Response to editor decision received 06/01/2025

Dear Nanna B. Karlsson

My co-authors and I thank you again for your communication. We have addressed the comments from referee #1, with our responses detailed below.

I have uploaded a new version of the manuscript, alongside a version with changes highlighted, and an updated version of the supplementary material.

We look forward to the next steps for the manuscript

Kind regards

Piers Larkman

Response to Reviewer #1 – reviewer text in black, response in blue line numbers refer to lines in the manuscript version with markup

Thank you for the revised manuscript and responses to the comments. The manuscript has been improved well. I agree with the addition of Section 4.3.3 (Calibrated signals). Calibration of experimental data is important for the practical application. However, I suggest some modifications (clarification) to the description in Section 4.3.3, Table 3 and S3 to make it easier for the reader to understand. Once these minor revisions are made, I would recommend it for acceptance.

Thank you for your comments, we agree that the revised manuscript was improved based on the previous round of review. We also agree that section 4.3.3, which discusses some calibrated data, was an important addition prompted by comments made by both first-round reviewers. In the context of improving this section, the reviewer's comments are addressed below in blue, alongside the review text.

Questions and suggestions for improvement

Please provide a short overview of the calibration in the present manuscript, even if it was detailed in Bohleber et al. (2024).

L371- 376 of the revised manuscript contains a short description of the calibration process implemented by Bohleber et al. (2024), which utilises small droplets with known elemental concentrations to produce a calibration curve which is subsequently applied to calibrate LA-ICP-MS maps.

L379:
What does "The main effect of the calibration is to reduce the distance between the grain interior and boundary distributions" mean? I can understand this reduces the variability magnitude between interior and boundary, and signal MADs. Why is it necessary to reduce the distance between the grain interior and boundary distributions?

The interpretation presented here is the intended interpretation. The revised manuscript removes the use of the word 'distance' and states on L384-385 that "The main effect of the calibration is to reduce the magnitude of variability between the grain interior and boundary distributions"

Table 3:
I think the explanation of Table 3 (caption and mention in the main text) is not enough. I did not understand the meaning of Table 3. Are the values in the table results from a specific samples or an overall average? Please explain how these values were estimated and what the values claim (differences in values depending on methods, and between LGP and Holocene).

The following points relating to the table have been clarified, with additions made to both the caption the written text. L396-397 states that the results are from specific samples and provide

only snapshots. The figure caption specifies the methods used by Bohleber et al. (2024) to arrive at the 2D LA-ICP-MS map concentrations, by averaging over pixels in the measured maps, and bulk measurements, through use of discrete ICP-time-of-flight-MS measurements. The original text on L400-404 notes that difference in values between methods could be due to a number of factors (including 2D maps not fully representing bulk impurity content, spatial offsets in measurements, and grain size variability within a sample) and illustrates the need for more research to unify impurity measurements across scales and dimensions. Discussing the relevance of measured difference in concentrations between the Holocene and LGP samples is beyond the scope of this study.

S3 Calibrated data results:
It would be useful to place the calibrated and uncalibrated data side by side to understand the effect of calibration. With the present manuscript, it is difficult to know how calibration will change the result.

We agree that comparison between the two sets of results – calibrated and uncalibrated – is important. Therefore figures S15 and S20 (a) have been updated to include the non-calibrated intensity scale alongside the calibrated data.