12 June 2023

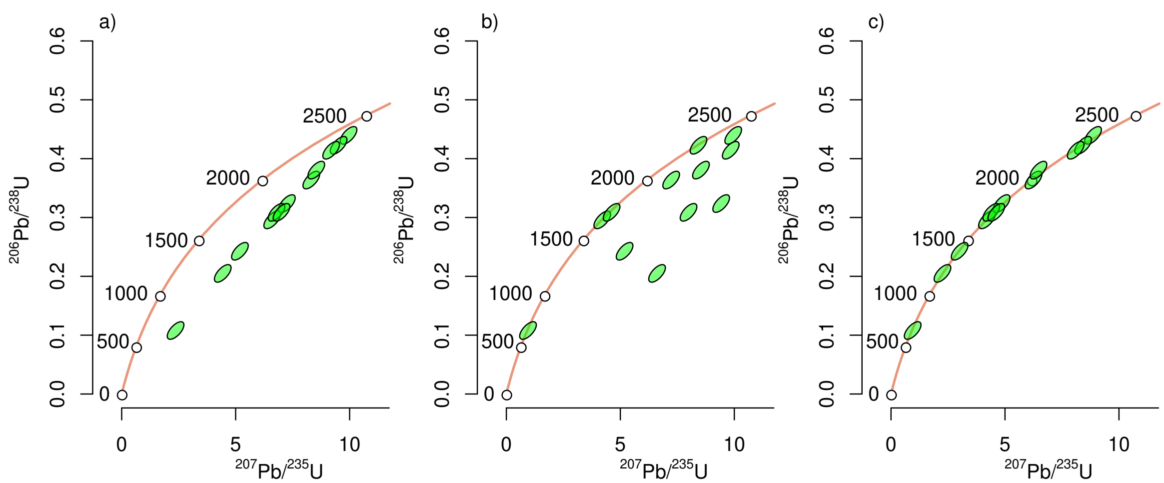
Dear Dr. Sharman and Dr. Malkowski,

I would like to thank you for submitting your work to *Geochronology*. Your manuscript entitled "Modeling apparent Pb loss in zircon U-Pb geochronology" discusses an important subject that is appropriate for the journal. Your proposed solution (deconvolution of crystallisation and Pb-loss distributions) holds promise. However, the reviewers have identified some fundamental issues with this solution, which make the paper unsuitable for publication in its present form. Your response to the reviewers addresses some of their concerns, but not all of them.

Reviewer 1 notes that your method uses dates, not atoms. It (intentionally?) ignores the physics of Pb-loss. Your paper does not include a single concordia diagram, reflecting the fact that these are not part of the algorithm. Reviewer 1 points out that the ^{238}U-^{206}Pb and ^{235}U-^{207}Pb systems respond differently to Pb loss. Your response to this comment comprises two parts. First, you propose to apply the deconvolution algorithm to the ^{206}Pb/^{238}U ratios instead of the ages. Second, you argue that ^{207}Pb/^{235}U ratios should be ignored because they cannot be measured with the same level of precision as the ^{206}Pb/^{238}U ratios.

Reviewer 2 raises an important concern, which you did not address in your response: "numerous processes [...] can lead to the same distribution".

The implications of both comments on your algorithm can be illustrated with the following three examples:



These three synthetic samples have a very different geological significance, but are characterised by identical ^{206}Pb/^{238}U ratio (and, hence, date) distributions. Plugging them into your deconvolution algorithm will produce three identical solutions.

The example in panel b) could be dismissed as a manifestation of the "garbage in, garbage out" phenomenon. However, the example of panel c) is more troubling. Subjecting this concordant dataset (containing a detrital or xenocrystic age signature, say) to the deconvolution algorithm will produce a mixture of two meaningless distributions ("good data in, garbage out").

As a basic sanity check, one would expect that, in the absence of Pb-loss, the deconvolution algorithm should yield the raw age distribution. Your method does not pass this sanity check. It would be dangerous to release such an algorithm into the wild.

Another aspect of Reviewer 2's comment is that the solutions produced by your deconvolution algorithm are *non-unique*. The paper claims that the Weibull distribution best describes the Pb-loss distribution, but does not offer any explanation why this is the case. Your algorithm tries 11 distributions. Why stop there? The space of probability distributions is infinite. For each dataset, there is a frequency distribution that completely describe it. In fact, there are infinitely many of them. In its present form, there does not appear to be any mechanism in your algorithm to constrain the functional form of the proposal distributions, or the number of parameters needed to describe them. This lack of theoretical justification further reduces the scientific interpretability of the algorithm's output.

In light of these issues, I am unfortunately unable to accept your paper for publication in *Geochronology*. However, should you find a way to address the above concerns, then I would be happy to reconsider a suitably revised version of the paper in the future.

Please do not hesitate to contact me directly should you have any questions about this decision.

With kind regards,

Pieter Vermeesch