

The manuscript describes a method to calculate the level of glutamine deamidation for several peptides by modelling the convoluted isotope distributions and combine the results to give an overall estimate of the deamidation for a parchment sample. The method is described as a simple three-step workflow, where the modelling occurs in the second and the third steps. The modelling of individual peptides uses weighted least squares regression which seems sensible for the limited data although the weights are calculated according to the noise level estimated from at most 3 observations. The details of the regression in this section somehow suggest more data than is available, possibly due to the matrix  $X$  (line 208). It could be worth linking back to the fact that  $n$  is at most 3 (sometimes just 1), and (according to Figure 2)  $m$  is at most 6 (should this be 5?). The notation is confusing as  $x_{ij}$  suggests the element in the  $i$ th row of the  $j$ th column in the matrix  $X$ , but in fact this is used to denote the model for the  $i$ th isotopic peak in the  $j$ th replicate. Perhaps the latter could be denoted  $y_{ij}$ . Also  $\gamma$  is of length  $k$  rather than  $k+1$ .

The third step of the process uses a mixed effects model to combine the individual deamidation estimates and the mathematical details are provided. My worry is that, for all the statistical theory, the results may not be meaningful if the data are poor. The authors perform residual analysis and apart from a few badly fitted values, seem to have obtained good results. However, these results check the fit of the model to the peaks extracted from (as the title advertises) low quality data. This depends entirely on the first step in the workflow which involves pre-processing the MALDI spectra and the only way to assess the performance here is from Figure 2. Of the 5 randomly chosen spectra, 4 appear to show only noise in all but the first peptide in Figure 2A. Maybe the much greater intensities for sample 67\_19\_2 make the other spectra look worse than they are. In any case, apart from this sample, Figure 2B shows just how few peaks were identified (assuming that the solid circles indicate identified peaks) and it is difficult to see why the peaks in other samples were identified. In the case of 59\_1\_4\_2, the intensities are just too low to see anything yet peaks were identified for several peptides. Maybe the weighted (normalised) spectra should be shown instead? However, even for sample 67\_19\_2, the pre-processing for the last peptide does not look convincing. Are these typical spectra?