

Dear Matt Fox and coauthors,

Thank you for your responses to the five original reviews that your manuscript received, and for patience as I sought additional reviewer feedback to the revised manuscript, which was provided by Olivia Thurston.

Thank you for helping to get this moving. It has been a long review. We also apologise that we took too long to get the revised manuscript back.

In general, I think you all have done a good job responding to the original reviews, although some points raised by the reviewers were left unacknowledged, and I think the revised manuscript is stronger than the original manuscript.

I do not necessarily agree with reviewer Thurston that you must use the revised Guenthner (2021) version of ZRDAAM that incorporates new data from the Ginster (2019) thesis into the analysis presented here, particularly considering when the original modeling efforts for this manuscript were performed. However, it appears that a Ginster-based model is available in recent versions of QTQt. If the authors prefer to use the Guenthner et al. (2013) model, they need to provide a rationale in the manuscript (e.g., because this is the model that the papers about Great Unconformity papers used, for example). Regardless, I do think it is appropriate for this manuscript to cite the Ginster (2019) thesis and the Guenthner (2021) incorporation of its results in places where the manuscript calls for new diffusion experiments kinetics data (e.g., lines 399-402). This would acknowledge that newer data do exist even if they have not been incorporated into the specific modeling efforts of this paper.

We have said that we use the Guenthner (2013) model because this is the one that was used in the papers we cite. We apologise for the oversight here. The main conclusions do not depend on the model at all. Instead, they depend on the new inversion and our ability to directly propagate uncertainties from the diffusion models to the thermal histories. This has not been possible before.

I encourage you to consider the other comments from reviewer Thurston about using more specific language in places. I will leave it to the authors' discretion about whether to add more discussion explicitly about implications for the timing and significance of the Great Unconformity. In my opinion, the general conclusion that uncertainties in the kinetics parameters make it challenging to argue for one Great Unconformity hypothesis or another is sufficient. In addition, please consider the following line comments from my read of the revised manuscript, in addition to the line comments from reviewer Thurston. The current manuscript is a little bit informal and unpolished in places. I have tried to provide comments on where things can be tightened up, but I also encourage the authors to do a close read when submitting their revisions.

We thank Dr Thurston for her helpful suggestions and have included most of these in the revision.

Lines 14-15: Rather than ‘...geological feature that has eliminated a time period of almost a billion years...’ I recommend phrasing as ‘...geological feature representing a period of missing time of almost a billion years...’

Changed.

Lines 28-30: I recommend adding a few key references here.

We have cited Rieners and Brandon here.

Line 30: I recommend deleting the sentence beginning ‘in many cases’ as the manuscript has moved away from describing the Great Unconformity as an event.

Deleted.

Line 49: Remove samarium as this is generally considered negligible and not measured for the ZHe system.

Deleted.

Lines 57-58: Rephrase from ‘the erosional event’ to just ‘erosion’.

Corrected.

Lines 38-40: Something is missing/wrong in this sentence. Maybe the word ‘for’ is needed between ‘however’ and ‘many’? Also, reheating events are not required for different thermal histories to yield the same thermochronometric age.

We have clarified this sentence.

Line 59: This sentence as written implies that McDannell et al. (2022) is the only recent work to use thermochronometry to understand the origin of the Great Unconformity. Please reframe/rephrase and include other relevant references.

We have added a few more references here.

Lines 67-69: This sentence reads out of place here between two sentences focused only on ZHe/ZRDAAM. Consider moving this sentence before the previous one.

This was suggested here by the previous reviewers and we feel that it fits in ok.

Line 96: Consider adding ‘based on the same underlying data’ after ‘present a new calibration’ at the end of this sentence.

Added this suggestion.

Lines 146-147: Can you give an example of a parameter/aspect of ZRDAAM that you are thinking about here?

WE have added “these” so that it is clear that we are talking about the 4 damage parameters described directly above.

Line 169-170: I believe it be more consistent to say ‘minimally damaged and extremely damaged’ rather than ‘undamaged and damaged’ here.

Corrected.

Line 205: specify ‘extremely’ damaged N17 crystal

Corrected.

Lines 208-209: I agree with reviewer Thurston that this sentence is vague; can you be more specific about what you mean by ‘not accurately predicted’?

We have changed this sentence to this. “If this uncertainty is not decreased, the frequency factor and activation energy of this crystal are not predicted.”

Line 229: These parameter values need to be reported with units.

Added units.

Lines 230-243: Please provide headers/labels for each column and row in the covariance matrix and correlation matrix to make these tables more readable.

Corrected and made these into Table 1 and Table 2.

Lines 269-271: I don’t understand what the authors are trying to describe in this sentence. Please clarify or refer to a figure where the reader can see what you are describing.

We have clarified this.

Lines 286-287: ...such as? Please give an example of the important factor(s) or remove this sentence.

We have added some of the main causes of dispersion here.

Line 292: Reference should be Guenthner et al. (2013).

Corrected.

Line 295: Rephrase ‘The assumption of Gaussian’ to ‘The assumption of a Gaussian distribution’.

Corrected.

Lines 301-303: Please describe how these 25% uncertainties compare/differ from the reported uncertainties.

These are the same uncertainties as used by McDannell.

Lines 322-324: Here I tend to agree with one of the original reviewers Becky Flowers that it would be helpful to show how well the predicted and observed data agree, particularly if you are going to discuss differences in the fit between observed and predicted dates. Or, if you don't want to show this comparison, providing the likelihood/log likelihood values for these different model results would be helpful.

This plot has already been shown in the original papers.

Lines 347-349: This sentence reads awkwardly and informally; please revise.

We have modified this sentence.

Line 368: Specify figure 6C.

Corrected.

Line 370: 40 or more crystals.

Corrected.

Line 380: Replace '(RAMPED)' with an actual reference.

Oh, yes. We apologise for this oversight. We have written "One promising approach to estimate the amount of zonation and anomalous diffusion behaviour could be to screen crystals using ramped degassing experiments (Idelman et al., 2018; Guo et al., 2024)."

Lines 399-407: The authors have left out what to me is an alternative but important solution: obtaining grain-specific diffusion parameters, i.e., performing zircon $4\text{He}/3\text{He}$, rather than having to obtain many dozens or hundreds of dates across a wide range of eU. Consider mentioning $4\text{He}/3\text{He}$ in this discussion.

We have included a reference to "Tripathy-Lang, A., Fox, M., and Shuster, D. L.: Zircon $4\text{He}/3\text{He}$ thermochronometry, *Geochim. Cosmochim. Acta*, 166, 1–14, <https://doi.org/10.1016/j.gca.2015.05.027>, 2015." here.

Code availability: Is it not possible to provide input files for the QTQt model simulations in Figure 5? It would also be helpful to be more specific about code access, as I'm assuming that readers would specifically need to contact Kerry Gallagher for a version of QTQt that allows for the diffusion kinetics parameters resampling (as well as the standard version of QTQt).

We have added “Please contact Kerry Gallagher to obtain the version of QTQt with our new damage parameters and uncertainty estimates.”

Figure 1 caption: Please specify that the data are represented by circles and that the lines represent model fits, and that both the data and model fits are colored by alpha dose.

We have added this suggestion.

Figure 2 caption: Use extremely damaged and minimally damaged rather than amorphous and undamaged. Also in the Figure 3 caption.

We have changed this to minimally damaged. But it is actually an undamaged crystal.

Figure 3: Change y-axis units to kJ/mol to be consistent with figure 2.

We use J/mol here to be consistent with the covariance matrix.

Figure 4 caption: Cite Guenthner et al. (2013) as the ‘canonical values’. The caption explains what the red line is but not what the color map represents – please include this.

“The colours highlight the relative probability of obtaining a specific age for a given [eU] value.”

Figure 5 caption: Please specify that the McDannell et al. (2022) data are the inputs for these models. Please specify Guenthner et al. (2013), not just “Guenthner diffusion parameters.” Last sentence, specify ‘relative to the maximum posterior and likelihood models.’

Corrected.

Figure 6 caption: Explain that the tan region in panels A and B corresponds to the range of eU values shown in panel C. I don’t follow the argument of the last sentence in this caption – please clarify.

Corrected.

The manuscript “Uncertainty in Helium Diffusion in Zircon Limits Thermochronologic Resolution: Application to The Great Unconformity” by Fox et al. highlights one of the ongoing weak spots within the (U-Th)/He zircon thermochronometry system in that the underlying diffusion kinetics are underdeveloped. Though the paper does not provide direct updates to the diffusion kinetics in the form of diffusion experiments, it does provide a way to quantify possible uncertainties within the overall precision of thermal histories that currently rely on the diffusion and annealing kinetics coded into ZRDAAM (Guenthner et al., 2013). The authors highlight that larger uncertainties in temperature and timing for specific thermal events, particularly the Great Unconformity, may be

more appropriate than the timing of events that have been reported in the literature using (U-Th)/He zircon thermochronometry and ZRDAAM modeling. Though this is a valid critique that is supported by the figures presented in the paper, the authors do not expressly state how the new results impact the interpretation of the Great Unconformity. The authors also opt for brevity and ambiguous language in some areas that would benefit from more direct descriptions of the exact parameters utilized by the authors to determine thresholds within their resampling and propagation of uncertainty within the development of thermal histories. I would like for the authors to address these areas of ambiguity before publication of the final manuscript.

We have not discussed the Great Unconformity in more detail here. We do not want to distract from the key message – that uncertainties are important and underestimated in most thermal history models.

The first area that could use more clarity within the text is what version of ZRDAAM the authors utilized. Since the updated version of ZRDAAM (Guenther, 2021) incorporates more recent annealing kinetics from Ginster et al. (2019) than those used in the original 2013 version (Guenther et al., 2013), it seems imperative that the authors use this more updated and accurate version in their modeling efforts. Without the use of the most up-to-date version of ZRDAAM, the magnitude of uncertainties produced by the technique presented by the authors may be grossly inaccurate.

We have described why we use the original Guenther model in the revised manuscript. We did not use the Ginster data because it is not published. Our main point is that we have provided a means to propagate uncertainty from the diffusion experiments to the data prediction and then to the thermal model interpretations. This aspect of our work is not grossly inaccurate and can be easily adapted to any dataset.

The second area of concern is a lack of disclosure on the exact metrics used to determine thresholds within the new code presented by the authors. Within the text the authors use wording such as “efficient balance,” “not accurately predicted,” and “good enough” that leave some ambiguity as to precision used in developing the code. I assume that the authors did in fact choose statistically significant metrics for all aspects of their code, however, direct reporting of those metrics is needed for transparency and robust academic discussion of the new coding presented by the authors. The authors also use imprecise language when describing radiation damage levels of the zircon crystals used to develop both ZRDAAM and their own updated code. Given that both annealing and diffusion are dependent on the level of radiation damage present within the crystal, it is critical that the radiation damage level be understood and reported correctly.

We have attempted to correct these here. We have clarified that we use exactly the same radiation damage values as Guenther et al., 2013. We apologise for this oversight.

Finally, the authors do not make a clear through connection with the Great Unconformity from beginning to end of the paper, making the abstract and title feel

misleading. The authors do a good job of explaining and addressing the role that uncertainty propagation within the (U-Th)/He zircon modeling may play in the resolution of the timing of the Great Unconformity in the first half of the paper, however, the concept and term Great Unconformity are not mentioned in the results or implications of the paper. The paper would greatly benefit from the authors highlighting the role of the Great Unconformity in their choice of case study for their new code and in the implications of their code on the exact timing of the Great Unconformity as shown by their case study. I can appreciate that this paper is not an attempt to rewrite the results of the chosen case study, but implications of the propagation of uncertainty performed by the new code should be highlighted in reference to the Great Unconformity specifically as suggested by the title and abstract.

We do not want to do this and leave this up to other people. An updated version of QTQt has been developed and this could be used to update all the Great Unconformity results and conclusions. It could also be used to update other thermal histories one uncertainty has been added to the RDAAM and AFT systems.

Here I provide line-by-line comments to improve the clarity and flow of the text.

Line 17: Delete “in” from “(U-Th)/He in zircon.”

We have always used “in” as this is the convention.

Line 30: Sentence is vague, consider adding more context.

We have deleted this sentence.

Line 40: “Same age” is ambiguous. Same age as the measured value, same age as purely cooling histories?

It is not clear why this is ambiguous. This is why the thermal histories are non-unique.

Line 44: Insert “(4He)” after “helium” to connect back to abstract.

Corrected.

Line 47: Annealing should be mentioned along with radioactive decay even if it is not the focus of the remainder of the paragraph.

Corrected.

Line 49: Include element symbols (U, Th, Sm) to connect back to name of method.

Corrected.

Line 52: Clarify that diffusion rate is a kinetic parameter earlier in paragraph to ensure a clear through line.

Diffusion rate is not a kinetic parameter.

Line 82: Provide temperature system.

Corrected.

Line 132: Provide metric you use to determine radiation level. Are you relying on FWHM or do you introduce an alpha-dose calculation?

We now specify this and write “We use the same diffusion dataset, alpha damage values and parameterisation as Guenthner et al. (2013).” on line 163-164.

Line 195: How is 20% determined to be an area of efficient balance?

This is a standard acceptance rate for optimal MCMC algorithm performance. We have added a reference here.

Line 208: How are you determining accuracy?

We now specify “within error”.

Line 213-214: Stating that your model provides a better fit here contradicts the stated intent of the figure in the figure caption.

We have changed the figure caption to “The model parameters are the minimum misfit model parameters and represent a single realization of the parameter set we infer. A key point of the figure is to show the fits to the data and the data that are used to predict the model parameters, and to highlight that both models do not predict all the data, high-lighting the source of uncertainty.”

Line 218: Amorphous crystal refers to a metamict crystal, but line 132 refers to the crystal as “extremely damaged.” These are two different states for the crystal. Ensure consistency throughout the paper and with the measured radiation value of the original sample because the shift to fully metamict or amorphous has real implications for annealing and other kinetic processes.

Corrected.

Line 265: Spell out numbers used to start sentences. “Twenty” not “20.”

Corrected.

Line 269-271: Refer reader to corresponding figure/table.

We now refer to Figure 4.

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