

Comments and responses - Round 2

Reviewer

C: The authors made several improvements to the text which clarify the methods and results. The authors also corrected the names of several peptides, although errors remain in the images of Figure 2 and 4, as well as in the supplementary materials.

R: We thank the reviewer for pointing out the errors. Changes have been made in Figures 2, 4, and 5 (in the main text) and Figure 1 (in the supplementary material).

C: Researchers often face limitations in both availability of material and the quality of data that can be recovered from historical and archaeological materials and so I agree with the authors' sentiment that it is important to have tools which can provide as much information as possible with the data that is available. I am still concerned, however, with the utility of the index when more than 50% of the data fall outside the theoretical range for the index. The authors explain this as a problem of accurate baseline correction, but I think it would be worth elaborating on this point in the discussion as to why this would/would not unduly impact the interpretation of the results.

R: Our model generates some PQI values greater than 1, which we attribute to the problem of accurate baseline correction and noise. We note that replicates generally produce concordant deamidation estimates, and different peptides from the same sample generally produce highly correlated PQI values. It is worth noting that (Wilson et al., 2012) reported a similar problem and used truncation to solve for deamidation fractions > 1 . Ultimately these MALDI (MS1) based estimates of PQI will have to be ground-truthed by direct measurements of parchment deamidation during liming fully characterised by MS2. This is, outside the scope of the present study.

We have made the necessary additions in the Discussion section.

References

Wilson, J., van Doorn, N. L., & Collins, M. J. (2012). Assessing the extent of bone

degradation using glutamine deamidation in collagen. *Analytical Chemistry*, 84(21), 9041–9048.