SUMMARY

The authors use sp-ICP-TOFMS to analyze 18 ice core samples from EPICA Dome C (covering 10 climatic periods) to gain insights into the englacial geochemical processes that occur as a function of depth and temperature. Their sp-ICP-TOFMS results document notable differences in the composition of impurities in the deepest sections of the ice core. That is, the upper layers of the ice core contain material that is compositionally consistent with a crustal origin and aeolian deposition whereas the deepest ice layers contain material that is best explained by post-depositional changes. They present evidence of the dissolution of Al- and K-bearing primary minerals and the neoformation of alunite supergroup compounds (such as jarosite) and possible Fe-(oxyhydr)oxide coatings which act as cementing agents that lead to the agglomeration of particle clusters. The in situ production of alunite-like compounds in localized acidic micro-environments is further supported by an increase in the number of I-, As-, and Pb-bearing particles, as these chemical species are known to adsorb to (or become incorporated within) alunite/jarosite and related secondary minerals. These results demonstrate the importance of considering englacial geochemical processes when studying deep ice core records, and, by extension, very old ice.

I believe the authors do an excellent job summarizing the scope of the study and its key findings in the Abstract. Likewise, the structure of the manuscript is logically organized and the content of the article is generally very well-written. The discussions are thorough, the limitations of the study are not omitted, and the authors' main conclusions are supported by the evidence presented in the manuscript. I believe this study provides novel and scientifically significant insights into the geochemical environments of deep ice (which is an important topic right now within the ice core community), and as such I think the manuscript will be suitable for rapid publication following a few modifications.

SPECIFIC COMMENTS

I have three "major" comments (perhaps "moderate" is a better word in this case because they are not severe criticisms) that I would like the authors to address before proceeding.

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.

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.

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.

GENERAL COMMENTS

I also have several minor comments (some might even be considered trivial) that I think will be helpful to the authors to improve the quality and readability of the manuscript.

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.

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.

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).

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.

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.

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

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".

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).

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.

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.

14. Line 283 - "hearafter" is a misspelling. It should be "hereafter" or "henceforth".

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".

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.

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.

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.

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

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”.

FINAL THOUGHTS

Lastly, I would like to congratulate the authors on their study. This is a very solid manuscript that I believe will be beneficial to the glaciological and ice core communities.