Response to editor decision

Dear Nanna B. Karlsson

On behalf of myself and my co-authors, thank you for the invitation to upload a version of the manuscript revised based on reviewers' comments.

This revised version addresses the comments made by the reviewers. These changes are detailed below, directly following the responses shared in the interactive discussion, with specific detail on which additions have been made to address the reviewer's comments.

The revised manuscript has been uploaded, alongside a version with tracked changes. A revised supplement has also been provided.

I look forward to hearing back from you regarding the manuscript.

Kind regards

Piers Larkman

Relevant changes based on reviewer comments. Excerpts from the review comments are in black, revisions are shown in blue. **Line numbers refer to the version of the revised manuscript with visible tracked changes**.

Amendments made based on to review 1

The present manuscript focuses on sodium, which tends to accumulate at grain boundaries. In contrast, certain ions, such as chloride, dissolve and substitute for H_2O molecules within the grain interior. In these cases, I suspect that the impact of grain size on measurement resolution (or the difference between 1D profiles and CFA) may not be as pronounced compared to impurities that accumulate at grain boundaries.

The archetypical nature of Na as a soluble impurity is further noted on L65, and L246-247.

The fact that further considerations are required for modelling insoluble impurities in grain interiors is noted on L71, and L247

The authors concluded that signal deviation increases with larger grain sizes. I wonder whether this deviation is influenced solely by grain size. It seems likely that impurity type (whether substituted within the grain interior or concentrated at grain boundaries) and climate period (glacial or interglacial ice) could also play a role in signal deviation. If the authors have insights on these factors, I suggest them to share their perspectives, as it would be beneficial for readers.

Additional factors influencing signal deviation have been listed on L315 - 317

The variability between different impurity types has been noted on L42

Such complexities in grain boundaries and size distribution are observed not only in deep ice but also in ice samples deformed under high temperature and stress, such as those from the EastGRIP ice core.

A note has been included on L409-410, specifically referencing EGRIP, noting high deformation sites require further consideration.

L7 in the abstract: What does “high-frequency component of signals” mean?

This term has been revised on L7 of the abstract

L15 in the abstract: *This approach guides collecting layer-representative signals from LA-ICP-MS line profiles and may help to bridge the scale gap between LA-ICP-MS data and data collected from meltwater analysis*.

As mentioned, this approach could assess the scale gap. On one hand, how does this approach specifically bridge the scale gap? I think this is the most interesting for general glaciologist.

In the introduction of the revised manuscript, the scale gap discussion has been introduced on L44, and stressed on L57-59

L79 (Table 1) In the EDC ice samples, mean grain size in LGP is larger than that in Holocene? In Figure 3, grains in EDC LGP ice sample look small.

This has been clarified in figure 3’s caption.

L63: What does “The computational representation uses the arguably most simple manifestation of a climate signal, a constant signal” mean? How does the climate signal mode affect the results and discussion of the present study such as experimental measurements and ice structure generation? Please provide brief explanation for general cryosphere’s readers.

Section 2.1 has been reworked to clarify this. Specifically the paragraph at L83 – 88 now introduces this idea

L103: I understand that the modelling of 3D ice structure is useful. However, I didn’t see how 3D ice structure helped the verification of results and discussion. Figures 4 to 8 indicate 1D or 2D results. 2D ice matrix model is not sufficient?

The added discussion in the new section 4.3.3 relies on a 3D structure to predict bulk concentrations.

Points regarding the additional value of the 3D model have been added on L156, L294-295

L188: *These modelled signals show the same general features as experimentally measured signals, with large spikes in intensity where profiles intersect grain boundaries*.

It is difficult to determine whether experimental and modelled profiles signals have similar behaviors. Replication of experimental results by means of a model is, in my view, important in the present study. Please provide a comparative figure between the experimental and modelled results.

The plots in figure 5 (a) and figure 7 (a) are equivalent for experimental and modelled results. This is highlighted in the caption for figure 7, and on L244

Figure and table:

Table 1: Please provide explanation for “Profile lateral separation (mm)”.

This value has been illustrated in figure 6 (in addition to the redesign suggested by both reviewers). The table 1 caption directs the reader to this illustration.

Figure 3: Redesign is required.

Three images (optical image grain boundary segmentation, and chemical map) are not identical at RECAP ice samples. For example, in Holocene sample, grains in the middle and right images are elongated vertically. In LGP sample, grains in the left image are elongated vertically. Please modify.

Aspect ratio and covered areas in figure 3 have been revised and the plot re-made

The optical image of EDC Holocene sample is low resolution, it is difficult to distinguish grain boundaries.

A smaller image has been provided to aid readability, although this image is still at low resolution

Size of EDC LGP samples is too small. Image sizes should be the same for all samples.

Plot sizes have been unified

Figure 6: (a and c) It is difficult to see grain boundaries, please make contrast clearer.

Contrast for this figure, and all similar, have been increased

(b and d) What do the color difference of grains mean?

The colour scheme has been updated and clarified in the figure caption.

Figure 7: Why do the smoothed profiles (d) and (e) appear to be different? (It doesn't even look similar to panels a to c)

This is also the case in Figures S6 and S9.

This is explained on L218 – 222, and the y-axis change stressed in the captions of figures 5 and 7

Figure 8: Bottom two panels are labeled with (a) and (b). Please modify. Additionally, the spot size in panel b and caption is shown as 280 um, but the main text explain 260 um. Please correct value.

This is updated on L 227

Amendments made based on to review 2

The authors describe in the introduction that LA-ICP-MS analyses are necessary to reconstruct climate records in deep ice and use this as a primary reason for this study. However, the authors use the most basic structure of ice in this model. This is understandable given the continued questions around methodologies for LA-ICP-MS, however, as the work currently stands there are limited implications for deep ice studies.

The requirement for extension for deep ice application is clarified on L 69 - 71

Additionally, while there is an attempt to quantify the CFA results using modeled results, no actual experimental data is provided to ground truth the model's ability to reconstruct CFA results. Additionally, as no concentrations are provided and no calibration was conducted, it is difficult to see whether the modeled data that this project hinges on are realistic or comparable. As a result I recommend the manuscript be reconsidered after major revisions.

Section 4.3.3. has been added that adds further ground-truth examples, including reference to calibrated data

Specific comments:

L188: The statement “These modelled signals show the same general features as experimentally measured signals, with large spikes in intensity where profiles intersect grain boundaries.” is not proven as the experimental signals are not shown for comparison.

This statement has been clarified on L210- 211

Figure 6: greater contrast is needed to see grain boundaries in a) and c). It is also very difficult to see the blue and red lines. Please make these thicker or choose different colors.

The colours used in the figure have been revised to make the features clearer.

Figure 7: There is no explanation I can find for why 7d and 7e show opposite profiles. Please provide more information. This is particularly important for the author’s claim that this model can be used to compare between LA-ICP-MS and CFA results.

This is explained on L218 – 222, and the y-axis change stressed in the captions of figures 5 and 7

Line 222: I’m unclear why no calibration is used here. Particularly for comparing LA-ICP-MS to CFA signals or comparing between analyses, and ground truthing the model, this is important to understand how well the parameterized model works.

The added section 4.3.3. discusses this work in the context of limited available calibrated data

Line 254: “In this context, the framework presented here can allow improved comparison between the outputs of different experimental setups and can form an essential foundation for inter-technique comparisons, first and foremost with CFA.” This has not been proven. The authors themselves mention that day-to-day comparisons are not comparable, and as no calibration or concentration data is provided this remains conceptual.

This statement has been revised on L284

Line 264: “Furthermore, the simulation of a CFA signal allows a direct comparison of LA-ICP-MS and CFA signals which is only possible as this is a 3D model” This has not been shown in this paper as no comparison to experimental CFA data is provided to show these are comparable.

This statement has been revised on L294

Line 264: What does “This facilitates a direct comparison that is not currently possible for physical ice samples as the outer portion of ice measured using CFA is not measured to avoid contamination (Dallmayr et al., 2016).” Mean? I’m unclear how direct comparison is only possible with a 3D model here and why contamination control procedures impede this.

This has been re-addressed on L297 – 301