

Dear Referee 2,

We sincerely thank you for your careful, thoughtful, and highly constructive review. We fully agree with your suggestions and have incorporated all of them into the revised manuscript. All changes are marked in blue in the attached PDF.

Our responses to your twenty comments are given below, with *your comments in italics* and **our replies in blue**. Should anything remain unaddressed or require further clarification, please do not hesitate to let us know.

1. *The classification of minerals is described in just a single sentence clause on Lines 139-140: "[Our data processing scheme follows...] a stoichiometric analysis of co-detected elements within the [single particle] data to infer specific mineralogical groups." Given the major significance of mineralogy to the study, this simple description is relatively inadequate. Did the sp-ICP-TOFMS software perform the stoichiometric analysis on behalf of the authors? Or did the authors process this data separately using their own classification scheme? If the latter, then I highly recommend that the authors present their classification criteria, ideally in a table in the Methods section or in the supplementary materials. If the complexity of the classification criteria is too complicated to be summarized in a table, then the authors should include a detailed description of the methodology behind their stoichiometric approach and/or make available an Excel / Python / Matlab etc. file that can be used to reproduce the mineral classification workflow. Granted, more details on the authors' stoichiometric approach are provided later in the manuscript (for example, Lines 355-357 clarify that hornblende-like primary aluminosilicates are classified based on the co-detection of Al, Fe, Ca, and Si; and, Lines 390-391 clarify that alunite-like minerals are identified by 3:1 Al/K, Fe/K, and (Al+Fe)/K molar ratios), but I believe this information should also be presented in the Methods section.*

We agree that the mineralogical classification deserves a fuller description, given its central role in this study.

To clarify your question: the stoichiometric analysis was not performed automatically by the sp-ICP-TOFMS software. The classification was carried out by the authors, and we have now added a dedicated subsection (Section 2.2.4, lines 235–283) together with Table 3, which presents the representative chemical formula, the diagnostic elemental ratios, and the co-detected elements used to identify each mineral phase (ferro-hornblende, alunite, jarosite, and the alunite–jarosite mixture). The criteria previously given only in the Results (lines 355–357 and 390–391 of the original manuscript) are now consolidated in Table 3. We would also like to explain why our classification is based on a combination of criteria rather than stoichiometry alone. Several factors limit what stoichiometric ratios can resolve on their own: different minerals may share similar elemental ratios; elemental substitution introduces variability within a single mineral group; and single-particle detection by sp-ICP-TOFMS does not capture the complete elemental composition of a particle, as O, H, C, and S are not currently quantified and Si is determined only qualitatively. We therefore characterize mineralogical changes using the 22 quantifiable elements (Table B1) in combination with elemental particle number concentrations, particle size distributions from the Bin Count Analysis, the anomaly in particulate-to-dissolved elemental mass ratios, and mineralogical information reported for other ice cores and comparable environments.

We note that our analysis was not restricted to the mineral phases named above. We examined elemental ratios across all pairwise combinations of the 22 quantifiable elements, including non-target phases, and found that most element pairs displayed a pronounced dissolution signature. Because these signatures are also evident in the element-specific Bin Count Analysis and in the particulate-to-dissolved mass ratio anomalies, we present a selected set of representative minerals rather than the full set of elemental pair ratios, which would not add further information.

2. *Code and Data Availability* – The data sets provided by the authors for peer review are adequate. I was successfully able to reproduce most of the main figures using the provided files. What I think is missing is a corresponding code file(s) that may help future researchers apply the techniques used in this study to their own work. These would not need to be the exact files used by the authors in their analysis, but something that provides a framework for future researchers to build upon. For example, it may be beneficial to include a Python script or Excel file (or something similar) to guide readers through the processing of sp-ICP-TOFMS data using the bin count analysis (BCA) protocol that the authors introduced. I generally encourage authors to make code files available when they use or develop helpful techniques. It makes it easier for future researchers to copy the methods, and it enhances transparency with regard to data reproducibility.

We appreciate this suggestion. Although the BCA protocol is straightforward to implement, providing it as code should make it considerably easier for other researchers to adopt and build upon the approach, and we agree that this is worth doing.

We have therefore made our processing code publicly available on GitHub

(<https://github.com/Learner-2021/sp-icptof-bin-counts/tree/v1.0.0>), archived on Zenodo (<https://doi.org/10.5281/zenodo.22208733>) together with documentation on its use. The repository provides a working framework for processing sp-ICP-TOFMS data using the bin count analysis, which future users can adapt to their own datasets. The link has been added to the Code and Data Availability section (lines 676–677).

3. *The References section requires a lot of attention. There are issues with many citations, including formatting inconsistencies, repetitive hyperlinks, and misspellings. For example, many of the citations contain multiple DOIs, and most (none?) of the citations use journal abbreviations, which is a requirement according to the publication guidelines* (<https://www.the-cryosphere.net/submission.html#references>). *Each of the references in the bibliography needs to be assessed carefully before publication.*

We apologize for the inconsistencies in the reference list and thank you for pointing them out. We have carefully reviewed the entire bibliography and corrected the formatting inconsistencies, duplicated DOIs, and misspellings. Journal names are now abbreviated in accordance with the journal's submission guidelines. The revised reference list is given in lines 710–1033.

4. *Line 7* – *In the abstract, the authors write that the acid dissolution in the deep ice environment leaves behind “refractory mineral phases,” but the term “refractory mineral” is not used elsewhere in the manuscript. I would either change the wording in*

the abstract or edit the main manuscript in the relevant locations to include the term “refractory mineral” so that it does not only exist in the abstract.

We agree that the term "refractory mineral" was insufficiently defined and appeared only in the abstract. We have replaced it with "resistant", which we consider more appropriate in this context, and have ensured that the term is now used consistently in both the abstract and the main text.

Specifically, the revised Discussion (lines 470–473) describes the residual silicates and links their persistence to the relative increase in Si-bearing particle number concentrations shown in Fig. 1, explicitly framing these as dissolution-resistant minerals.

5. *The authors use both "ice-core" and "ice core" throughout the manuscript. Either spelling is acceptable, although for consistency only one spelling should be used. Throughout the manuscript, we now use “ice core” as a noun and the hyphenated “ice-core” as an adjective (e.g., “ice-core records”).*

6. *Lines 25-26 should include a citation(s) referencing the seminal works related to the discovery of the glacial-interglacial periodicity shift at the Mid-Pleistocene Transition (the MPT).*

We agree that these references should be included. We now cite Lisiecki and Raymo (2005) and Elderfield et al. (2012) for the shift in glacial–interglacial periodicity across the Mid-Pleistocene Transition, and Fischer et al. (2013) for the ice-core community's efforts to recover ice spanning this interval (lines 26–27).

7. *Table 1 – Consider adding an asterisk to the δD value for bag 1994 to direct the reader to the note at the bottom of the table. I understand that the authors probably omitted this in order to keep the decimal places aligned, but hopefully the editorial team will be able to fix any alignment issues during the proofing stage.*

We agree that this improves clarity, and an asterisk has been added to the δD value for Bag 1994 (Table 1). The decimal alignment has been preserved.

8. *Line 115 – I may be wrong, but since Teflon is a brand it should theoretically include a trademark symbol (Teflon™). Alternatively, the authors could simply write “PTFE” to describe the plastic. I would also recommend checking for other brands used throughout the manuscript to see if ™ or ® symbols are needed.*

We agree, and have replaced "Teflon" with "PTFE" (line 125). We have also checked the manuscript for other brand names and have adopted a consistent approach of not using trademark symbols: instrument and reagent names are given in the conventional format with the manufacturer and location indicated (e.g. the laminar flow bench and the ICP-TOFMS system), and we have accordingly removed the registered trademark symbol from "Certipur®, Supelco, Germany" (line 118). We would of course be happy to reinstate these symbols if the referee or editor prefers.

9. *Line 123 – There is a space between the end of the word "standard" and the comma that follows it.*

The extra space has been removed (line 136).

10. Line 140 – The abbreviation "SP" is not technically defined until the following line, and so the authors should forego the use of the abbreviation and instead write "single particle".
Corrected. We now write "single particle" in full at this point, before the abbreviation is defined (line 153).
11. Lines 143-145 – If the "a" term corresponds to a one-sided 95% confidence interval, shouldn't $a=1.645$ instead of 3.29? I would think that $a=3.29$ would correspond to the 99.95% confidence level (<https://zscorecalculator.net/blogs/complete-guide-to-z-scores-for-confidence-intervals>).
We agree with the referee that the z-score (k) corresponding to a one-sided 95% confidence interval is 1.645. We realize our initial text lacked sufficient detail, leading to this confusion. Please note that a is defined as $2k$ (3.29), which accounts for both false positives ($\alpha = 0.05$) and false negatives ($\beta = 0.05$) in the detection limit formulation. Furthermore, b is defined as k^2 (2.71), which is the necessary correction term to restrict the false negative probability under non-constant variance (Poisson) conditions. We have revised the text to explain these definitions more clearly to avoid any ambiguity (Lines 157–159).
12. Equations 1 and 2 (Lines 166 and 170) – I don't think the commas at the ends of the equations are necessary. Similarly, the periods and commas at the ends of Eqs. 3, 4, 7, 8, and R1.
The commas and periods following Eqs. 1-4, 7, 8, and R1 have been removed (lines 180, 184, 198, 200, 559, 569, 585).
13. Section 2.2.5 Bin count analysis (BCA) - Traditionally, when I think of "bins" in the context of particle analysis I think of bins as size ranges (for example, 1-2 microns). But, if I understand correctly, in this work the "bins" refer to the moments in time in which a signal is detected. I found this confusing at first and I had to read through the description of BCA a few times to understand the significance of a bin. I think this can be easily clarified to the reader by changing Lines 232-233 to say: "To circumvent this limitation, we introduce BCA as a relative size classification approach that utilizes the number of consecutive acquisition bins [(i.e., consecutive time slices)] over which individual particles are distributed." Or something similar that helps re-iterate to the reader what a bin actually represents.
We understand how the term "bin" could be confused with the size bins conventionally used in particle analysis. Following your suggestion, we have revised the sentence to read "consecutive acquisition time bins (i.e., consecutive time slices in the time series)" (line 289).
14. Line 283 - "hereafter" is a misspelling. It should be "hereafter" or "henceforth".
Corrected (line 297).
15. Figure 2 – There is a typographical error in the color bar label. It should read "Anomaly in particulate-to-dissolved elemental mass ratios", but it says "particular".

The typographical error in the color bar label has been corrected to "particulate-to-dissolved" (Fig. 2, page 18).

16. *Figure 4 – The tick marks on the color bar are scaled logarithmically, but the color gradient appears linear. If so, then the visualization is somewhat misleading. Even though the units for Number Density are arbitrary, I think the tick marks should be labeled to make the scaling of the data clear to the reader. I recommend normalizing the Number Density units so that they fall in the range [0,1] if that makes labeling the tick marks easier.*

We agree with the referee. The color bar had originally been adjusted to keep pixels of very low density visible, but we accept that the mismatch between the logarithmic tick marks and the linear color gradient was potentially misleading. Following your recommendation, we now show a normalized particle density, and the color scale and tick labels are consistent with one another. We have also removed the faintest colors that were barely distinguishable. The figure and its caption have been revised accordingly (Fig. 4, page 20).

17. *Figure 7 – This figure is first referenced on Line 486 and then again on Line 493, long before BELDC is ever mentioned or defined (that is, BELDC is not officially described until Lines 539-540). There needs to be at least some mention of BELDC before Fig. 7 is referenced. As it stands currently, the first mention of BELDC is in the caption for Fig. 7, but without any relevant context.*

We agree that referring to Fig. 7 before BELDC has been introduced could confuse the reader. We have therefore added a brief description of BELDC in the Introduction, where the abbreviation is now defined, so that it is familiar to the reader well before Fig. 7 is first referenced (lines 28–29).

18. *Line 558 – “Neoformation” is a very apt term for describing the genesis of minerals within the ice, and I think it is worth keeping. But, in the main text, the first (and only) use of “neoformed” occurs on Line 528 and “neoformation” does not appear until the conclusions section on Line 558. This is odd considering the importance of the word in context of the study’s scope, especially given that “neoformation” is used in the article’s title and in the abstract (“neoformation” also appears in the caption for Fig. 6 on page 22, but this is still very late in the manuscript). I would highly recommend introducing the term earlier and more frequently in the manuscript so that it does not appear so abruptly at the end.*

We agree that the term deserves to be introduced earlier and used more consistently. We have replaced "precipitation" and "formation" with "neoformation" wherever the latter conveys the intended meaning more precisely, so that the term now appears throughout the manuscript rather than only in the concluding sections (lines 42, 51, 52, 152, 427, 434, 438, 488, 538, 583, 602, 603).

19. *Lines 542 – 543: There is an extra right-parenthesis in the citation that should say: (Lecavalier et al., 2023; Fig. 7).*

The redundant parenthesis has been removed, and the sentence has been rephrased using an em dash for clarity (lines 620).

20. Line 550: I suggest writing out “particle number concentrations” because many readers will likely skim or skip straight to the Conclusions, and therefore they won’t know what PNC means. I recommend the same thing for the use of “SP” on Lines 558 and 584 in the Conclusions. Readers who skip to the conclusions may not intuitively know that “SP” stands for “single particle”.

We agree that readers who go directly to the Conclusions should not encounter undefined abbreviations. We have therefore written out "particle number concentrations" and "single particle" in full in the Conclusions (lines 652, 660).

On behalf of all authors,
Geunwoo Lee