Authors' Response

We thank the reviewers for their feedback. Below, we have copied the reviewers' comments (in bold) as well as our responses submitted on the interactive open discussion forum (in italic). In most cases, the proposed revisions were already discussed or addressed in the interactive comments, so we have simply added "Revised accordingly" in this document below each comment. We have highlighted any additional comments/changes related to each comment.

Additionally, as suggested by the editor, we have reduced the number of significant digits displayed in the supplementary table pdf to make it more readable, and made corrections at Line 567 for consistent reference to (U-Th)/He dating, and spelled out effective uranium concentration and sphere-equivalent radius at their first appearance in the manuscript.

"One abiding issue that the authors could discuss more are the ramifications of not knowing when the fracture took place – was the grain broken during the mineral separation process, or was already a fragment in the rock. A further complication to the latter case is whether it was an isolated fragment in the rock, or a part of a larger, fractured crystal, where ejection would be matched by implantation across the fracture interface, but the fracture would also serve as a grain boundary for diffusive loss. How does uncertainty on this point affect the correction? One idea might be to run cases similar to Fig 2 and 5, such as one where "broken grains" were broken pre-deposition and thus should ideally not be corrected; the new protocol increases the overcorrection, resulting in a less salutary histogram of deviations. Accordingly, it would also be beneficial to also discuss, what are the textural clues for discerning when the fracture took place, and how reliable they are likely to be? There are some easy ones, like rounding suggesting transport, but perhaps the authors have additional experience to convey."

As Dr. Ketcham's comment suggests, it is not possible to definitively assess when the fracture took place, and this is a fundamental problem for both the new and old protocols. Whether or not to apply a fragmentation correction is a question that precedes what protocol to apply. Nevertheless we agree that this is a significant issue that warrants additional discussion in the manuscript, and will make the appropriate revisions, as discussed below.

For igneous samples, any crystals with existing fractures prior to cooling would most likely still be intact, from an alpha-ejection perspective (i.e. the fragments are immediately adjacent, so long alpha-stopping distances implant He across fractures). This means that the fragmentation correction to a grain that has lost one or two termination(s) would still need to be applied. On the other hand, the question of when the fracture occurred matters for He diffusion. Depending on the thermal history, and whether the fracture pathway was a free surface for He loss, the He concentration profile of the fragment would be the result of some combination of alpha ejection having acted on external non-fragmented surfaces and diffusion having acted on all free surfaces. An additional complication, for certain thermal histories (e.g. partially reset samples), is that a broken face that is still adjacent to the other

broken side so that it has not experienced alpha-ejection helium loss, may experience more diffusion at this fracture plane than the exposed external faces where diffusion is inhibited by the lower He concentration at the crystal boundary. We will revise the text to address this, but it would be beyond the scope of this manuscript to quantitatively model the effect of diffusion along uncharacterized fracture pathways whose diffusive properties are unknown.

For detrital samples the situation is more complicated. For samples such as modern river sands derived from crystalline rocks, in our experience an assumption that c-axis-perpendicular breakage occurred after cooling (through approximately-closure-temperature temperatures) can be reasonably based on textural clues, particularly the contrast between detrital apatite grains that are rounded or abraded by transport and the sharp faces and corners of fragment surfaces that have not. For sedimentary samples that have not been clearly buried and reset, e.g. sedimentary rocks not buried more than couple km, the timing of fragmentation would be more ambiguous, unless the age of the deposition is known (assuming that the age of the deposition is close to the age of erosion and transport, and therefore the timing of fragmentation). It would be difficult to illustrate this type of uncertainty in a histogram such as Fig. 2 or 5 because the deviation from the "true" age, i.e. the extent of the overcorrection, would depend on the unknown period during which the fragmented surface has had the opportunity to experience alpha ejection. The "true" age would lie anywhere between the corrected date with fragmentation correction (minimum date) and the corrected date without fragmentation correction (maximum date). We could arbitrarily specify a sediment age, for example, and show the resulting histogram, but we think that may be a misleading characterization of the uncertainty, which would depend on geologic constraints specific to a sample. Instead, we propose to add a discussion of this problem, as follows:

Analogous to problem of unknown timing of abrasion for rounded detrital apatite and zircon grains (Rahl et al., 2003; Thomson et al., 2013; Reiners et al., 2018), if the timing of fragmentation is unknown, we could define a maximum date A_{fc}, the FT-corrected date without fragmentation correction, corresponding to fragmentation before or immediately after cooling; and a minimum date A_{ffc}, the fully FT-corrected date with fragmentation correction, corresponding to fragmentation during laboratory mineral separation. If there is sufficient geologic context, we can take the date of the sediment A_s to be the latest time at which the fragmentation occurred, such that the minimum date would be:

$$A_{min} = (A_{ffc}) + (A_{fc}-A_{ffc})(A_s/A_{fc}).$$

A conservative approach would be to display a "plot date" that is the mean of the maximum and minimum possible dates, and an error bar depicting the possible range of dates (Thomson et al., 2013).

In the case of detrital samples for which the timing of fragmentation is unknown, and that have also experienced non-monotonic cooling so that there has been significant diffusive loss after fragmentation (e.g. a fragment in a sedimentary rock that has been partially reset by burial-induced heating), fragmentation-corrected dates will be systematically younger than the corrected dates of whole, unbroken grains. For example, the corrected dates of

any whole crystal may reflect a date-Rs relationship, and naively applying the fragmentation correction to fragments will lead to corrected dates that all lie below this the corrected date-Rs relationship of unbroken crystals. In this case, a plot date with a maximum and minimum date could still be calculated, as defined above. The plot dates would reflect a similar date-Rs relationship: i.e. if fragmentation occurred soon after cooling and significantly before partial reheating, the maximum date A_{fc} would be the closest to the corrected date of an equivalent unbroken grain (except that it would be younger than the equivalent unbroken grain to the extent that the unbroken grain is less affected by diffusion due to its size).

In all cases where significant diffusive loss complicates the application of fragmentation correction, the best way to approach the problem may be to consider a combination of factors in deciding whether to apply the fragmentation correction or calculate a plot age that is combination of the fragment-corrected date and the normal FT-corrected date: a.) whether there is a date-size correlation of corrected dates of unbroken grains, and b.) whether the fragment-corrected dates are systematically younger than the corrected dates of unbroken grains, and c.) whether there is geologic context to suspect earlier fragmentation.

We have added a section in the Discussion (Section 4.3) that addresses these issues, in line with our comments above (as posted in the interactive open review forum of GChron). We have also emphasized this problem in Lines 89-90.

One other possible improvement is extending their thinking to the case of modeling: how should a broken grain be entered into thermal history modeling software? Does their reasoning on FT correction also extend to the proper effective radius for diffusion modeling on a sphere? For a grain broken on both sides, an infinite cylinder calculation would probably be the most appropriate, although the proper ejection profile for a cylinder would have to be derived. But, if for now we're stuck with converting everything to a sphere, what is the appropriate conversion?

For the purpose of thermal history modeling (in particular, QTQt, which implements the modelling approach of Brown et al., 2012), apatite helium dates can be input in their uncorrected form, and the fragment dimensions can also be directly entered, without conversion to effective radius. Although Dr. Ketcham's point is certainly valid, because we focus solely on protocols for Ft-corrected dates, we prefer to avoid the considerable additional complexity that would come from discussing thermal history modelling (and the complex additional considerations from timing of breakage on He diffusion effects) in this manuscript.

Regarding the appropriate geometric approximation: the fragmentation correction proposed in the manuscript does not introduce any additional uncertainty that is not already a problem for non-fragments, and potential implications of this for modeling related to this issue have been addressed in the literature (e.g., Gautheron and Tassan-Got, 2010). For consistency, we opted not to introduce a different geometric approximation only for broken grains. Furthermore, because the difference between the beta-FT relationships of various geometric approximations diminishes with lower beta (cf. Fig. 2 of Farley 1996), our

proposed correction leads to a slightly smaller difference between the spherical approximation vs. cylindrical/other approximations, compared to the original fragmentation correction.

Regarding the conversion of sphere-equivalent radius for the purpose of evaluating any date- size relationships when fragments are involved, one possibility would be to use the half-width and an assumed aspect ratio for typical crystals to calculate an assumed sphere-equivalent radius (Rs'). Another alternative would be to use a sphere-equivalent radius based on FT (Cooperdock et al., 2019), using the alpha-ejection-affected-FT value proposed here. The rationale would be analogous to the case of the fragmentation FT correction: the alpha-ejection-affected surface-to-volume ratio of a broken crystal is a good proxy for the available-for-diffusion-surface-to-volume ratio.

In Section 4.3 we discuss the issue of the appropriate sphere-equivalent radius, as discussed in the last paragraph above.

Finally, if the broken grain boundary is inclined with respect to the c-axis rather than perpendicular, what is the appropriate length – the long edge along the c-axis, the short edge along the c-axis, or their average (which is roughly equivalent to the length along a central axis, a reasonable estimate for the axis of symmetry)? In most cases, it probably amounts to a negligible difference, but for the sake of completeness it would be good to consider and discuss.

Because fragmentation of apatite is almost always along a crystallographically controlled cleavage, it is uncommon to see obliquely broken fragments. In the rare cases of such fragments, the appropriate length measurement to use would simply be the length along the central axis. i.e. assuming that the crystal is symmetrical with respect to the c-axis, an obliquely fragmented crystal of average length 85 um (e.g. long edge 90um, short edge 80um) would have exactly the same fraction of helium retained as a perpendicularly fragmented crystal of length 85 um.

We will clarify this in the revised manuscript.

We added a sentence at Line 175: "In the unlikely case of an oblique fragment, L should be the average of the longer and shorter sides."

[line 30] Delete "both"

[line 93-94] Delete "since the widespread application of the technique"

[line 296] Does this sentence refer to the error bars? Maybe better to say it directly: "Error bars in date-elevation profile represent dispersion of dates corrected with new protocol only."

[line 333] Spell out XRCT

We agree with the above four line-by-line comments and will revise as suggested.

Revised accordingly.

Response to Reviewer 2

This manuscript presents a new fragmentation Ft calculation that improves (U-Th)/He date accuracy and precision for broken apatite crystals. They demonstrate this using a previously published synthetic dataset by using two fragmentation correction approaches – the "old" method and their "new" method – and then estimate the variation in their datasets. This is a relevant topic to the (U-Th)/He community, as apatite crystals often do not have terminations, either naturally from growth or from breaking during mineral separations. It is my opinion that this work is most appropriate as a Technical Note in Geochronology.

Although the approach seems straightforward, I found myself struggling to understand exactly what the authors did at certain steps. As such, I have several specific suggestions for places that the text and figures can be made clearer. I will list these below based on the location in the text.

We appreciate the constructive criticism and will revise the text and figures accordingly to try to clarify our approach (see specific comments below)

Line 64: "For apatite, these errors are usually minor (<1.5% for 80% of apatite crystals, and <9.5% for 95%), … Furthermore, the errors are usually symmetrically distributed, with apatite populations … (Ault and Flowers, 2011)." These lines state that apatite do not typically exhibit "extreme zonation" and that zonation is evenly distributed based on Ault and Flowers, 2011. Ault and Flowers, 2011 demonstrated this for a large number of apatite crystals from very old, cratonic crustal samples. Perhaps we do not know this statement to be true for all apatite across all rock types. For example, apatite from ore or metamorphic systems may exhibit different

zonation distributions and concentrations. I suggest rewording this to not sound so definitive, unless there are other studies to cite to back it up.

We agree and will revise to qualify the statement to the following:

For apatite, these errors are usually minor (<1.5% for 80% of apatite crystals, and <9.5% for 95%), because apatite crystals in most cases do not typically exhibit extreme zonation of parent nuclides (Ault and Flowers, 2011, in the case of old cratonic samples). The data from that study suggests that the errors are usually symmetrically distributed, with apatite populations not exhibiting bias towards either rim-enriched or rim-depleted grains, though this may not be the case for those from rocks that experienced metamorphism or hydrothermal alteration.

Revised at Lines 81-85

Line 160: Why are equations at Line 160 different from the equations in Figure 1? Should they be the same?

Equations at Line 160 are the same as the right hand side of the equations in Fig. 1 – with very slight rearranging of the terms. We will modify the equation in the text for consistency, so that the equation reads " 2R + …." Instead of " … + 2R".

Revised accordingly.

Lines 170-174: Here you describe in words how your approach differs from Farley, 2002. To more effectively drive this important point home and to help those more inclined to visual understanding, I suggest you provide a schematic like Figure 1 (or modify Figure 1) that shows the different approaches, including the difference in the proportion of helium added based on the methods.

This is a good suggestion and we will revise Fig. 1 with a new column to show the difference between the old and new protocol, as well as a schematic illustration of the rationale from the point of view of the fraction of helium retained. (Suggested figure attached).

Revised Fig. 1 accordingly.

Figure 2/Section 3: The information provided in Section 3 and the caption of Figure 2 can be reorganized to more clearly outline the methods used. This should describe the synthetic dataset in more detail for the reader and the approach that was taken. How exactly were the 'old protocol' and 'new protocol' implemented? This information is introduced starting at Line 190, but should be introduced and explained more in depth before summarizing the results.

This is a good suggestion and we will re-organize and revise Section 3 to more clearly state the steps and assumptions of the method, before introducing the results.

We have reorganized and re-written Section 3 from Lines 240-270 to more logically introduce the methods and results as suggested.

Line 196: "crystals that are <20 µm from the tip,"/ Rephrase to "crystals broken <20µm from the tip"

Agreed.

Revised accordingly.

Line 197: "apply both protocols to all fragments as we would in routine laboratory analyses."

This phrasing is tripping me up. I'm not sure what you are referencing with regard to routine laboratory analyses here. Are you talking about the assumptions you are using, or the way you are measuring the fragments? This paragraph could be clearer.

We are referring to the way we calculate FT corrected dates in routine laboratory analyses, not the measurement of the fragments. Will revise to clarify that:

We assume uniform spatial distribution of the parent nuclide, and apply both protocols to all fragments exactly as we would calculate F_T corrected dates in routine laboratory analyses: i.e. we assume no knowledge of the original length and thermal history of the crystals to compute the corrected age, and use only the raw date, length and width of the broken crystals, and the number of terminations present for the calculation.

Revised accordingly.

Line 203: "In a more realistic scenario, when the two assumptions are relaxed, the proposed fragmentation correction results in a broader range of uncertainty (+0.7% ±205 4.2%), but it is nevertheless more accurate and more precise than the old protocol (+2.9% ± 5.0%)." What does "when these assumptions are relaxed" mean with regard to how you implemented it in your model? Exactly which 2 assumptions are you referring to? My reading is that Fig 2b doesn't exclude crystals broken <20µm from the tip? If that is correct, can it be more clearly worded in the text and labeled on the figure?

Will clarify text to emphasize what these 2 assumptions are, and will revise the caption to the figure to clarify what is included/excluded from Fig. 2b.

Revised accordingly at Lines 235-237. Revision of Section 3 also addresses these points.

Line 206: "These results are based on the full fragment dataset from Brown et al., 2013, which includes a representative range of thermal histories that are more

complicated than simple rapid cooling (i.e. slow, monotonic cooling; prolonged isothermal residence in the partial retention zone followed by rapid cooling; a mix of slow cooling and isothermal holding in the partial retention zone; and gradual reheating (e.g. burial) followed by rapid cooling; cf. Wolf et al. 1998)."Earlier you say you used 2 thermal histories – what thermal histories did you use, and what do you mean here when you say that "these results are based on…a representative range of thermal histories …" ? Are your results not comparable to the Brown results?

Will revise to clarify that Fig. 2a includes the two simple thermal histories (fast/slow monotonic cooling) and Fig. 2b includes in addition the complex thermal histories involving prolonged residence in the partial retention zone and/or reheating.

Revised accordingly; revision of Section 3 also addresses these points.

Figure 3: I find this figure confusing. Since L x 2 is the same for the old and new method, is it necessary to have it for both? Perhaps having a difference in the symbols between singly broken and doubly broken would be helpful to emphasize how they are generating different "answers". The red arrows are too difficult to see to realize they are arrows. Similarly, the dashed vs solid line arrows were only apparent when I zoomed in really close. The light gray bars saying "difference between corrections" is both hard to see, and unclear which points are being connected. Why do the black lines roll over at length = 20x width? Perhaps this would do better as two plots with single terminations and double terminations separated?

This is a good suggestion and we will make the stylistic revisions to the figure as recommended (for example, plotting singly and doubly broken grains as graphically representative symbols). We think it will be best to keep this as a single figure for ease of comparison. We also think it will be best to keep the two lines for both the old and new method to clarify that one represents the correction for singly broken grain and the other for doubly broken grains.

The FT value at 20x is approximately the same as the asymptotic value of infinity.

Revised accordingly; Fig. 3 has been updated with more intuitive symbols, larger arrow/font sizes, and we removed the dashed asymptote lines to de-clutter the figure.

Line 258: "two ideal assumptions relaxed" Please state the 2 assumptions here so that there is no ambiguity.

Agreed.

Revised accordingly.

Figure 7: The figure caption says this is schematic, but some of the curves are apparently based on real data too It is kind of hanging in the text and not so clear how it contributes to the rest of the paper (particularly D). The figure seems to show

the "direction" and spread of error from different factors, all scaled the same(?), but without any quantitative indication of the degree of the error introduced. The text says this shows the expected distribution based on different factors, but it's unclear what this expected distribution is based on (i.e., some constrained examples, or completely a cartoon?).

We agree with this comment and will revise the caption and text to more clearly indicate which curves are schematic and which curves are based on actual data, and to better integrate the figure into the text. The point of this figure is simply to show the potential of fully characterizing the uncertainties involved in apatite He dating, and how this and future work can help us interpret large intrasample date variations.

Revised accordingly at Lines 515-520.

Line 347: – "and in certain cases up to 20%"

What cases had 20% older dates? Please be more specific.

These cases were drum-shaped fragments with no terminations that experienced gradual reheating. Will revise appropriately.

Revised accordingly.

One other comment: There is no mention of zircon in the paper, but presumably a similar approach could be taken there? Can the authors add a brief discussion of whether this could be applied to zircon and what the expected outcome might be? I suggest this because I can imagine that labs that adopt this practice are likely to adopt it across any of the minerals they analyze, and it would be helpful to have a reference for why it would or would not be appropriate.

This approach would apply analogously to zircon, with exactly the same reasoning; however, it is much less common to see similarly fragmented zircon because it does not have the well-expressed basal cleavage that apatite does. For completeness, we will include an additional statement addressing zircon.

Revised accordingly.