R1: I have read this manuscript with great interest, particularly the geochemical analysis using elemental data from sp-ICP-TOFMS, which complements the authors' previous research. I fully agree that ice core samples should be analysed using multiple techniques to extract climatic signals from polar ice from different perspectives. Since no single method can capture the complete climatic record stored in ice, this study serves as an excellent example of utilizing multiple analytical approaches for dust analysis in the same Greenland ice cubes. This comprehensive investigation can provide valuable multi-dimensional insights into Earth's climate variability.
However, upon closely examining the sp-ICP-TOFMS measurement results, I was highly concerned about the data quality. This raises substantial concerns regarding potential contamination of the ice cube samples used in this study. Below, I outline the reasons for my concerns regarding data quality related to sample contamination. Until the authors address these contamination issues clearly, I am unable to provide a complete review of this manuscript.

A: We thank the reviewer for accepting this review and are grateful for the comments. However, we respectfully disagree with the reviewer, in particular regarding the data quality and decontamination. We can address the comments as we outline below. In doing so, we hope to improve the manuscript´s quality and avoid misunderstandings. Below, we will reply in detail to the raised issues. We quote the reviewer here to emphasise the objectives of this study:" I fully agree that ice core samples should be analysed using multiple techniques to extract climatic signals from polar ice from different perspectives. Since no single method can capture the complete climatic record stored in ice, this study serves as an excellent example of utilising multiple analytical approaches for dust analysis in the same Greenland ice cubes." In summary, we aim to display a multi-method approach to analyse ice samples using several methods; only the final stage contains single particle analysis, but we demonstrate here that this is possible at no expense of the data quality. This objective should be considered when discussing the data. We do not aim to produce a stand-alone data set comparable to a CFA record but to compile complementary data from sophisticated analyses on one sample set. For this purpose, ice core samples dedicated to CFA analysis cannot be used because they are destroyed by the melting of the samples. Thus, other approaches, including using different core parts, must be explored - especially for analysing precious, highly thinned layers of million-year-old ice.

R1: **Potential contamination issues**
The methodology employed in this study is novel and has not been previously established. Given that trace metal analysis of polar ice cores requires extraordinary precautions to prevent contamination, additional examination is necessary. The authors used the "physical property" sections of the EGRIP ice core—these outermost sections are typically used for physical property measurements, where contamination from external materials (e.g., drilling fluid) is less critical. However, for trace metal analysis, contamination risk must be minimized.
The manuscript describes a decontamination process, but given the dimensions of the ice cube samples (1 cm × 1 cm × 1 cm, 1 mL of ice), I am concerned that this procedure is insufficient to effectively eliminate contamination. Established decontamination protocols for ice cores retrieved from fluid-filled boreholes require an initial acetone rinse to remove drilling fluid, followed by ultrapure water rinsing until 20–60% of the original ice volume has melted (Boutron and Batifol, 1985; Delmonte et al., 2002; Gaspari et al., 2006). The authors should provide a more detailed description of their decontamination procedure and critically assess its suitability for trace element analysis in polar ice cores.

A: We are unsure which methodology is addressed here: decontamination or single particle analysis. In particular regarding the second issue, we respectfully disagree. The decontamination of discrete samples with MQ water (e.g., Delmonte et al. 2004) under an air flow bench using gloves is an established technique, e.g., in dust particle research using the coulter counter. We further applied all the knowledge and experience of our co-authors, analytical chemists working on nano- and single particles, who are well-trained in ultra-trace analysis, in the lab. The used samples are always from the inward-facing side of the physical properties (PP) piece at its thickest location and thus never had contact with the outside of the core or the drilling liquid. For the previously conducted studies on these samples, it was essential to use intact samples containing no micro-cracks, which was ensured by microstructural analyses utilising Large Area Scanning Macroscopy and Microstructure Mapping (Stoll et al., 2021, 2022, 2023). This makes it highly unlikely that drill liquid has reached our samples. If this is assumed, the adjacent surface of the CFA and gas piece would also be contaminated (see figure below). Initial sample decontamination included several steps of microtoming for

Raman spectroscopy and later decontamination with ceramic knives for laser ablation measurements. Thoroughly rinsing of the sample with MQ water from all sides and melting the outer layers was the final step. Thus, the initial ice sample volumes were melted to at least 60%, which is in agreement or higher with the studies mentioned by the reviewer. It should further be acknowledged that drill liquid quality has improved over the last decades, and the EGRIP driller used state-of-the-art drill fluid (M. Hüther, pers. communication 03.03.25). Additionally, the analyses conducted before the SP-ICP-MS analysis (cited in the manuscript) did not show unexpectedly high values of any element analysed, supporting that no initial contamination took place or occurred due to sample storage and handling.

We suggest to provide a more detailed description of the decontamination method in the revised version. We would add additional text regarding the potential risks of the chosen samples, highlighting that using other ice core pieces, such as CFA, would be preferable. Unfortunately, this remains unlikely due to the limited availability of samples, but it could spark meaningful discussions and collaborations in the community for future studies.

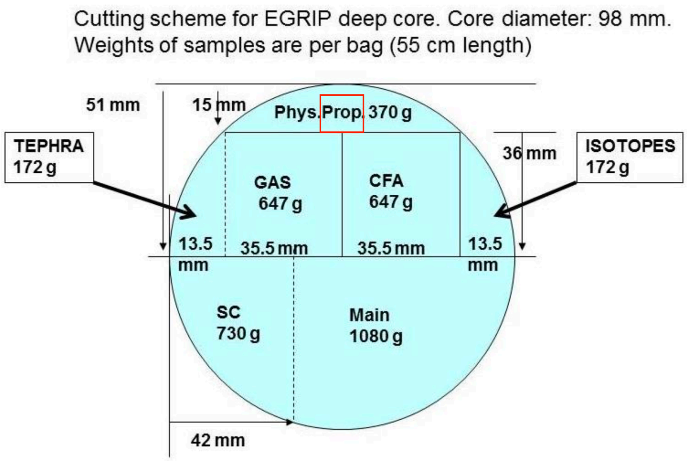

Figure 1: Cutting scheme of the EGRIP ice core. The red box displays the approximate position of the used samples overlapping with CFA and Gas pieces.

R1: Concerns about sample cleanliness become even more pronounced when comparing the ionic Fe background thresholds in Table A1 with total Fe levels reported in two well-aligned Greenland ice core studies: North Greenland Eemian Ice Drilling (NEEM) and EGRIP (Burger et al., 2021; Erhardt et al., 2019).

Typically, total Fe concentrations (including both particulate and dissolved Fe) should be significantly higher than ionic Fe background thresholds. However, the reported values in this study appear unexpectedly high.

First, the unit for ionic Fe background thresholds is missing from Table A1. Assuming it is in ppb (ng/g), which is a common convention, the lowest and highest Fe ionic backgrounds in this study are 9.803 ppb and 424.5 ppb for the cold climate samples G9 and YD3, respectively. These values are alarmingly high compared to the total dissolvable Fe concentrations of 2.9–146.4 ppb reported by Burger et al. (2021) after a month-long acidification process. Despite differences in sampling locations within Greenland, such high background thresholds may suggest contamination.

Furthermore, dissolved Fe background levels in a 3-m EGRIP ice core from the Holocene typically range from sub-ppb to 6 ppb, with non-dusty sections near 0 ppb and dusty sections around 2–3 ppb (Erhardt et al. 2019). The authors attribute the discrepancy between these two studies to methodological differences and different climate periods (line 303), but given that both Holocene EGRIP ice core samples exhibit similar dust particle concentrations (~1000 particles/mL, as measured by a laser particle counter (Abakus)), the observed Fe background levels in this study remain questionable.

A: This comment is based on a misunderstanding, which we hope to clarify here. The mentioned thresholds are applied to make quantitative sample comparison possible, which is a big challenge in single particle analysis of samples with vastly different concentrations, as found in, e.g., Holocene and Younger Dryas ice. Thresholds are defined based on a statistical analysis of the data set, in which the mean value of the data set is considered and compound Poisson statistics are applied. These statistics are used to determine an alpha value of 10^-6 (meaning a false positive detection rate of 1 in a million detection events), which provides the “threshold” over which a detection event is considered a particle signals. As such, the threshold is a statistically derived parameter for pinpointing particulate signals and does not provide quantifiable particle parameters besides the size detection limit. This threshold only influenced by the sample mean value and therefore, when comparing data sets with different ionic Fe levels, thresholds change. This results in different size detection limits which complicates the comparison of numbers and sizes of found particles across a sample set. To enable a consistent comparison, we used a conservative measure, in which the highest threshold in a sample set was used across all other samples to enable a comparison. More detailed information on this procedure can be found in the cited references. In summary, the thresholds do not reflect mean Fe concentrations. However, we would like to point out that this study specifically chose dust-rich samples based on CFA and visual stratigraphy data (Stoll et al., 2021, 2022, 2023), it is thus expected that these samples have comparably high concentrations.

The comparison to the Fe values obtained by Burgay et al. (2021) is inept. As displayed in section S1 and table S2 (Burgay et al., 2021), the authors analyse four small parts of a very shallow (100.8 m) and, thus, very young (maximum age: 1363 CE, oldest analysed sample 1611-1596 CE) core. Derived data is thus not comparable to our samples from the Younger Dryas or glacial often containing cloudy bands and thus a much higher insoluble particle content. On the opposite, this strengthen the motivation of our study as we explicitly mention that data is challenging to interpret as so little data from deep ice cores is available resulting in the need for more single particle studies on ice.

We suggest enhancing the clarity of the text in a revised version to avoid further misunderstandings regarding the applied thresholds. We will make sure to clarify that the applied thresholds are intensity values (in counts) and not concentrations.

Below is a figure displaying the thresholds and the ultra-pure blanks measured in the same analysis runs. The black line indicates the compound Poisson threshold (critical limit for a=10^-6) for the respective element in the blank. The dashed blue line indicates the highest threshold across all samples (taken from tables A1 and A2), which we used for all data evaluation.

Both combined display that our sample handling, ultra-pure water and the instrument itself do not create any particle artefacts for the chosen thresholds, which are comfortably high. There are zero data points above the dashed line, meaning we virtually do not expect any false positive events from the "instrument/chemical" blank.

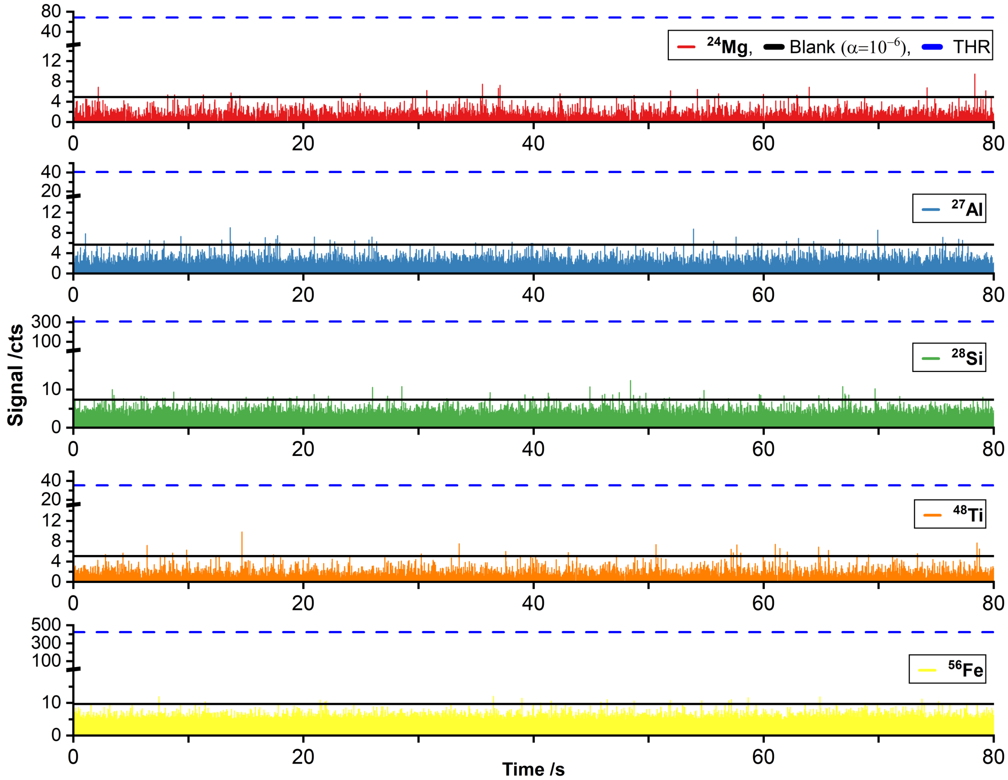

Figure 2: Comparison of ultra-pure blanks (black) and chosen thresholds (blue dashed line). No contamination is visible.

In addition to the high Fe background levels, the inconsistency in particle counts and sizes for the five major elements in Figure 5 further suggests a high probability of contamination. Since Mg, Al, Si, Ti, and Fe primarily originate from mineral dust, their dissolved and insoluble concentrations should be positively correlated unless there is a specific event providing an element only without the others. For instance, if a sample exhibits a high dissolved Fe background, a correspondingly high Fe particle concentration would be expected. Similarly, increased dust content should be reflected in Al and Mg concentrations due to their co-occurrence in mineral dust, and vice versa. However, the observed discrepancies in detection trends—for example, high Al but low Fe detections in sample H1 compared to samples G5 and G7—suggest inconsistencies in the data that are difficult to be produced with natural sources alone.

A: The difference in particle counts and sizes displayed in Fig. 5 is due to the applied thresholds using the highest respective thresholds to enable the quantitative comparison between samples with very different particle concentrations. For example, the high threshold for ^{56}Fe derived from YD3 (Table A1) is applied to all other samples (while the ^{27}Al threshold is much lower) thus explaining the difference in the number counts in Fig. 5 (see also previous comment).

The use of physical property sections—directly exposed to drilling fluid, uncleaned processing and packaging materials—introduces a high risk of contamination, making these samples unsuitable for trace-level analyses without strict decontamination procedure. Additionally, given that these samples have been handled in different environments over the years for multiple analyses, the risk of contamination is further increased. Due to their small size, effectively decontaminating 1 cm^3 ice samples for trace metal analysis is particularly challenging.

A: As explained above, we disagree with these statements. The used samples were never directly exposed to drilling fluid. Further, the processing and packaging in the field procedures were the same for all EGRIP ice core pieces. We can assure you this as the lead author has been involved in four EGRIP field seasons and leading the science trench twice. We conducted strict decontamination procedures at the University of Graz under the supervision of established analytical chemists. Handling and decontaminating small ice samples is challenging but was successfully done, as displayed by the ultra-pure blanks, which we handled likewise. In fact, it should be considered an important result of this work that such sequential multi-method studies are possible, even when highly-sophisticated analyses are involved. In addition, spICP-TOFMS analysis works well for small sample volumes of less than 500 µL while enabling analysis of all chemical elements. We will extend the text to stress this point. At the same time, we see greater risk for introducing biases to the SP-ICP-MS results if samples are stored in vials for prolonged periods, refrozen, or exposed to even mild acids. Based on or our approach we have successfully avoided all of these potential pitfalls.

This proof-of-concept study displays the idea of several cascading measurements enabling as much data as possible from the same sample. In future studies, starting with samples of larger volume would be beneficial to make decontamination easier. However, this has the drawback that analyses take longer to achieve representative results, especially for time-consuming analyses such as confocal cryo-Raman spectroscopy, which must be focused on every single particle inside the ice sample. The recent progress in analytical techniques and multi-method analyses of ice core samples and the needed strengthening of a holistic microstructural and geochemical analysis evokes rethinking and updating historically grown ice core sample routines, which might not apply to the same degree any more.

Conclusion

The study presents a good case for multi-technique ice core analysis. However, the potential contamination issues outlined above need to be thoroughly addressed. The authors should provide clearer details on their decontamination process, re-evaluate its effectiveness, and discuss how contamination might have influenced their results. Without these clarifications, the reliability of the data remains uncertain, preventing further evaluation of the manuscript.

A: We thank you again for this review and hope we could clarify the addressed points of criticism. We are happy to include more details on the decontamination, the specific origin of the sample, and previous contamination steps. We will further discuss the advantages and disadvantages of the used samples. These clarifications will help focusing on the main objectives of this study, the presentation of a novel multi-method approach and the merit of single particle analyses on ice cores for a vast field of research fields.

References

Delmonte, B., I. Basile-Doelsch, J-R. Petit, V. Maggi, Marie Revel-Rolland, A. Michard, E. Jagoutz, and F. Grousset. "Comparing the Epica and Vostok dust records during the last 220,000 years: stratigraphical correlation and provenance in glacial periods." *Earth-Science Reviews* 66, no. 1-2 (2004): 63-87.