mean snow height with respect to the typical AA conditions, were induced by strong cold polar vortex triggered by a net positive Arctic Oscillation (AO) phase, and recorded in the 2019-20 winter season". It would be more valuable if you could **provide some number (mean temperatures, anomalies, deviations) to provide a quantitative framework** to support your description.

A4. *We thank the Reviewer for raising this aspect. We first opted for putting meteorological information in Table S3 (cited in the text), but we saw the Reviewer's point and we provided quantitative information to support the description in the main text (L324-329).*

R5 - About Pb: you make some discussion about the period presenting anomalously high Pb concentration, assuming an enhanced atmospheric transport from pollution sources (related to the weakening of the polar vortex) as the most likely cause. **It would be interesting to see the behavior of other element typically related to atmospheric pollution to see if something similar is observe (As, Zn, and others,** you measured a lot!).

A5. *We appreciate the Reviewer's suggestion to explore the behaviour of other elements typically associated with atmospheric pollution (such as As, Zn, and others). The relationship between Pb and other anthropogenic species is recognized with HCA where As and Zn have been shown short linkage distance. We have chosen to focus primarily on Pb in this manuscript due to its clear role in the observed anomalous concentration period. Moreover, adding other species in Figure 3, already very complex, would generate more confusion without adding new significant information. Given the already discussed tracers of atmospheric transport, we feel that the current discussion provides already significant insights into the potential link between enhanced pollution sources and the meteorological-climatic conditions.*

R6 - You refer to many Figures reported in the Supplementary material, while in the end you only included a few figures in the main text. This is not very helpful as the reader has continuously to shift from the main text to the supplement. I suggest to **move some of the supplementary figures in the main text** and eventually to limit the number of figures in the supplementary.
A6. *We see the Reviewer's point, and we moved Figure S1 to the main text (now Figure 1), after modifying it as suggested by Reviewer's 1. We also decided to add a new Fig. 5 (correlation plots) in the manuscript, in spite of placing it in the SI.*

Considering the above, I can't recommend this manuscript for publication at this stage. From my point of view major revisions are needed.

Please find below some more specific comments:

R7 - Figure1: I can't see solar radiation, but this is reported in the caption. Please add something to indicate the years shown in the graphs.

A7. *Actually, in the last version of the manuscript, we didn't report solar radiation in the caption. Caption claims: "**Figure 1.** AO Index, air temperature (°C), precipitation (mm), snow height (cm), Δh snow height (cm), wind speed (m sec^{-1}), and wind direction (°) from the NCEP/NCAR Reanalysis data. The green horizontal line above the wind speed graph indicates the 5 m sec^{-1} threshold, above which wind drift may occur on surface snow layers. The colour of the line refers to the Δh color scale, which indicates negative values of Δh, NOAA Physical Sciences Lab's daily composites tool was used to calculate the near-surface air temperatures across the Northern Hemisphere from October to May. Grey bands indicate the winter periods."*

We also reported the years in x-axis already.

R8 - Figure 3 and related discussion about clustering: I can't see in the list all the elements that you measured (as reported in the introduction of the manuscript. For example Zn).

A8. *Also in this case, we already modified the Figure during the first round of revisions. The referee 2 evaluated the previous version of the manuscript.*

R9 - Line 103: "this is the snow layer most influenced by the aerosol-cryosphere exchanges" not very clear, maybe "this is the snow layer most affected by exchanges involving snow, atmosphere and aerosol"?

A9. *We see the point of Reviewer2, and we agreed to slightly change this sentence. We rephrased as follows: "*this is the snow layer most impacted by aerosol deposition and exchange processes at the snow-atmosphere interface".

R10 - Line 208-215: it is not clear the reason why you choose to apply the test. You want to highlight the difference among two different data populations right? Maybe it would be better to clearly report what the two populations are.

A10. *We appreciate the reviewer's comment. To clarify, our aim was indeed to compare the ionic loads at Ny-Ålesund and Gruvebadet, which represent the two data populations under study. We applied the Wilcoxon rank-sum test (Mann-Whitney U test) to assess whether there was a statistically*

significant difference in contamination levels between these two sites. We revised the manuscript accordingly, to state this more clearly.

R11 - Line 231: it would be interesting to know what ions showed significant differences among the two trends and provide a little bit of discussion about this.

A11. *Wilcoxon test highlighted significant differences among the two populations of samples (NyÅ and GSRS) only for Na^+, K^+, NO_3^-, Br^-, and MSA, which are all marine species, likely most influenced by the closeness of NyÅ site to the coast.*

R12 - Line 238-239: what % data refer to? To total ionic load?

A12. *% refer to the concentrations, not the ionic loads.*

R13 - Line 277: it is clear what you want to say, but "Arctic type" is not very rigorous. I suggest to rephrase

A13. *We appreciate the Reviewers' feedback. However, we already clarified this aspect during the previous round of revisions, and no more "Arctic type"expressions have been used within the main text.*

R14 - Line 295-296: but you said before that sulphates are not always dominated by ss fraction, so I would add something to say that sulphate are not always dominated by marine aerosol.

A14. *Also in this case, we already clarified this aspect during the previous round of revisions.*

R15 - Line 304-306: how the presence of drift ice can explain a higher deposition of geogenic elements? The thing is clear for marine-related species but I can't understand for the geogenic ones.

A15. *We thank the Reviewer for this insightful comment. While statistical analysis did not show significant differences for the geogenic elements (Al, Ca, Mn, Sr), except for Fe, which weakly correlated with air temperature, we hypothesize that their increase may still be linked to the overall atmospheric conditions during the late spring 2020 period, although correlations may not have been captured fully in the statistical analysis. For example, these conditions could have promoted the resuspension of dust and sediment from the local environment, such as from coastal areas, which could have been transported by stronger winds to the study area. To avoid overinterpretation of the text, we decided to modify this paragraph as follows "These conditions likely enhanced the production of sea spray aerosols, which, when carried by winds, may have increased the deposition of marine species onto the snowpack. The increased deposition of geogenic elements might also have*

been influenced by low temperature anomalies (as seen with Fe), and/or by stronger wind speeds, although significant correlations have not emerged in this preliminary statistical analysis."