Manuscript gchron-2021-28 - Revision 1

Short communication concerning experimental factors affecting fission-track counts in apatite

Carolin Aslanian, Raymond Jonckheere, Bastian Wauschkuhn, and Lothar Ratschbacher

Replies to the reviewers' comments

Overview of substantial corrections

As explained in our detailed replies to reviewers *Drs. M. Tamer* and *H. Iwano*, we accepted almost all their comments. The few exceptions concern matters of phrasing and one Figure. Instead, we added two Figures, replaced Figure 4, and provided three more in supplement. Our main corrections are:

1. We added an image (Figure 2) of a basal section illustrating the loss and gain of tracks from different causes.

2. We made minor changes to Figure 2 (becomes Figure 3), to its caption and its discussion in the text.

3. We corrected and added some detail to Figure 3 (becomes Figure 4). This forms the basis for a comparison of the measurement data with numerical predictions based on the etch model of Aslanian et al. (2021). The model predictions are explained in Figure 5, which replaces our previous Figure 4. The new Figure illustrates the same principle as the previous in greater quantitative detail.

4. We added significant details to Figure 6 (becomes Figure 7), which enable us to address the matter of transmitted-light vs. reflected-light counts in a more detailed and convincing fashion (see replies to Dr. *H. Iwano's* comment to Figure 5). We added Figure 8, illustrating the extended discussion.

5. We added the missing data on the sizes of the etched-track openings to the data supplement.

6. We provide reflected- and transmitted-light images illustrating the loss and gain of tracks with etch time in supplement.

The reviews of *Drs. M. Tamer* and *H. Iwano* offered us an opportunity to take a fresh look at our data. This permitted us to pull together further strands of numerical evidence, and to present a fuller interpretation and a much stronger case for our conclusions than before. We expect that the complete corrections will lengthen our text by 50%, and therefore ask the editor to consider the suggestion of one reviewer to treat our manuscript as a full article instead of a short communication.

Freiberg, 7 December 2021.

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Reviewer #1 (Dr. M. Tamer)	Replies
This manuscript reports two different experiments on track counting in Durango apatite and uses a recent etch model to explain how the track counts change in different observation criteria. The first experiment is track count and etch pit measurement analysis of fossil tracks in three different sections of Durango apatite; the second one is a further counting analysis of unannealed, annealed, induced and fossil tracks in c-axis parallel section of Durango apatite.	We are grateful to the reviewer for emphasizing the merits of our work and supporting its publication.
The text is well written, the figures are clear, the tables and supplementary file contain almost all the analysis data, excluding etch pit/track opening length measurements.	We included the missing data in the supplement. This occasioned us to correct the calibration of our measurements of the etched-track openings in the different apatite faces. This, in turn, led us to expand our discussion and to relate the numerical values to our published etch model.
Analyst bias and track selection criteria have significant impact on fission track data analysis and ultimately affect the modelling results. After adding suggested figures, a new table and supplementary data for etch pit/track opening lengths and rearranging the format from short communication to regular manuscript (with more discussion and conclusion part), this work should be resubmitted as a regular manuscript. Please find the specific comments and suggestions in annotated pdf.	We address the reviewer's specific comments below, including those on the annotated manuscript.
Line 4: Institute fuer Geologie, Technische Universität, Bergakademie	Corrected to: Institut für Geologie
Line 30: Time-temperature sounds better than temperature-time, maybe I am just use to hear the first one.	This is a matter of preference; we believe temperature-time is more used in most geological papers.
Line 30: frequently?	Another matter of preference: "a much-used … tool" sounds better to us.
Line 31: The dating method rests on counting and the modeling on length measurements.	It is not so straightforward; an age without lengths can be a formation, cooling, mixed or a reset age, with uncertain or no geological meaning; a modelled T,t-path without an age can

	extend over any length of time. Our emphasis on "*counting and measuring*" is deliberate and, we believe, right.
Line 32: Latent?	Corrected.
Line 34: unable to be observed under optical microscope	Another matter of preference: "*too thin to observe with an optical microscope*" sounds better to us.
Line 47: and Tamer et al. 2019	Corrected; we cited the wrong reference: Ketcham and Tamer (2021) should indeed have been Tamer et al. (2019).
Line 56: Just like Figure 5 showing the images of tracks, maybe a figure can be made to show the step-etched tracks at the exact same locations for three Durango sections in three different etch times.	Corrected; transmitted and reflected-light images of the step-etched tracks in a basal, prism and intermediate face have been added in supplement, because they would overwhelm the text elsewhere.
Which light source was used for counting?	Corrected; we explain that we used both reflected and transmitted light (cfr. images in supplement).
The experimental procedure includes mounting, polishing and etching. The reader can expect that the tracks being counted are only spontaneous tracks but maybe the word spontaneous or fossil can be added in this sentence, referring the type of tracks.	Corrected: we specified "spontaneous tracks".
Figure 1: Comparisons on the 1 to 1 lines already show the increase and decrease in track counts with the step-etch experiments, however, an addition of a simple density vs time plot may help.	We do not object to adding this simple figure but would prefer not do so because we already have a lot of figures in relation to the text, and need to add others in response to the reviewers' comments. We prefer to add the more important figures rather than this not so important one.
Although it is written in the text and figure caption, maybe it would be good to include "basal face", "prism face" and "intermediate face" to the corresponding figures at top left.	Corrected; we added "basal face", "prism face" and "intermediate face" to Figure 1.
Table 1: what is TL? Transmitted light? Did you use transmitted light only for counting?	TL means transmitted light. We used reflected light for measuring the track openings and transmitted light for counting the track channels. We now explain this in the caption to Table 1, to which we have also added basic statistics related to the measurements of the track openings.
Table 1: Something unimportant: the densities reported in the table as tracks/cm^2. Maybe it would be good to use cm^2 to describe the area of the field instead of μm2.	Corrected; $3.815\ 10^{-4}\ cm^2$
Line 96: in?	Corrected.

Line 102: etch	Corrected.
Line 108: A figure showing a (some) track(s) disappear in the same field with the increasing etch time can turn this proposition truth. Maybe you can add such a figure. But there is also an inevitable possibility that you may missed some tracks to count, which is part of the fission track counting. Maybe you can mention this too.	Corrected. We added a figure showing the merger and gradual loss and gain of tracks in a basal face. We also use this figure to illustrate the discontinuous track terminations in relation to experiment 2. As the images in supplement illustrate, in step-etch experiments on Durango apatite, there is little room for accidental miscounts. Although it is not possible to exclude them (or anything else) formally, emphasizing such a hypothetical effect would detract from the results. We do not deny that accidental miscounts are real, but they are surely more important for routine counts of geological samples where neither the apatite, polishing nor etching are as in this work.
Line 131: The vast majority of the papers in fission track methodology is on the tracks parallel to c axis. A new figure that is suggested in comment #9 may visually assist the reader to understand the difference of the track openings in these 3 different samples.	Corrected. We added a figure in response to this comment, showing tracks in a basal, prism and intermediate face.
I could not see any data in the tables or in the supplementary excel file regarding the track opening measurements. How many openings were measured? Were the opening measurements executed consistently on the same openings from 10s to 30s or random openings were measured each time? What are the long axis in basal and in 30 degree section? Maybe a figure would be good.	Corrected. We included the measurements of the track openings (>18.000 in total) in the data supplement. The openings were measured in the images used for the track counts. Although overall the same tracks were re-measured after each etch step, we did not trace the individual openings from one step to the next. As the reviewer demonstrated (Tamer and Ketcham, 2020) and our etch model implies (Aslanian et al., 2020) their rate of growth is constant.
Line 158: I did not notice that there is a second experiment until here.	That is deliberate. To describe both experiments at the start and then return to each in turn for discussion would confuse the reader, as the two experiments are in part similar and in part dissimilar.
Figure 3: Visual differentiation of the shades of gray is a little narrow. Different shades can be distinguished in the histograms but it is somewhat harder for Figure 4d. Maybe using further ends of shades in the spectrum would be better.	Corrected. We improved the shading contrast in Figure 3. We also extended the discussion of the track openings, comparing them with numerical predictions based on Aslanian et al. (2020).
Line 165: Maybe the experimental details and the results can be divided, or at least the experimental details of the both experiments can be pointed out first. Both experiments include Durango apatite underwent same type of polishing and microscopy routines, same type of etchant with a little difference in etch times. The light source used in the first experiment is not pointed out. Merging the experimental details can reduce the repetition of the routine procedural descriptions, cover missing descriptions and pre-inform the reader about the number of experiments.	The problem is that we report two experiments on the same topic, stated in the title. One is a step-etch experiment and the other an annealing and counting experiment. The material and initial sample preparation are indeed similar, but that is what makes it difficult to distinguish between them. After considering the alternatives, we chose to treat the experiments in separate blocks, so that the reader does not have to switch back and forth between them. After reconsidering, we believe that it is the best solution, as the repetition amounts to a couple of lines at most. We made it clear at the start that we report two experiments in succession.

Line 185: Did you use isolate the counting only RL and only TL in the approach or did you switch the light sometimes? How about a third approach of counting by switching the light source constantly? For example, the TL and RL counts in sample 313 °C are 1450 and 7852 but if you use a mixed light approach with high switch frequency (maybe 30-40 switches per field counted, depending on the degree of annealing track density and other features), you may achieve a new number, probably close to 7852 but likely to be higher.	We separated the TL and RL track counts with the explicit intention to avoid biasing one or the other. We first performed the TL counts without use of RL and then the RL counts without use of TL. Although we did so a field at a time (instead of finishing all the TL counts before doing the RL counts), the TL count betrays next to nothing about the RL image. Conversely, with few exceptions the track channels cannot be distinguished in RL (Figure 5). A practised track counter will know from experience that it is impossible to keep in mind a previous image while counting the next. Thus, to all intents and purposes, the TL and RL counts are independent. Even if there were some inadvertent bias, that would have no effect on the rather obvious fact, illustrated in Figure 5, that there is a discrepancy between the TL and RL images. We explain this in detail in our revised manuscript.
Figure 5: Instead of not annealed, maybe unannealed or control would be better?	Another matter of preference: "*not annealed*" seems to us as good as "*unannealed*", which spelling checkers signal as incorrect.
Figure 5: maybe (t,T)?	Most co-authors have used (T,t) for years; we do not understand the need to change it now.
Figure 6: Why does this figure remind me the length vs density relationship figures in Green 88? The same dog leg pattern is visible here. Maybe Jonckheere 2003 and Green 1988 can be considered in the discussion part to point out these similarities.	We agree that the similarities between Figure 6 and those of Green (1988) and others are not coincidental, the common factor being the break-up of the tracks, which causes the TL track densities to collapse but not the RL track densities or the mean confined track lengths. We expanded our discussion and added a new Figure showing how these observations are related.
Line 201: Maybe this common knowledge shouldn't be in the discussions and conclusions?	Every manuscript contains some common knowledge. In this case, it is a single sentence introducing the topic of etching, which is the subject of our research. "*Measuring and counting fission tracks requires etching to make them accessible for microscopic examination. Track etching is …*". The discussion would be ill-structured and difficult to follow if we had no such sentences.
Line 218: It is understandable that there is no solution for these issues but maybe some further speculation can be added in the discussion part.	This comment refers to: " *Our findings provide no solution*". This is a warning that our results refer to our experimental conditions and cannot be extended to others. In contrast, our approach, based on step-etching, measurements of the track openings, and attention to polishing and etching as contributing factors, can be useful in searching for a solution. In particular efforts to automate track counts could benefit from considering these factors. We prefer to desist from speculation other than the references to ongoing research mentioned in our manuscript.

Freiberg, 16 November 2021.

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Reviewer #2 (Dr. H. Iwano)	**Replies**
This paper reported experiments concerning the effects of grain orientation, polishing, etching and observation on fission-track counts in apatite. The paper also showed results were systematically obtained and theoretically explained in the framework of a recent etch model. I think this is an article worth reading for FT researchers, therefore my recommendation would be publish this paper as a short communication. The following comments should be further developed for publication.	We are grateful to the reviewer for supporting the publication of our manuscript. We have acted on all comments, as discussed below, and expanded the discussion of our etch model and track counts.
Figure 1. Explanation for regression line is needed in the caption.	Corrected; the caption to Fig. 1 now refers to the regression lines, and to their slopes and intercepts in Table 2.
Table 1. The track densities of B00 and B60 are clearly smaller than those of P00. Is this a difference due to U concentration? If not, which one is closer to the true track density?	Indeed; the sections were cut from different crystals. The higher track counts for P00 are due to its higher U-content; the differences between the samples are well within the range of uranium variation in Durango apatite and immaterial to the dependence of the track counts on etch time.
Figure 5. I am amazed at the number of track-shaped pits in the reflected image. Of course the authors counted them as a fission track. What are the criteria for track identification? Please describe them for each (TL and RL). If there are several etch pits detected by apatite that is totally annealed at 450 ° C, I think it can be set as the minimum noise to identify (count) fission tracks. Additional images of totally annealed samples are needed and helpful.	Corrected; we clarified our track counting criteria. In both transmitted light (TL) and reflected light (RL), we counted each distinct feature as a track that was not identifiable as a polishing feature or another defect; we do not recall observing the latter in these samples. We guess that the reviewer suspects that the features counted in RL but not in TL are not fission tracks. This is reasonable, as such features have not been reported before. But that is not difficult to understand. First, the shallow etch pits observed in RL (Figure 5) develop in the slowest-etching faces (Jonckheere et al., 2019; Jonckheere et al., in press). The principle illustrated in our Figure 2 explains this. Some prism faces etch >15 % faster than the slowest. Shallow etch figures are therefore less prominent or absent in faster etching prism faces. A second reason why the shallow etch pits have not been reported before is that, as far as we know, apatite surfaces are rarely polished to the standard of our sam-

ples, i.e. a nano-polish with 0.04 µm silica suspension until no scratches are visible in reflected light, even using Nomarski differential interference contrast. The scratches visible in Figure 5 (re-)appeared after etching even though they are caused by polishing. The shallow etch figures would not be distinguishable in less well polished surfaces. The third reason why the shallow etch figures have not been reported is that they cannot be counted in transmitted light, while track counts in reflected light are uncommon. Moreover someone observing them would be inclined to dismiss them, either as not being identifiable as tracks or as too difficult to count with confidence. None of this is reason to conclude that they are not tracks, however.

The reviewer proposes to etch an apatite annealed 450 °C as a decisive test. We have not done that because it would not prove what one might expect. Numerous experiments attest that at 450 °C all tracks are erased *in TL*, but TEM shows that short track segments survive (Paul, 1993; Li et al., 2010; 2011; 2012; 2014). So we can expect to see no tracks in TL, but what of RL? If we see nothing, we could conclude that the small etch figures were associated with defects with identical annealing kinetics as fission tracks, and thus likely also tracks. If we do observe shallow etch pits when we see no tracks in TL, then do they correspond to defects that are more stable than fission tracks or to short surviving fragments of tracks? Neither outcome establishes if the RL features correspond to fission tracks or not.

In our revision, we explain our reasons for interpreting them as fission tracks, or sections of tracks. The first is that we did in fact perform 450 °C annealing experiments, eleven even, with the difference that we irradiated the annealed sections to create the induced tracks. Now, if the RL-features (for short) are not fission tracks what is the likelihood that they occur in a constant proportion with the TL-tracks (for short) in all eight pre-annealed and irradiated samples (except three later re-annealed to $\rho/\rho_0 < 0.70$) with induced track densities 10-20 times higher than the fossil track densities (before partial annealing)? We further note that for those eight samples as well as the four containing fossil tracks annealed to $\rho/\rho_0 > 0.70$ there exists an almost perfect correlation (r = 0.995) between the TL and RL counts.

All the ρ_{TL}/ρ_{RL}-ratios before the break-up point are of the order of ~0.9, a value consistent with independent estimates of the track counting efficiencies in transmitted light (ηq_{IS}; Jonckheere and Van den haute, 2002; Enkelmann et al. 2005; Soares et al., 2013; Iwano et al., 2018).

At $\rho/\rho_0 < 0.70$, the $\rho_{TL}/\rho_{TL,0}$-ratio collapses to zero, while the $\rho_{RL}/\rho_{RL,0}$-ratio *remains constant*. The first is interpreted as a result of a break-up of the tracks into segments too short to be distinguished in TL. The second observation can be accounted for in the same way but only if the RL-features are sections of broken-up fission tracks. The last empirical fact which puts it beyond doubt that the RL features are indeed tracks or track sections is that the track counts including both the TL tracks and RL features have standard deviations which in all investigated samples are close to those of a Poisson distribution (Table 3). That could happen once by accident but not 18 times and for close to 1000 counted areas. Strong claims require strong evidence, which we believe we supplied in this case. But the

	most convincing argument of all is that all FT labs are in possession of four samples identical to ours. Everyone wishing to investigate our conclusion can go to the microscope and put it to the test.
Figure 6. Between 0.7 and 1.0, track density for RL is higher than TL. This means that the track identification criteria are different. Please describe the identification criteria for minimum track, at least.	Corrected; we explain our track counting criteria, which are straightforward, in the revised manuscript
Table 3 and Figure 6. Are there data at 450 ° C for total annealing? I am very concerned about the density of track-like defects.	Yes and no. All the samples with induced tracks had been pre-annealed at 450° before neutron irradiation. We did not put a sample apart before the neutron irradiation because the sections were intended for a different experiment. As we explained above, an annealed but un-irradiated sample would not be conclusive, and we gave compelling reasons for our interpretation of the RL features as fission tracks ($1 > \rho/\rho_0 > 0.70$) or track sections ($0.7 > \rho/\rho_0 > 0$). We question, however, that the RL features need to be a cause for great concern, as lossless fission-track counts were never possible, as the reviewer's own work on standardless FT dating shows.

Freiberg, 16 November 2021.

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