Response to Reviewer 2 (Karl Lang) Comment on gchron-2021-22

We thank Karl Lang for his comments that help us to clarify issues concerning differences between the LA-ICPMS and EDM AFT analysis methods. It is not our intent to suggest that the LA-ICPMS AFT method is superior to the EDM method nor to promote the idea that people should stop using EDM in favour of LA-ICPMS. Indeed, we have plenty of quality EDM data for multikinetic AFT samples and AFTINV can accommodate either type of data. That said, it is equally important to note that the opposite is also true, that the EDM method is by no means superior to the LA-ICPMS approach that we chose to utilize. Based on the many thousands of analyses that our primary analyst has generated by both LA-ICPMS and EDM, it is clear that a properly-trained analyst can produce data of equal quality using either method.

Reviewer 2: Karl Lang

Use of “detrital” was a little confusing to me at first, since many applications of detrital thermochronology are now also focused on interpreting cooling histories of source rocks prior to deposition, and not simply the common cooling history of detrital minerals in a sedimentary rock after deposition. This is a semantic difference, but perhaps adding a sentence to state this explicitly at the beginning of the manuscript might clear up any confusion amongst readers.

We use detrital in the normal geological sense of “pertaining to particles of rock derived from the mechanical breakdown of preexisting rocks by weathering and erosion.” We avoid the term, detrital thermochronology, because of the connotation it carries. From the title it should be evident that we are talking about sedimentary basin thermal histories. However, we could change the first sentence in the Abstract to read “…cause for AFT age dispersion in sedimentary samples” rather than “…cause for age dispersion in detrital AFT samples.” Elsewhere in the text, detrital is used to describe the nature of the grain and so this should be clearly understood. If the confusion is related to the use of “detrital sample” then this could be replaced with sedimentary sample, for example.

Why does the manuscript include a vigorous preference of LA ICPMS over EDM approach? This seems unrelated to the central motivation of the paper and, in my opinion, is largely unsupported (see comments by line). The authors should explain why they chose to use LA-ICPMS instead of EDM, but they should avoid generalized claims about the relative efficacy of one method over the other (e.g. “The LA-ICP-MS method has some distinct advantages compared with EDM” [117]).

The manuscript does not include a “vigorous preference” of one method of data generation over another. Rather, the comments within the manuscript made in favour of the LA-ICPMS method used here provide exactly the requested “explanation” of why we chose to use LA-ICPMS instead of EDM. EDM is well established and has been around for many decades and its “perceived” advantages are well documented. For instance, as long as they had access to a research reactor and funding to pay for irradiations (heavily subsidized for academic labs), a trained analyst using EDM could create a fission track lab with not much more than an optical microscope and an automated stage. In contrast, the LA-ICPMS method is relatively new and requires access to more expensive and sophisticated equipment than EDM (not including the access to a research reactor). As a result: 1) only a limited number of labs have incorporated this newer approach, which directly measures parent U instead of using a proxy for U, and 2) the LA-ICPMS method is less understood within the thermochronology community.

This lack of familiarity with the LA-ICPMS AFT method has led some to question if the LA-ICPMS method provides data of the same quality (e.g., see community comment on paper). Comparisons of EDM and LA-ICPMS AFT data have shown that the latter method may yield data with more age dispersion. In some cases, χ^2 statistics and age dispersion have been interpreted as data quality indicators to infer that there are analytical problems with LA-ICPMS data such as U zoning. Certainly zoning exists in nature but rigorous and consistent analytical practices can mitigate problems with zoning. If there are differences between EDM and LA-ICPMS AFT data, the tendency is to automatically look for problems in the latter data. However, there are multiple causes of age dispersion and these statistics must be used with caution because it can be misleading to infer data quality on that basis. There are other issues that may contribute to differences between EDM data such as grain selection and counting bias or differences in the precision of U values used for age calculation. High age dispersion is the norm for our multikinetic samples yet age dispersion is significantly reduced when grains are binned into different kinetic populations using elemental data.

In closing, we contend that both methods can yield data of similar quality but they are different methods with different advantages and disadvantages. In the paper, we are just pointing out some relative advantages of the LA-ICPMS method compared to EDM that are pertinent to this particular discussion. This doesn't preclude that EDM has other advantages relative to LA-ICPMS.

118. It has not been my experience that analytical costs are lower for LA-ICPMS than for the EDM when measured on a per grain basis. If you can measure 100 grains per mount and 50 mounts fit in a $1000 irradiation package, that's $5/grain. By comparison, LaserChron (probably cheapest option in US, at least) charges $9-16 per grain for 100 grain samples, not including costs for CL imaging. Also, throughput is not necessarily higher for LA if you have to wait several months for lab time to become available. In my experience the analytical time to produce a complete fission-track dataset is comparable regardless of the analytical approach. I worry that comments like this will gradually discourage scientists from using the EDM, which is a well established and data-rich method.

In terms of the wording within the manuscript, we offer our thanks for pointing this out. Lab costs are highly variable and depend on many factors, some of which are discussed in our reply to reviewer 1. We can say in our situation that overall costs were lower when we switched from using EDM to LA-ICPMS. However, that may not be the case for others so we will omit reference to costs. We maintain that AFT analysis is faster for the LA-ICPMS method versus EDM under normal conditions for the reasons given in the paper. Of course, there will be delays if equipment needs servicing and lab schedules are backed up but the same could be said for EDM if there are issues accessing a reactor and shipping samples. It is a safe bet that access to facilities would have been taken care of prior to any lab instigating the use of the LA-ICPMS approach; either by purchasing the necessary equipment (expensive), or having reached an agreement for access with an existing LA-ICPMS facility. It is an equally safe bet that any lab using the EDM method would have taken care of any questions surrounding access to irradiations.

The review comment above seems to be based on the costing related to the academic pricing for a ZrnUPb DZ analysis provided by LaserChron instead of a direct comparison to LA-ICPMS analysis for AFT (i.e., AFT does not require additional costs for CL imaging). If correct, and if LaserChron does charge between $900-$1,600 for 100 spots ($9-$16/spot), then this is not the cheapest option in the US as the price we were charged for the use of a laser ablation and mass spec system to complete the AFT grain-age analyses presented in this manuscript was on the order of $2.12/grain. Even if the values presented by the reviewer were pertinent to an AFT analysis using the LA-ICPMS approach, the quoted cost of $5/grain would seem to be incorrect. Fifty mounts of 100 grains each (5,000 grains) should work out to $1,000/(50x100grains) = $0.2/grain. The subsidized $1,000/irradiation for 50 mounts makes it more affordable for users but this does not reflect the true cost of the service. Considering that most AFT labs provide analyses with up to 25 rather than 100 grains per mount, an analyst would produce data from a total of 1,250 age grains per $1,000 spent for each irradiation – at a cost of $0.8/grain or 4x the reviewer's estimate. It is evident that when looking at just analytical costs on a per-grain basis, subsidized irradiations are cheaper. However, simply not having to deal with all the rules and regulations associated with holding a radiation licence more than makes up the difference when evaluating the costs.

137-138. Wouldn't observer bias have a greater impact on age determination when it is only accounted for in spontaneous track counting? It seems to me that observer bias may actually be reduced when it is accounted for in both the spontaneous and induced track counts, rather than just in spontaneous counts. Either way, I don't consider it fair to say that LA-ICPMS is "more objective" if it still relies on user interpretation and collection of spontaneous track data.

Unconscious bias is an established phenomenon in cognitive research. The extent to which it may influence the selection and counting of grains in AFT analysis is unknown, but its existence has been established previously. We believe that it is valid to point out this fact and that it may have an influence on grain selection, especially if only a subset of potential age grains are counted. Under these circumstances, the age data may be of good quality but it may not be capturing the full range of compositional variation within a sample, which is the most important issue for multikinetic samples.

Unlike the LA-ICPMS FT method, all the information for U content and age determination is available at the time of measurement using EDM. The counting of spontaneous and induced tracks relies on the judgement and experience of the analyst to select which areas to count to yield reliable age data. An advantage of this approach is that areas with significant U zonation can be avoided. We accept that good data are obtained for the grains that are counted. However, Ns and Ni data are accumulating as tracks are being counted and the human brain is well adapted to pattern recognition. If multikinetic populations are present but cannot be resolved using the conventional kinetic parameters, Dpar and Cl, then there is not a strong incentive to count a large number of grains and add to the sample age dispersion. In this context, a dominant population may be counted with other grains viewed as outliers.

For the LA-ICPMS method, a measurement area is selected and spontaneous tracks are counted first. Subsequently, laser spot analysis within the predetermined track count area yields a U/Ca ratio for age determination. For our multikinetic samples, we tried to avoid grain selection bias by counting a broad range of grains including lower quality, more difficult to analyse smaller grains. From a multikinetic perspective, it is desirable to count enough grains to sample different age populations if they are present. We believe that EDM and the LA-ICPMS method will yield similar results for multikinetic samples if a large and diverse range of grains are analysed. Conversely, if too few grains are counted, then results will be more sensitive to the grain selection process and both methods may yield apparently different results.

140. This is not an inherent limitation of the EDM, simply a choice by the operator to count fewer grains. Many detrital studies regularly count more than 100 grains per sample with the EDM.

Yes. For thermal history studies involving sedimentary samples, we recommend that a more diverse and larger number of age grains be counted than is typically done for EDM, increasing the probability of higher age dispersion and χ^2 failures. If the age dispersion is related to compositionally-controlled multikinetic annealing then more information will be obtained from a sample. We believe that EDM and LA-ICPMS will yield similar results for multikinetic samples if the goal is to count a large enough number of grains to represent the true age and compositional variation within a sample.

142-143. It is convenient to make this argument here, but one could also make an alternative argument that the induced track print actually allows for more robust data collection because you can avoid the zonation issues you mention to be a problem on line 128-130. I don't understand why this is cast as an example of making the EDM less objective.

We raise the issue because of past concerns regarding higher age dispersion with LA-ICPMS data. We just want to make the point that this is not necessarily a bad thing and that other factors may be contributing to apparent differences between EDM and LA-ICPMS data.

145. Again, this is not an inherent problem with EDM it is a choice by the EDM user.

Yes. Results obtained from both EDM and the LA-ICPMS methods will depend on choices made by the analyst. It is important that a large enough number of grains are analysed to better represent the true age variation within a multikinetic sample. Our point is that higher age dispersion is likely to be encountered using an LA-ICPMS approach that tries to capture variability in single grain ages by analysing a sufficiently large sample size. Many applications of EDM have analysed a smaller number of age grains and this can lead to lower age dispersion. With fewer grains counted and decisions made on what to count after all the analytical data has been gathered, there may be a tendency to choose similar grains, which can result in lower age dispersion for multikinetic samples. The solution is not to abandon EDM but to count more grains with a broader range of characteristics to better represent different age populations.