Dating circulations of hydrothermal fluids in the crystalline basements of unconformity-related metallic deposits using in situ Rb/Sr geochronology : proof of concept

Pont-by-point reply to to the reviews:

Dear Editor (Klaus Mezger), Associate Editor (Noah M. McLean), and Reviewers,

We thank you for your careful evaluation of our manuscript and for your constructive comments. We have addressed all the technical points you raised. Please find below the point-by-point reply to your comments. Our replies are in blue. The line numbers in the replies refer to the version with the tracked changes.

Editor: Klaus Mezger

The language is ok, but not good enough to make an easy read for any reviewer. The text has too many grammar and wording problems that obscure your message. Widespread problems are the lack of definite and indefinite articles, mixing of times (past tense-present tense) for no obvious reasons, mismatching singular-plural for verbs, nouns. I suggest you use DeepL to correct the text. However, read it carefully afterwards to make sure it says what you want it to say.

The language has been systematically improved throughout the entire manuscript. We carefully corrected widespread grammatical and stylistic issues, including missing definite and indefinite articles, inconsistencies in singular-plural agreement for nouns and verbs, and unnecessary mixing of past and present tenses. Verb tenses were harmonized following standard scientific conventions, with the present tense used for established facts and interpretations, and the past tense reserved for specific results and methodological descriptions. These revisions were implemented manually and checked in detail to ensure clarity, readability, and preservation of the intended scientific meaning.

A serious problem is that you accept initial $^{87}Sr/^{86}Sr$ ratios below ~0.70. The lowest value in the solar system is 0.698, nothing can be below this value, as radiogenic isotope ratios go up with time, never down. The explanations given are thus impossible. Possibilities are rotations of the isochron by a geological process (Rb-loss proportional to Rb/Sr) or systematic miscalibration of Rb/Sr ratio measurements. This part needs to be reworked as it is impossible for a journal on isotopes to promote impossible isotope ratios.

We have completely reworked the treatment and interpretation of low $^{87}Sr/^{86}Sr$ intercept values. Values below the minimum solar-system ratio are no longer interpreted as physically meaningful initial isotopic compositions. Instead, they are now explicitly described as "regression intercepts" resulting from disturbed or open-system Rb-Sr behavior. The revised discussion explains that such intercepts arise from isochron rotation under non-conservative Rb-Sr redistribution during fluid-rock interaction, partial isotopic resetting, or proportional Rb loss relative to Sr. We further clarify that analytical or calibration-related biases can be excluded based on reference material reproducibility and internal consistency of the datasets. As a result, the manuscript no longer promotes impossible isotopic ratios, and the geological significance is now restricted to the regression slopes, which may still record meaningful timing information, consistent with established Rb-Sr literature.

Report data properly. Enough and not more decimal places so the error is in the last digit or last two digits. Errors and data must match: something like 1914 ± 43.78 does not make sense, it is either 1914 ± 44 (preferred) or 1914.00 ± 43.78 (too many digits). Fix this problem in the paper AND the Supplement!

All reported numerical data have been carefully revised to ensure consistency between values and uncertainties. Excessive decimal places have been removed, and uncertainties are now reported such that the error affects the last one or two digits, in accordance with standard geochronological practice. These corrections were applied consistently in both the main text and the Supplement.

In the supplement: what is this?: Rb85 mesuré. Is this a concentration, cps????? Isotopes are written as superscripts in front of the chemical symbol, not behind.

The Supplement has been clarified and corrected. Ambiguous labels (e.g., "Rb85 mesuré") have been properly defined, isotope notation has been standardized using superscripts preceding chemical symbols, and all tables and descriptions have been revised for clarity and consistency.

Associate Editor: Noah M McLean

Dear authors,

Thank you for your submission to GChron. This manuscript requires only a few technical corrections before heading out for review.

- Notably, the manuscript includes several extremely long paragraphs. See, for instance, lines 119-159, 186-242, 630-665, 752-825 (one paragraph across two pages), and 826-889. These may be the product of a copy/paste mistake when formatting the article for GChron. Please evaluate the article and ensure its paragraphs follow standard scientific writing style, referring to other articles from this journal if examples are required.

Lines 119–159 (now lines 119–162)

The paragraph originally spanning lines 119–159 has been reorganized and subdivided into several shorter paragraphs in order to comply with standard scientific writing practices and improve readability.

Lines 186–242 (now lines 159–247)

The long paragraph originally spanning lines 186–242 has been restructured into multiple paragraphs with clearer thematic separation.

Lines 630–665 (now lines 607–673)

The paragraph originally located between lines 630–665 has been shortened and reorganized to better follow the formatting style of GChron.

Lines 752–825 (now lines 688–757)

The very long paragraph originally spanning lines 752–825 (across two pages) has been divided into several shorter paragraphs to improve clarity and manuscript structure.

Lines 826–889 (now lines 760–898)

The paragraph originally spanning lines 826–889 has also been subdivided into shorter paragraphs following standard scientific writing conventions.

- On line 131, there is an “(e.g.,” without its closure.

Line 131 (now lines 131-132)

At line 131, the incomplete expression "(e.g.," resulted from missing bibliographic references. The omitted references have now been added and the sentence has been corrected accordingly.

- There is a duplicated sentence at line 218.

Line 218 (now lines 221-226)

The duplicated sentence originally present at line 218 has been removed from the manuscript.

Overall, the highly constructive and detailed referee comments agree that this contribution is of broad interest, presents high-quality measurements, and that its geological interpretations are appropriate and defensible. I agree with both reviewers that the submitted manuscript is thoughtfully and thoroughly referenced, and its detailed methods and results are sufficient to judge its conclusions. Author responses to the reviewers are also thoughtful and thorough. In particular:

The authors' proposed clarification to the propagation of uncertainty related to the reproducibility of the La Posta secondary standard and its communication in the manuscript is excellent. I'm not sure what the "weighted mean statistics, along with their dispersion" means precisely, but the ~1.3% (1σ) magnitude they report in 'AC1' looks about right. I look forward to reading the details in the Methods section of the revised manuscript. As the authors indicate, adding the analytical uncertainty (in quadrature) to the reproducibility of the secondary reference material ensures that the reported uncertainty best reflects the total uncertainty in these analyses.

Thank you for this positive assessment. We agree that the previous description was insufficiently explicit. The Methods section has been expanded to provide a detailed explanation of the procedure used to assess long-term reproducibility from repeated analyses of the La Posta secondary reference material over a three-year period. We now specify that for each analytical session, an isochron age was calculated independently. The resulting session ages were then combined using a weighted mean, in which each session age was weighted by the inverse square of its internal uncertainty. The dispersion of individual session ages around this weighted mean, quantified as the standard deviation of the weighted residuals, was used to estimate an external reproducibility of approximately 1.6% (2σ). We also clarify that final age uncertainties incorporate both internal analytical uncertainty and external reproducibility through quadrature propagation (σ_total = √(σ_internal2 + σ_reproducibility2)), ensuring that the reported uncertainties reflect the total analytical uncertainty of the method.

Adding detailed concentration calculations to the revised Methods section would be a welcome addition, considering that they differ from standard practice.

We appreciate this suggestion and have expanded the Methods section accordingly. A dedicated paragraph now describes in detail the procedure used to calculate 87Rb, 86Sr, and 87Sr concentrations from LA-ICP-MS/MS isotopic measurements. The revised text explains each calculation step, including the correction for natural isotopic abundances, derivation of Sr concentrations from measured isotopic ratios, and application of the matrix correction factor. For clarity and reproducibility, the corresponding equations have been added to the manuscript.

I appreciate the discussion in the reviewer comments and replies about where and how to address the apparent low initial 87Sr/86Sr isochron intercepts. The proposed changes, in response to Reviewers 1 and 2, appear adequate to address these concerns. I echoed Reviewer 1's surprise at the ± 1.7 Myr and ± 1.2 Myr uncertainties on the high-MSWD fits for altered muscovite in figure 7C-D. The text makes it clear that you do not interpret these uncertainties as geologically meaningful. I recommend eliding the last decimal places and the uncertainty on these high-MSWD fits to help others better understand your interpretation; for instance, reporting both as “ca. 1635 Ma”.

Thank you for this helpful recommendation. We agree that reporting highly precise internal uncertainties for regressions characterized by elevated MSWD values may inadvertently imply a level of geological significance that is not warranted. Accordingly, the manuscript has been revised to report these regressions as ca. 1635 Ma and ca. 1636 Ma respectively, consistent with our interpretation that the regressions define a broad age cluster rather than precise geological ages. We have also retained the discussion explaining that the elevated MSWD values indicate substantial excess scatter and limit the geological significance of the formal regression uncertainties.

Attaching the proviso about BIC-selected Gaussian mixture models to the revised manuscript is prudent and sufficient in this context. I agree that more sophisticated statistical treatments are beyond the scope of this paper. I look forward to seeing the proposed KDE plots for the uncertainty-binned data.

Thank you for your assessment. The manuscript now explicitly includes the caveat regarding the limitations of BIC-selected Gaussian mixture models and emphasizes that the resulting age populations should be regarded as exploratory statistical groupings rather than uniquely defined geological events. In addition, the KDE plots for uncertainty-binned datasets have been added to the revised manuscript.

The authors’ responses to Reviewer 2’s alternative regression considerations and suggestions are all reasonable. I appreciate the constructive back-and-forth on this topic from the authors and the reviewer.

We thank you for this positive evaluation. We found the discussion with Reviewer 2 highly constructive and have incorporated the agreed clarifications and explanations into the revised manuscript where appropriate.

The authors proposed changes to typographic errors and color schemes, as suggested by both reviewers, are all excellent.

We appreciate this comment. All typographic corrections identified by the reviewers have been implemented. In addition, the color schemes of the relevant figures have been revised to improve readability and visual consistency throughout the manuscript.

In addition to the reviewer comments: I appreciated the clarity and detail in Figure 9. Both reviewers commented that this contribution is “somewhat dense,” and I’d echo this sentiment. However, the text is not duplicative, and the sections are well-organized. Before resubmission, I’d recommend reading through the text once more to identify long paragraphs that might be divided into two, with the new paragraph getting a summative topic sentence. This revision helps readers navigate longer, more technical prose. I believe there is a place for detail-oriented science writing as you’ve provided here, but I think the easier you can make it to read, the more your article will be read and cited.

Thank you for this thoughtful suggestion and for your positive assessment of the manuscript structure and Figure 9. We carefully reread the manuscript with readability in mind and revised several of the longer paragraphs, particularly within sections 5.2 and 5.3 of the Discussion. Where appropriate, lengthy paragraphs were divided into shorter thematic units, and additional topic sentences were introduced to improve logical flow and guide the reader through the more technical sections of the text. These modifications were implemented without reducing the scientific detail of the discussion.

Overall, I suggest only minor edits, as outlined by the authors and discussed above, are required for publication of this manuscript in Geochronology.

Best wishes,

Noah McLean, AE

Referee#2: Jarred Lloyd

The contribution, while somewhat dense in some sections, and needs refinement to the ordering of some content, is a comprehensive and timely contribution to the geochronology community. In general it is very well written, although I am unconvinced by some statements made by the authors; however, these concerns do not detract from the overall rigour of the paper nor the overall conclusions. I appreciate the attention to detail and prior literature. This assessment is based on the current version of the preprint, prior to the changes the authors have stated they've implemented in their reply to referee 1.

From my reading of the contribution, the aim is to investigate the utility and application of the Rb–Sr decay system geochronology in micas as a time record of hydrothermal events. This is done using samples from a Proterozoic basin in Canada that is economically important for uranium and is known to have been subject to long lived hydrothermal events.

On the use of 1s/sigma level - I note that the other referee had queried this as well, and the authors have replied to this. I would prefer to see these uncertainties referred to at 2s level so they represent the uncertainty parameter at the interval in which ~ 95 % of any random sample within the distribution μ ± 2s would fall, rather than ~ 68 %. I take the point that the GMM (and all statistical tests for that matter) require the uncertainty at 1s during computation; however, IsoplotR does this conversion so long as the appropriate input uncertainty level is noted. It can also be output as 2s or 95 % CI with the appropriate flag. Given you are also comparing Rb–Sr to existing geochronology from other studies that used different decay systems, it is appropriate to use 2s plus the decay constant uncertainty for comparison. You also do quote 2s uncertainties for other data in the paper.

Thank you for this comment. We agree that reporting uncertainties at the 2σ level is more appropriate for comparison with geochronological data from other studies and more informative in terms of confidence intervals. We have revised the manuscript accordingly and converted all uncertainties to 2σ throughout the text. This conversion was performed directly from IsoplotR outputs using the appropriate 2σ flag and does not affect the underlying statistical treatment of the data, which was performed at the 1σ level as required by the GMM algorithm.

On the 5 populations and BIC. IsoplotR's documentation notes that increasing sample size often causes the implemented algorithm to overestimate the number of populations as sample size increases, "This option should be used with caution, as the number of peaks steadily rises with sample size (n). If one is mainly interested in the youngest age component, then it is more productive to use an alternative parameterisation, in which all grains are assumed to come from one of two components, whereby the first component is a single discrete age peak (e^m, say) and the second component is a continuous distribution (as described by the central age model), but truncated at this discrete value."

The BIC itself probably shouldn't be used here as some of its assumptions break down in this particular type of modelling. A much more robust measure like the Bayes Factor (although computationally heavy) should probably be used here; however, this particular aspect is beyond the scope of your manuscript.

Instead, you should visually assess your distributions, taking note of the peakfit algorithm's comment quoted here. I'd interpret your data (notably in fig 8), as log-normal distributions of data, or as normal distributions with continuous tail, so at most 2 distributions, not 5.

Thank you for this constructive comment.

We fully acknowledge the limitations of the BIC-based approach for selecting the number of Gaussian components in this context, and we agree that more robust criteria such as the Bayes Factor would be preferable in principle, although this is beyond the scope of the present manuscript.

We also acknowledge the warning in IsoplotR's documentation regarding the tendency of the peakfit algorithm to overestimate the number of populations with increasing sample size. This is a valid concern that we have taken seriously.

However, we wish to clarify several important points:

First, the five-component GMM was not used as the sole basis for identifying discrete geological events. As explicitly stated in the manuscript: 'the components should be regarded primarily as probabilistic centers, which we attempt to compare with published datasets from the literature.' We therefore do not claim that five discrete hydrothermal episodes are unambiguously resolved by the data alone.

Second, the five GMM components and radial plot populations are not interpreted independently, their geological significance is assessed through their correspondence with independently dated tectono-hydrothermal events documented in the regional literature (Fig. 10). Each age population is discussed in the context of existing geochronological constraints from U–Pb, K–Ar, and Ar–Ar chronometers applied to minerals from the same geological setting. It is this multi-system convergence, rather than the statistical decomposition alone, that underpins our geological interpretations.

Third, the KDE distributions shown in the revised Figure 8A are consistent with log-normal or asymmetric normal distributions with continuous tails. The dominant population at ca. 1640 Ma is robust and clearly identifiable regardless of the number of components assumed. The subordinate populations at ca. 1680, 1600, 1550, and 1467 Ma are acknowledged to carry greater uncertainty and are explicitly discussed as potentially reflecting either discrete hydrothermal pulses or artificial segmentation of an otherwise continuous distribution.

In response to this comment, we have added the following clarification to the manuscript: "It should be noted that BIC-based GMM algorithms tend to overestimate the number of populations with increasing sample size (Vermeesch, 2018), and that the present data are equally consistent with a smaller number of broader, partially overlapping distributions. The age components identified here are therefore not interpreted as discrete geological events but as probabilistic centers whose significance is evaluated through their correspondence with independently dated tectono-hydrothermal events documented in the regional record."

The uncertainty quartiles - why did you choose to plot these as histograms rather than density plots? Density plots aren't as sensitive to bin-width for visualisation (and you may as well fit distributions to them giving you mean and std). It looks like Q1 and Q2 have peaks (c. 1620 and 1645 Ma respectively) that correspond to the bimodal distribution of Q3, and then Q4 has a bimodal distribution with the lower peak on Q2 peak, and an additional peak at ~ 1800 Ma.

Thank you for this suggestion. We agree that kernel density estimates (KDE) are more appropriate than histograms for this type of visualization, as they are not sensitive to bin-width choice and provide a smoother representation of the underlying distributions. We have replaced the histograms in Figure 8A with KDE curves.

I'll note here, the "samples" data provided in the supplementary showed different single spot age ranges (overall 1353.6 to 1876.7 Ma) compared to fig 8(A) and in the text when I filtered by "Altered" and computed the quartiles based on this - did I miss a step in how you chose the data subset?

Thank you for catching this discrepancy. Upon careful review, we identified an inconsistency between the age values reported in the supplementary material and those used to generate Figure 8(A), which arose during the preparation of the supplementary file. The supplementary data have now been corrected to accurately reflect the dataset subset used in the analysis, and all reported age ranges are consistent throughout the manuscript. We apologize for any confusion this may have caused.

The low Sr/Sr - Firstly, the explanation should come prior to the discussion of their values, I went through a lot of the paper thinking these values are exceptionally low and not feasible for well defined Sr concentrations.

I find it difficult to reconcile these values with well-defined Sr concentrations, and cannot exclude the possibility of an analytical artefact (or artificially induced isochron rotation due to data processing), partly because the supplementary data I have does not contain the La Posta data, and I know (https://doi.org/10.5194/gchron-6-21-2024) using MicaMg increases the overall uncertainty of samples (unnecessarily) and incurs its own matrix offset from micas. I think there has been some misinterpretation of Dodson's paper in regard to the emphasis on radiogenic Sr - yes it controls the Rb–Sr geochronometer, but this is true of any radiogenic parent-child pair. My understanding of the statements in Dodson is they are more generally talking about the mobility of child nuclides (and those elements, e.g. Sr relative to Rb). Regardless of this, the observations the statements are made on (the Alps study) have since been shown to be a conflation of true volume diffusion (i.e. thermal closure) and hydrothermal alteration (e.g. https://doi.org/10.1046/j.1365-3121.1998.00156.x, https://doi.org/10.1144/jgs2021-098), and we still don't really have a good handle on Sr diffusivity in mica (very few studies, only one may not have conflated fluid-mediated influence with pure thermal diffusivity). In any case, IF the assumption that 87Sr is moved preferentially (I haven't got an issue with this, 87Sr that is a product of 87Rb decay will generally occupy the I site in a mica, and will be more readily moved out of that site) it still would not explain how you get to such low initial values. "Common" Sr is likely to be in the M-site (octahedral) rather than I-site. For Sr in the M-site, I see no reason for 87Sr to be preferentially removed in place of 86Sr or 88Sr, both of which should be more abundant (relative to "common" 87Sr) anyway. Removing Sr from the I-site (likely to be radiogenic 87Sr generated in the mica) should then bring the predicted initial 87/86Sr back to a feasible (i.e. chondritic or higher) value, unless the true initial Sr concentrations were close to zero (highly radiogenic, Sr poor micas (e.g. polylithionites) can show this effect). Most of your arguments suggests that alteration could slightly increase 87/86Sr in the presence of open system behaviour, and this is generally what is observed throughout the literature.

We thank you for these detailed and constructive comments, which we address point by point below.

1. Anomalously low intercept values

We agree that the anomalously low 87Sr/86Sr intercept values (~0.67) obtained from preserved muscovite regressions are physically impossible as true initial isotopic compositions, as they fall below the minimum value of the solar system (~0.698; Papanastassiou and Wasserburg, 1969). As stated in the manuscript, these intercepts are explicitly not interpreted as meaningful initial 87Sr/86Sr ratios, but rather as mathematical artefacts arising from open-system Rb–Sr behavior and isochron rotation under non-conservative redistribution of Rb and Sr, consistent with the framework discussed by Brooks et al. (1976), Villa (1998), and Faure and Mensing (2013). We acknowledge that the explanation of this interpretation should appear earlier in the manuscript, prior to the discussion of the intercept values themselves, and we have restructured the relevant section accordingly.

2. Use of MicaMg as primary reference material

We acknowledge the limitations associated with the use of MicaMg as a primary reference material, including the matrix offset relative to natural micas and the additional uncertainty it introduces, as documented by the study cited. However, we wish to emphasize that MicaMg currently represents the only widely available and internationally distributed nano-powder reference material specifically designed and validated for in situ Rb–Sr dating by LA-ICP-MS/MS. In the absence of a commercially available mica reference material with better matrix-matching properties and similarly well-constrained Rb–Sr isotopic composition, its use remains unavoidable. We have added a statement in the methods section acknowledging this limitation explicitly.

3. Interpretation of Dodson (1973)

We thank you for this important clarification. Upon re-reading Dodson (1973), we agree that his statements on the control of the geochronometer by radiogenic nuclides are made in a general sense applicable to any parent-daughter isotopic system, and are not specifically directed at the mobility of 87Sr relative to other Sr isotopes. We have revised the relevant sentence in the manuscript to avoid any misinterpretation of Dodson's original work.

4. Crystallochemical argument regarding site occupancy

We thank you for this particularly insightful comment, which we find scientifically compelling. You are correct that radiogenic 87Sr produced in situ by decay of 87Rb occupies the interlayer (I-site) in mica, substituting for monovalent cations such as K^+ and Rb^+, whereas common Sr (86Sr, 88Sr) is more likely to be hosted in the octahedral (M-site), where divalent cations such as Ca^{2+} are stable. As you rightly point out, preferential removal of radiogenic 87Sr from the I-site should, in principle, drive the regression intercept back toward chondritic or higher values rather than toward the sub-chondritic values we observe. To reach intercepts as low as ~0.67, the muscovites would need to have had near-zero common Sr concentrations from the outset, which is not the case here.

We therefore agree that the sub-chondritic intercepts are better explained as a consequence of isochron rotation under open-system conditions involving non-conservative, grain-scale redistribution of both Rb and Sr, a process well-documented in the literature (Brooks et al., 1976; Villa, 1998; Eberlei et al., 2015; Glodny and Grauert, 2009) — rather than as a direct result of selective 87Sr loss from the I-site alone. We have revised the discussion accordingly to reflect this more nuanced interpretation, removing the over-reliance on the Dodson framework and replacing it with a clearer discussion of open-system isochron rotation as the primary mechanism responsible for the anomalous intercept values.

With that said, I still think your overall interpretations and conclusions are valid and supported by the data available, and your explanation of how you handle these anomalous values is reasonable. I am just hesitant on the absolute ages derived from your data. Fixing the intercept to 0.71 +/- 0.03 for your "preserved metamorphic micas" and recalculating the age takes it from ~ 1782 Ma down to 1737 Ma (using a Mahon type regression) and does not increase the reduced chi-squared value to unacceptable values.

We thank you for this thoughtful comment and for performing an independent age calculation. We would like to address several methodological and geological points that we believe are critical for interpreting this result.

1. Regression method

Our isochron ages were calculated using the York regression as implemented in IsoplotR (Vermeesch, 2018), which is the current community standard for Rb–Sr geochronology and accounts for uncertainties in both 87Rb/86Sr and 87Sr/86Sr ratios as well as their error correlations. The Mahon-type regression differs methodologically from the York regression, and direct comparison of ages derived from these two approaches requires caution, as they may yield systematically different results depending on the data structure and the weighting of individual data points.

2. Anchoring the intercept

We respectfully argue that anchoring the regression intercept to a fixed value of 0.71 ± 0.03 is not appropriate in our geological context. As clearly demonstrated by Rösel and Zack (2022), fixing the initial 87Sr/86Sr ratio converts an isochron regression into an anchored single-spot-type calculation. This is fundamentally different from a free isochron, where both the age and the intercept are simultaneously and independently constrained by the data themselves. Rösel and Zack (2022) explicitly show that anchoring the intercept introduces an age bias that scales with the fraction of common 87Sr and with the deviation between the assumed and the true initial 87Sr/86Sr ratio. The larger this deviation, the larger the age bias introduced.

3. The choice of initial 87Sr/86Sr in a Paleoproterozoic polymetamorphic basement

This point is of particular importance in our study. The rocks of the Wollaston-Mudjatik Transition Zone are Archean to Paleoproterozoic crustal lithologies that have experienced multiple tectono-metamorphic events between ca. 1840 and 1720 Ma, as well as subsequent hydrothermal fluid circulations. In such a geological context, the initial 87Sr/86Sr ratio at the time of muscovite crystallization (ca. 1780 Ma) cannot be assumed a priori to be 0.71. Crustal rocks with long and complex pre-histories of Rb/Sr fractionation commonly develop elevated initial 87Sr/86Sr ratios well above mantle or primitive crustal values. As discussed by Rösel and Zack (2022), for crustal rocks the appropriate model range for the initial 87Sr/86Sr is 0.730 ± 0.030, reflecting the inherently large variability in evolved crustal sources. Imposing a fixed value of 0.71 without independent constraint from a co-genetic low-Rb phase would therefore likely underestimate the true initial 87Sr/86Sr ratio, resulting in a systematic underestimation of the calculated age, which is precisely what the independent recalculation appears to demonstrate.

4. Geological consistency of the obtained age

Our isochron age of 1780.8 ± 32 Ma (2σ, combined internal and external uncertainty) is fully consistent with independent geochronological constraints from U–Pb dating of monazite and zircon from the same basement terranes, which constrain the M2–D2 metamorphic event to ca. 1813–1770 Ma (Annesley et al., 1992, 1997, 1999; Jeanneret et al., 2017; Toma et al., 2024), thereby confirming the geological significance of our Rb–Sr age.

For all of the above reasons, we maintain that the free isochron approach using the York regression in IsoplotR, without anchoring the intercept to an assumed initial 87Sr/86Sr composition, is the most appropriate and unbiased method for our dataset, and that our reported age of 1780.8 ± 32 Ma (2σ, combined internal and external uncertainty) is robust and geologically meaningful.

Minor corrections

Terminology - as per Zack & Gilbert (2024) (https://doi.org/10.1016/B978-0-443-18803-9.00014-6), I strongly encourage the use of LA-ICP-MS/MS (yes the Agilent machine is named QQQ-MS, but it is generally configured as a QOQ-MS) to cover all varieties of ICP-MS with controlled mass shifting capabilities.

Thank you for this comment. We have updated the terminology throughout the manuscript from QQQ-MS to LA-ICP-MS/MS, in line with Zack & Gilbert (2024).

Consistency of Ma/Ga - I'd understand switching to Ga use if you were generally talking about < 1000 Ma and then a much larger value above this, but Ga / Ma are used inconsistently throughout. I'd stick with Ma as the uncertainties on Rb–Sr dates are usually around 1–5% and ± 20 Ma is easier to read than ± 0.02 Ga.

Thank you for this comment. We have revised the manuscript accordingly and converted all age values to Ma throughout, ensuring full consistency in the use of time units.

Stylistic, but ranges or from-to should use an en-dash, not hyphen, e.g. Rb–Sr should be Rb–Sr, 2050-1860 Ma should be 2050–1860 Ma...

Thank you for this comment. We have carefully revised the manuscript and replaced hyphens with en-dashes throughout for all ranges and intervals (temperatures, pressures, ages, isotopic systems, etc.), in line with standard typographic conventions.

Units: while bar is a metric unit, Pa (MPa) is the SI unit and should be used preferentially. I understand bar may have been used for historic reasons, but in the SI system and other scientific fields its continued use is discouraged. All other units used are SI units anyway.

Thank you for this comment. We have updated all pressure units throughout the manuscript to SI units: bar and kbar values have been converted to Pa and GPa respectively, in line with SI conventions.

L530 - The first part of this sentence should be in the methods (I believe you may have made this change already) and "using and applied" is probably meant to be "using an applied". If this is a true "matrix correction factor" not just a mass bias correction, I'd be very careful about using MicaMg for micas in this way as MicaMg is not crystallographically similar to a mica, and as per previous, incurs its own offset. It would be much simpler to calculate these concentrations using NIST SRM 610 and an internal standard as per Iolite, LADR, literature etc.

Thank you for this comment.

The first part of this sentence has been moved to the methods section (section 3.2.4), as suggested.

The typographic error 'using and applied' has been corrected to 'using an applied' in the revised manuscript.

Regarding the matrix correction factor, we acknowledge that the use of MicaMg for calculating elemental concentrations introduces an approximation, as MicaMg is a nano-powder reference material and is not crystallographically identical to natural muscovite. However, this correction factor was applied consistently across all samples and reference materials, and its influence on the calculated concentrations is proportional and reproducible. The long-term reproducibility of La Posta biotite over three years of analytical sessions, yielding ages and intercepts in excellent agreement with published ID-TIMS values, independently confirms that no significant systematic bias is introduced by this approach. We note that the concentration data are used exclusively for illustrative purposes in this study and do not form the basis of any geochronological interpretation, which relies solely on the measured isotopic ratios.

L620 - "Some of these elements remain in and are ... whereas the rest leave ..." (suggested change for grammar)

L650-652 Thank you for this suggestion. The sentence has been corrected accordingly: "Some of these elements remain in the system and are in situ transferred to the newly-formed clays, whereas the rest leave the system."

Wish list

As per the guidance on Geochronology's author guide, figures should endeavour to use colour schemes that are colour vision deficiency friendly. This should be best practice for authorship in the modern era, as free tools are readily available for the generation and use of CVD-safe colour schemes which also enhance the accuracy of perception/interpretation.

Figure 1 this may be more difficult as TMI/gravity images sourced from government surveys tend to be rainbow coloured, but it isn't impossible. The use of red lines to annotate in panel A further decreases the accessibility of this figure for people with CVD. Also, I'm assuming the CRS is WGS84 - please put the correct CRS in the caption or on the maps - without a CRS, coordinates can be impossible to accurately reproduce.

Thank you for this helpful comment. We agree that the original use of red lines and the lack of coordinate reference information reduced the accessibility and reproducibility of the figure. We have revised Figure 1 accordingly: the graphitic conductors are now displayed in magenta to improve accessibility for readers with color vision deficiency, and the figure caption now explicitly states that the map is projected in WGS 84. In addition, we have updated the color palette of the RMI images to a Viridis-based scheme to improve overall accessibility and ensure better perceptual uniformity.

Figure 3 - being true colour polarising microscopy images the concerns re. colour schemes are not applicable here.

Figures 5 & 6 - rainbow colour schemes really aren't appropriate here for count intensity. Firstly, they are not accessible, and secondly they actively hinder easy, intuitive and accurate interpretation by the reader; what is green, yellow etc - we perceive certain colour wavelengths better than others giving a false perception of greater range. Simple grey scale images would suffice here, but if you want to have them coloured use a single colour per image and map dark to low count, light to high counts.

Thank you for this helpful and important comment regarding the color schemes used in Figures 5 and 6.

We agree that the original rainbow color scale was not optimal in terms of accessibility and could hinder accurate visual interpretation of count intensity. In response, we have revised both figures accordingly. The updated versions now use a more appropriate color mapping that improves readability and ensures a clearer, more intuitive representation of the data.

Specifically, we have replaced the previous rainbow scheme with a perceptually consistent scale, mapping lower counts to darker tones and higher counts to lighter tones, as suggested. This change enhances both accessibility and interpretability for the reader.

We appreciate your suggestion, which has significantly improved the quality of the figures.

Referee#1: Anonymous Referee

There is a lot to unpack in this contribution and I commend the authors on a comprehensive synthesis of this timely topic. The writing is excellent if somewhat dense in spots. I appreciate the attention to previous literature. The design of the project is thoughtful, and the data is collected using highly suitable methods. Incorporating textures and compositional data to support in situ Rb-Sr is essential and the authors do an overall excellent job of contextualizing the isotopic data. This is absolutely required in a setting like the Athabasca Basin where prolonged high-T to low-T hydrothermal activity has created a wide range of alteration assemblages during basin formation and readjustment. Integrating the in situ Rb-Sr data with models for U deposition is very novel indeed. Also particularly noteworthy is the suggestion, supported by the data, that partly altered white micas underwent preferential liberation of 87Sr* relative to non-radiogenic Sr. This could be a excellent case to follow-up using atom probe measurements to examine the distribution of Rb vs. 87Sr* and 86Sr in the mica lattice.

The only suggestion I can make is that the order of presentation of some important text could be improved. For example we read through all of the results and note that the initial Sr intercepts are impossibly low. I was questioning to myself if this was an analytical bias. It is only in the discussion that the robustness of La Posta secondary material is described which allays some fears of analytical bias. So perhaps it would be good to state more clearly the reproducibility of La Posta age AND initial Sr intercept at the end of the methods section.

We thank the reviewer for this constructive suggestion regarding the presentation of the La Posta secondary standard. In response, we have added a dedicated paragraph to the Methods section detailing the long-term reproducibility of both the isochron age and the initial 87Sr/86Sr intercept of La Posta. This section now includes a compilation of analytical sessions acquired over a three-year period. For each session, isochron ages and corresponding initial Sr isotopic ratios were calculated, and the resulting values were subsequently combined using weighted mean statistics, along with their associated uncertainties and dispersions. These results demonstrate robust long-term reproducibility of both age and initial Sr isotopic composition, and support the absence of instrumental drift, calibration bias, or matrix-dependent effects. We believe that this addition clarifies the reproducibility of La Posta prior to the Results section and addresses the reviewer’s concern.

Additionally, we provide the full results of this compilation as supplement attached to this response, in order to further document and illustrate the absence of analytical bias.

There are a few other minor things I’ve highlighted below. Notwithstanding these the manuscript is in very good shape and illuminates some very important processes affecting a world-class U camp.

Line 154: switching here from xxxx Ma to x.xx Ga.

We have revised Line 154 to report the ages in Ma rather than Ga, in order to maintain consistency with the other age values expressed in Ma throughout the manuscript.

Line 167: please consider italicizing P and T here and elsewhere

We have italicized P and T in Line 167 and throughout the manuscript for consistency with standard scientific notation.

Line 336-340: La Posta is listed as a secondary standard. But it is not obvious from the text how well it was reproduced based on NIST610/MicaMg-NP external calibration? I also note that no additional error was added to the final isochron ages and they are quoted as 1σ starting on line 456. This is a bit unconventional, with most studies using in situ Rb-Sr citing 2SE and including additional error reflecting the long-term reproducibility of secondary standards. Is the 1σ required to use the Gaussian Mixing Model? (this looks a lot like Isoplot 'unmix age' routine). What happens to this unmixing exercise if you use 2SE rather than 1σ? This gets back to La Posta results. What was the measured error on La Posta in this study? We are only given the Zack & Hogmalm 2016 reported value.

First, regarding the 1σ uncertainties reported for the isochron ages: in IsoplotR, the Gaussian Mixture Model (GMM) routine requires that each data point be associated with a 1σ uncertainty. These internal uncertainties are therefore used in the GMM analysis to weight contributions of individual analyses and to model the age distributions statistically. While some studies report 2SE to provide a conservative estimate of uncertainty, using 1σ is required for the GMM routine, and it accurately reflects the internal precision of the regression.

Second, to demonstrate long-term reproducibility, we compiled results from repeated analytical sessions of the La Posta secondary standard acquired over a three-year period. For each session, isochron ages and corresponding initial 87Sr/86Sr intercepts were calculated, and the resulting values were combined using weighted mean statistics, along with their dispersion. These results confirm stable and reproducible measurements. The total uncertainty on the reported isochrone ages is obtained by combining the internal regression error (1σ) with the external reproducibility derived from La Posta (~1.3%).

These additions are now described in a dedicated paragraph in the Methods section, making clear that the measured 87Sr/86Sr ratios and derived ages are robust, and that the reported 1σ uncertainties are both appropriate for the GMM analysis and consistent with long-term analytical reproducibility.

Line 499: Here and elsewhere in Results section I'm surprised to see some methodology details related to concentration calculations. Methods for calculating concentrations should be in an earlier section. If NIST610 was analyzed it would be a simple matter using the Trace Elements DRS in Iolite4.

We thank the reviewer for raising the point regarding the calculation of concentrations. We acknowledge that standard practice often employs the Trace Elements DRS routine in Iolite to derive concentrations automatically. However, in this study we calculated ^{87}Rb, ^{86}Sr, and ^{87}Sr concentrations using a rigorous, reproducible workflow based directly on measured isotopic ratios, regression outputs from Rb–Sr diagrams, and matrix corrections derived from the internal standard MicaMg.

To ensure transparency and clarity, we will add a dedicated paragraph in the Methods section detailing this calculation procedure, including the derivation of 87Rb from 85Rb, the calculation of 86Sr and 87Sr from measured ratios, and the application of the matrix correction factor. We believe that this addition will fully address the reviewer's concern while demonstrating that our concentration calculations are scientifically robust and conform to standard practices in LA-ICP-MS geochronology.

Line 523: Is 1634.7 ± 1.7 Ma - an error of 0.1% - realistic? Again, how well as the secondary standard measured?

We thank the reviewer for raising this important point regarding the apparently small uncertainty associated with the isochron age.

The reported uncertainty of ±1.7 Ma corresponds to the 1σ internal regression error calculated by IsoplotR from 224 individual analyses defining the isochron. Although individual single-spot ages carry uncertainties on the order of ~20 Ma (1σ), the uncertainty on the isochron age reflects the statistical precision of the regression slope, which improves with the number of analyses. For a dataset of this size, the expected reduction in uncertainty follows approximately $1/\sqrt{n}$. Given ~20 Ma single-spot uncertainties and n = 224 analyses, the theoretical reduction in uncertainty yields: $20 / \sqrt{224} \approx 1.3$ Ma, which is consistent with the reported ±1.7 Ma. Therefore, the small reported internal uncertainty arises from the strong statistical constraint on the regression slope provided by the large number of analyses, rather than from unrealistically low analytical uncertainty on individual measurements.

We also note that the internal regression uncertainty does not account for long-term analytical variability. To provide a realistic estimate of the total uncertainty, the external reproducibility derived from the La Posta secondary standard (~1.3%, 1σ) is propagated. Combining the internal regression error with this external uncertainty yields a more representative total uncertainty on the isochron age, which in this case corresponds to approximately ±21 Ma (1σ). This ensures that reported ages reflect both the statistical precision of the regression and the long-term reproducibility of the analytical protocol.

Suggested statement for the manuscript:

"The Rb–Sr regression yields an age of 1634.7 ± 1.7 Ma (1σ internal regression error; the total uncertainty including external reproducibility is ~±21 Ma, 1σ)."

We have clarified in the revised manuscript that the quoted uncertainty represents the internal regression error (1σ) derived from the isochron fit.

Line 645: "…the mobility of radiogenic 87Sr* is significantly greater than that of Rb and non-radiogenic Sr…" The argument is that 87Sr* is preferentially located in interlayer site which is where alteration processes might start. Preferential removal of 87Sr* would, therefore, lower the 87Sr/86sr to values unsupported on earth. But the least-altered muscovite in basement gneiss also gives initial of 0.6709 ± 0.0115. This doesn't make sense. The authors claim to have ruled out instrumental effects but is there some cryptic matrix-mismatch adversely affecting corrected 87Sr/86Sr? Some statement about the anomalously low initial in least-altered muscovite would be warranted (unless I missed it somewhere in the text)

Thank you for this comment. We believe there may be a misunderstanding, as the explanation for the anomalously low intercepts in the least-altered muscovite is already provided in the manuscript (Lines 686–705).

In that section, we explicitly state that the regression intercepts (e.g., 0.6704 ± 0.0075) fall below the minimum solar system 87Sr/86Sr value (~0.698) and therefore cannot represent physically meaningful initial isotopic compositions. We clearly indicate that these values are not interpreted as true initial 87Sr/86Sr ratios, but rather as apparent intercepts produced by disturbed Rb–Sr systematics.

We further explain that the well-correlated regressions, combined with low MSWD values (<1) and high p-values (p >> 0.05), indicate strong internal coherence despite the anomalous intercepts. We interpret this behavior as characteristic of rotated or disturbed isochrons formed under open-system conditions, where non-conservative redistribution of Rb and Sr (e.g., proportional Rb loss, selective Sr mobility, or partial isotopic resetting) modifies the intercept while preserving a geologically meaningful slope.

Line 697: I see here that the authors explain that age and initial intercept for La Posta were within accepted range and that anomalously low 87Sr/86Sr are not analytical artifacts. It would be nice to know this before launching into the results section where these anomalously low 87Sr/86Sr raise flags about this possibility.

Thank you for this constructive suggestion.

We agree that informing the reader earlier about the reproducibility of the La Posta secondary standard would improve clarity and help avoid potential concerns when encountering the anomalously low $^{87}Sr/^{86}Sr$ intercepts in the Results section.

Accordingly, we have added a dedicated paragraph to the Methods section explicitly detailing the analytical performance of the La Posta biotite secondary standard. This addition specifies both the reproducibility of the obtained age and the consistency of the initial 87Sr/86Sr intercept relative to accepted values. By presenting this information upfront, prior to the Results section, we aim to clearly demonstrate that the anomalously low intercepts observed in some samples are not attributable to analytical bias, calibration issues, or standardization problems.

We believe this modification strengthens the manuscript structure and improves the logical flow between the Methods and Results sections. Thank you again for highlighting this important point.

Line 723: typo in isochrone

The typo in "isochrone" (Line 723) has been corrected in the revised manuscript.

Line 728: back to x.xx Ga rather than xxxx Ma ages…

We have revised Line 728 to report the ages in Ma rather than Ga, in order to maintain consistency with the other age values expressed in Ma throughout the manuscript.

We believe that these revisions have further strengthened the manuscript and that it is now ready for publication in Geochronology. We sincerely thank you again for your insightful comments and for your positive assessment of our work.

Yours sincerely,

Quentin Boulogne and co-authors