Answers to comments on manuscript Confined fission track revelation in apatite: how it works and why it matters

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Our answers are below in *red italics*.

Answers to comments by Raymond Jonckheere and Bastian Wauschkuhn

We should declare that we have a competing model, and thus a possible prejudice against, or in favour, of the reviewed manuscript. It was our intention not to review it, but we accepted at the insistence of the authors, and because of its interest to those in our group involved in related studies. Our evaluation is critical but we believe not unfair. The public comments will set us right, if need be.

General comments

This manuscript reports calculations and numerical simulations of fission-track etching in apatite based on a variable track etch rate vT(x), isotropic apatite bulk etch rate vB, and certain track selection criteria. It concludes with a discussion of the implications for apatite fission-track da-ting and modelling.

An astounding fact considering the first author's impressive publication record is that the manuscript is almost unreadable. Even with foreknowledge of the concepts involved and of earlier publications, we struggled to understand certain sections. This was not helped by the constant use of ill-defined or undefined notions and meaningless word strings (e.g., semi-track penetration calculation). There appears to be no attempt to be understood, let alone to be exact, and readers less familiar with the subject can make heads nor tails of the manuscript, much less evaluate its significance.

We apologize for the difficulty caused; perhaps we can attribute it partly to Covid-19-related stresses. In any event, we acknowledge, as in our response to the other reviewer, that some things could be explained more clearly, and in a paper like this a small hang-up can greatly affect the reader's ability to see how the steps lead to the larger picture.

A second striking fact is that the manuscript presents a track etch model that appears to exist in a vacuum, disconnected from all existing knowledge of latent tracks, etching of tracks and minerals, and from all published step-etch data, except for those of Tamer and Ketcham (2020), e.g., for the 5.5 M etch (Carlson et al., 1999; Jonckheere et al., 2007, 2017; Tamer et al., 2019), the comparable 5.0 M etch (Laslett et al., 1984; Green et al., 1986; Barbarand et al., 2003; Moreira et al., 2010), and, given that the model is a first-order approximation, for lower etchant concentrations as well.

We're not entirely sure what the reviewers are asking for on this point – a review of all existing knowledge on latent tracks and etching? The reviewers themselves have produced thorough reviews, in their papers, and we refer to those. With the exception of Jonckheere et al. (2017), all prior step-etch studies have measured a fresh, random sample of tracks after each etching step, and we created our model to specifically interrogate data in which the same set of tracks is measured in each step, as such data provide a much clearer signal. It probably would not be too hard to extend our approach to earlier etching studies, but would require additional assumptions, most notably the need to estimate, and perhaps evolve, the analyst's decision-making process at every etch step, rather than only the first, which both of our reviews agree is fraught enough as it is.

One respect in which we found the manuscript lacking is a post-factum assessment of what it all signifies. The known statistical properties of surface tracks and confined tracks are a basic ingredient of the model. A second ingredient is the principle that to etch a track-in-track, the etchant penetrates down a host track, crosses to the confined track and etches it from their intersection point outwards. It seems not unlikely that this explains the broad traits of Figures 2-4, and, with scaling based on measured lengths, some experimental results as well, without the need for a specific vT(x) model. A foreseeable consequence is an excess of "under-etched" tracks which is culled down to ≪10% based on for the most part unexplained ad hoc selection criteria. This results in etch rates (vTmax) ranging by a factor of ~3 and core lengths (ΔxTmax) by a factor of >50 between samples. Dazed by the mathematical acrobatics, the reader is left wondering what it all means. Is this a hard numerical result, and which of the several factors above weighed most on the outcome, the fundamental geometrical configuration, the etch rate model, or the selection criteria?

We can add such a summary.

From our standpoint, the proposed model is too approximate and dependent on questionable assumptions to be confident about the numerical results. On the other hand, it highlights the paramount importance of etching protocols and selection criteria for apatite fission-track dating and modelling.

Again, we're not sure what the reviewers are getting at, or asking for. A sensitivity analysis of all model inputs?

Specific comments

4.1. Etching structure. Given the title: "*Confined fission track revelation …*", and Figure 1, showing a confined track with two endpoints, one could be forgiven for thinking that the calculations refer to etching of confined tracks. It turns out, after a full page of baffling equations suspended in mid-air, that the calculation was not that of the lengths of confined tracks at all but of those of a semi-tracks. The confined track length is then obtained as the sum of complementary semi-tracks. One needs to inform the reader from the start about what is going on, i.e. how one is going about solving a problem. With that information and a full explanation of all the symbols (and simpler notation) one need not bother about (1)-(5) as the conclusions are rather obvious from the outset.

We agree that we can improve how we introduce our approach to the problem. We do explain all of our symbols; we don't know which explanations the reviewers are finding insufficient. We're not sure what line of reasoning indicates that we "need not bother" with equations 1-5; are the reviewers suggesting that they should just be entirely relegated to the Appendix? Similarly, we have no idea what the reviewers mean by "the conclusions are rather obvious from the outset."

Figure 2 suggests a more interesting observation that is not discussed: the results for the "Constant-core model" (A, C) and the "Linear model" (B, D) are identical. It is possible that this is due to a conservation principle, in the sense that if A and B are far enough separated, then the time to etch from A to B is independent of the etch rate function vT(x) between A and B, but only depends on the area under vT(x). A consequence of this observation is that if the two end-member models predict the same evolution of (semi-)track length with etch time (cf. Figure 2), then how are step-etch mean-length data going to distinguish between them, or between them and other models?

The two models are similar, but not identical. It's actually the area under 1/vT(x) that must be the same, and if there is only one etching step then the models are indeed indistinguishable. However, if there is a second etching step (leading to points A+dA and B+dB), and then a third, etc. then the structure imposed by the two models constrains how these additional increments can be accommodated. The same goes for a different track, etched to two different points (C and D), perhaps over a different time, etc.

4.2. Semi-track penetration and confined track revelation. Much as the preceding discussion of the track etch rate and apatite etching, the lead up to Figure 3 (line 150) reads as a perfunctory dismissal of the accomplishments of earlier scientist as too trivial to cite. This entire section could have read: "*Semi-tracks have a sin(2δ) dip angle distribution and a homogeneous etchable length distribution (Dakowski, 1978; Laslett et al., 1982)*". Then some modelling happens and something is plotted in Figure 3: relative semi-track penetration and relative confined track revelation against depth (below the initial or etched surface?). Even with foreknowledge it is not possible to guess at the significance of Figure 3. What is concluded from Figure 3 does not seem to require modelling at all: "*~10% of tracks reach ≲1 μm*" and "*252Cf tracks are shorter, … but efficient*". Yes, see Dakowski (1978): ¾ of semi-tracks reach ≤½ a full track length deep, and most have near-zero depths.

Yes, we could have made this section briefer and just referred to the equations, but we wanted to take it a little bit slower for the sake of readers not as familiar as the reviewers with the mathematics of track revelation. Figure 3 is developing the first stage of the overall model; the curves are *dependent not only on the known mathematics (and we can cite Dakowski here), but also the vT(x) structure. We are not really trying to conclude anything from these figures. However, they do provide, in our opinion, an interesting visualization of semi-track penetration, and confined track revelation, through time, which helps develop an intuition for not only where tracks reach, but when they get there. These figures are also helpful in understanding the subsequent contour diagrams (Fig 4, 7).*

4.3 Confined track intersection. Line 158: "*weighted as cos δ*": see Dakowski (1978). It is hard to comment; on the one hand the concept is simple (hence likely the shorthand reporting) but, on the other, it is impossible to puzzle out what exactly has been done or what exactly is shown in Figure 4 (CDF?). Its discussion again refers to issues that seem to require no modelling at all: "*only about half of the tracks remain after surface intersecting track are excluded*". Yes, a slab of thickness L below a unit of surface contains L N tracks of which ½ L N intersect it (Fleischer et al., 1975).

The reviewers are incorrect in their translation here, as the model shown here only includes tracks within the angular interval the analyst might measure; and we are also including tracks with a range of depths, but very much weighted toward tracks near the polished surface, because that's where intersections are concentrated as semi-track etching progresses (see Figure 3).

We also note that in these critiques the reviewers are making the same omission in earlier treatments that we want this contribution to begin to correct: the element of time, and the fact that all aspects of the track intersection and etching process evolve in time, not just track length and shape. This has been known for a long time, of course (e.g., Jonckheere et al. 2007) but this paper is the first we're aware of that puts the entire process together. The straightforward reading of Figure 4 is: here is where and when track intersections occur.

4.4 Confined track selection. Fleischer et al. (1969), Paretzke et al. (1973) and a great number of other scientists published calculations of etch-time dependent track profiles for varying vT(x) and isotropic vB.

We mention Fleischer et al. (1969) in Section 2, and mistakenly omitted re-citing that work in section 3. We consider the model in section 5 rather primitive, and exclusively done for illustration sake.

Line 185: the isotropic-vB case is not "simplified" compared to Aslanian et al. (in press), but contradicts it, as well as fundamental etching theories (e.g., Heimann, 1975). The definition of "bulk etch rate" is different.

The reviewers are argumentative here; one person's simplification is another's heresy. We are already saying that their work will provide a better answer, but because they are still using undescribed "graphical construction methods" (Aslanian et al., 2020) rather than providing a time-evolving calculation, we could not reproduce their work for this illustration.

Line 198: "*we constructed … an operator bias function*". Despite the reserve one has to ask: where does it end? It brings to mind Murphy's variable constant, which multiplied with anything gives the desired result. How decisively does Figure 6A differ from a straight ramp, or even a step function? If there is an actual basis for it, please discuss it; if not, all that follows from 6A is a possible artefact.

As also expressed in our other response, here we are faced with a problem of quantifying something nobody has seen the need to try to quantify before: which barely-etched tracks will an analyst see and select, and which ones will either be missed or rejected. Unfortunately, if one wants to reproduce a measured track length distribution based on latent track etching structure, there is no getting around somehow accounting for which tracks are measured and which ones are not.

Furthermore, we believe we make a fairly convincing case that lack of a quantitative treatment of the issue of which tracks are measured and which are not underlies the AFT community's difficulties with reproducibility of length measurements. This may also get to the crux of the mental barrier that both reviews appear to reflect – they substitute virtue (implicitly: "good analysts only measure fully etched tracks") for empiricism ("what, exactly, is a fully etched track?").

As outlined in the text, we took the first-step measured data, and constructing a function that reproduced the left, or short, side of the track length distribution, and we furthermore used the same function for all such measurements in this study to limit the degree of license. The vT(x) model is responsible for the right, long side of the distribution, and so the results have an anchor in reality – too long, or too short, and we cannot match the data. Furthermore, with subsequent etching steps, both the left side and the right side of the distribution has to be generally reproduced to match the mean measured length; another anchor.

We thus reject the reviewers' exaggerated dismissal.

Figure 5 is to all intents and purposes identical with Figure 4 of Fleischer et al. (1969); the equations are also not dissimilar. It is worth noting that the assumed gradual increase of track length is contradicted by the step-etch data of Jonckheere et al. (2017, in part measured by the present co-author), demonstrating that confined track lengths increase in fits and starts, reflecting a discontinuous latent-track structure, as reported in numerous apatite studies (Paul and Fitzgerald, 1992; Paul, 1993; Li et al., 2010; 2011; 2012; 2014, etc.), as well as nigh the entire literature on latent ion tracks (not counting the amusing contributions from the Canberra group who posit a cylindrical track and then shoot their ions straight through the crystal so that the track ends are missing). One can of course think of vT(x) as averaging over a certain section, but this should be stated explicitly.

We can state this explicitly.

Figure 6. It is worth commenting on the true significance of the observed "excellent agreement". It would appear that the invasive, ad hoc filters (6A and 6D) would transform just about any underlying distributions (light blue in 6B and 6E) into almost negligible residual fractions (dark blue in 6B and 6E) with the general Gaussian appearance of the measurements presented for comparison.

As expressed above, and in the text, this is not the case. The "ad hoc" filters are only responsible for the left side of the distributions; the right side is an outcome of the vT(x) model.

Figure 7 is interesting, but, of course, underlies the combined reservations concerning the steps leading up to it.

One can also look at Figure 7 as a set of potentially testable predictions.

A puzzling fact about section 4 is that 4.1 sets up two competing ad hoc models for the variation of the etch rate vT(x) along the tracks (in fact one end-member and an generalized member of a single model, which is assumed to be the same for tracks produced by all fission fragments and energies). Sections 4.2 to 4.4 then present modelling results, which seem not to require the equations in 4.1, and never again mention which model (or parameter fit) was used, much less how the models differ.

This was indeed an unfortunate omission, which we will address. Frankly, in these figures, the differences between model forms is not really recognizable (as was largely the case in Figure 2). The principal difference is in their ability to reproduce the measured length data. The equations in section 4.1 are required to etch both semi-tracks and confined tracks as a function of time.

4.5 Fitting step-etching data. This section presents a complicated account of different modelling issues. It would help to reproduce Figure 1 of Tamer and Ketcham (2020) to present the reader with visual map of the data. It is worrying that there appears to be a need to exclude one data point, that the iterative fitting procedure does not seem to converge towards unique best-fit solutions, and that such solutions appear to not be strictly reproducible. Please discuss what that actually signifies.

We can add the figure, and write a short paragraph summarizing how sections 4.1-4.4 go into a model realization. It "signifies" that we were not able to come up with a set of mathematical expressions that deterministically generate a track length distribution as a function of etching structure and time, and thus used a Monte Carlo approach.

5. Results. One cannot help thinking that the discussion of Figures 8-11 could be so much better structured if, instead of the artificial opposition of "two models", one of which is an end member of the other, there would have been just one constant-core model with two end members, a linear-vT(x) model (ΔxTmax = 0) and a constant-vT(x) model (ΔxTmax = Llat). The latter is at least relevant to the ceremonial dismissal of the model unfairly attributed to Laslett et al. (1984) in Tamer et al. (2019).

We recognize the reviewers' desire for more order, but we disagree with the prescription – we are responding to what the data say about these two possibilities. Why would we want to force an end-member that doesn't fit most of the data?

The reviewers should re-read part 8 of Laslett et al. (1984). It refers specifically to a single vT, and says the track ends are "first resolved" when the "full etchable range" is reached, and that "in practice most confined tracks are slightly over-etched." These statements are all, strictly speaking, incorrect. We believe that this helped develop a misplaced confidence and complacency concerning our ability to identify a "fully etched" track, which in turn has led to poor reproducibility.

6. Discussion. "*It is remarkable that we are able to reproduce … using these simple models of etching structure*". Yes indeed, on condition (1) that the predictions in the first place depend on the assumed model, and in negligible measure on the geometrical framework and assumptions concerning track selection, (2) there is a negligible self-referral between the data used for calibration of the models and those adduced for testing them. Perhaps the careful term "reproduce" reflects concern about the fact that these overlap, which, in our view, affects the significance of the results.

We made the statement in (a) some amazement that everything worked, and (b) recognition that there is much that can be improved. One has to start somewhere.

"*We take these successes as an indication of the overall correctness of our characterization of confined track etching*". This is too facile; it could be a Ptolemaic model, which, admittedly based on false assumptions, nevertheless manages to account for isolated appearances (individual experiments) given suitable eccentricities and epicycles. The authors need to make a serious effort to make transparent the network of mutual influences between the geometrical framework, the actual etch model, the various selection criteria, calibration data, and the data used for verification, so as to to reassure the reader that their model is not in effect a self-sustaining mirage hovering in mid-air.

We were transparent in the one "self-referral", which was to estimate the left/short side of the length distribution for the first etch step based on observations, and we minimized the influence of this by using the same rules across all experiments. We will clarify this. Concerning Ptolemaicism, every other aspect of our model is based on accepted physical and mathematical principles of fission track behavior.

It could be argued in favour of the model that, following annealing and etching together, pre-annealed induced tracks are longer than fossil tracks (Wauschkuhn et al., 2015; Figure 15). Perhaps they etch faster?

Certainly, etching faster should cause some difference, but we don't yet have the information to estimate how much of one – in other words, any such estimate would be highly artificial. First, we do not know the vT/vB of the analyst (or, if vT/vB is the optimal way for parameterizing analyst *track selection; it's more a proposal at this point). Second, we have no data yet on how laboratory annealing affects the etching velocity in spontaneous tracks.*

Line 358: "*A thorough re-evaluation of this biasing based on measurements…*". This appears to overlook the fact that the (length) bias function implemented in modelling programs is based on experimental data.

Experimental data of laboratory-annealed induced tracks; this is why we have to study fossil tracks.

Lines 370-380. "*As vT/vB rises the proportion of accepted tracks increases …*". Do we interpret this accurately as: "Including shorter tracks decreases the mean length and increases the standard deviation (range)"?

Somewhat accurately – one of the points of Figure 15A to provide some more specificity on the estimated trade-off.

Line 386: "*… the mean latent track length of 17 μm indicated by our data and model*". More reliable estimates bracket the average latent track length in apatite between ~18 and ~21 μm (Jonckheere, 2003a; 2003b; Jonckheere et al., 2017). A discussion of established concepts such as etching threshold and range deficit relevant to this issue. On a related point: one could cite Figure 1 of Jonckheere (2003a) in support of the linear model, on the assumption that vT(x) is proportional to dE/dx.

We generated a new model using the current version of SRIM, which we can add as a figure (while citing previous related work); the profile actually looks a bit more like a cross between linear and constant-core. We also note that the "more reliable estimates" only characterize the track in the moment of formation, and ambient-temperature annealing may affect it to some degree. We can add such discussion.

Line 406: "*tip appearance depends on … mount preparation, polishing, and cleaning technique, microscope optics, and captured image quality*". This should include etching protocol and apatite composition.

We will add these.

Lines 450-455: "*These differences in latent versus measured lengths …*". The discussion omits established facts such as the etching threshold and range deficit (Fleischer et al., 1975; Iwano and Danhara, 1998).

We can add such text, but elsewhere; we were taking the shortcut of considering the "latent" track as the zone of enhanced etchability, a shortcut that has also been used by our forebears (e.g., Laslett et al., 1984). But we can be more precise, or specify the sense in which we are using it.

Lines 450-455: "*By stopping etching as soon as the curve of length versus time in step-etch experiments was reached …*". No, … as soon as the data showed a constant rate of increase of the mean track length, taken to be the average bulk etch rate. The same principle used for calculating your bulk etch rate.

An incomplete sentence on our part; we'll fix it.

Lines 460-462: "*… a rate defined by vB, which may be obtainable from etch figure measurements parallel and perpendicular to the c axis (Tamer and Ketcham, 2020a)*". Long-established etching theories hold that measurements of surface features are not suitable for estimating etch rates (Jonckheere et al., 2019).

We are utilizing an empirical approach to address a practical problem.

Lines 497-498: "*Figure 17 shows the near-surface portion of the penetration model …*": Figures 3 and 17 remain obscure; they could be etch-time-dependent depth distributions, but a better ex-planation is needed.

Lines 524-525: "*If … geological annealing results in different etching rates than laboratory annealing …*". It could be argued that, after simultaneous annealing, pre-annealed induced tracks are longer than fossil tracks (Wauschkuhn et al., 2015; Figure 15), perhaps because they do etch faster?

Again, we do not yet have data on how high laboratory annealing temperatures affect etching in fossil tracks. But, this is a good reason to get such data.

Lines 530-549: The outlook section reads as an appeal for more and better data and community involvement, but is rather short on scientific detail and tends to drift off into a dreamy utopia. It can be deleted. It is at this stage more important to look back and re-evaluate the results, than to look forward.

We disagree, completely. Getting more data on more types of tracks and more types of apatite, is not dreamy – it's work that we feel is well justified.

Line 551: "*a range of detailed step-etching data …*". This is a rather exaggerated designation of a handful of mean track lengths (Table 1) and some intersection depths. The imbalance between the "comprehensive" model and the limited data for calibration and testing is a conspicuous weakness of this work.

We note here that the reviewers criticize our amount of data in the next breath after they criticize our recommendation to get more and better data. We also note that the measurements we present constitute a lot of work.

Line 554: "*Along-track etching velocity … varies … among fission tracks …*". The intended meaning is probably among different types of tracks (spontaneous, induced, annealed), not between individual tracks.

Actually, all individual tracks, especially fossil ones which have each undergone different amounts of annealing.

Line 563: "*Most variation …*". Barring sample preparation accidents, does one need an etch model to conclude that lower mean lengths and higher standard deviations result from including shorter tracks? The vT/vB criterion is not progress, because it applies to isotropic detectors, not to apatite.

Having something where previously there was nothing is progress. We leave the door wide open for something better, but it's also possible that a simple solution will be the best.

Lines 575-613: "*Appendix: vT(x) Etching Model Equations*". This appendix contains a long list of repetitive piecewise integrals of simple functions, that are not used to obtain the results in this manuscript, instead produced by numerical simulations. The appendix thus seems superfluous to the manuscript, and may be either deleted or included as a supplement. We have not checked these equations. It is on reflection possible that the modelling uses the equations but this is no-where made clear.

This is a truly bizarre comment. Of course they are used to obtain the results of this manuscript; they are how the numerical simulations calculate etching of each semi-track and confined track as a function of time. They define the variables we use to fit the data, and plot in Figures 9-11.

Is this nowhere made clear? Line 138-139 states, for example, "The semi-track penetration calculation thus consists of randomizing some number (typically 10^5) lengths, dips, and x_{int} points, and then using the etching model to trace each semi-track's penetration into the grain surface." We can make it yet more explicit ("… etching model defined by Equations 1 and 2 and a given v_{Tmax}, L_{lat}, and Δx_{Tmax}…"), but what other etching model did the reviewers think we were using?

Matters of detail relating to the writing, figures and captions are too numerous to be addressed at this stage.