GChron-2024-33: Technical Note: Improved calculation of volume, FT correction, and other derived data for polished zircon (U-Th)/He thermochronology
Author's Response to Reviews, Iteration: Minor Revision
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AC: Firstly, I would like to thank the reviewers for agreeing to review a manuscript that had already been heavily revised since initial submission and taking the time to review the first-round manuscript and reviewer comments in addition to providing new feedback on the revised version. I appreciate their feedback on where the manuscript still needs clarification and have adopted the majority of their suggestions. Responses to specific feedback are below.

RC3 Reviewer comments:
RC3: Having read the manuscript and the two previous reviewers' comments I feel that the author made the necessary changes requested by the reviewers and editor. I believe that the clarity of the manuscript has been much improved. This improvement was required to ensure readers understood the experimental process suggested by the author which could be followed using the program and equations within.

However, I do have some minor corrections that if applied I believe would improve the manuscript. I leave whether they are done or not to the discretion of the editor and author.

RC3: Line 9: *obtain the maximum geologically relevant information.* Could this be clarified, does this sentence mean the most relevant information or the largest amount of relevant information?

AC: The sentence was meant to refer to the latter reading. Based on this comment and a comment from RC4, I have modified the sentence to be more explicit (Lines 8-10).

"Polishing mounted zircon crystals prior to bulk grain (U-Th)/He thermochronology analysis provides opportunities for characterizing and subsampling each grain via in situ methods to obtain additional information relevant for (U-Th)/He date interpretation and the broader geologic questions of interest."

RC3: Line 110: *This calculation requires additional measurements of the polished grain surface: length (LP) and width (WP) of the polished face.*

If the grinding depth is assumed to be the amount of crystal removed and the crystal is assumed to have been polished parallel to the c-axis then the grain surface does not need to be measured and can be calculated from the assumed geometry. This can be done for both the ellipsoid and the tetragonal prism. I think this could lead to improvements to the manuscript in three ways.

AC: The reviewer is correct that this can be done for tetragons, and this is in fact done and reflected in Table 1, Appendix A, and the accompanying R script. The notations on Figure 1 were mistakenly left from an earlier version of the manuscript and this has now been corrected. Cylinders and ellipsoids need the additional measurements of the polished face to account for the curvature of the exterior surface which is not calculable from the four orthogonal measurements L1, L2, W1, and W2 alone due to the fact that $a \neq b$. I have clarified this in Line 125:

"This calculation requires additional measurements of the polished grain surface for ellipsoid and cylinder geometries…"

RC3: Firstly, as the author themselves appears to state, it is often difficult to differentiate faces from crystal exteriors using light microscopy. This would reduce the burden on the experimentalist and speed up the process of measuring crystal sizes which is a significant time sink.

AC: I'm not entirely sure what the reviewer is referring to with this comment. In Line 129 I state that L_P and W_P are often indistinguishable from L_1 and W_1 but this is unrelated to crystal faces. This is just referring to the fact that these measurements are often similar/overlap. It is easy to imagine situations when this would not be the case though, such as when a very large grain is only abraded a small amount. I have edited Line 131 to try to clarify this:

"In practice, L_P and W_P are often indistinguishable from L_1 and W_1 for small and medium grains, but for larger grains, the difference between the polished face and total axis measurements can be much greater."

In regards to time, the experimentalist is already making 4 measurements per grain so adding 1-2 more for a subset of grains does not really add that much time to the total endeavor.

RC3: Secondly, because it is often indistinguishable, then calculating these values from a grinding depth which is used anyway, would increase the accuracy of the resultant model. Unless this measurement is in fact being used to account for the fact that the model geometries might not match with the actual geometries in the crystal in reality. In which case this should be stated.

AC: As noted above, measurements of the face are needed to calculate the values a_P, b_P, and c_P in cases when L_P and W_P are not indistinguishable from L_1 and W_1. The only way to determine if they are indistinguishable is to measure the face, so personally I think it is better to have these measurements be a standard part of the protocol.

RC3: Thirdly it might mean that in Figure 1, the measurements of Lp and Wp could be removed from the figure, and make it easier to read.

AC: The L_P and W_P measurements on the tetragon subfigure in Figure 1b were included by mistake and have been removed. As explained above, these measurements are needed for ellipsoids and cylinders and are retained.

RC3: Line 206: *For the most symmetric grains...*
Here I would suggest replacing symmetric with equidimensional or equant. As crystals could be equant but not symmetric, such as would be the case with tetragonal prisms with a single termination. This would lead to changes in lines 213 and figure captions 2 and 3.

AC: This suggestion has been adopted throughout Section 4 (Lines 244, 251, 254, 278, 281, 283 and Figure 2 and 3 captions).

RC3: Figure 2: Do the 'least symmetric' grains which pass the requirement of Ft > 0.5 have quite variable shapes? I think it would be nice if those shapes were added as a 3D mini drawing to the graphs. This might help contextualise the 'least symmetric' data.

AC: The least symmetric grains do vary in terms of relationship between the a, b, and c axes. I have added a cartoon depicting this as Figure 2b.

RC3: Figure 3: In the caption it is written that a) is for equant crystals and that section b) is for minimally equant crystals – could it also be put on the figure? Again, maybe as shape drawings?

AC: I have referenced the new Figure 2b regarding the variability in shapes in the Figure 3 caption.

RC3: I also think that there are several assumptions that underlie the method that are not explicitly stated, or perhaps I have missed them. This list is not exhaustive but some might be:

a) That the spheres used for measuring grinding depth and the zircon crystals are at the same depth under the epoxy surface

AC: Yes, this is assumed and is generally a good assumption based on how these mounts are made. The grains are also assumed to be at the same level (touching) the epoxy surface. I have modified Figure 1a to include a glass bead for reference that shows this orientation.

RC3: b) That the zircon crystal is not at an angle to the polishing surface

AC: Yes, this is also assumed but I think this is communicated by the fact that the protocol is explicitly designed for grains polished parallel or perpendicular to the crystal c axis as shown in Figure 1 and discussed in Section 3. I have added a sentence to be explicit about the classification of polishing orientation, Line 106:

"Polishing orientation is also classified as perpendicular or parallel to the crystallographic c-axis based on visual inspection of the grain fragment."

RC3: c) That the crystals have homogenous parent isotope concentrations

AC: Yes, this is an assumption, which is made commonly for single-aliquot (U-Th)/He. Deviation from this assumption could be problematic but no more so than for whole grains. I have added a sentence being explicit about this to Section 3 (Line 144):

"Like most whole-aliquot (U-Th)/He thermochronology data reduction, the grains are assumed to have homogenous parent isotope concentrations (e.g., no zonation). Deviation from this assumption would impact the calculated dates in similar ways to zonation in whole grains (e.g., Danišík et al., 2017; Hourigan et al., 2005)."

RC3: d) That the polynomial coefficients used to calculate Ft from SA/V are used while perhaps actually deviating from the geometries which they were defined from (this assumption was highlighted by reviewer 2 and the author addresses it in Lines 137- 140)

AC: As the reviewer notes, this assumption has already been addressed.

RC3: My personal preference is for assumptions like these, whether they are likely to cause inaccuracies or not, to be focused in one area of the method section.

AC: These assumptions are addressed in Section 3.

RC4 Referee Report:
RC4: This manuscript highlights the importance of accurate Ft and aliquot volume calculations in (U- Th)/He Thermochronology. The author provides improved equations to calculate Ft and grain volume accounting for grain polishing required to make in situ thermochronologic measurements. The author greatly improved the manuscript between the first and second round of revisions. I greatly appreciate the author's addition of a comprehensive synthetic dataset to demonstrate the effectiveness of the proposed protocol compared to former methods. I believe this work represents a substantial contribution to our field, and should be published following minor revisions to address readability.

Overall, the subject matter and scientific process in this manuscript are well presented. The manuscript could benefit from some minor edits to improve the flow. The introduction section, largely changed from the original submission, is somewhat disorganized. Much of the information presented in this section focuses on the technicalities of other methods for measuring V and Ft, which may be better discussed in section 2.

AC: I appreciate the reviewer's time providing text edits and feedback. While I agree that the level of technical background presented in the Introduction is high, I believe that this is appropriate for a Technical Note paper as it serves to explain why the new method presented is novel. It was also my hope to make this manuscript accessible to thermochronology users who may be less familiar with the mechanics of F_T corrections and (U-Th)/He thermochronology derived data calculations more generally. Many geoscientists who use thermochronology data get their data already processed from the lab that generated it and the knowledge of how this was done may not be front of mind when assessing if the new method presented here is relevant for them. However, these thermochronology users can also benefit from the methodology presented in this manuscript.

RC4: Line 9: "...maximum geologically relevant information" wording is clumsy and missing a preposition?

AC: Based on this comment and a comment from the other reviewer, I have modified the sentence to be more explicit. (Lines 8-10)

"Polishing mounted zircon crystals prior to bulk grain (U-Th)/He thermochronology analysis provides opportunities for characterizing and subsampling each grain via in situ methods to obtain additional information relevant for (U-Th)/He date interpretation and the broader geologic questions of interest."

RC4: Line 17: You do not specify the acronym eU as effective uranium in your definition (Line 12)

AC: Thank you for pointing out this oversight, the acronym has been specified in Line 12.

RC4: Line 75-77: It is unclear what the subject of this sentence is. What is being directly compared to Ft corrections of whole grains?

AC: This sentence has been edited for clarification (Line 64):

"These contributions have largely focused on direct comparisons between F_T corrections for polished grain fragments and F_T corrections for corresponding whole crystals from which polished grains were derived…"

RC4: Lines 74-104: This paragraph covers a lot of information (some of which might do better in the discussion, i.e. lines 87-89), and could be broken down to streamline the story. I recommend having a paragraph that outlines previous work and limitations followed by a paragraph describing your proposed methodology and how it addresses these limitations.

AC: I agree this paragraph could be broken up for easier readability but I disagree that this information does not belong in the beginning of the paper. As noted above, since this is a technical note, I think it is necessary to establish how the new method differs from previous ones, which is difficult to do effectively without providing some background on previous methodology.

I have moved the bulk of Lines 74-104 to a new Section 2, "Existing methods and limitations for polished grains" (Lines 60- 86) and revised the end of the Introduction (Lines 50-59):

"Although some previous work has addressed the effect of grinding and polishing on F_T corrections (He and Reiners, 2022; Marsden et al., 2021; Reiners et al., 2007), these contributions do not address other data derived from volume and surface area and apply to specific cases that do not reflect the full range of real zircon samples or sample preparation. To address the lack of a comprehensive approach to volume-derived data for polished zircon, this contribution presents a protocol and set of equations (Appendix A) coded in an R script (Code Availability) that can be integrated with existing workflows for grain characterization and (U-Th)/He thermochronology data reduction and interpretation. Values calculated under this protocol include V, SA_α, volume-to-alpha-ejection-surface-area-equivalent spherical radius (R_{SV}), mass (M), parent isotope concentrations, eU, F_T, and F_T-equivalent spherical radius (R_{FT}). Results of using the protocol are evaluated using a synthetic dataset encompassing a range of possible grain geometries, sizes, polishing orientations and grinding depths (Section 4) and application to a real detrital zircon dataset (Supplementary Text, Table S2)."

RC4: Table 1: Caption should include mention of where on the grain 2D measurements are taken on the grain (the polished surface?).

AC: Most measurements are made parallel to an orthogonal set of crystal axes (e.g., c and a). This is shown in Figure 1b and has been clarified in Line 105:

"two orthogonal sets of length and width measurements (L_1, W_1 and L_2, W_2) parallel to orthogonal crystal axes, are made by rotating the grain fragment (Fig. 1b)."

Measurements L_P and W_P are made on the polished surface, which is explained in Line 132 and shown in Fig. 1b.

The caption for Table 2 has been updated to reference Fig. 1b.

RC4: Line 524: Have you tried incorporating measurements of tetragonal grain "tippiness" or the pyramidal length measured parallel to c-axis from tip to base of the pyramid? The aspect ratio of these pyramidal terminations can greatly affect grain volume and surface area calculations.

AC: I agree that "tippiness" can greatly influence these values, however, I have not incorporated this as the F_T polynomial coefficients assume symmetric terminations that are slope 45° to the a-b crystal plane (Ketcham et al., 2011). This simplification likely explains some of the difficulty in fitting F_T to real values for tetragonal grains, both for whole grains, as documented by Ketcham et al., and for the polished grains presented here. I do not think this manuscript can really provide a better way for dealing with this limitation, but I agree it should be discussed. I have added it to discussion of results (Lines 233-236):

"Terminations are approximated using a uniform assumption of symmetric pyramidal terminations sloped 45° to the prismatic core of the grain (Ketcham et al., 2011) which is also likely responsible for some of the unexpected behavior of these grains, as in reality this angle can vary from zircon to zircon."

RC4: Line 587: Section numbering is incorrect

AC: This has been corrected to Section 5.

Additional Changes:
Remarks from the preceding review file validation:
Please number the sections of the supplement according to the guidelines (see "Supplements" at https://www.geochronology.net/submission.html#assets), i.e. "S1" instead of "1".

AC: This change has been made in the supplemental text file and in references to the supplement in the main text.