

## Experimental constraints on the sources of lithium-rich granites and pegmatites

Corresponding Author: Mr Bence Horányi

**This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.**

**Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.**

This manuscript has been previously reviewed at another Nature Portfolio journal. This document only contains reviewer comments and rebuttal letters for versions considered at Communications Earth & Environment.

Version 0:

Decision Letter:

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Dear Mr Horányi,

Your manuscript titled "Lithium enrichment in granites and pegmatites: a reflection of their crustal source" has now been seen by 3 reviewers, and we include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication. In particular please ensure that you:

**\*\*Strengthen your argument for the anatectic model by addressing the detailed comments of Reviewer #1\*\***

**\*\*Fully address the bauxite in the context of feasibility as a source for pegmatites due to its rarity\*\***

**\*\*Expand on the viability of staurolite to participate as a reactant to melt generation\*\***

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

Please submit your point-by-point responses as a separate file, distinct from your cover letter where you can add responses to the Editors' comments that you do not want to be made available to the reviewers. Word files are preferred. We recommend that any figures, tables or graphs that are included in the response to reviewers are also included in the main article or Supplementary Information.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and the completed checklist:

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We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

Please do not hesitate to contact us if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Evan Hastie, PhD  
Editorial Board Member  
Communications Earth & Environment  
[orcid.org/0000-0001-9647-2034](https://orcid.org/0000-0001-9647-2034)

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## REVIEWER COMMENTS:

### Reviewer #1 (Remarks to the Author):

Lithium enrichment in granites and pegmatites: a reflection of their crustal source

Horányi et al.

The following text in italics was extracted from "Granite-Related Ore Deposits" by Černý et al. (2005), which appeared in the 100th anniversary volume of Economic Geology. I was a distant fourth author on that work, but the extracted comments below are Černý's alone.

About the anatectic model, Černý et al. (2005) said this:

"Anatectic proposals, applied to the origin of individual LCT pegmatites, collide with numerous problems: (1) extremely low percentage partial melting of protoliths would be required to generate elevated concentrations of rare elements in the anatectic magmas, and concentrations encountered in pegmatite deposits would be impossible to achieve (London, 2005a); (2) low-percentage melting would involve mainly quartzofeldspathic components and muscovite of the protoliths but most mafic phases that contain HFSE would remain (or, in the case of biotite, re-equilibrate) in the residuum (e.g., Icenhower and London, 1995); (3) protoliths enriched in rare elements are occasionally invoked (such as evaporites), but they are rare, poor in HFSE, subject to depletion at low grade metamorphism (Li, B) and virtually absent in Archean shields (e.g., Černý, 1991b, 1992a); (4) metamorphism of typical host rocks, low-pressure upper-greenschist to medium amphibolite facies, is too low to generate granitic pegmatites in situ, and temperatures of the host rocks at the time of pegmatite emplacement are very much below those of the peak conditions of the low-grade metamorphism observed (Morgan and London, 1987; Chakoumakos and Lumpkin, 1990); (5) vapor-absent to water-saturated melting styles in higher grade rocks, capable of generating diverse granitic magmas, do not show enrichment in LCT elements comparable to the concentrations found in appreciably fractionated pegmatites, either in experimental work or in nature (e.g., migmatites; summarized in Černý, 1998); (6) elevated concentrations of rare elements could be achieved by very low percentage anatexis, but the volumes of source protoliths would need to be enormous and the problem of extracting the melt insurmountable (Černý, 1992a); (7) the saturation concentrations of beryl, columbite-group minerals, cassiterite, pollucite, and LiAl silicates in quartzofeldspathic melts are very high and impossible to be achieved without protracted fractionation of the parent medium (e.g., London, 2005a); (8) the general purity of rare element pegmatites is at odds with a potential history of melts seeping through country rocks during a largescale segregation; chemical and phase contamination would be expected but is not observed (Černý, 1992a); (9) stable and radiogenic isotope signatures of rare element pegmatites contradict direct anatexis from, or equilibration with, enclosing country rocks (e.g., Longstaffe et al., 1981; Taylor and Friedrichsen, 1983; Tomascak et al., 1992, 1998); and (10) anatectic generation of rare element pegmatites would locally produce rocks corresponding to arrested, incomplete evolution, but such cases of failed genesis are not known."

Černý became adamant about the granite-pegmatite paradigm (Černý et al., 2005) in the face of the emergent anatectic model:

"The generation of pegmatites by magmatic differentiation is supported by field-based physical evidence, geochemical criteria, and experimental petrology, including: (1) granite-cum-pegmatite suites have commonly intruded into low-pressure greenschist- to amphibolite-facies country rocks, above the regional depth of anatectic conditions (e.g. Beus, 1966; Černý, 1989b, 1990); (2) rare element pegmatites form aureoles of marginal and exterior dikes surrounding granite intrusions (e.g., Černý, 1992b); (3) highly evolved exterior pegmatite dikes are locally physically linked to the interior or marginal parts of granitic plutons (e.g., Beus, 1948; Černý, 1991c); (4) interior pods and dikes of pegmatites enclosed in, and evolved from, a parental granite are commonly impossible to distinguish from exterior dikes, in terms of structure, mineralogy, and geochemical features (e.g., Černý et al., 1981, 2005); (5) continuity of geochemical evolution is documented in numerous cases of granite-to-pegmatite suites (e.g., Černý et al., 1981; O'Connor et al., 1991; Trumbull, 1995; Mulja et al., 1995); (6) the fractionation path shown by granite-to-pegmatite sequences is matched by observations on rhyolite and ongonite suites (e.g., Kovalenko and Kovalenko, 1976; Pichavant et al., 1987; London, 1995; Raimbault and Burnol, 1998); (7) bulk compositions of rare element pegmatites are generally granitic and correspond to the minima established in relevant experimental systems (Ab-Qz-Or, Ab-Ecr (eucryptite)-Qz, with or without Rb, Cs, B, F, and/or P added (e.g., Stewart, 1978; summarized in Černý, 1991b; cf. Table A6); (8) all paragenetic categories of the complex pegmatites are easily explained by relatively minor shifts in pressure and temperature conditions (London, 1984) or in chemical potentials of a few components in their parent melts (e.g., London, 1982; reviewed in Černý, 1991b); (9) saturation in beryl, pollucite, tourmaline, and LiAl silicates can be achieved only by protracted, even multistage fractionation of granite-to-pegmatite suites (summarized in London, 2005a); and (10) experimental evidence shows that liquidus and solidus of pegmatite-forming melts decrease with increasing contents of Li, Rb, Cs, F, B, P, and H<sub>2</sub>O (reviewed in Černý, 1991b), which supports the contention that batches of progressively fractionated magma can be generated by protracted differentiation of a relatively primitive starting melt."

I'll get down to the review.

(1) The premise of the article is that if one partially melts a highly anomalous, lithium rich rock (a bauxite!), then one can obtain an anomalously lithium-rich partial melt. This does not seem to be a novel finding. Lithium-rich pegmatites as so common in so many terrains of different ages that their generation cannot be dependent on a pre-enriched source as rare as meta-bauxite.

(2) There is nothing wrong with the experimental work or its analysis as far as I can see, but I didn't look closely. I did see that they took some care to spread out the EMPA beam current. The approach to equilibrium partitioning of trace elements only matters for the crystallization of minerals from melt, but not to partial melting of crystalline rocks, as Bea (1996) pointed out.

(3) Fernando Bea's (1996) publication on the trace element budgets of partial melts was insightful, and the authors should read it if they haven't. To wit: why measure equilibrium partition coefficients between a partially melted residuum and its partial melt? They are not at equilibrium at all. The minerals that participate in melting dissolve from their surfaces, meaning congruently more or less with respect to trace elements, such that the effective bulk distribution coefficient for trace elements is unity in relation to the proportions of the trace elements in the minerals that participate in melting.

(4) Analyses of trace elements in crustal rocks that might serve as sources of partial melts have been measured from numerous sample sources, including international rock standards (e.g. Hu and Gao, 2008) and metamorphosed schists (e.g., Bebout et al., 2007). Shale, greywacke, and their metamorphic products reach the conditions of the amphibolite facies at which anatexis might begin with no more than tens of parts per million of boron and the rare alkalis.

(5) Matrix glasses within aluminous restite enclaves that were erupted with dacites from El Hoyazo, southeastern Spain, offer the most authoritative guide to the trace element concentrations in partial melts derived from muscovite schists (Acosta-Vigil et al., 2012). Those first-formed melts contained less than 200 ppm each of Li, B, Rb, and less than 25 ppm Cs.

(6) Macdonald et al. (1992) tabulated the trace element compositions of 317 rhyolite obsidians that were mostly obtained from subduction-related and back-arc volcanics. These obsidians can be construed as the late-stage fractionated melts from far more voluminous magma bodies. The average beryllium content of 158 samples was only 4 ppm Be (Macdonald et al., 1992). The average concentrations of rare alkalis and fluorine in the silicic obsidian data base are all below 100 ppm.

(7) line 42 "Recent studies have proposed that RMGPs form in a two-stage process, involving both anatexis and fractionation." The authors are referring to the model by Koopmans et al. (2024), in which they proposed two stages of melting – one at source to produce a granite, then remelting of that crystalline granite to achieve a higher concentration of trace elements that approach the threshold of pegmatite compositions. The proposal is flawed for the same reason as given by Bea (1996); that is, the assumption that equilibrium partitioning of trace elements pertains to the remelting of the granite. Upon the remelting of granite, the trace element contents of the second melt are determined by the dissolution of minerals at their surfaces. As an example, the authors can see Figure 2 of Acosta-Vigil et al. (2006) that is a visual depiction of how melting proceeds in a granitic rock. In that case, the trace element content of the second melt is that of the minerals that participate in their proportion, and for each mineral, the effective partition coefficients are unity.

The model of Koopmans et al. (2024) requires a source of heat to remelt the granite, which is not known and can only be inferred, unless one does calculations similar to Bea (2012) to calculate the heat produced by radioactive decay of the pertinent minerals of the granite. An alternative model, which is well documented, is the sequential extraction of fractions of melt from a crystalline mush to generate successive aliquots of more chemically fractionated melt. This is the model that has been widely adopted to explain the derivation of chemically evolved melts from large-volume rhyolite magma chambers (Hildreth and Wilson, 2007), and from smaller plutons (Bea et al., 1994). In the case of beryllium, London and Evensen (2003) showed that starting with the average Be contents of silicic rhyolites (Macdonald et al., 1992), two stages of melt extraction at 75% crystallization would yield a melt whose composition reached the beryl saturation threshold. Bachmann and Bergantz (2004) demonstrated numerically that it should be possible to extract crystal-free melts at that level of crystallization within a pluton.

(8) line 109 "Staurolite-bearing metasedimentary rocks are considered to be fertile sources of lithium-rich, felsic melts due to the enrichment of lithium in staurolite relative to other pelitic minerals, including cordierite, biotite, muscovite, garnet, and tourmaline." Staurolite is long gone by the point that metapelites reach anatexis. It disappears at the first sillimanite reaction. Lithium-rich pegmatites are hosted by staurolite-bearing rocks, which means that staurolite did not contribute to their lithium content.

(9) Pegmatites are rocks defined by texture, not by composition. Because pegmatites are expressions of texture, they are petrologic systems, not geochemical ones, and not connected directly with their source. The defining texture of pegmatite is graphic granite.

Graphic granite, aplite, and granophyre are all prevalent textures in the outer zones of pegmatites. There is now a body of experimental evidence beyond any reasonable doubt that these textures arise when high-silica melts are cooled hundreds of degrees below their liquidus temperatures before the onset of crystallization along the margins. In order to be undercooled by that extent along their contacts, their host rocks must be hundreds of degrees cooler. Lithium-rich pegmatites are emplaced along the margins of their pegmatite group, in host rocks that did not exceed the limit of the greenschist facies (500-550°C: Moody et al., 1983). They are emplaced at depths of 7-9 km, for which a continental geothermal gradient (18°C/km in Archean terrain: Burke and Kidd, 1978, and in the modern Himalayas, Catlos et al., 2001) would put the host rocks at 150-185°C. With a liquidus temperature of 700°C, the pegmatite-forming melts could not be sourced at these conditions. The heat necessary to initiate partial melting of juvenile crustal rocks at this temperature can only be attained at the base of tectonically thickened crust (~ 65-70 km: Bea, 2012). That puts ~ 60 km between the possible sources of pegmatite-forming melts and the level of their final emplacement.

12 June 2025

Reviewer #2 (Remarks to the Author):

13 June 2025

Dear Dr. Evan Hastie

Enclosed is my review of manuscript COMMSENV-25-2064-T entitled "Lithium enrichment in granites and pegmatites: a reflection of their crustal source" by B. Horányi, A. Gion, F. Gaillard, E. Gloaguen, A. Plunder, J. Melleton, A. Moradell-Casellas, J. Garde, S. Erdmann, and I. Di Carlo.

I recommend the manuscript for publication with minor revisions.

The article brings much needed experimental data to answer the question as whether or not all rare-metal granitoids originate from fractionated crystallization from a sizeable peraluminous parental granite. The authors relied on an integrated study using experimental work to calculate partition coefficients that subsequently feed melt-production modelling. Note that as I reviewed this paper from a metamorphic petrologist perspective and not as an experimental petrologist. Nonetheless, my expertise is relevant since the authors experimented on metamorphic rocks as potential sources for rare-metal granitoids. The demonstration is compelling and in line with recent studies and support the conclusion that the anatexis of rare-metal-enriched metapelites and other metasedimentary rocks of more exotic compositions such as bauxite is a suitable mechanism to produce rare-metal granitoids. For metabauxites, because of their rare occurrence in nature, the frequency at which this material would melt may be overstated.

The manuscript is well written and well organized. My comments are minor and mostly deal with wording and adding some clarification over a few points, particularly in the method section.

I hope my review will be helpful in reaching a decision.

Sincerely yours,

Dr. Manuel Duguet

General comments:

Recent work (laboratory experiment and field studies) called into question whether rare-metal granites and pegmatites originated exclusively from parental peraluminous granites by fractionated crystallization. Age discrepancies between putative parental granites (when present) and the rare-metal granites and the level of fractionation required to reach lithium saturation for spodumene crystallization led researchers to consider anatexis of a suitable protolith (i.e., enriched) as an alternate viable mechanism. Although many field studies documented the possible relationships between anatexis and rare-metal granites, the feasibility of such a genetic link still needed to be demonstrated on the petrogenetic front. This was only addressed very recently by several laboratory experiment studies testing the partial melting of diverse enriched sources (e.g., granitoids). In addition of testing these sources, these studies aimed also at improving our knowledge of melt-minerals partitioning coefficients for rare metals that are indispensable for the accuracy of melt modelling.

This article explores the suitability of metasedimentary rocks moderately to highly enriched in rare metals as potential source for rare-metal granites and pegmatites. 3 samples were studied; two are staurolite-bearing micaschists enriched in rare metals compared to the average metapelites, the third one is a metabauxite. Chemical composition of these samples is compared to those in a voluminous database assembled by the authors and covering sediments (pelites, bauxites), sedimentary rocks and their metamorphic equivalents and rare-metal granitoids. This problematic is addressed in two consecutive steps. The authors present first results related to their partial melting experimental work on these samples which objective is to assess mineral-melt partition coefficients for the elements of interest (Li, Rb, Nb, Cs, Ta, W) with a clear emphasis on lithium. Both fluid-absent and fluid-present melting configurations are investigated for a range of pressures commonly encountered in the continental crust (200 MPa to 1000 MPa). Using newly acquired melt-minerals partitioning coefficients, the authors model the composition of melt generated by crustal anatexis of aluminum-rich metasedimentary rocks and granitoids. Without surprise, low-degree partial melting experiments (less than 10%) produce the most enriched melts. Passed that threshold, enrichment of the melt in these elements may drop in some cases by an order of magnitude. The most enriched protolith in elements such as lithium (i.e., bauxite) will yield melts that require little to no fractionation to reach the saturation point in lithium for spodumene to crystallize.

Results of mineral-melt partition coefficients presented in this paper do not deviate substantially from previous studies for some of the silicate phases. The novelty is clearly in the type of material, both rocks and mineral phases (e.g. staurolite), that is investigated. The silicate phases present in the starting material are ubiquitous in metamorphosed metapelites and the case of staurolite is particularly interesting because it can accommodate up to several 1000s ppm of lithium making it a primary source for lithium in the melt. However, only rocks displaying anomalous compositions in zinc, aluminum and lithium would stabilize staurolite at pressures and temperatures at which it would participate as a reactant to melt generation and not reacting out before the solidus at lower temperature (metastable staurolite). Another way to involve staurolite in melting-reaction would be to lower the solidus by having unusual high-fluid content. Very few studies have addressed the possibility of existence of suprasolidus staurolite, either being the product of melting reactions or participating to them (see for instance Spear et al. 1999 and Carcía-Casco et al. 2003). This is a problematic that is not currently discussed or taken into account (regrettably so) in thermodynamic modelling for metamorphic rocks.

For this experiment, staurolite-melting reaction could only be texturally documented for the metabauxite sample (Barr888). Bauxite is not the most abundant nor is it the most survivable material in geological formations. Beside their very peculiar chemistry, their preservation before erosion and potential burial necessitates a specific set of circumstances that are rarely met. Erosion of such material and deposition in turbiditic sequences of highly aluminous metapelites is a viable alternative that was not mentioned. In addition, such sources would not be suitable to generate the numerous LCT-type pegmatites in

the Archean because aluminum-rich pelites do not appear in significant amount before the Paleoproterozoic. There were other possible scenarios with less enriched metapelites such as metastable staurolite at P-T conditions at which partial melting place or melt infiltration (disequilibrium melting of staurolite) that could have extended the pool of fertile material. It is argued that the small volume of enriched sources such as metabauxites and staurolite-melting reactions may be under documented; especially for the former, I am not fully convinced by this. This is one of the very few criticisms I would have for this paper. However, it must be pointed out that the authors try to address this issue by assembling a voluminous rock geochemistry database (several 10000s of chemical analyses) to assess the representativeness in term of frequency and composition of the chosen samples. The other objective was to compare the chemical compositions of experimental melt products to those encountered in natural enriched rocks (i.e., rare-metal granitoids). Discussion on the experiment results is sound and compared to different sources (e.g., orthogneiss). For fluid-absent experiments, some elements (Li, Cs, Rb) in the produced melt are a good match with natural product composition whereas others (Nb, Ta, W) are not. These discrepancies are appropriately pondered and discussed in the light of other studies. It is particularly relevant because the peritectic phases crystallizing with the melt play a significant role in this. This can only be explored in the light a careful examination of the textures of the melting experiment from which melting reactions are deduced. This contribution executed this part of the study appropriately.

This experimental work and modelling study succeed at demonstrating the viability of anatexis of aluminous metasedimentary rocks enriched for producing melts enriched in rare metals. The geochemistry of the experimental melt falls within the range of those of natural rocks and are enriched enough in rare metals that lower amount of fractionation compared to the parental granite model is required to achieve observed ore-grade deposits. The behavior of lithium partitioning depends strongly on what phases crystallize during melting and this complexity is properly discussed by the authors. Similar discussions, although shorter, are conducted for the other elements. Melt modelling from the newly acquired partitions coefficients strengthens conclusions presented in this paper.

In conclusion, this article is a valuable and necessary contribution to our understanding of the petrogenesis of rare-metal granitoids. Length wise the article falls within the parameters allowed by Nature Communications with the following tally: abstract: 163 words, body of the text: 5873 words, methods: 2044 words.

#### Specific comments:

Most of my specific comments are embedded in the manuscript and the supplementary note. I will address in this section corrections related to the tables of the supplementary material.

- Regarding the excel files of the supplementary material, I suggest that you add in the supplementary note a list detailing the names of the files and what they contain (i.e., sheet tabs). For instance, In the excel file 12179\_0\_data\_set\_10715251\_svhfqm, the names of the different sheet tabs are imprecise because excel is quite limited in the number of characters you can input in these. As an example, the second sheet is titled Major elements- Metasediments whereas minerals chemistry-major elements-metasediments would be more accurate. I believe it is a necessity considering the number of tables that you have. You need to make it easy for a reader to navigate through them.

- In some tables (body of the article and supplementary material alike): parameter "n" is present but is never explicated. I assume that you talk about the number of analyses here. This should be added to the relevant tables. It may sound like a mere technicality, but every acronym and abbreviation should be explained.

#### - Supplementary Table 5:

For FM1 sample: longitude for a sample located in Spain should be 8.11W, therefore -8.11 according to your convention. for ST19.03 sample, Lat-Long coordinates were likely inverted. A latitude of -2.97 would place that sample close to the equator.

- Supplementary Table 5: 2 typos need to be corrected at the bottom line: Institut instead of Institute and Grenoble instead of Gr  noble

- Supplementary Table 11: Title is obviously incomplete and needs to be fixed.

- In the excel file 12179\_0\_data\_set\_10715250\_svhfqm: in the sediments sheet, please specify the unit of the "age" column. I guess they are in millions of years (Ma).

#### Reviewer #3 (Remarks to the Author):

Dear Editor, dear Authors,

In the manuscript submitted under the title "Lithium enrichment in granites and pegmatites: a reflection of their crustal source" the authors explore the impact that source rocks may exert on the production of rare-metal rich granites and pegmatites. Through partial melting and crystallization experiments under controlled conditions of metasedimentary rocks, the authors conclude that melting of Li-rich metasedimentary protoliths is key to produce these Li-rich granitic melts, which should undergo further enrichment through moderate fractional crystallization. The authors also produced mineral-melt partition coefficients, which are then integrated in their discussion and properly discussed and compared with previous data, mostly on natural systems, with their inherent geologic limitations (e.g., late metasomatism).

I found the science behind this manuscript is excellent, as is the writing, to the point, and illustration. Excellent and exhaustive is also the presentation of data in very complete supplementary material. This is one of those rare cases where the authors offer what appears to be a manual on how to prepare research and publish results. I have enjoyed reading it and it is clear that this work will receive significant attention from the scientific community and will be widely cited. I am not an experimental petrologist so there might be some technical aspects that escape me, so I strongly recommend that the manuscript is also revised by some expert in this field.

While the results and discussion are very robust, I note that the conclusions differ from those we recently reached in Ramírez-Briones et al. (2025 – Journal of Petrology, <https://doi.org/10.1093/petrology/egaf041>). From an "empirical" study we found that Li-rich volcanogenic rocks from Macusani of ca. 8.9 Ma did not have a different crustal source than the nearby ca. 24 Ma granites connected to tin mineralization (e.g., San Rafael deposit), which only have moderate contents of Li. High degrees of fractional crystallization (>70%, probably >80%) was deemed as needed to attain the high Li values found in these tuffaceous rocks and cospatial macusanite glass. The authors might be interested in reading this article, which is relevant as an example of Li-rich tuffaceous mudstone, with Li contents as high as 4160 ppm. This value is higher than the maximum value considered for tuffaceous sediments in the text (L417: "tuffaceous sediments (up to 3300 ppm)"), which should be adapted accordingly.

I recommend acceptance after some very minor changes. For the consideration of authors:

1. Concentration vs. content - which seem to be indistinctly used in the manuscript. I have been recommended a number of times to follow the following rule: it is recommended that the term "content" be employed when discussing solids and minerals, as it pertains to the amount per mass (ppm). Conversely, the term "concentration" is more appropriate when referring to solutions, as it is indicative of the amount per volume.
2. "Sediments" and "metasediments" - please, change to sedimentary rocks and metasedimentary rocks, respectively, in text, tables, and figures. Note that in Geology sediments are recent deposits while sedimentary rocks are lithified sediments.
3. L34: The statement "Approximately 60% of the global lithium supply is sourced from RMGPs" is misleading. Actually, a 100% of this 60% is sourced by pegmatites alone (from Australia in particular), not granites. Please, reword accordingly.
4. L175: "The partial melts are enriched in SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and the alkalis, whereas they are depleted in TiO<sub>2</sub>, FeO, MgO, and MnO, which is characteristic of evolved granitic rocks (Table 2)." Enriched and depleted, compared to what? Please, explain.
5. Regarding text in L304: we found Cs (probably Li-Cs) salt inclusions in Li-F micas in Macusani (Torró et al. 2025 – Scientific Reports <https://doi.org/10.1038/s41598-025-96173-6>). They are minute, so we found them only through TEM, but they are there!
6. In figure captions, I suggest sorting the mineral abbreviations alphabetically.
7. In the Supplementary Tables including references, please add the respective reference list (e.g., Supplementary Data 2).

Congratulations on the excellent work. Hope to see it published soon.

Lisard Torró  
July 5, 2025

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Decision Letter:

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Dear Mr Horányi,

Your revised manuscript titled "Experimental constraints on the sources of lithium-rich granites and pegmatites" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment.

We therefore invite you to revise your paper one last time to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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Best regards,

Evan Hastie, PhD  
Editorial Board Member  
Communications Earth & Environment  
[orcid.org/0000-0001-9647-2034](https://orcid.org/0000-0001-9647-2034)

Alireza Bahadori, PhD  
Senior Editor  
Communications Earth & Environment  
Consulting Editor  
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#### REVIEWERS' COMMENTS:

Reviewer #2 (Remarks to the Author):

16 September 2025

Dear Dr. Evan Hastie,

Enclosed is my follow-up review of manuscript COMMSENV-25-2064-T entitled "Experimental constraints on the sources of lithium-rich granites and pegmatites" by B. Horányi, A. Gion, F. Gaillard, E. Gloaguen, A. Plunder, J. Melleton, A. Moradell-Casellas, J. Garde, S. Erdmann, and I. Di Carlo.

After a second pass on this article, I am pleased to say that the authors adequately and thoroughly addressed my recommendations.

First and foremost, the new section about the involvement of staurolite in melting reactions in the supplementary material is a welcomed addition that addresses pending questions and uncertainties that I had pointed out in the first version of the article. Diverse possible scenarios (i.e., melt flux, staurolite metastability, influence of chemistry on staurolite stability...) are explored.

The authors also improved the discussion on the potential enriched sources for RMGPs and their relative contribution in the genesis of these melts (e.g., bauxite). They are weighted in the light of the partial melting of standard metasedimentary rocks.

Other recommendations brought forward by the 2 other reviewers were well taken into consideration and the ad hoc add-on and modifications have much improved the manuscript. The discussions about equilibrium and natural versus experimental partition coefficients are now a lot more robust and bring clarity to the topic at hand. Limitations of both anatectic and parental granite models are better explained and the authors made a better demonstration at emphasizing that an enriched source may be a prerequisite to form economical rare-metal granitoids.

I hope it is helpful in reaching your final decision.

Sincerely yours,

Dr. Manuel Duguet

Reviewer #3 (Remarks to the Author):

I recommend publication. The authors implemented all the requested changes.  
Congratulations on the excellent piece of work.

LT

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# **Response letter to the first revision of “Experimental constraints on the sources of lithium-rich granites and pegmatites”**

Manuscript Number: COMMSENV-25-2064-T

Title: Experimental constraints on the sources of lithium-rich granites and pegmatites

Authors: Bence Horányi, Austin M. Gion, Fabrice Gaillard, Éric Gloaguen, Alexis Plunder, Jérémie Melleton, Alban Moradell-Casellas, Josselyn Garde, Saskia Erdmann, and Ida Di Carlo

Corresponding Author: Bence Horányi (bence.horanyi@cnrs-orleans.fr)

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## 1. Response to the Editor

We sincerely thank the Editor and the Reviewers for their thorough evaluation of our manuscript, including their constructive comments and recommendations. We have written a detailed response to each comment addressed by Reviewer #1 (section 2), Reviewer #2 (section 3), Reviewer #3 (section 4), as well as the excerpt from Černý et al. (2005) in the comments of Reviewer #1 (section 5). Furthermore, we discuss the main concerns raised by the Editor and the Reviewers in section 1 of this letter. Each response is supported by appropriate line numbers where changes have been implemented in the manuscript. The references cited in this response letter are listed in the manuscript or the Supplementary Material. We hope that our response is deemed satisfactory for the Editor and the Reviewers.

### *1.1. Major comments*

**\*\*Strengthen your argument for the anatectic model by addressing the detailed comments of Reviewer #1\*\***

Please see section 2 of this response letter for a thorough reply to the comments of Reviewer #1.

Reviewer #1 expresses the significance of fractional crystallisation to reconcile the geochemical and textural features of rare-metal granites and pegmatites (RMGPs). The basis of this review is fully consistent with our conclusions, where we emphasise that partial melts produced by the anatexis of enriched crustal rocks must get extracted and undergo moderate fractional crystallisation to produce RMGPs (e.g., lines 463-474). To avoid ambiguity, we have further highlighted some of the limitations of anatectic models (e.g., lines 442-445) and emphasised the significance of fractional crystallisation (e.g., lines 422-423, 455-457) in the

updated manuscript. We have also re-worded the conclusive paragraph to emphasise the significance of crustal source enrichment and fractional crystallisation during RMGP petrogenesis. The title of the manuscript has been changed to “Experimental constraints on the sources of lithium-rich granites and pegmatites” to better reflect the conclusions of this study.

In our response to Reviewer #1, we highlight the limitations of models invoking the fractional crystallisation of conventional peraluminous granitic melts that are widely observed in nature (comments 4-7). Instead, we demonstrate that lithium enrichment in RMGPs begins with the anatexis of enriched crustal rocks (>300 ppm lithium; Fig. 7c). We emphasise that the partial melts must get extracted, transported, fractionated, and emplaced in the upper crust to reconcile the geochemical and textural characteristics of rare-element pegmatites (lines 422-423; comment 9).

The conclusions of this study are supported by trace element modelling using newly-constrained mineral-melt partition coefficients. However, Reviewer #1 argues that partition coefficients should not be used to model trace element behaviour during crustal anatexis due to the apparent disequilibrium between residual minerals and partial melts in nature (comments 2 and 3). In our response, we demonstrate that equilibrium is attained in experiments and in natural anatectic settings due to the fast diffusivity of lithium (see Supplementary Discussion).

**\*\*Fully address the bauxite in the context of feasibility as a source for pegmatites due to its rarity\*\***

For an exhaustive response to this comment, we direct the Editor and Reviewers' attention to section 3.1.2. of the response letter. We further address this in our response to comment (1) of Reviewer #1.

Reviewer #1 and Reviewer #2 both highlight the caveats of metabauxites being the sources of RMGPs. This is consistent with our interpretations in the '*Discussion*' section of the original manuscript. Here, we noted that the high-grade equivalents of lithium-rich bauxites are scarce, small in volume, and are interlayered with carbonate rocks (lines 427-435); therefore, they are unlikely to be the principal sources of RMGPs. We expanded on this following the perceptive comments of Reviewer #2 to highlight the poor preservation potential of metabauxites and their absence in Archean (lines 427-428). Instead, we discuss other potential sources, including pelitic rocks formed by the erosion of bauxites (lines 437-439), the metamorphic equivalents of lithium-rich sedimentary deposits (lines 439-442), moderately enriched metasedimentary rocks observed in nature (lines 403-415), and Archean gneisses enriched in lithium (lines 445-449). We ponder on the merits and caveats of each of these sources and conclude by stating the importance of further exploring unconventional metasedimentary rocks in nature (lines 450-462).

**\*\*Expand on the viability of staurolite to participate as a reactant to melt generation\*\***

Reviewer #1 and Reviewer #2 point out that staurolite typically breaks down below the solidus; therefore, it would not contribute to melting reactions during crustal anatexis.

However, there are several lines of evidence to suggest that staurolite can remain stable or metastable during the onset of anatexis and contribute to melting reactions, particularly in lithium-rich metasedimentary rocks. We have given a detailed explanation of these conditions in our response to Reviewer #2 (section 3.1.1.), Reviewer #1 (comment 8), the '*Introduction*'

section of the manuscript (lines 74-78), and the Supplementary Discussion. In the trace element model, we emphasise the assumption that staurolite remains stable or metastable during the onset of crustal anatexis (lines 397-398).

## 2. Response to Reviewer #1

We would like to extend our gratitude to Reviewer #1 for their insightful and thought-provoking comments on the petrogenesis of lithium-rich granites and pegmatites. Reviewer #1 firmly demonstrates that fractional crystallisation is a necessary step to reproduce the geochemical signature and the textural features of RMGPs. These suggestions are wholly consonant with our conclusions, where we highlight that partial melts produced by crustal anatexis must get extracted and undergo fractional crystallisation, prior to their emplacement in the upper crust. Our conclusions differ from the interpretations of previous studies by emphasising the significance of an enriched crustal source. Below, we have written a detailed response to the concerns addressed by Reviewer #1, which we implemented into the manuscript to emphasise the significance of fractional crystallisation. Our response also includes a point-by-point reply to the pioneering review paper of Černý et al. (2005), which was kindly pointed out by Reviewer #1 (see '*Response to Černý et al. (2005)*' in section 5 of this review).

### 2.1. Major comments

(1) The premise of the article is that if one partially melts a highly anomalous, lithium rich rock (a bauxite!), then one can obtain an anomalously lithium-rich partial melt. This does not seem to be a novel finding. Lithium-rich pegmatites as so common in so many terrains of different ages that their generation cannot be dependent on a pre-enriched source as rare as meta-bauxite.

Reviewer #1 correctly highlights that metabauxites cannot be the principal sources of lithium-rich granites and pegmatites, which is consistent with the conclusions of our study. In the

original version of our manuscript, we emphasise that metabauxites are unlikely to be the principal sources of RMGPs due to the caveats addressed in the ‘*Discussion*’ section (lines 427-435). We have further explained additional limitations of metabauxites being the sources of RMGPs, following the perceptive comments of Reviewer #2 (lines 428, 442-445; see section 3 for a detailed response to the concerns of Reviewer #2). Instead, we explore alternate sources, including pelitic sequences formed by the erosion of bauxites (lines 437-439), the metamorphic equivalents of lithium-rich sedimentary rocks (lines 439-442), and moderately enriched metapelites and paragneisses observed in nature (lines 403-415; 445-450). We ponder on the merits and caveats of each of these sources, and conclude by stating that future studies must investigate the occurrence of lithium-rich metasedimentary rocks to better constrain RMGP petrogenetic models (lines 450-462).

The scarcity of unconventional metasedimentary rocks in nature may also reflect the under-representativeness of metasedimentary rocks in geochemical databases ( $n = 1626$ ; see Supplementary Data 2) compared to peraluminous granites ( $n = 3059$ ; see Supplementary Data 2). There is also a considerable sampling bias associated with RMGPs due to their economic significance. We have added a sentence to the Supplementary Note (lines 37-40) to emphasise this.

Nevertheless, the experimental and modelling results demonstrate that the anatexis of conventional or moderately enriched crustal rocks ( $<300$  ppm lithium) and the subsequent fractional crystallisation of extracted partial melts is unlikely to produce RMGPs (see Supplementary Discussion). Therefore, we conclude the manuscript by emphasising the importance of enrichment in the crustal source ( $>300$  ppm lithium) in order to produce RMGPs by a two-stage process (anatexis and fractionation; lines 463-474). Such numerical calculations may not capture all the complexities of natural systems; therefore, we underscore

the necessity of further exploring unconventional metasedimentary rocks in nature as well as their relation to RMGPs (lines 472-474).

(2) There is nothing wrong with the experimental work or its analysis as far as I can see, but I didn't look closely. I did see that they took some care to spread out the EMPA beam current. The approach to equilibrium partitioning of trace elements only matters for the crystallization of minerals from melt, but not to partial melting of crystalline rocks, as Bea (1996) pointed out.

Reviewer #1 highlights the insightful study of Bea (1996), where the author emphasises that mineral-melt partition coefficients of REEs, Y, Th, and U should not be used for trace element modelling during crustal anatexis. These trace elements are primary constituents of several accessory minerals in migmatites (e.g., zircon, monazite, xenotime); therefore, the partition behaviour of REEs, Y, Th, and U does not obey Henry's law (Bea, 1996). However, lithium behaves as a trace element and obeys Henry's law in all major and accessory phases that are observed in our experiments as well as in lithium-undersaturated rhyolites, migmatites, and metapelites in nature (see lines 85-88 in the Supplementary Discussion). Bea (1996) also states that trace elements can only be used for modelling if they do not become mobilised by hydrothermal fluids following post-magmatic emplacement. Indeed, this is an important reason why  $D_{Li}^{Mineral/Melt}$  derived from granites and pegmatites should not be used for trace element modelling (lines 275-281). However, our study deals with experimental partition coefficients, which provide a direct insight into magmatic processes that take place at high pressure and temperature. Therefore, based on the study of Bea (1996), the  $D_{Li}^{Mineral/Melt}$  used in our study can be applied to model the anatexis of crustal rocks.

(3) Fernando Bea's (1996) publication on the trace element budgets of partial melts was insightful, and the authors should read it if they haven't. To wit: why measure equilibrium partition coefficients between a partially melted residuum and its partial melt? They are not at equilibrium at all. The minerals that participate in melting dissolve from their surfaces, meaning congruently more or less with respect to trace elements, such that the effective bulk distribution coefficient for trace elements is unity in relation to the proportions of the trace elements in the minerals that participate in melting.

In the second part of the comment, we believe that Reviewer #1 refers to the study of Acosta-Vigil et al. (2006), where the authors argue that the partial melting of minerals is limited to their interface and produces disequilibrium melts. The authors support this conclusion by the heterogeneous composition of the partial melts produced in their experiments, as well as the lack of compositional or textural changes observed in the residual minerals. For this reason, Reviewer #1 argues that bulk partition coefficients of trace elements between minerals and melts should be  $\sim 1$ .

Contrary to the study of Acosta-Vigil et al. (2006), we note that the composition of partial melts are homogeneous in our experiments of varying timescales (Supplementary Fig. 3). We also highlight that we observe clear textural (e.g., Fig. 3) and compositional (e.g., Table 3, Supplementary Tables 6-11) differences in the residual minerals compared to their starting composition (see lines 66-92 in the Supplementary Discussion). For example, we observe that rare metals are generally depleted in all residual minerals relative to their starting composition (lines 208-209). Therefore, bulk partition coefficients are consistently less than 1 (Table 2). These observations are supported by a plethora of experimental studies, which also demonstrate homogeneity in melt composition, as well as textural and compositional changes

in residual minerals (e.g., Holtz and Johannes, 1991; Patiño Douce and Johnston, 1991; Patiño Douce and Harris, 1998; Castro et al., 1999; García-Casco et al., 2003; Gion et al., 2021; Michaud et al. 2021). These studies demonstrate the incongruent melting of minerals, and infer that bulk partition coefficients are not in unity.

The applicability of experimental studies to natural systems and the attainment of chemical equilibrium in migmatites has been debated in the literature for several decades (e.g., Sawyer, 1991; Brown et al., 1995; Bea, 1996; Nabalek, 1999; Solar and Brown, 2001; Acosta-Vigil et al., 2006; Watson and Müller, 2009; Holness et al., 2011; Gion et al., 2021). However, there are several lines of evidence to suggest that rare metals (Li, Rb, Cs, Nb, Ta, W) are in equilibrium between residual minerals and melts in migmatites or the effects of disequilibrium in nature are negligible for certain elements (see lines 93-111 in the Supplementary Discussion). Brown et al. (1995) proposed that trace elements may achieve equilibrium distribution between residual minerals and melts if their diffusivity is faster than  $10^{-17} \text{ m}^2 \text{ s}^{-1}$  at 800°C. The alkali metals (Li, Rb, and Cs) diffuse rapidly at  $\sim 10^{-10} \text{ m}^2 \text{ s}^{-1}$  to  $\sim 10^{-16} \text{ m}^2 \text{ s}^{-1}$  in felsic melts at 800°C (Jambon, 1982); therefore, equilibrium partitioning is inferred. Lithium is a light element that diffuses particularly fast in felsic melts. At 800°C, the diffusion distance of lithium in partial melts is >26 cm/year (Zhang et al., 2010; Michaud et al., 2021), which is significantly faster than the rate of residual melt extraction in migmatites (1-2 cm/year; Sawyer, 1991). The attainment of equilibrium between minerals and melts is further supported by the fast diffusivity of lithium in residual phases. For example, Charlier et al. (2012) calculated that lithium equilibrates in plagioclase on a scale of minutes to hours at 737°C in crystals that are up to  $\sim 500 \text{ }\mu\text{m}$  in size. Therefore, lithium is inferred to be in equilibrium between residual minerals and melts during crustal anatexis. This interpretation is consistent with our experimental results, where we observe that staurolite is metastable due to

its slow dissolution (Pattison and Spear, 2018), yet the partition coefficients of lithium are equilibrated (Fig. 5).

In contrast to the alkali metals, Nb, Ta, and W diffuse significantly slower (lines 105-111 in the Supplementary Discussion). However, these high-field strength elements are primarily hosted in newly crystallising minerals (magnetite and rutile), wherein equilibrium partitioning is inferred (Bea, 1996;). Therefore, the effect of disequilibrium partitioning between residual minerals and melts on the Nb, Ta, and W content of the extracted partial melts is inferred to be negligible.

The attainment of equilibrium between partial melts and residual minerals in nature is further supported by experimental studies. The segregation of partial melts from residual minerals in migmatites takes approximately 23 years (Sawyer, 1991); therefore, the attainment of equilibrium in experimental studies that last <1 month can be used to infer equilibrium conditions in nature. Evidence for equilibrium includes the comparable partitioning behaviour of rare metals in residual and newly crystallising biotite and the homogeneous composition of minerals and melts over varying timescales (lines 66-92 in the Supplementary Discussion).

Lastly, the attainment of equilibrium in nature is demonstrated by the partially melted metapelitic enclaves in El Hoyazo, SE Spain (Acosta-Vigil et al., 2010; Holness et al., 2011; lines 82-83 in the Supplementary Discussion). These enclaves were entrained in a dacite during its eruption; therefore, partial melting textures in the enclaves (melt inclusions and matrix glasses) were preserved during the rapid ascent and cooling of the dacite (Acosta-Vigil et al., 2010; Holness et al., 2011). The El Hoyazo enclaves contain matrix glasses that are thoroughly distributed throughout the metapelite, demonstrating that melting was not limited to crystal rims, as previously suggested by Acosta-Vigil et al. (2006) and Reviewer #1. Furthermore, the matrix glasses and the melt inclusions contain up to 276 ppm lithium,

whereas the residual minerals typically contain less than 100 ppm lithium (Acosta-Vigil et al., 2010). The enrichment of partial melts relative to residual minerals demonstrates that the bulk partition coefficients of trace elements are not in unity, in contrast to the suggestion of Reviewer #1.

To conclude, experimental, analytical, and numerical studies demonstrate that equilibrium is attained between residual minerals and partial melts during crustal anatexis. Therefore, the partition coefficients determined in our study are representative of natural systems and are applicable to trace element modelling. We have summarised our response to this comment in the ‘*Supplementary Discussion*’ section of the Supplementary Material, where we address the attainment of equilibrium in detail (lines 66-111).

(4) Analyses of trace elements in crustal rocks that might serve as sources of partial melts have been measured from numerous sample sources, including international rock standards (e.g. Hu and Gao, 2008) and metamorphosed schists (e.g., Bebout et al., 2007). Shale, greywacke, and their metamorphic products reach the conditions of the amphibolite facies at which anatexis might begin with no more than tens of parts per million of boron and the rare alkalis.

Indeed, conventional metasedimentary rocks that may source granitic melts have a median lithium content of only ~20 ppm (Fig. 1b) and only up to tens of ppm of Rb, Cs, Ta, Nb, and W (Fig. 4). The partial melting of such metasedimentary rocks produces a melt with up to ~120 ppm lithium during low-degree anatexis (Supplementary Fig. 7). This mechanism summarises the petrogenesis of conventional, lithium-poor peraluminous granites, which are ubiquitous in the crust (lines 383-386). The extreme fractional crystallisation (>90%) of partial melts produced by the anatexis of conventional metasedimentary rocks (~120 ppm

lithium) produces residual melts that contain only ~720 ppm lithium (Supplementary Fig. 7), which is an order of magnitude lower than in economic-grade RMGP deposits (>5000 ppm lithium). In fact, the extraction of these highly fractionated melts, followed by a second stage of extreme fractionation (>90%) can only produce a melt with a maximum lithium content of ~4320 ppm lithium, which is again, depleted relative to economic-grade RMGPs (Supplementary Fig. 7). Therefore, RMGPs are unlikely to be sourced from conventional metasedimentary rocks in nature (~20 ppm), even following low-degree partial melting (<10%), and two-stages of extreme fractional crystallisation (>90%). Therefore, we propose that the crustal source of RMGPs must be pre-enriched. Using trace element modelling, we demonstrate that metasedimentary rocks should contain at least 300 ppm lithium to produce RMGPs by low-degree anatexis (~10%), followed by moderate fractional crystallisation (<75%) of the partial melts (Fig. 7c). We have summarised this response in the Supplementary Discussion (lines 113-136), where we highlight that the anatexis of conventional metasedimentary rocks (~20 ppm lithium) and the subsequent fractional crystallisation of extracted partial melts produces lithium-poor peraluminous granites in the crust. We support this claim by the addition of Supplementary Fig. 7, where we simulate the anatexis of conventional metasedimentary rocks and the subsequent magmatic differentiation of extracted partial melts.

The anatexis of conventional metasedimentary rocks and the genesis of lithium-poor peraluminous granites is a ubiquitous process, that happens on a large-scale during orogenic events. The petrogenesis of RMGPs on the other hand, is much more scarce and occurs on a smaller scale (<1 km<sup>3</sup>); therefore, these melting reactions will be inherently less represented in nature. Nevertheless, we demonstrate in the manuscript that there is evidence for metasedimentary rocks that contain up to 600 ppm lithium, which may serve as sources of RMGPs (lines 469-472; Supplementary Data 2).

(5) Matrix glasses within aluminous restite enclaves that were erupted with dacites from El Hoyazo, southeastern Spain, offer the most authoritative guide to the trace element concentrations in partial melts derived from muscovite schists (Acosta-Vigil et al., 2012). Those first-formed melts contained less than 200 ppm each of Li, B, Rb, and less than 25 ppm Cs.

We direct the attention of Reviewer #1 to comment (4) for a detailed response. The restite enclaves reported by Acosta-Vigil et al. (2010) contain matrix glasses and melt inclusions with 22-276 ppm lithium. These partial melts were likely derived from conventional metasedimentary rocks, containing up to few tens of ppm lithium (Supplementary Fig. 7). Reviewer #1 correctly states that these enclaves provide an authoritative insight into the trace element content of partial melts derived from typical crustal processes (i.e., the anatexis of conventional metasedimentary rocks). However, these enclaves are not considered to be representative of parental melts that source RMGPs (see response to comment (4)). The extraction of these partial melts, and their subsequent two-stage fractional crystallisation (>90%) is unlikely to produce economic-grade RMGP deposits (Supplementary Fig. 7). Instead, the fractional crystallisation of the partial melts reported in Acosta-Vigil et al. (2010) would produce weakly to moderately enriched peraluminous granites that are commonly observed in nature (Fig. 1b). In this study, we demonstrate that RMGPs are sourced from enriched crustal rocks (>300 ppm lithium), which are poorly represented in the geological record due to their inherent scarcity. Such metasedimentary rocks have been recognised in nature (up to 600 ppm lithium); however, it is imperative to continue exploration for lithium-rich sequences (up to 1000 ppm lithium) in order to further constrain the potential sources of RMGPs (lines 450-462, 472-474). We have summarised this response in the manuscript, along with our response to comment (4), to highlight the anatexis and fractionation of

conventional metasedimentary rocks and peraluminous felsic melts, respectively (lines 383-386). We have also given a detailed response to this comment in the Supplementary Discussion (lines 113-136), which is supported by trace element modelling (Supplementary Fig. 7).

(6) Macdonald et al. (1992) tabulated the trace element compositions of 317 rhyolite obsidians that were mostly obtained from subduction-related and back-arc volcanics. These obsidians can be construed as the late-stage fractionated melts from far more voluminous magma bodies. The average beryllium content of 158 samples was only 4 ppm Be (Macdonald et al., 1992). The average concentrations of rare alkalis and fluorine in the silicic obsidian data base are all below 100 ppm.

The premise of this response is similar to comments (4) and (5). Indeed, highly fractionated rhyolites in the crust typically contain <200 ppm lithium (e.g., Macdonald et al., 1992). These evolved, felsic melts may end up crystallising to produce lithium-poor peraluminous granites, which are omnipresent in the crust (see Supplementary Data 2). The low lithium content of these highly fractionated rhyolites demonstrates that fractional crystallisation alone is unlikely to produce RMGPs. The database of Macdonald et al. (1992) also highlights several samples that contain hundreds and even thousands of ppm of lithium. Previous studies have demonstrated that these lithium-rich rhyolites are sourced from enriched crustal rocks (e.g., Pichavant et al., 2024; Ramírez-Briones et al., 2025). Pichavant et al. (2024) and Ramírez-Briones et al. (2025) demonstrate that enriched parental melts (up to ~700 ppm lithium) undergo moderate fractional crystallisation (50-70%) to produce lithium-rich rhyolites observed in nature (lines 408-411). Alternatively, these melts may get arrested in the crust and crystallise to produce RMGPs.

Again, we state that the two-stage extreme fractional crystallisation (>90%) of lithium-poor parental melts (<200 ppm lithium) is unlikely to produce economic-grade RMGPs (>5000 ppm lithium; Supplementary Fig. 7). Instead, RMGPs are likely sourced from enriched parental melts, akin to the lithium-rich obsidians in the database of Macdonald et al. (1992). We have incorporated our response alongside with our response to comments (4) and (5) into the Supplementary Discussion (lines 113-136). We also direct the attention of the Editor and the Reviewers to Supplementary Fig. 7 for the trace element modelling that was performed to support this claim.

(7) line 42 “Recent studies have proposed that RMGPs form in a two-stage process, involving both anatexis and fractionation.” The authors are referring to the model by Koopmans et al. (2024), in which they proposed two stages of melting – one at source to produce a granite, then remelting of that crystalline granite to achieve a higher concentration of trace elements that approach the threshold of pegmatite compositions. The proposal is flawed for the same reason as given by Bea (1996); that is, the assumption that equilibrium partitioning of trace elements pertains to the remelting of the granite. Upon the remelting of granite, the trace element contents of the second melt are determined by the dissolution of minerals at their surfaces. As an example, the authors can see Figure 2 of Acosta-Vigil et al. (2006) that is a visual depiction of how melting proceeds in a granitic rock. In that case, the trace element content of the second melt is that of the minerals that participate in their proportion, and for each mineral, the effective partition coefficients are unity.

The model of Koopmans et al. (2024) requires a source of heat to remelt the granite, which is not known and can only be inferred, unless one does calculations similar to Bea (2012) to calculate the heat produced by radioactive decay of the pertinent minerals of the granite. An

alternative model, which is well documented, is the sequential extraction of fractions of melt from a crystalline mush to generate successive aliquots of more chemically fractionated melt. This is the model that has been widely adopted to explain the derivation of chemically evolved melts from large-volume rhyolite magma chambers (Hildreth and Wilson, 2007), and from smaller plutons (Bea et al., 1994). In the case of beryllium, London and Evensen (2003) showed that starting with the average Be contents of silicic rhyolites (Macdonald et al., 1992), two stages of melt extraction at 75% crystallization would yield a melt whose composition reached the beryl saturation threshold. Bachmann and Bergantz (2004) demonstrated numerically that it should be possible to extract crystal-free melts at that level of crystallization within a pluton.

For a detailed response to the first part of the comment concerning the partitioning of trace elements between residual minerals and partial melts, please see our response to comments (2) and (3).

Reviewer #1 is referring to lines 42-43 in our introduction, where we introduce state of the art petrogenetic models of RMGPs that have been proposed in the literature. Throughout the manuscript, we mention that rare metals must become further enriched following crustal anatexis, which may involve the fractional crystallisation of extracted melts or the re-melting of granitic cumulates. Each of these mechanisms have their merits and caveats, which is widely discussed in the literature (e.g., Bea, 1996; Bachmann and Bergantz, 2004; Hildreth and Wilson, 2007; Dufek and Bachmann, 2010; Koopmans et al., 2024). We do not debate the mechanical constraints that enable these processes because this is not in the scope of this study. Instead, we propose both hypotheses and discuss how rare metals may become enriched during these processes (e.g., lines 42-48, 408-415, 419-423, 463-474).

Reviewer #1 highlights that beryllium may become enriched during two-stages of high-degree fractional crystallisation (~75%) to produce melts that are comparable to RMGPs. However, in the case of lithium, trace element modelling (Supplementary Fig. 7) demonstrates that the two-stage extreme fractional crystallisation (>90%) of partial melts (120 ppm lithium) sourced from conventional metasedimentary rocks (20 ppm lithium) is unlikely to produce economic-grade RMGPs that are saturated in lithium-bearing minerals (>5000 ppm lithium). We summarise this response in the Supplementary Discussion (lines 113-136) to underline the significance of pre-enrichment in the crustal sources of RMGPs.

Trace element modelling using newly constrained partition coefficients ( $D_{Li}^{Mineral/Melt}$ ) from this study demonstrates that lithium enrichment during anatexis and fractionation is not as pronounced as previously inferred using  $D_{Li}^{Mineral/Melt}$  derived from natural granitic and pegmatitic samples (Supplementary Fig. 6). Therefore, we propose that RMGPs are sourced from enriched crustal rocks (>300 ppm lithium), which may undergo anatexis to produce lithium-rich felsic melts (>1500 ppm lithium). We emphasize that these partial melts must get extracted and undergo moderate fractional crystallisation (<75%) to produce RMGPs (lines 463-474).

(8) line 109 “Staurolite-bearing metasedimentary rocks are considered to be fertile sources of lithium-rich, felsic melts due to the enrichment of lithium in staurolite relative to other pelitic minerals, including cordierite, biotite, muscovite, garnet, and tourmaline.” Staurolite is long gone by the point that metapelites reach anatexis. It disappears at the first sillimanite reaction. Lithium-rich pegmatites are hosted by staurolite-bearing rocks, which means that staurolite did not contribute to their lithium content.

We direct the attention of Reviewer #1 to our detailed response to Reviewer #2 (section 3.1.1.), where we further discuss the role of staurolite during melting reactions. We have also dedicated a section of the Supplementary Discussion to discuss the stability of staurolite during crustal anatexis and its implications for the lithium budget of metasedimentary systems (lines 43-64). In summary, staurolite may remain stable above the solidus (up to 855°C) or contribute to melting reactions, particularly in lithium-rich aluminous rocks (Dutrow et al., 1986; Vielzeuf and Montel, 1994; Montel and Vielzeuf, 1997; Fockenberg, 1998; Patiño Douce and Harris, 1998; Spear et al., 1999; Chopin et al., 2003; García-Casco et al., 2003; Konzett et al., 2018; Knoll et al., 2023). The contribution of staurolite to melting reactions is particularly significant during fluid-present melting, due to the pronounced effect of circulating fluids on the solidus of metapelitic systems (<700°C, García-Casco et al., 2003). Furthermore, staurolite may remain metastable following its subsolidus breakdown and hence contribute to melting reactions if the temperature difference between staurolite breakdown and the solidus is low (e.g., Knoll et al., 2023).

Reviewer #1 further suggests that staurolite does contribute to melting reactions because RMGPs are hosted by staurolite-bearing rocks. However, the host rocks of RMGPs are not representative of the crustal rocks that source granites and pegmatites. Crustal anatexis in late-orogenic settings typically takes place in the middle or lower crust (e.g., Plunder et al., 2022). The partial melts get extracted and they are transported several kilometres through the lithosphere before they are emplaced in the upper crust (e.g., Plunder et al., 2022). We emphasise in the discussion that there is a complex spatial, structural, and temporal relationship between RMGPs and their source; therefore, the host rocks of peraluminous granites should not be regarded as their crustal source (lines 453-460).

(9) Pegmatites are rocks defined by texture, not by composition. Because pegmatites are expressions of texture, they are petrologic systems, not geochemical ones, and not connected directly with their source. The defining texture of pegmatite is graphic granite.

Graphic granite, aplite, and granophyre are all prevalent textures in the outer zones of pegmatites. There is now a body of experimental evidence beyond any reasonable doubt that these textures arise when high-silica melts are cooled hundreds of degrees below their liquidus temperatures before the onset of crystallization along the margins. In order to be undercooled by that extent along their contacts, their host rocks must be hundreds of degrees cooler. Lithium-rich pegmatites are emplaced along the margins of their pegmatite group, in host rocks that did not exceed the limit of the greenschist facies (500-550C: Moody et al., 1983). They are emplaced at depths of 7-9 km, for which a continental geothermal gradient (18C/km in Archean terrain: Burke and Kidd, 1978, and in the modern Himalayas, Catlos et al., 2001) would put the host rocks at 150-185C. With a liquidus temperature of 700C, the pegmatite-forming melts could not be sourced at these conditions. The heat necessary to initiate partial melting of juvenile crustal rocks at this temperature can only be attained at the base of tectonically thickened crust (~ 65-70 km: Bea, 2012). That puts ~ 60 km between the possible sources of pegmatite-forming melts and the level of their final emplacement.

The comment of Reviewer #1 is consistent with our conclusions, stating that partial melts must get extracted and undergo fractional crystallisation before getting emplaced in the upper crust. We have added a sentence to emphasise that the textural features of pegmatites form during the undercooling and disequilibrium crystallisation of felsic melts (e.g., London, 1992; lines 422-423). We again emphasise that the host rocks of RMGPs are not representative of their crustal source (lines 453-460). Crustal anatexis in late-orogenic settings typically takes place in the middle or lower crust (e.g., Plunder et al., 2022). The partial melts get extracted

and they are transported several kilometres through the lithosphere before they are emplaced in the upper crust (e.g., Plunder et al., 2022).

To clarify that pegmatites are defined by texture, we have replaced the term ‘pegmatites’ with ‘potential pegmatite-forming melts’ whenever we discuss the petrogenesis of pegmatites by crustal anatexis and fractional crystallisation.

### 3. Response to Reviewer #2

We thank Reviewer #2 for their thorough and constructive feedback on our manuscript. We are particularly grateful for Reviewer #2 sharing their expertise on metamorphic petrology, which helped us to refine the petrogenetic model of RMGPs in the ‘*Discussion*’ section of our manuscript. We have given a detailed response to each comment to address the concerns of Reviewer #2. The ‘*Major comments*’ were further subdivided to appropriately answer the insightful comments on a) the contribution of staurolite to melting reactions and (section 3.1.1.) b) limitations of bauxites as sources of RMGPs (section 3.1.2.).

#### 3.1. *Major comments*

##### 3.1.1. *The contribution of staurolite to melting reactions*

However, only rocks displaying anomalous compositions in zinc, aluminum and lithium would stabilize staurolite at pressures and temperatures at which it would participate as a reactant to melt generation and not reacting out before the solidus at lower temperature (metastable staurolite). Another way to involve staurolite in melting-reaction would be to lower the solidus by having unusual high-fluid content. Very few studies have addressed the possibility of existence of suprasolidus staurolite, either being the product of melting reactions or participating to them (see for instance Spear et al. 1999 and Carcía-Casco et al. 2003).

There were other possible scenarios with less enriched metapelites such as metastable staurolite at P-T conditions at which partial melting place or melt infiltration (disequilibrium melting of staurolite) that could have extended the pool of fertile material.

We thank Reviewer #2 for their thought-provoking comment on the potential subsolidus breakdown of staurolite during crustal anatexis, which warrants further discussion in the manuscript. Indeed, previous studies have demonstrated that staurolite typically breaks down at  $\sim 650^{\circ}\text{C}$  during Barrovian metamorphism as it intersects the sillimanite isograd (e.g., Pattison and Spear, 2018). However, there are several lines of evidence to suggest that staurolite can contribute to melting reactions, particularly in lithium-rich metasedimentary rocks. Firstly, Reviewer #2 kindly pointed out that Al, Zn, and Li-rich staurolite can remain stable at unusually high pressure and temperature conditions, which may subsequently contribute to melting reactions (e.g., Dutrow et al., 1986; Chopin et al., 2003; García-Casco et al., 2023). Lithium-rich metasedimentary rocks (e.g., metabauxites) typically contain staurolite that is enriched in Zn ( $\sim 12$  wt%), Al (55 wt%), and Li ( $\sim 3400$  ppm) relative to lithium-poor metapelitic rocks (Supplementary Tables 7 and 8). Secondly, several experimental (Vielzeuf and Montel, 1994; Montel and Vielzeuf, 1997; Patiño Douce and Harris, 1998; García-Casco et al., 2003; Konzett et al., 2018) and numerical studies (Fockenberg, 1998; Spear et al., 1999; Knoll et al., 2023) have demonstrated that staurolite can contribute to melt genesis or remain stable above the solidus (up to  $855^{\circ}\text{C}$ ) during fluid-present and fluid-absent melting. The contribution of staurolite to melting reactions is particularly significant during fluid-present melting, due to the pronounced effect of excess fluids on the solidus of metapelitic systems ( $< 700^{\circ}\text{C}$ , García-Casco et al., 2003). Thirdly, staurolite may remain metastable following subsolidus breakdown due to its sluggish dissolution (Pattison and Spear, 2018), akin to the partial melting experiments in our study. Therefore, metastable staurolite may contribute to melting reactions if the temperature difference between the sillimanite isograd and the solidus is low (e.g., Knoll et al., 2023). In summary, experimental, analytical, and numerical studies demonstrate that staurolite can contribute to melting reactions during crustal anatexis, particularly in lithium-rich

metasedimentary rocks. This reaction is likely under-represented in the geological record due to the narrow stability field of staurolite and its progressive consumption during crustal anatexis (e.g., García-Casco et al., 2003; Konzett et al., 2018).

In the Supplementary Discussion, we have dedicated a section to argue for the contribution of staurolite to melting reactions (lines 43-64), which we also summarise in the ‘*Introduction*’ section of our manuscript (lines 74-78). We have also emphasised in the ‘*Discussion*’, that our models assume that staurolite remains stable or metastable during the onset of partial melting (lines 397-398). Again, we sincerely thank Reviewer #2 for facilitating this fruitful discussion.

For this experiment, staurolite-melting reaction could only be texturally documented for the metabauxite sample (Barr888).

In the partial melting experiments, staurolite melting reactions could only be texturally documented for the Barr888 metabauxite (Fig. 3e). Nevertheless, the crystallisation experiments further provide evidence for the partial melting of staurolite in the FM1 micaschist. In these experiments staurolite is observably replaced by hercynite, sillimanite, and melt pseudomorphs. We have added a figure to show this (Supplementary Fig. 2 and we have emphasised this in the manuscript (line 161).

### *3.1.2. Limitations of bauxites as sources of RMGPs*

For metabauxites, because of their rare occurrence in nature, the frequency at which this material would melt may be overstated.

Bauxite is not the most abundant nor is it the most survivable material in geological formations. Beside their very peculiar chemistry, their preservation before erosion and potential burial necessitates a specific set of circumstances that are rarely met. Erosion of such material and deposition in turbiditic sequences of highly aluminous metapelites is a viable alternative that was not mentioned.

It is argued that the small volume of enriched sources such as metabauxites and staurolite-melting reactions may be under documented; especially for the former, I am not fully convinced by this.

We thank Reviewer #2 for pointing out some of the limitations of metabauxites being the sources of RMGPs. In the original version of the manuscript, we emphasised that metabauxites are unlikely to be the principal sources of RMGPs due to their small volume and their interlayering with carbonate rocks (lines 427-435). Following the recommendation of Reviewer #2, we have also added a comment on the low preservation potential of such deposits (line 428) and the scarcity of metapelitic rocks in the Archean (see next comment, lines 442-450). In the discussion, we postulate that RMGPs are sourced from the metamorphic equivalent of lithium-rich clays (up to 2400 ppm lithium; *Bowell et al., 2020*), tuffaceous sedimentary rocks (up to 4160 ppm lithium, *Ramírez-Briones et al., 2025*), or Jadar-type deposits (8400 ppm lithium; *Bowell et al., 2020*). Alternatively, RMGPs may be sourced from pelitic rocks that formed by the erosion of bauxites and their deposition in turbiditic environments, following the perceptive suggestion of Reviewer #2 (lines 437-438). Lastly, we propose that gneisses are likely to be significant sources of Archean RMGPs. Paragneisses in particular have a comparable lithium content to metapelites (up to 560 ppm lithium; see Supplementary Data 2) and they are abundant in Archean terranes (see next comment, lines 442-450). In the discussion, we highlight that unconventional metapelites, gneisses, and metavolcanic rocks containing >1000 ppm lithium have not yet been identified in nature and

must be further explored (lines 450-462). Therefore, we propose that RMGPs likely form by the anatexis of enriched crustal rocks observed in nature (>300 ppm lithium), followed by the moderate fractional crystallisation (<75%) of the extracted melts (lines 463-474).

In addition, such sources would not be suitable to generate the numerous LCT-type pegmatites in the Archean because aluminum-rich pelites do not appear in significant amount before the Paleoproterozoic.

We thank Reviewer #2 for pointing out this caveat in our model. The occurrence and distribution of aluminous metapelites in the Archean can be inferred from strongly peraluminous granites (SPGs), which primarily form by the partial melting of metasedimentary rocks (Bucholz and Spencer, 2019). Strongly peraluminous granites become widespread in the late Archean (2.8-2.5 Ga), which suggests that highly aluminous metapelitic rocks also became more abundant around this period (lines 442-443; Bucholz and Spencer, 2019; Frost and Da Prat, 2021; Mallick and Bose, 2025). Therefore, late Archean RMGPs could have been sourced from metasedimentary sequences. However, Reviewer #2 is correct by highlighting that such rocks are unlikely to have sourced the numerous LCT-type pegmatites in the Archean, especially some of the oldest RMGPs observed in nature (~3 Ga; Gardiner et al., 2024). Therefore, we highlight in the manuscript that less aluminous paragneisses and orthogneisses are also likely to be important sources of several Archean RMGPs due to their widespread distribution in Precambrian terranes (lines 445-450; e.g., Kinny et al., 1990; Černý, 1991). Paragneisses in particular can have a notably high lithium content (up to 560 ppm), which is comparable to metapelitic rocks (up to 600 ppm; see Supplementary Data 2). We have updated the manuscript to highlight these points (lines 442-450).

### 3.2. *Minor comments*

Lines 37-38: with the extreme degree of fractionated crystallization of residual melts extracted from a parental granite.

The sentence has now been changed to ‘The elevated lithium content of RMGPs has been historically associated with the high-degree fractional crystallisation of residual felsic melts extracted from a parental granite’, following the recommendation of Reviewer #2 (lines 37-38). We refrain from using the term ‘extreme’ in the manuscript due to its ambiguity, emotive tone, and lack of consistency with established nomenclature.

Line 40: Since you mention metasedimentary rocks, gneisses should be renamed orthogneisses. A few studies focused on the low partial melting of lower crust mostly composed of meta-igneous rocks.

We thank Reviewer #2 for the clarity. We have updated the sentence to emphasise ‘orthogneisses’ (line 40).

Line 57: Don't need the hyphen here. Rare metal is not used as an adjective here.

Line 191: don't need the hyphen here.

Line 199: don't need the hyphen here.

We thank Reviewer #2 for pointing out our grammatically erroneous use of the term ‘rare-metal’ throughout the manuscript. We have updated this term in the manuscript and the Supplementary Material to ‘rare metal’ when the term is not used as an adjective.

Lines 78-80: Something is missing here: how did you determine the mode of the different products of the experiments? The same goes for modal determination for the initial material.

We have highlighted that the modal mineralogy of the experimental run products was determined by mass balance calculations (lines 97-98). We further elaborate on these calculations in the ‘*Methods*’ section of our manuscript, where we emphasised that these calculations were performed to determine the mode for the starting materials, as well as the run products (lines 586-587).

Line 96: are composed of an assemblage of

We changed ‘comprise phenocrysts’ to ‘are composed of an assemblage of’ to improve the clarity of the writing (line 107).

Lines 98-99: enriched compared to what? average continental crust, sediments, gneisses?

We have added ‘relative to the median composition of metasedimentary rocks in nature’ (line 111). We notice that the same mistake was made on our behalf in the next sentence, so we again added ‘relative to conventional metasedimentary rocks’ to rectify this (lines 113-114).

Lines 109-11: I would phrase this differently. Staurolite is a bearer of lithium but this only works if there is already enough lithium present in the metasedimentary rocks. Its contribution for enriching the melt in lithium is what is relevant here.

Great comment. If the lithium content of the metasedimentary rock is low, then the presence of staurolite alone would not contribute to the genesis of lithium-rich, felsic melts during crustal anatexis. We have rephrased this sentence to emphasise the partial melting of enriched, staurolite-bearing metasedimentary rocks (lines 78-80).

Line 183: late

‘metasomatic alteration’ has been changed to ‘late-stage metasomatic alteration’ (line 189).

Line 191: Fig 4 title especially mentions low-degree partial melting (fluid-absent experiment?). It seems to be a required factor to fully benefit of the enrichment of the melt of these elements.

We discuss the effect of melt fraction on the degree of rare metal enrichment later in the paragraph (lines 205-208); therefore, we avoided repetition here.

Line 244: This would only work for staurolite being involved in partial melting reactions. Even though it has been documented, these are rare occurrences.

In a Barrovian facies metamorphism for average metapelites, staurolite reacts out before the rocks hits the solidus.

The reaction is: Staurolite + Quartz + Muscovite  $\rightleftharpoons$  Garnet + Biotite + Kyanite + H<sub>2</sub>O

In metagreywacke, the reaction becomes: Staurolite + biotite + plagioclase = garnet + Kyanite (sill) + quartz + H<sub>2</sub>O.

The real question here is what happens to the Li in sub-solidus staurolite-consumption reactions.

In order to have staurolite being melted in an average metapelite, you would need a set of specific circumstances such as metastable staurolite up to the melting point or melt infiltration.

We thank again Reviewer #2 for pointing out the potential subsolidus breakdown of staurolite in metapelitic systems. For a detailed response, we direct the Editor' and Reviewers' attention to '*The contribution of staurolite to melting reactions*' part of the '*Major comments*' (section 3.1.1.). We have included a summary of this response in the updated version of the manuscript in the 'Introduction' (lines 74-78), as well as the Supplementary Discussion (lines 43-64). Here, we discuss evidence for the occurrence of suprasolidus staurolite under certain conditions, as well as the fate of lithium during the partial (metastable) or complete breakdown of staurolite below the solidus. We have also clarified the concern of Reviewer #2 by highlighting that lithium enrichment is only enhanced if staurolite remains stable or metastable during the onset of melting (e.g., lines 357, 397-398).

Lines 372-373: When I checked these 2 publications and their supplementary material, I could not find these sample numbers until I realize that TK and LK are the initials of the senior authors of these 2 publications.

Maybe I am wrong but it looks like to me that you renamed these samples. Unless there is a good reason (that you didn't explicit), you should refrain doing this and reference the original sample number. I spent a significant amount of time figuring things out and I can guarantee that many readers will encounter the same problem. If you want to keep these names, you need to properly reference them from the source material (i.e., table number, sample number, etc.)

We understand the concern of Reviewer #2, and we have renamed the samples accordingly. The anatectic granite from Koopmans et al. (2024) is purely from a numerical study; therefore, there are no sample names given by the authors. Instead, we decided to refer to this sample as 'Granite – Greywacke source' which is what Koopmans et al. (2024) named this in their Supplemental Material (line 390). We initially had the same issue for Knoll et al. (2023), hence the shortened sample name. After further inspection of their Supplementary Material, we have identified a sample name for the staurolite-bearing metapelite 'RS34/01', which we subsequently updated throughout the manuscript (line 389) and in Fig. 7c.

Line 375: Fig. 1b ?

Initially, we were referring to Fig. 1a, which contains information on the highest recorded lithium content in metapelites and gneisses. However, this is probably ambiguous; therefore, we directed the readers' attention to Supplementary Data 2 instead (line 392)

Line 415: (<10%)

We changed 'low-degree (<10%) anatexis' to low-degree anatexis (<10%) in line 436.

Lines 416-418: This sentence does not make much sense, especially if we consider what you said in the previous one. Such enriched rocks would not need to be present in as much quantity (i.e., 10km<sup>3</sup>) to produce an intrusion with the size and chemical characteristics of the Beauvoir granite.

We thank Reviewer #2 for pointing out the issues concerning the clarity of this sentence. We have specified that the metamorphic equivalent of metamorphic rocks contain less lithium than their sedimentary counterpart due to metamorphic devolatilisation (line 442). The values indicated in the indices refer to the lithium content of sedimentary or volcano-sedimentary sequences.

Line 424: In my opinion, there is a significant break between the Archean and the Proterozoic. Although, pelitic metasedimentary rocks do exist in the Archean, this is in the Proterozoic that you start seeing highly aluminous metapelites (>22-23% wt Al<sub>2</sub>O<sub>3</sub>; see for instance the McKim formation in the Huronian Supergroup). One must seek other sources than aluminum-rich metapelites and metabauxites to explain the LCT-type pegmatites present in great number in the Archean.

We invite Reviewer #2 to refer to the '*Limitations of bauxites as sources of RMGPs*' section of the '*Major comments*' for a thorough response to this comment (section 4.1.2.). We highlight that metapelites start to become widespread by the late Archean (2.8-2.5 Ga; Bucholz and Spencer, 2019; Mallick and Bose, 2025); therefore, these rocks could have sourced several Precambrian RMGPs observed in nature (lines 442-445). However, it is unlikely that unlikely that highly aluminous metapelites could have sourced the oldest

RMGPs observed in the Archean (~3 Ga; lines 442-445). Instead, we propose that orthogneisses and paragneisses (up to 560 ppm lithium; Supplementary Data 2), likely played an important role in the petrogenesis of Archean RMGP due to their abundance in Precambrian cratons (445-450). However, we do not delve further into such melting reactions because crustal conditions were strikingly different in the Archean compared to the present-day crust, and thus diverges from the scope of this paper. We thank again Reviewer #2 for this insightful and thought-provoking comment.

Lines 450-451: What quantity of crushed material are we talking about for each capsule? If quantities vary, maybe an extra column in table 1 would work better.

We specified that 30-50 mg of crushed material was added to each capsule to improve the clarity of the sentence (line 486). We also specified the mass of fluid added to the capsules (3-5 ml; line 489) and the mass of the crushed starting materials (~200 g; line 485). Quantities do not vary significantly between experiments; therefore, we did not add an extra column to Table 1.

Line 455: The error bar should be of the same unit as the value (i.e., MPa, not bar).

Reviewer #2 is correct. We have changed 1 bar to 0.1 MPa (line 492).

Line 539: By what means the bulk composition was analyzed? By classic methods such as XRF for major, ICP-MS for traces?

It seems that for Barr888, the analyses were performed either by EMPA or LA-ICP-MS. Was it the case for the 3 samples?

What laboratory performed these analyses?

If I understand the rationale of having the initial material entirely melted before being analyzed for whole rock chemistry, I wonder how this bulk chemical composition compares to the ones from the natural samples with classic methods (providing you had about 150 mg of powdered material of each to be analyzed or the analyses were already available).

We clarified the sentence to highlight that the bulk composition of all three starting materials was determined by major (EPMA) and trace element (LA-ICP-MS) analyses of glasses produced by the high-temperature (1200°C) melting experiments at ISTO-CNRS-BRGM (A1, BH\_MSed1, BH\_MSed4, lines 576-579).

Reviewer #2 raises a valid concern regarding the representativeness of this method for bulk rock analyses. To ensure that the powders were homogeneous, we finely milled, ground, and crushed 200 g of each sample. The experiments were performed on the same batch of rock powders which were later used for the partial melting experiments; therefore, the bulk analyses by this method are representative on the experimental scale. We have added a sentence to emphasise this (lines 579-581).

Line 546: You need to provide more detail here.

In your supplementary table 3, you also have the modal composition of the starting materials, but there is no mention on how they were calculated.

Was the modal composition for these calculated from the original material or from the high-temperature experiment?

I see that you have the chemistry of the minerals of the starting materials in supplementary Table 6.

Have you consider using EDS analyses to estimate the proportions of the different phases? If not, why?

I feel this could have been a good way for crosschecking the mass balance calculations.

We have clarified in the text, as well as in the table captions that the modal mineralogy of the metasedimentary rocks and the run products was determined by mass balance calculations (lines 586-587). The bulk rock compositions of the metasedimentary rocks were determined from the high-temperature melting experiments (see previous comment), whereas the major element compositions of the minerals and glasses were determined by EPMA. We specified in the text that mineral, glass, and bulk rock compositions are all required for such calculations (line 589). We presume that by ‘EDS analyses’, Reviewer #2 is referring to SEM-EDS imaging techniques to determine the modal mineralogy. However, we decided not to use this method, or other methods involving 2D imagery, due to the complex 3D structure of the starting materials and the run products. For example, micaschists are highly heterogeneous and comprise micaceous and quartzofeldspathic layers. Therefore, 2D imaging techniques may underestimate or overestimate the proportion of different phases, depending on where the image was taken. Therefore, we have decided to use mass balance calculations to account for the heterogeneous nature of the samples (lines 587-588).

Lines 813-814: Are you referring here to the fluid-absent experiment? It is clear in the text but it should be explicitly said in the figure captions as well.

The text has been reworded to 'low-degree, fluid-absent partial melting experiments' (lines 857-859).

Line 834: crystallised

'crystallising' has been changed to 'crystallised' to improve grammatical accuracy (line 879).

Line 837: Same comment as for Fig1.

Are you referring here to the fluid-absent experiment?

It is clear in the text but it should be explicitly said in the figure captions as well.

The text has been reworded to 'low-degree, fluid-absent partial melting experiments' (line 884).

Line 839: , respectively

','respectively' has been added to the end of the sentence to improve clarity of the writing (line 884).

Line 862: of crustal rocks of diverse compositions

‘of a diverse composition of crustal rocks’ has been changed to ‘of crustal rocks of diverse compositions’ (line 908).

Line 867: What 'n' stands for?

‘n’ denotes the number of analyses. We have clarified this in all tables and supplementary tables.

### *3.3. Supplementary Material*

Line 40: How would you account for residual phases inf the equilibrium is reached or nearly reached?

We have re-worded this section to focus on the attainment of equilibrium between residual minerals and melts (lines 66-111). Residual phases, such as staurolite, remain metastable due in experiments and in natural anatexis settings to their slow dissolution (e.g., Pattison and Spear, 2018; line 56-57). Even though staurolite is metastable in our experiments, there are several lines of evidence to suggest that chemical equilibrium is attained with the coexisting melt (lines 66-111). Therefore, the rare metal content of minerals and melts is a reliable ‘snapshot’ of equilibrium processes during crustal anatexis, before the complete breakdown of residual minerals. The persistence of residual minerals in experiments is consistent with the short timescale of melt extraction inferred from migmatites in nature (lines 99-101; e.g., Sawyer, 1991; Brown et al., 1995; Solar and Brown, 2001).

Line 80: In your experiments, plagioclase is a reactant. I assume that the analysis of the "pure plagioclase" was performed on relict plagioclase. Whether or not this is the case, it should be explicitly said.

Indeed, the term 'pure plagioclase' is ambiguous. By this, we meant plagioclase that is not contaminated by any crystals or melt inclusions during LA-ICP-MS analyses. We clarified that these analyses were performed on residual plagioclase (line 163).

Line 118: chloritoide

We apologise for the spelling mistake. 'chlortoise' has been changed to 'chloritoide'.

Regarding the excel files of the supplementary material, I suggest that you add in the supplementary note a list detailing the names of the files and what they contain (i.e., sheet tabs). For instance, In the excel file 12179\_0\_data\_set\_10715251\_svhfqm, the names of the different sheet tabs are imprecise because excel is quite limited in the number of characters you can input in these. As an example, the second sheet is titled Major elements-Metasediments whereas minerals chemistry-major elements-metasediments would be more accurate. I believe it is a necessity considering the number of tables that you have. You need to make it easy for a reader to navigate through them.

Following the recommendation of Reviewer #2, we have added a new sheet to each excel file to highlight what the Supplementary Data files entail (file names and sheet tabs).

In some tables (body of the article and supplementary material alike): parameter “n” is present but is never explicated. I assume that you talk about the number of analyses here. This should be added to the relevant tables. It may sound like a mere technicality, but every acronym and abbreviation should be explained.

‘n’ denotes the number of analyses. Reviewer #2 is correct, this should be explained in the tables. We have updated each table to specify this.

Supplementary Table 5:

For FM1 sample: longitude for a sample located in Spain should be 8.11W, therefore -8.11 according to your convention.

for ST19.03 sample, Lat-Long coordinates were likely inverted. A latitude of -2.97 would place that sample close to the equator.

We thank Reviewer #2 for this keen observation. We have updated the coordinates in Supplementary Table 5 to be accurate.

Supplementary Table 5: 2 typos need to be corrected at the bottom line: Institut instead of Institute and Grenoble instead of Grénoble

We have corrected the spelling mistakes in Supplementary Table 5 following the observant comment of Reviewer #2.

Supplementary Table 11: Title is obviously incomplete and needs to be fixed.

Supplementary Table 11 is entitled ‘Representative major and trace element compositions of minerals in the Barr888 experimental run products’. We believe that this is representative of the table and what it entails.

In the excel file 12179\_0\_data\_set\_10715250\_svhfqm: in the sediments sheet, please specify the unit of the “age” column. I guess they are in millions of years (Ma).

The ‘age’ column in the ‘sediments’ sheet of Supplementary Data 2 was updated to specify the units (millions of years, Ma)

#### 4. Response to Reviewer #3

We sincerely thank the constructive comments and positive remarks of Reviewer #3, which helped us to considerably improve the manuscript. We have given a line-by-line response to these comments, supported by appropriate line numbers to highlight where we implemented the changes in the manuscript.

##### *4.1. Major comments*

While the results and discussion are very robust, I note that the conclusions differ from those we recently reached in Ramírez-Briones et al. (2025 – Journal of Petrology, <https://doi.org/10.1093/petrology/egaf041>). From an "empirical" study we found that Li-rich volcanogenic rocks from Macusani of ca. 8.9 Ma did not have a different crustal source than the nearby ca. 24 Ma granites connected to tin mineralization (e.g., San Rafael deposit), which only have moderate contents of Li. High degrees of fractional crystallization (>70%, probably >80%) was deemed as needed to attain the high Li values found in these tuffaceous rocks and cospatial macusanite glass. The authors might be interested in reading this article, which is relevant as an example of Li-rich tuffaceous mudstone, with Li contents as high as 4160 ppm. This value is higher than the maximum value considered for tuffaceous sediments in the text (L417: "tuffaceous sediments (up to 3300 ppm)"), which should be adapted accordingly.

We have updated the manuscript (line 440) to highlight that tuffaceous sediments may contain up to 4160 ppm lithium, following the study of Ramírez-Briones et al. (2025).

We apologise for the ambiguity induced by the term ‘moderate fractional crystallisation’ in our manuscript. Here, we refer to moderate fractionation as less than 75% crystallisation, which has been previously considered as the threshold for crystals becoming interlocked, and

forming a rigid framework (Vigneresse et al., 1996). Several studies in the literature have proposed that melt extraction is inhibited by this crystalline framework following subsequent magmatic differentiation (lines 50-54; e.g., Marsh, 1995; Vigneresse et al., 1996; Bachmann and Bergantz, 2004; Dufek and Bachmann, 2010). A limitation of previous RMGP petrogenetic models (e.g., Ballouard et al., 2023; Knoll et al., 2023) is that they invoke the extraction of low-degree partial melts (<10%) following extreme fractional crystallisation (>90%; lines 42-48). In our study, we demonstrate that the moderate fractional crystallisation (<75%) of pre-enriched felsic melts (sourced from unconventional metasedimentary deposits) can produce RMGPs without the need to invoke extreme fractional crystallisation (>90%; lines 411-415).

Pichavant et al. (2024) and Ramírez-Briones et al. (2025) demonstrate that the geochemical signature of rare-metal rhyolites (i.e., the Macusani volcanics) can be largely reproduced by the moderate fractional crystallisation (50-70%) of a pre-enriched parental melt. These results are consistent with our trace element modelling, which we emphasised in the revised version of the manuscript (lines 408-415). A notable inconsistency between our study and the models of Pichavant et al. (2024) and Ramírez-Briones et al. (2025) is the degree of fractionation required to reproduce the lithium content of rare-metal rhyolites. As Reviewer #3 kindly pointed out, the models of Pichavant et al. (2024) and Ramírez-Briones et al. (2025) require high-degree fractional crystallisation (>80%) to produce melts with a lithium content that is comparable to the Macusani volcanics. However, we believe that these models underestimate the degree of lithium enrichment due to the use of comparatively high  $D_{Li}^{Mineral/Melt}$  for biotite ( $D_{Li}^{Bt/Melt} = 3.66$ ), muscovite ( $D_{Li}^{Ms/Melt} = 1.12$ ), and quartz ( $D_{Li}^{Qz/Melt} = 0.21$ ). As highlighted in the manuscript (lines 341-343),  $D_{Li}^{Mica/Melt}$  derived from rhyolitic samples is likely overestimated due to the entrapment of exsolved, lithium-rich fluids during the low-

temperature crystallisation of micas (Ellis et al., 2022; Torró et al., 2025). The use of experimental partition coefficients (this study; Icenhower and London, 1995; Maneta and Baker, 2014; Pichavant et al., 2016; Pichavant, 2022) for biotite ( $D_{Li}^{Bt/Melt} = 0.25-1.70$ ), muscovite ( $D_{Li}^{Ms/Melt} = 0.12-0.82$ ), and quartz ( $D_{Li}^{Qz/Melt} = 0.001-0.027$ ) would yield significantly lower bulk partition coefficients than what is used by Pichavant et al. (2024) and Ramírez-Briones et al. (2025). Therefore, the degree of lithium enrichment is likely underestimated in previous trace element models that derive  $D_{Li}^{Mineral/Melt}$  from natural rhyolitic samples (Pichavant et al., 2024; Ramírez-Briones et al., 2025), which we have now emphasised in the manuscript (lines 343-345). In contrast, using  $D_{Li}^{Mineral/Melt}$  calculated from granitic and pegmatitic samples has the opposite effect, leading to an overestimation of lithium enrichment (e.g., Ballouard et al., 2023; Knoll et al., 2023; Koopmans et al., 2024; lines 343-345). We have added a supplementary figure to demonstrate the variable degree of lithium enrichment during crustal anatexis as a function of  $D_{Li}^{Mineral/Melt}$  derived from experiments, granitic rocks, and volcanic rocks (Supplementary Fig. 6).

Nevertheless, the degree of fractionation required to produce RMGPs depends on the lithium content of the anatectic melts. Assuming that the partitioning behaviour of lithium is similar during anatexis and fractionation, the fractional crystallisation of parental melts containing 1000 and 2290 ppm lithium by ~90% and ~70% crystallisation, respectively, can produce a melt with a lithium content that is comparable to the most enriched unit of the Beauvoir granite (5800 ppm lithium; Fig. 7c). The feasibility of the former remains debated as it invokes the extraction of low-degree partial melts (<10%; Vigneresse et al., 1996). Ramírez-Briones et al. (2025) postulates that fluxing elements (Li, P, F, B) may enhance melt segregation during extreme fractionation (>90%) due to the pronounced effect of these elements on the viscosity of the melt (e.g., Bartels et al., 2013; Bartels et al., 2015). The effect

of fluxing elements on melt extractability remains elusive; therefore, further experimental and numerical studies are required to investigate the extraction of partial melts following extreme fractional crystallisation (>90%).

#### *4.2. Minor comments*

1. Concentration vs. content - which seem to be indistinctly used in the manuscript. I have been recommended a number of times to follow the following rule: it is recommended that the term "content" be employed when discussing solids and minerals, as it pertains to the amount per mass (ppm). Conversely, the term "concentration" is more appropriate when referring to solutions, as it is indicative of the amount per volume.

We thank Reviewer #3 for pointing out our erroneous use of the terms ‘concentration’ and ‘content’. We have changed ‘concentration’ to ‘content’ throughout to manuscript when we are referring to minerals or bulk analyses.

2. "Sediments" and "metasediments" - please, change to sedimentary rocks and metasedimentary rocks, respectively, in text, tables, and figures. Note that in Geology sediments are recent deposits while sedimentary rocks are lithified sediments.

We have changed ‘sediments’ and ‘metasediments’ to ‘sedimentary rocks’ and ‘metasedimentary rocks’, respectively, following the insightful advice of Reviewer #3.

3. L34: The statement "Approximately 60% of the global lithium supply is sourced from RMGPs" is misleading. Actually, a 100% of this 60% is sourced by pegmatites alone (from Australia in particular), not granites. Please, reword accordingly.

Fair point. We have changed ‘RMGPs’ to ‘granitic pegmatites’ (line 35).

4. L175: "The partial melts are enriched in SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and the alkalis, whereas they are depleted in TiO<sub>2</sub>, FeO, MgO, and MnO, which is characteristic of evolved granitic rocks (Table 2)." Enriched and depleted, compared to what? Please, explain.

Great comment. We have re-worded the sentence without the use of the terms ‘enriched’ or ‘depleted’ to avoid ambiguity (lines 181-182).

5. Regarding text in L304: we found Cs (probably Li-Cs) salt inclusions in Li-F micas in Macusani (Torró et al. 2025 – Scientific Reports <https://doi.org/10.1038/s41598-025-96173-6>). They are minute, so we found them only through TEM, but they are there!

We thank Reviewer #3 for pointing out this critical piece of evidence that further explains the elevated  $D_{Li}^{Mica/Melt}$  observed in natural rhyolitic samples. We have cited Torr  et al. (2025) to support our hypotheses (line 311).

6. In figure captions, I suggest sorting the mineral abbreviations alphabetically.

Great suggestion. We have sorted the mineral abbreviations alphabetically in the captions of Fig. 2, Fig. 3, Table 1, and Table S5.

7. In the Supplementary Tables including references, please add the respective reference list (e.g., Supplementary Data 2).

We have updated Tables S5, S12, and S14 to highlight whether the references are from the manuscript or the Supplementary Material.

## 5. Response to Černý et al. (2005)

### 5.1. Limitations of the direct anatectic model (Černý et al., 2005)

(1) Extremely low percentage partial melting of protoliths would be required to generate elevated concentrations of rare elements in the anatectic magmas, and concentrations encountered in pegmatite deposits would be impossible to achieve (London, 2005a).

Indeed, the mechanism of low-degree melt extraction remains highly debated in the literature (e.g., Marsh, 1995; Vigneresse et al., 1996; Bachmann and Bergantz, 2004; Dufek and Bachmann, 2010; Plunder et al., 2022). Therefore, the applicability of models invoking low-degree anatexis (<10%) or extreme fractional crystallisation (>90%) to natural systems remains enigmatic (lines 50-54). Assuming that the threshold of melt extraction and the partitioning behaviour of lithium is the same during both processes, the degree of rare-metal enrichment is inferred to be comparable between anatexis and fractionation. Therefore, the anatexis of conventional metasedimentary rocks or the fractional crystallisation of conventional peraluminous granitic plutons (both have a median lithium content of 20-25 ppm) would produce a melt with only <120 ppm lithium (Supplementary Fig. 7). In fact, the extraction of such partial melts and their subsequent two-stage extreme fractional crystallisation (>90%) is unlikely to produce economic-grade RMGP deposits (>5000 ppm lithium; Supplementary Fig. 7) Therefore, partial melts and their metasedimentary source must be pre-enriched in order to produce RMGPs (Fig. 7c).

(2) Low-percentage melting would involve mainly quartzofeldspathic components and muscovite of the protoliths but most mafic phases that contain HFSE would remain (or, in the case of biotite, reequilibrate) in the residuum (e.g., Icenhower and London, 1995).

Partial melting experiments from Michaud et al. (2021) demonstrate that the incomplete breakdown of biotite does not inhibit the enrichment of Ta, Nb, and W. Instead, the enrichment of high-field strength elements primarily depends on the presence of Fe-Ti oxides (ilmenite, rutile, magnetite), and hence the oxygen fugacity of the system (lines 221-234). The anatexis of enriched crustal rocks at reducing conditions (i.e., absence of magnetite) can produce melts with niobium and tungsten signature that is comparable to RMGPs (Fig. 4). Nevertheless, similar to lithium, we emphasise in the discussion that further fractional crystallisation is needed to further enrich all rare-metals, including Li, Rb, Cs, Nb, Ta, and W (e.g., lines 467-469).

(3) Protoliths enriched in rare elements are occasionally invoked (such as evaporites), but they are rare, poor in HFSE, subject to depletion at low grade metamorphism (Li, B) and virtually absent in Archean shields (e.g., Černý, 1991b, 1992a).

Unconventional metasedimentary rocks are scarce in nature, similar to RMGPs, when compared to conventional peraluminous granites in the crust (see response to comments (1) and (4) of Reviewer #1). Lithium-rich metasedimentary rocks remain poorly explored in nature relative to RMGPs, which may reflect a sampling bias associated with economically important pegmatite deposits.

Lithium-rich metasedimentary rocks are enriched in other alkali metals (Rb and Cs) and high-field strength elements (Nb, Ta, W). This includes unconventional metasedimentary sequences such as metabauxites (e.g., Barr888; Fig. 4), as well as more commonly occurring metapelites and paragneisses (Supplementary Data 2). Lithium, and other rare metals, may become depleted in sedimentary rocks following high-grade metamorphism (20-50% loss; Gloaguen et al., 2023 and references therein). However, the occurrence of high-grade

metasedimentary rocks, such as the Barr888 metabauxite, demonstrates that rare metals can be retained in minerals such as staurolite following high-grade metamorphism.

Černý et al. (2005) highlights that metasedimentary rocks are scarce in the Archean; therefore, they are unlikely to source RMGPs during this period. We thoroughly address this comment in section 3.1.2. of this response letter, as well as lines 442-450 of the manuscript. In summary, we propose that gneisses, in addition to Late-Archean metasedimentary rocks, likely source Precambrian RMGPs.

(4) Metamorphism of typical host rocks, low-pressure upper-greenschist to medium amphibolite facies, is too low to generate granitic pegmatites in situ, and temperatures of the host rocks at the time of pegmatite emplacement are very much below those of the peak conditions of the low-grade metamorphism observed (Morgan and London, 1987; Chakoumakos and Lumpkin, 1990).

The observations of Černý et al. (2005) are correct. However, the host rocks of RMGPs are not representative of the crustal rocks that source felsic melts. Following crustal anatexis, partial melts must get extracted, transported, fractionated, and emplaced in the upper crust, several kilometres above the anatectic front (e.g., Plunder et al., 2022). Therefore, the host rocks of RMGPs cannot be used to infer their crustal source (lines 453-462). Please see our response to comment (9) of Reviewer #1 for a thorough explanation.

(5) Vapor-absent to water-saturated melting styles in higher grade rocks, capable of generating diverse granitic magmas, do not show enrichment in LCT elements comparable to

the concentrations found in appreciably fractionated pegmatites, either in experimental work or in nature (e.g., migmatites; summarized in Černý, 1998).

Since the publication of this paper, several experimental (our study; Michaud et al., 2021), analytical (Ballouard et al., 2023; Knoll et al., 2023), and numerical studies (Koopmans et al., 2024) have been performed which demonstrate that crustal anatexis can lead to the considerable enrichment of rare metal in partial melts, comparable to RMFGs (e.g., Fig. 1b and Fig. 4).

(6) Elevated concentrations of rare elements could be achieved by very low percentage anatexis, but the volumes of source protoliths would need to be enormous and the problem of extracting the melt insurmountable (Černý, 1992a).

We addressed this comment in the ‘*Discussion*’ section of the original manuscript (lines 435-437). Here, we highlight that approximately  $\sim 10 \text{ km}^3$  of a metasedimentary sequence (1000 ppm lithium) is needed to produce  $1 \text{ km}^3$  of a potential pegmatite-forming melt, akin to the B1 unit of the Beauvoir granite (5800 ppm lithium). The anatexis of thin layers of a lithium-rich metabauxite may produce small-volume pegmatite-forming melts; however, it is unlikely to produce voluminous RMFGs such as the Beauvoir granite. However, the accumulation of  $\sim 10 \text{ km}^3$  of metapelitic, metavolcanic, or gneissic sequences is not unrealistic, especially considering the volume of their sedimentary equivalents (e.g., Benson et al., 2017). Therefore, the formation of potential pegmatite-forming melts by direct anatexis is considered to be a viable mechanism (lines 437-442). As previously highlighted, rare metals become further enriched during moderate fractional crystallisation ( $<75\%$ ) following the extraction of partial melts. The mechanism of low-degree melt extraction is further discussed in our response to comment (1) from Černý et al. (2005).

(7) The saturation concentrations of beryl, columbite-group minerals, cassiterite, pollucite, and LiAl silicates in quartzofeldspathic melts are very high and impossible to be achieved without protracted fractionation of the parent medium (e.g., London, 2005a).

The saturation of lithium-bearing minerals (>5000 ppm lithium; Maneta et al., 2015) may be achieved following the low-degree anatexis of unconventional metasedimentary rocks (~1000 ppm lithium; Fig. 7c). However, other rare metals must get further enriched during moderate fractional crystallisation (<75%; lines 419-421). The partial melts produced by the anatexis of enriched crustal rocks observed in nature (300-600 ppm lithium) must also become further enriched by moderate fractional crystallisation to produce lithium-saturated melts (Fig. 7c).

We emphasise in our conclusions that fractional crystallisation is a necessary step (lines 463-474). However, trace element modelling demonstrates that the two-stage fractional crystallisation of conventional peraluminous granites is unlikely to produce melts that are saturated in lithium-aluminosilicates (Supplementary Fig. 7). Newly-constrained mineral-melt partition coefficients from this study demonstrate that lithium is not as incompatible in felsic systems as previously inferred from granitic and pegmatitic samples (lines 337-339).

Therefore, previous models of crustal anatexis and fractional crystallisation (e.g., London, 2005), likely overestimate the degree of lithium enrichment (lines 343-345). Trace element modelling using the newly constrained partition coefficients demonstrates that the crustal sources of RMGPs must be pre-enriched (Fig. 7c).

(8) The general purity of rare element pegmatites is at odds with a potential history of melts seeping through country rocks during a largescale segregation; chemical and phase contamination would be expected but is not observed (Černý, 1992a).

Černý et al., 2005 highlights that RMGPs are sourced from metasedimentary rocks in the crust, which is consistent with our petrogenetic model. The fundamental difference between the two models is the lithium content of the crustal source. In both models, the partial melts get extracted, get transported through the crust, and undergo moderate (our study) or extreme fractional crystallisation (Černý et al., 2005) to produce RMGPs (lines 455-457). Therefore, if no chemical or phase contamination is observed during the anatexis of lithium-poor metasedimentary rocks, then the same would be expected during the partial melting of unconventional crustal rocks enriched in lithium. The reason for the lack of chemical and phase contamination in both models remains uncertain. This is not discussed in the manuscript because it is outside the scope of this study.

(9) Stable and radiogenic isotope signatures of rare element pegmatites contradict direct anatexis from, or equilibration with, enclosing country rocks (e.g., Longstaffe et al., 1981; Taylor and Friedrichsen, 1983; Tomascak et al., 1992, 1998).

We direct the attention of the Editor and the Reviewers to our response to comment (9) of Reviewer #1, where we emphasise that enclosing country rocks of RMGPs are not representative of their crustal source (lines 455-460). Crustal anatexis takes place in the mid- to lower crust, which is followed by the extraction and transportation of the partial melts through the crust (e.g., Plunder et al., 2022).

(10) Anatectic generation of rare element pegmatites would locally produce rocks corresponding to arrested, incomplete evolution, but such cases of failed genesis are not known.

Lithium-rich leucosomes in migmatites have not yet been observed in nature, which may reflect the sampling bias associated with economic-grade pegmatite deposits (see lines 37-40 in the Supplementary Note). Trace element analyses of migmatites are considerably underrepresented in geochemical databases compared to granites and pegmatites (see Supplementary Data 2).

## *5.2. Significance of fractional crystallisation (Černý et al., 2005)*

(1) Granite-cum-pegmatite suites have commonly intruded into low-pressure greenschist- to amphibolite-facies country rocks, above the regional depth of anatectic conditions (e.g. Beus, 1966; Černý, 1989b, 1990).

See our response to comment (9) of Reviewer #1 for a thorough explanation. In summary, the host rocks of RMGPs are not representative of their crustal source (lines 455-460). Crustal anatexis in the middle to lower crust produces partial melts that must get extracted, transported, fractionated, and emplaced in the upper crust, several kilometres above the anatectic front (e.g., Plunder et al., 2022).

(2) Rare element pegmatites form aureoles of marginal and exterior dikes surrounding granite intrusions (e.g., Černý, 1992b).

(3) Highly evolved exterior pegmatite dikes are locally physically linked to the interior or marginal parts of granitic plutons (e.g., Beus, 1948; Černý, 1991c).

(4) Interior pods and dikes of pegmatites enclosed in, and evolved from, a parental granite are commonly impossible to distinguish from exterior dikes, in terms of structure, mineralogy, and geochemical features (e.g., Černý et al., 1981, 2005).

(5) Continuity of geochemical evolution is documented in numerous cases of granite-to-pegmatite suites (e.g., Černý et al., 1981; O'Connor et al., 1991; Trumbull, 1995; Mulja et al., 1995).

(7) Bulk compositions of rare element pegmatites are generally granitic and correspond to the minima established in relevant experimental systems (Ab-Qz- Or, Ab-Ecr (eucryptite)-Qz, with or without Rb, Cs, B, F, and/or P added (e.g., Stewart, 1978; summarized in Černý, 1991b; cf. Table A6).

These observations are consistent with our conclusions, stating that partial melts must get extracted and undergo fractional crystallisation to produce RMGPs (lines 463-474).

Nevertheless, there are several occurrences of pegmatites in nature that are not associated with a parental granite (e.g., Simmons et al., 2016; Shaw et al., 2016; Müller et al., 2017).

Therefore, evidence for fractional crystallisation (i.e., comments (2), (3), (4), and (5) of Černý et al. (2005)) is not always observed in granitic pegmatites, which lead authors to speculate on the direct anatectic origin of RMGPs (Stewart, 1978; Simmons et al., 2016; Shaw et al., 2016; Müller et al., 2017; Gourcerol et al., 2019; Plunder et al., 2022). Here, we propose that RMGP petrogenesis is a combination of both anatexis and fractionation, beginning with the partial melting of enriched crustal rocks (>300 ppm lithium), followed by the moderate fractional crystallisation (<75%) of the partial melts (lines 463-474).

(6) The fractionation path shown by granite-to-pegmatite sequences is matched by observations on rhyolite and ongonite suites (e.g., Kovalenko and Kovalenko, 1976; Pichavant et al., 1987; London, 1995; Raimbault and Burnol, 1998).

Pichavant et al. (2024) and Ramírez-Briones et al. (2025) demonstrate that rare-metal rhyolites (i.e., the Macusani volcanics) are sourced from an enriched parental melt, which undergoes moderate fractional crystallisation (50-70%) to produce rare metal rhyolites. These studies emphasise the significance of pre-enrichment in the parental melt to reconcile the geochemical signature of the Macusani volcanics (Pichavant et al., 2024; Ramírez-Briones et al., 2025). These conclusions are consistent with the results of our study and with the two-stage petrogenesis of RMGPs (lines 408-415).

(8) All paragenetic categories of the complex pegmatites are easily explained by relatively minor shifts in pressure and temperature conditions (London, 1984) or in chemical potentials of a few components in their parent melts (e.g., London, 1982; reviewed in Černý, 1991b).

Indeed, the extracted anatectic melts must get transported and emplaced in the upper crust, akin to the conclusions of this paper (lines 455-457). Emplacement at different crustal depths can explain the diverse range of pegmatite group that are highlighted by Černý et al. (2005). Our paper focuses on the petrogenesis of pegmatite-forming melts; therefore, the emplacement of RMGPs is not discussed in detail.

(9) Saturation in beryl, pollucite, tourmaline, and LiAl silicates can be achieved only by protracted, even multistage fractionation of granite-to-pegmatite suites (summarized in London, 2005a).

Trace element modelling demonstrates that spodumene saturation ( $>5000$  ppm lithium) is unlikely to be achieved by the two-stage extreme fractional crystallisation ( $>90\%$ ) of conventional peraluminous granitic melts (Supplementary Fig. 7). Therefore, we propose that the crustal sources of RMGPs must be pre-enriched ( $>300$  ppm lithium). The partial melts produced by the anatexis of such metasedimentary sequences can become further enriched during moderate fractional crystallisation ( $<75\%$ ) to produce lithium-saturated RMGPs (lines 463-474).

(10) Experimental evidence shows that liquidus and solidus of pegmatite-forming melts decrease with increasing contents of Li, Rb, Cs, F, B, P, and H<sub>2</sub>O (reviewed in Černý, 1991b), which supports the contention that batches of progressively fractionated magma can be generated by protracted differentiation of a relatively primitive starting melt.

Fluxing elements (e.g., Li, B, P, F) likely enhance the segregation of partial melts which can produce progressively fractionated pegmatites (Černý et al., 2005). However, we demonstrate using trace element modelling, that lithium-saturated pegmatites are unlikely to form following the two-stage extreme fractional crystallisation ( $>90\%$ ) of conventional peraluminous granitic melts in the crust (Supplementary Fig. 7). Therefore, the crustal sources of RMGPs must be pre-enriched ( $>300$  ppm lithium). The partial melts may then get extracted and undergo fractional crystallisation to produce progressively fractionated pegmatites that are observed in nature (lines 422-423).

# Lithium enrichment in granites and pegmatites: a reflection of their crustal source

**Bence Horányi<sup>1\*</sup>, Austin M. Gion<sup>1,2</sup>, Fabrice Gaillard<sup>1</sup>, Éric Gloaguen<sup>1,3</sup>, Alexis Plunder<sup>3</sup>, Jérémie Melleton<sup>3</sup>, Alban Moradell-Casellas<sup>1,3</sup>, Josselyn Garde<sup>1</sup>, Saskia Erdmann<sup>1</sup>, Ida Di Carlo<sup>1</sup>**

<sup>1</sup>*Institute des Sciences de la Terre d'Orléans, UMR 7327, Univ. Orléans, CNRS, BRGM, OSUC, F-45071, Orléans, France*

<sup>2</sup>*Department of Earth Sciences, University of Oxford, OX1 3AN, Oxford, UK*

<sup>3</sup>*Bureau de Recherches Géologiques et Minières, F-45060, Orléans, France*

## Supplementary Note

**Geochemical database (Fig. 1b).** The geochemical composition of sedimentary rocks was compiled from the Sedimentary Geochemistry and Paleoenvironments (SGP) Project dataset<sup>1</sup> (Supplementary Data 2). The SGP database was filtered for siliciclastic, volcano-sedimentary, and evaporitic deposits (n = 10015). The volcano-sedimentary deposit in Jadar, Serbia (8360 ppm lithium<sup>2</sup>) and lithium-rich bauxites (up to 8200 ppm lithium<sup>3</sup>) were added to the database to highlight the most enriched sediments that are observed in nature (n = 628). The geochemical composition of metamorphic and granitic rocks was derived from the GEOROC database (<https://georoc.eu/>) on the 3<sup>rd</sup> of June, 2024. The metamorphic rocks were filtered for metasedimentary rocks and gneisses (n = 1675), which are considered to be fertile sources of peraluminous, granitic melts<sup>4,5</sup>. Enriched metabauxites (up to 2300 ppm lithium) from Goffé<sup>6</sup>, Franceschelli et al.<sup>7</sup>, and Verlaquet et al.<sup>8</sup> were also added to the database (n = 37). Lithium

may become enriched in metamorphic rocks during supergene processes<sup>9</sup>; therefore, altered metasediments were removed from the database. The granitic dataset was filtered for peraluminous granites (n = 3059), which may form by the partial melting of crustal rocks<sup>4,5</sup>. The composition of rare-metal granites was obtained from Linnen and Cuney<sup>10</sup> and references therein (n = 86). Only the peraluminous, high-phosphorous (PHP) rare-metal granites were considered, which are interpreted to have a crustal source<sup>10</sup>. Rare-metal pegmatites, including Tanco<sup>11</sup>, Greenbushes<sup>12</sup>, Chèdeville<sup>13</sup>, Wabouchi<sup>14</sup>, and Kamativi<sup>15</sup> were subsequently added to this dataset (n = 68). Rare-metal granites and pegmatites often undergo late-stage hydrothermal alteration, which may alter the lithium budget of the system<sup>16,17</sup>. The effect of hydrothermal alteration on the lithium budget of RMGPs is poorly constrained; therefore, altered deposits were also included in the RMGP dataset. In contrast, the extrusive equivalent of RMGPs, termed rare-metal rhyolites, are considered to be unaltered by late-stage hydrothermal processes<sup>18-20</sup>. There are only two known examples of peraluminous, high-phosphorous rare-metal rhyolites in nature, namely the Macusani volcanics<sup>18,19</sup> and the Richemont rhyolite<sup>20</sup>.

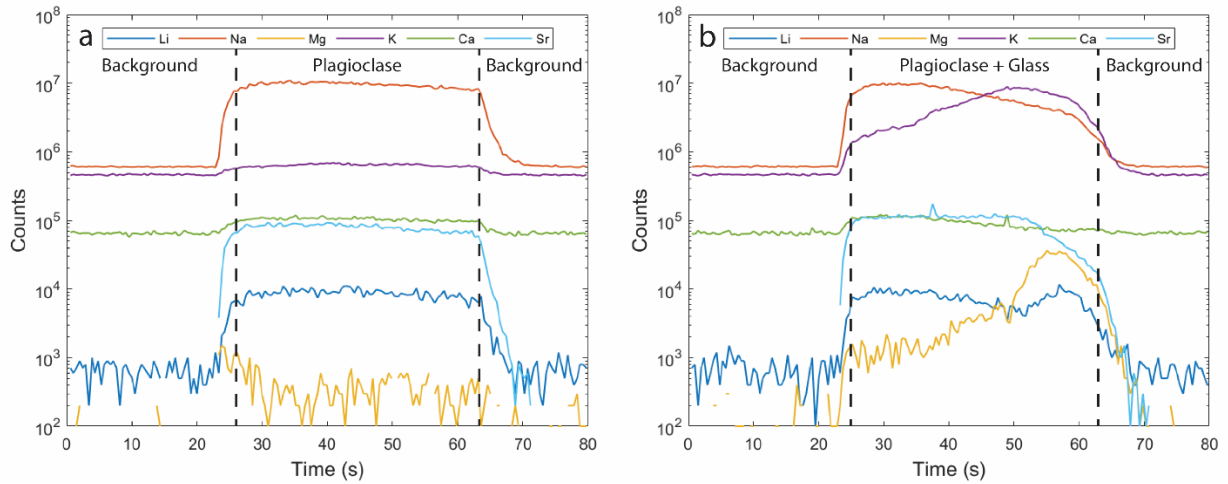
## Supplementary Discussion

**Approach to equilibrium.** Experimental studies at <800°C have demonstrated that kinetics are sluggish in low-temperature felsic systems<sup>21-25</sup>. Nevertheless, there are several observations which demonstrate an approach to equilibrium in the melting and crystallization experiments. Firstly, partition coefficients of rare-metals (e.g., Li, Rb, Nb, Ta, and W) are comparable in residual and newly crystallised biotite from GAR 01 and GAR 15, respectively (Table 3 and Supplementary Table 9). The partition coefficient of caesium between biotite and melt is slightly higher in the melting experiments than in the crystallization experiments; therefore, the caesium content of the GAR 01 glasses is interpreted to be a lower estimate of

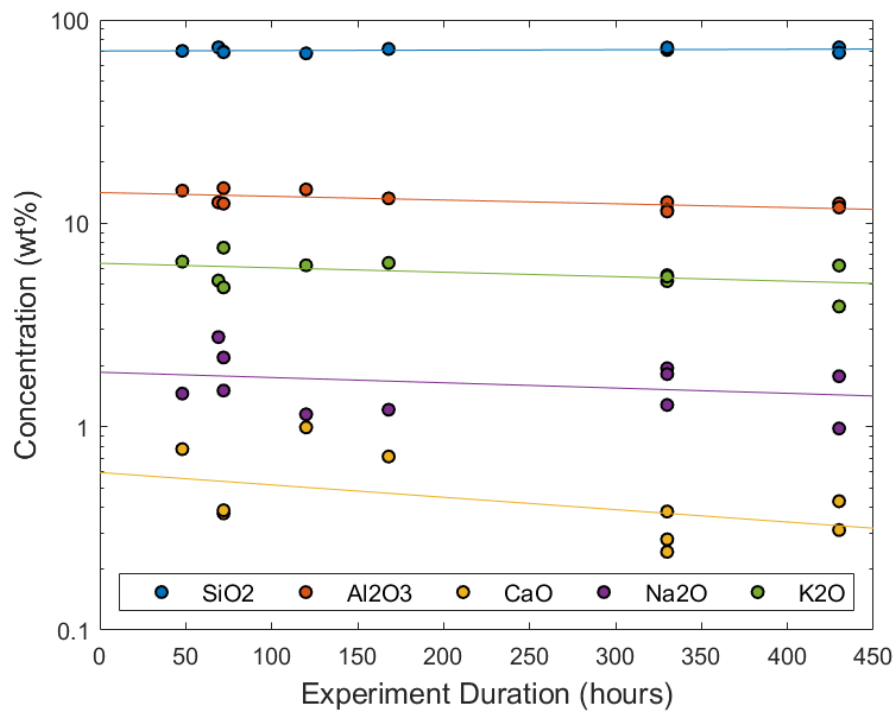
enrichment. Secondly, the major element composition of glasses is consistent between experiments of varying timescales (48 to 430 hours; Supplementary Fig. 2). Lastly, the trace element compositions of minerals and glasses are homogenous in the run products (Supplementary Tables 8 to 11). At 800°C, the diffusion distance of lithium in felsic melts is >10 mm over 14 days<sup>25</sup>. These calculations are consistent with the homogenisation of lithium in the experimental run products by diffusion.

**Trace element modelling.** The enrichment of rubidium and caesium during the anatexis of the FM1 micaschist was calculated using a modal batch melting model (equation (6)), assuming the complete breakdown of staurolite and biotite (Supplementary Fig. 5). The resulting bulk partition coefficients of rubidium and caesium range between 0.01-0.16 and 0.01-0.08, respectively (Supplementary Table 13). Following the complete breakdown of biotite, rubidium and caesium may become enriched by up to a factor of ~10 during low-degree (~10%) anatexis. The produced partial melts contain up to ~2500 ppm rubidium and ~600 ppm caesium, which is comparable to rare-metal rhyolites<sup>18-20</sup> and the most evolved unit of the Beauvoir Granite (B1)<sup>26</sup>. In contrast, the anatexis of weakly enriched crustal rocks, such as the Barr888 metabauxite (80 ppm rubidium and 8.1 ppm caesium) produces a melt with up to 800 ppm rubidium and 80 ppm caesium (assuming an enrichment factor of 10), which is depleted relative to high-grade RMGP deposits. The modelled results demonstrate the significance of rare-metal enrichment in the crustal source and the importance of biotite breakdown to maximise rubidium and caesium enrichment during crustal anatexis. Partial melts produced by the anatexis of weakly enriched crustal rocks (e.g., the Barr888 metabauxite), must become further enriched by fractional crystallization<sup>19</sup> or late-stage metasomatic processes<sup>27</sup> to produce RMGPs.

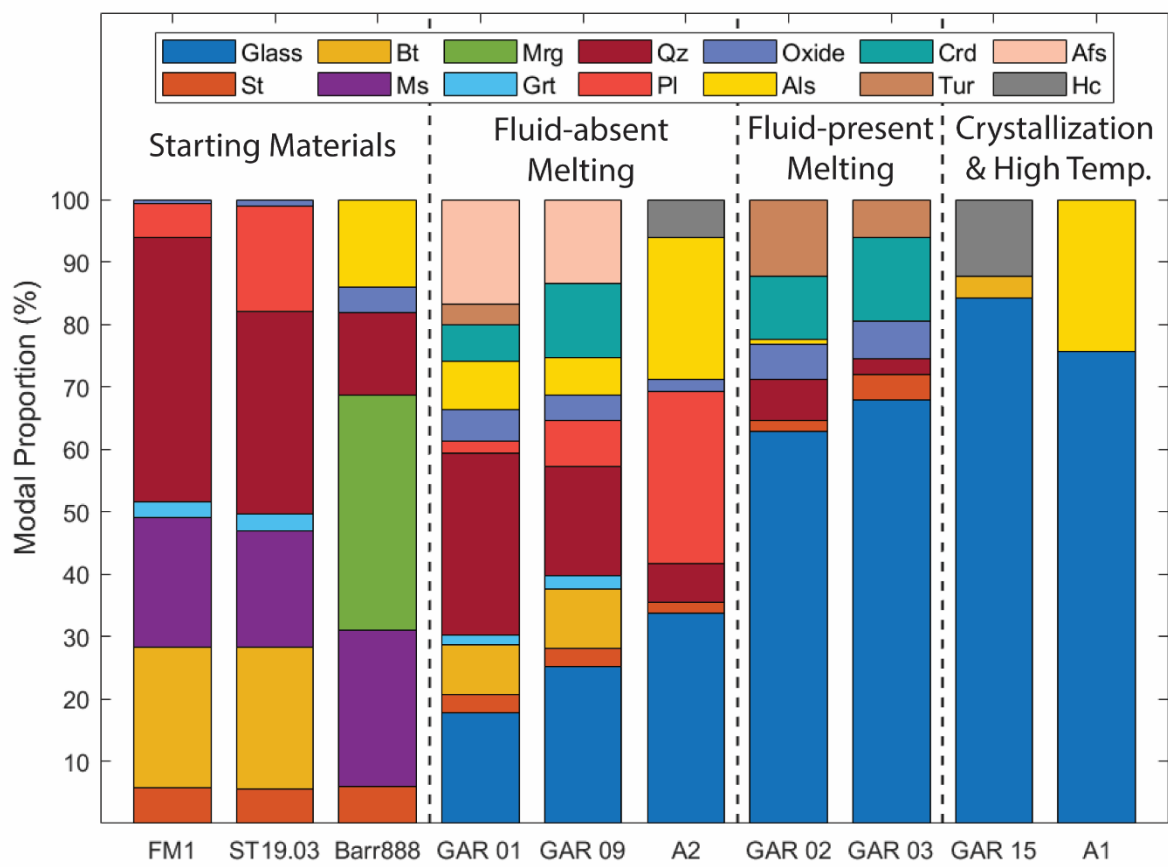
72           The enrichment or depletion of Nb, Ta, and W during anatexis was not considered in  
73 the trace element model due to the sensitivity of the calculations as a function of the modal  
74 proportion of oxides. Furthermore, partition coefficients of niobium and tantalum in rutile  
75 vary significantly as a function of rutile composition<sup>28</sup>, which results in high degrees of  
76 uncertainty in the model. Tin is also enriched in RMGPs; however, it was not considered in  
77 this study due to the loss of tin to capsule walls during the experiments<sup>29</sup>.



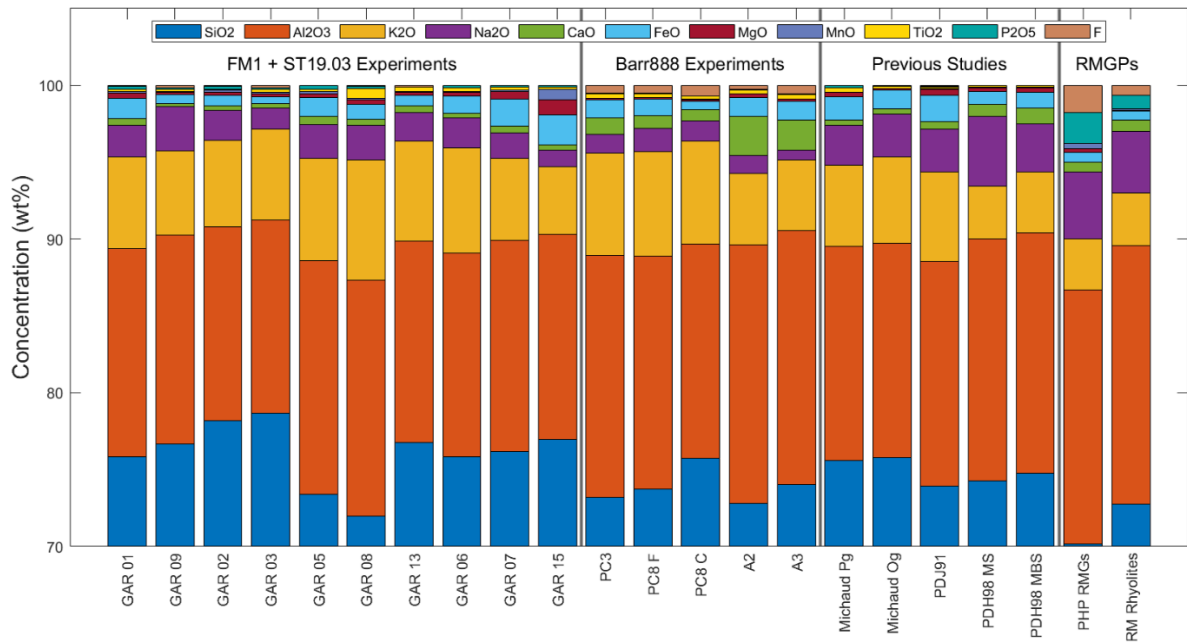
**Supplementary Fig. 1. Representative analyses of trace elements in plagioclase by LA-ICP-MS. a. Analysis of pure plagioclase. b. Contamination of plagioclase analyses by glass, which results in fluctuations of trace elements.**



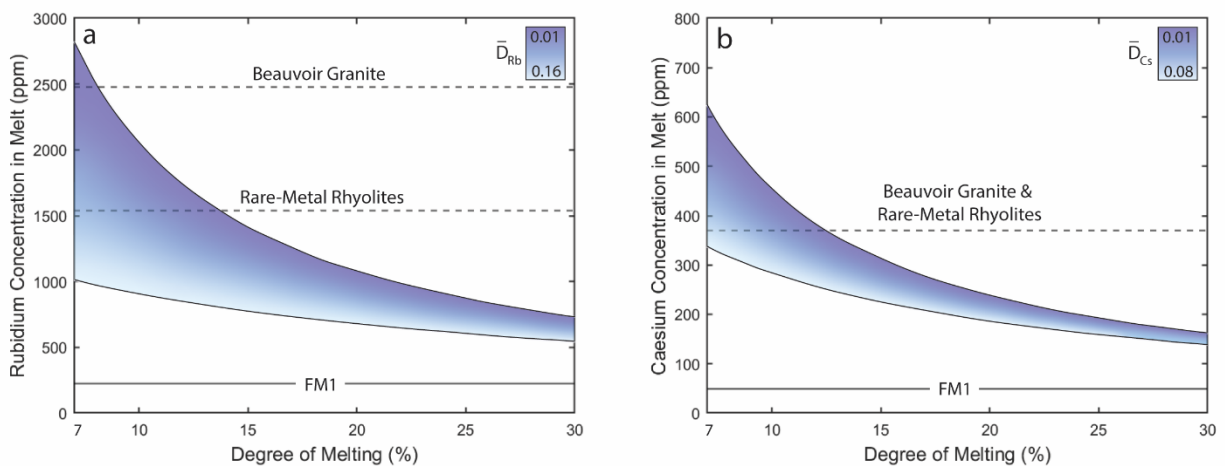
**Supplementary Fig. 2 The major element composition of glasses as a function of experiment duration. The lack of compositional variability over time demonstrates an approach to equilibrium.**



89 **Supplementary Fig. 3 The modal mineralogy of the starting materials and the**  
90 **experimental run products.** Mineral abbreviations are: St = staurolite, Bt = biotite, Ms =  
91 muscovite, Mrg = margarite, Grt = garnet, Qz = quartz, Pl = plagioclase, Als =  
92 aluminosilicates (including sillimanite, pyrophyllite, and corundum), Crd = cordierite, Tur =  
93 tourmaline, Afs = alkali feldspar, Hc = hercynite. The oxides include ilmenite, rutile, and  
94 magnetite.



**Supplementary Fig. 4 The normalised major element composition of the experimental glasses.** The composition of the glasses was compared with melt compositions from previous experimental studies<sup>5,21,25</sup>, rare-metal granites<sup>10</sup>, and rare-metal rhyolites<sup>18-20</sup> (Supplementary Data 2).



**Supplementary Fig. 5 The enrichment of rubidium and caesium during anatexis.** The enrichment of rubidium (a) and caesium (b) during the partial melting of the FM1 micaschist, assuming the complete breakdown of biotite and staurolite.

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# Lithium enrichment in granites and pegmatites: a reflection of their crustal source

**Bence Horányi<sup>1\*</sup>, Austin M. Gion<sup>1,2</sup>, Fabrice Gaillard<sup>1</sup>, Éric Gloaguen<sup>1,3</sup>, Alexis Plunder<sup>3</sup>, Jérémie Melleton<sup>3</sup>, Alban Moradell-Casellas<sup>1,3</sup>, Josselyn Garde<sup>1</sup>, Saskia Erdmann<sup>1</sup>, Ida Di Carlo<sup>1</sup>**

*<sup>1</sup>Institute des Sciences de la Terre d'Orléans, UMR 7327, Univ. Orléans, CNRS, BRGM, OSUC, F-45071, Orléans, France*

*<sup>2</sup>Department of Earth Sciences, University of Oxford, OX1 3AN, Oxford, UK*

*<sup>3</sup>Bureau de Recherches Géologiques et Minières, F-45060, Orléans, France*

## **Abstract**

Rare-metal granites and pegmatites (RMGPs) are enriched in critical metals, such as lithium (>5000 ppm), relative to conventional granites in the crust (<100 ppm). The petrogenesis of RMGPs has been historically associated with the high-degree fractional crystallisation of parental granites. However, the extent of lithium enrichment during this process remains debated due to uncertainties concerning the partitioning behaviour of lithium between minerals and felsic melts. Alternatively, lithium enrichment in RMGPs may reflect the composition of their crustal source. To test the anatectic origin of RMGPs, partial melting experiments were performed on enriched metasedimentary rocks (100-800 ppm lithium). The experiments produced felsic melts with a lithium content of 180-2200 ppm and constrained mineral-melt partition coefficients for granitic systems. Trace element modelling demonstrates that high-grade RMGP deposits (>5000 ppm lithium) are sourced from the

anatexis of enriched crustal rocks (>300 ppm lithium), followed by the moderate fractional crystallisation of the partial melts.

## Introduction

Granitic rocks have a diverse geochemical composition, which has been interpreted to reflect magmatic processes, such as fractional crystallisation, or the compositional diversity of their source material<sup>1,2</sup>. A notable example of this variability is the range of lithium contents observed in peraluminous granites. An end-member of this compositional range constitutes rare-metal granites and pegmatites (RMGPs; Fig. 1a), which are regarded as important deposits of lithium (up to 11800 ppm<sup>3</sup>) and other rare-metals (Rb, Cs, Ta, W, and Nb). Lithium is of particularly high economic interest due to its rising demand for modern-day technologies, such as batteries. Approximately 60% of the global lithium supply is sourced from RMGPs<sup>3</sup>. These deposits are enriched by over two orders of magnitude relative to the median composition of peraluminous granites in the crust (25 ppm; Fig. 1b). The elevated lithium content of RMGPs has been historically associated with the high-degree fractional crystallisation of a parental granite and the extraction of residual felsic melts<sup>4-6</sup>. Alternative hypotheses propose that RMGPs form by the direct anatexis of metasedimentary rocks or gneisses<sup>7-12</sup>. Thus, the enrichment of lithium in RMGPs may reflect the composition of their crustal source, in addition to magmatic evolution (Fig. 1a).

Recent studies have proposed that RMGPs form in a two-stage process, involving both anatexis and fractionation<sup>13-16</sup>. In the first stage of RMGP genesis, the low-degree (<10%) partial melting of metasedimentary rocks produces a lithium-rich, felsic melt<sup>13,14,16</sup>. In the second stage, these partial melts undergo high-degree fractional crystallisation (80-99%) to produce RMGPs<sup>13-14</sup>. Alternatively, the extracted melts may first crystallise to form a granitic

pluton and re-melt during a second anatectic event<sup>15-16</sup>. Low-degree (7%) partial melts may then get extracted and crystallise to form RMGPs.

Although these models constitute considerable progression in the understanding of RMGP genesis, several limitations impede their application. Firstly, the validity of these models relies on the accuracy of mineral-melt partition coefficients. Experimental partition coefficients of lithium between minerals and granitic melts are scarce and vary significantly as a function of crystallisation conditions, as well as the composition of minerals and melts<sup>17-24</sup>. The use of these partition coefficients may lead to strikingly different interpretations of lithium enrichment during RMGP genesis. Secondly, the geochemically diverse crustal source of peraluminous granites remains poorly explored. Previous studies have considered the anatexis of metapelites and gneisses that are moderately enriched in rare-metal (50-270 ppm lithium) relative to the median composition of metasedimentary rocks in the crust (20 ppm lithium; Fig. 1b)<sup>13,14,16,25,26</sup>. To better understand the enrichment of lithium during crustal anatexis, it is important to also consider the partial melting of enriched metasedimentary rocks that are observed in nature (Fig. 1b).

Lithium can accumulate in clay, evaporite, bauxite, and altered volcano-sedimentary deposits, which contain up to ~2500 ppm, ~2000 ppm, ~8200 ppm, and ~8400 ppm lithium, respectively<sup>3,27-29</sup> (Fig. 1b). These sediments may undergo metamorphism, during which approximately ~20-50% of the bulk lithium content can be removed by devolatilisation<sup>30</sup>. The medium to high-grade metamorphism of enriched protoliths produces gneisses, metapelites, and metabauxites with up to 560 ppm, 600 ppm, and 2300 ppm lithium, respectively (see Supplementary Note and Supplementary Data 2). The partial melting of these deposits may produce lithium-rich melts that crystallise to form RMGPs or rare-metal rhyolites<sup>31</sup>.

To test the anatectic origin of RMGPs, partial melting and crystallisation experiments were performed on two micaschists (ST19.03 and FM1) and a metabauxite (Barr888), with

bulk lithium contents of 102 ppm, 193 ppm, and 800 ppm, respectively (Fig. 1b). The partial melting experiments were conducted in either a piston-cylinder apparatus or an internally-heated pressure vessel at 750-800°C and 400-1000 MPa, for up to 430 hours (Table 1). Crystallisation experiments were performed at the same conditions to identify whether equilibrium has been attained. All experiments were performed either at fluid-absent (no added H<sub>2</sub>O) or fluid-present conditions (10 wt% added H<sub>2</sub>O) to investigate the effect of the degree of melting on lithium enrichment. Minerals and glasses in run products were imaged by scanning electron microscopy, whereas their major and trace element compositions were determined by electron probe microanalysis and laser ablation inductively coupled plasma mass spectrometry, respectively (Supplementary Tables 1 and 2). The experimental results demonstrate that the rare-metal content of partial melts varies as a function of the starting material composition, the degree of melting, and the partitioning of rare-metals into residual or newly crystallising minerals. The enrichment of lithium during crustal anatexis was further investigated by trace element modelling using newly constrained mineral-melt partition coefficients and the modal mineralogy of the experimental run products (Supplementary Tables 3 and 4). The modelled results are consistent with RMGPs being sourced from enriched crustal rocks. The partial melts produced by anatexis may become further enriched in lithium and other rare-metals during fractional crystallisation, the re-melting of granitic plutons, or late-stage metasomatic processes.

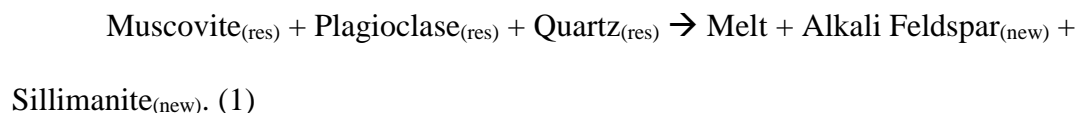
## Results

**Starting materials.** The FM1 and ST19.03 micaschists formed by the high-grade (amphibolite facies) metamorphism of a pelitic protolith in the Variscan orogenic belt<sup>32</sup>, whereas the Barr888 metabauxite formed at upper greenschist facies conditions in a subduction zone setting<sup>33</sup>. The metasedimentary samples ~~comprise phenocrysts~~ of staurolite +

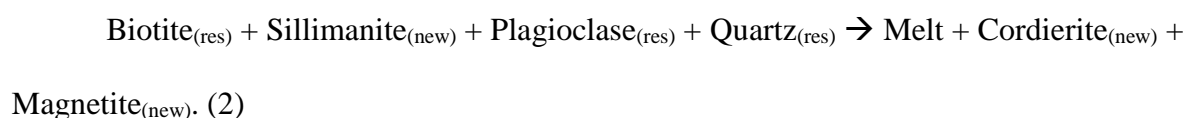
97 muscovite + quartz + rutile ± biotite ± margarite ± garnet ± plagioclase ± ilmenite ± kyanite ±  
98 pyrophyllite (Fig. 2; Supplementary Tables 5 and 6). The ST19.03 micaschist is weakly  
99 enriched in Li, Rb, Cs, Ta, Nb, and W, whereas FM1 is moderately enriched in all rare-metals  
100 (Table 2). In contrast, the Barr888 metabauxite contains relatively low concentrations of  
101 rubidium and caesium; however, it is highly enriched in Li, Ta, Nb, and W (Table 2). Lithium  
102 is primarily hosted in staurolite, followed by margarite, biotite, garnet, and muscovite (Fig. 2  
103 and Supplementary Table 7). In the micaschists, Rb, Cs, Nb, and Ta are principally hosted in  
104 biotite, whereas tungsten is predominantly found in muscovite (Supplementary Table 7).  
105 Ilmenite and rutile are enriched in Nb, Ta, and W, particularly in Barr888, wherein biotite is  
106 absent (Supplementary Table 7). In the metabauxite, rare-metals are almost exclusively hosted  
107 in staurolite, micas, and oxides, which are stable phases during high-grade metamorphism<sup>34</sup>.  
108 Thus, the bulk lithium content of Barr888 is interpreted to be representative of lithium  
109 enrichment in high-grade metabauxites. Staurolite-bearing metasedimentary rocks are  
110 considered to be fertile sources of lithium-rich, felsic melts due to the enrichment of lithium  
111 in staurolite relative to other pelitic minerals, including cordierite, biotite, muscovite, garnet,  
112 and tourmaline<sup>35</sup>. Metabauxites and metapelites are also more enriched in lithium (up to 2290  
113 ppm and 600 ppm, respectively) relative to paragneisses (up to 560 ppm), granulites (up to  
114 280 ppm), metavolcanic rocks (up to 260 ppm), amphibolites (up to 240 ppm),  
115 metagraywackes (up to 210 ppm), and orthogneisses (up to 160 ppm) in nature  
116 (Supplementary Data 2).

117  
118 **Experimental run products.** The experiments produced a felsic melt ± staurolite ± quartz ±  
119 plagioclase ± sillimanite ± rutile ± biotite ± cordierite ± tourmaline ± garnet ± magnetite ± Fe-  
120 Ti oxides ± hercynite ± gahnite ± alkali feldspar ± corundum (Fig. 3 and Table 1). The degree  
121 of melting varies from 15-35% in the fluid-absent melting experiments to 60-80% in all

crystallisation and fluid-present melting experiments (Table 2; Supplementary Tables 3 and 8). Partial melting of the metasedimentary rocks is primarily driven by the breakdown of muscovite ( $\pm$  margarite) in a schematic reaction that forms a melt, alkali feldspar ( $\pm$  plagioclase), and sillimanite<sup>36</sup> (Figs. 3a and 3b):



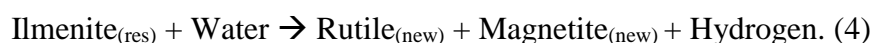
Newly crystallising biotite is absent in all partial melting run products, which is consistent with previous experimental studies conducted on metapelites<sup>36</sup>. Residual biotite ( $\pm$  garnet) partially breaks down in the fluid-absent melting experiments (Figs. 3c and 3d), which produces a melt + cordierite + magnetite ( $\pm$  tourmaline):



Staurolite is a residual phase in the fluid-absent and fluid-present partial melting experiments (Table 1). In the Barr888 experimental run products, staurolite partially breaks down to form a melt, sillimanite, and hercynite or gahnite (Fig. 3e):



In the FM1 and ST19.03 experiments, ilmenite breaks down to form intergrowths of magnetite and rutile pseudomorphs (Fig. 3f). This reaction is interpreted to be a function of the increasingly oxidising conditions of the experiments following the dissociation of water to H<sub>2</sub>, and the outward diffusion of hydrogen through the capsules<sup>37</sup>:



The Barr888 experiments were buffered at CCO by the presence of carbon and sulphur in the accessory minerals ankerite and pyrite, respectively<sup>37</sup>. Therefore, magnetite is absent in the reducing experiments performed on the Barr888 metabauxite.

In the fluid-present melting experiments, muscovite, biotite, and garnet completely break down, as illustrated by the schematic equations (1) and (2) (Fig. 3g). Plagioclase is also consumed in these reactions; therefore, feldspars are absent in the run products of the fluid-present melting experiments. Corundum crystallises following the complete or partial breakdown of pelitic minerals (micas, garnet, staurolite), which indicates that the melt is saturated in  $\text{Al}_2\text{O}_3$ .

Staurolite is absent in all crystallisation experiments (Fig. 3h), which demonstrates that it is unstable during low-temperature (750-800°C) crustal anatexis. The breakdown of staurolite results in the crystallisation of hercynite  $\pm$  gahnite  $\pm$  sillimanite  $\pm$  corundum (see schematic equation (3)). Garnet and feldspars are absent whereas biotite is stable in all FM1 crystallisation run products (Fig. 3i). Plagioclase  $\pm$  alkali feldspar  $\pm$  biotite + Fe-Ti oxides are also present in the crystallisation experiments of the Barr888 metabauxite (Table 1).

The melting reactions observed in the experiments (see schematic equations (1), (2), (3), and (4)) have a pronounced effect on the modal mineralogy of the run products. The partial melting of the metasedimentary rocks is associated with the decrease in modal proportion of residual staurolite, biotite, muscovite, garnet, plagioclase, quartz, and ilmenite, as well as the increase in the proportion of newly crystallising cordierite, tourmaline, alkali feldspar, hercynite, rutile, magnetite, and sillimanite in the run products compared to the starting materials (Supplementary Fig. 3 and Supplementary Table 3). The complete breakdown of biotite and garnet in the fluid-present melting experiments is consistent with the increased proportion of cordierite, tourmaline, and melt in the run products (Supplementary Fig. 3).

**Major element composition of partial melts.** The major element compositions of the glasses in the fluid-absent melting, fluid-present melting, and crystallisation experiments are comparable to the geochemical signature of RMGPs and melts produced in previous experimental studies<sup>26,34,36</sup> (Table 2 and Supplementary Fig. 4). All glasses are interpreted to be fluid under-saturated, containing <10 wt% water, which is consistent with anatectic granites observed in nature<sup>38</sup>. The partial melts are enriched in SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and the alkalis, whereas they are depleted in TiO<sub>2</sub>, FeO, MgO, and MnO, which is characteristic of evolved granitic rocks (Table 2). The melts are also highly peraluminous, with an aluminium saturation index (ASI, defined as the molar ratio of Al<sub>2</sub>O<sub>3</sub> to Na<sub>2</sub>O, K<sub>2</sub>O, and CaO) of 1.2 to 1.4, which is similar to the ASI of RMGPs (Table 2). The concentration of fluxing elements (F and P<sub>2</sub>O<sub>5</sub>) and the Na<sub>2</sub>O/K<sub>2</sub>O ratio of the experimental glasses is low compared to RMGPs; however, they are consistent with previous experimental studies<sup>34,36</sup>. The Na<sub>2</sub>O/K<sub>2</sub>O ratio of RMGPs is interpreted to be higher due to the crystallisation of alkali feldspar and biotite during fractionation<sup>39</sup> or the metasomatic alteration of alkali feldspar to albite<sup>40</sup>. Fluxing elements may also become further enriched during fractional crystallisation or late-stage metasomatic processes<sup>13</sup>.

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**The enrichment of rare-metals during crustal anatexis.** The partial melting of the metasedimentary rocks produced a melt that is enriched in Li, Rb, and Cs, whereas the glass is depleted in Nb, Ta, and W relative to the starting materials (Fig. 4). The experimental results demonstrate that rare-metal endowment in the partial melts is most pronounced during the anatexis of enriched crustal rocks. The enrichment of rare-metals can be further enhanced or

limited by the partitioning of trace elements between minerals and melts, which can be estimated using mineral-melt partition coefficients ( $D_i^{Mineral/Melt}$ ):

$$D_i^{Mineral/Melt} = \frac{C_i^{Mineral}}{C_i^{Melt}}, \quad (5)$$

where  $C_i^{Mineral}$  is the concentration of a trace element in the mineral and  $C_i^{Melt}$  is the concentration of a trace element in the melt. Partition coefficients of Li, Rb, Ta, Nb, and W are comparable between fluid-absent melting, fluid-present melting, and crystallisation experiments, which demonstrates an approach to equilibrium (see Supplementary Discussion and Supplementary Fig. 2). Due to the incompatible nature of the rare metals, the glasses produced by the low-degree, fluid-absent melting experiments are inherently more enriched relative to the high-degree, fluid-present melting and crystallisation experiments (Supplementary Table 8). Furthermore, residual minerals are generally depleted in Li, Rb, Cs, Ta, Nb, and W relative to the composition of the minerals in the starting materials. These elements either partition into the melt or into newly-formed minerals, such as cordierite, tourmaline, alkali feldspar, rutile, and magnetite.

The glasses in the fluid-absent melting experiments are enriched in rubidium (up to 432 ppm) and caesium (up to 130 ppm) relative to the starting materials by a factor of ~2-3 following the breakdown of white micas and the partial breakdown of biotite (Fig. 4 and Table 2). Enrichment in the melt is limited by the partitioning of rubidium and caesium into residual biotite ( $D_{Rb,Cs}^{Bt/Melt} = 1.8-2.0$ ), as well as newly crystallising alkali feldspar ( $D_{Rb}^{Afs/Melt} = 0.79$ ) and cordierite ( $D_{Cs}^{Crd/Melt} = 0.34$ ) produced by incongruent melting (Table 3). The rubidium and caesium content of the partial melts produced by the FM1 fluid-absent melting experiments is consistent with weakly enriched RMGPs; however, it is depleted relative to high-grade deposits (Fig. 4).

Tantalum (<1 ppm), Nb (<14 ppm), and W (<7 ppm) are depleted in the partial melts relative to the starting materials due to the crystallisation of rutile and magnetite in the FM1 and ST19.03 experiments (Fig. 4 and Table 3). Tantalum and niobium are highly compatible in rutile ( $D_{Ta}^{Rt/Melt} = 130$ ,  $D_{Nb}^{Rt/Melt} = 75$ ) and magnetite ( $D_{Ta}^{Mag/Melt} = 17$ ,  $D_{Nb}^{Mag/Melt} = 12$ ), whereas tungsten is also compatible in magnetite ( $D_W^{Mag/Melt} = 6.6$ ). In contrast to the Barr888, FM1, and ST19.03 experiments, the partial melting of an orthogneiss by Michaud et al.<sup>26</sup> produced a melt that is enriched in Ta, Nb, and W by a factor of ~1.5-3.5 relative to its starting composition. The enrichment of these elements is interpreted to be a function of the oxygen fugacity of the experiments. At reducing conditions in natural systems<sup>41</sup> and in the experiments of Michaud et al.<sup>26</sup>, ilmenite is stable, wherein Ta, Nb, and W are depleted relative to rutile and magnetite (see Supplementary Data 3). Therefore, the anatexis of enriched crustal rocks at reducing conditions can produce melts with a niobium and tungsten signature that is comparable to RMGPs (Fig. 4). In contrast, tantalum is depleted relative to RMGPs in all experimental run products.

**The enrichment of lithium during crustal anatexis.** The complete breakdown of white micas and the partial breakdown of staurolite ± biotite, produced a melt with 180-2200 ppm lithium (Fig. 1b). The enrichment of lithium is most pronounced in the fluid-absent melting experiments, wherein the glasses are enriched by a factor of ~2-3 relative to their metasedimentary source. Conversely, the glasses produced by all crystallisation and fluid-present melting experiments are enriched in lithium by a factor of ~1.1 to 1.5 (Supplementary Table 8) due to the dilution of incompatible elements during high-degree melting<sup>42</sup>. The degree of lithium enrichment is consistent between the reducing (Barr888) and oxidising (FM1 and ST19.03) experiments, which demonstrates that oxygen fugacity has a negligible

effect on lithium endowment during crustal anatexis. The lithium content of the glasses produced by the partial melting of Barr888 are comparable to moderately enriched RMGPs; however, they are depleted relative to high-grade deposits, such as the most evolved ‘B1’ unit of the Beauvoir granite (5800 ppm<sup>43</sup>) or the Tanco pegmatite (11800 ppm<sup>3</sup>). The enrichment of lithium in the partial melts may be furthered following the complete breakdown of rare-metal bearing minerals, such as **staurolite** and biotite. Lithium is enriched in these residual phases (along with garnet, and plagioclase) relative to newly crystallising minerals (e.g., cordierite, tourmaline, and alkali feldspar), which demonstrates that lithium primarily partitions into the melt during anatexis (Table 3; Supplementary Tables 9, 10, and 11). For example, the incongruent melting of staurolite produces minerals that are depleted in rare-metals (sillimanite ± hercynite ± gahnite; Table 3); therefore, lithium is predominantly incorporated into the melt. In comparison, the dehydration melting of muscovite and biotite (+ plagioclase ± garnet ± quartz ± sillimanite) produces cordierite and alkali feldspar (± magnetite ± sillimanite), which can sequester rare-metals in the run products (Table 3). Lithium also partitions into residual plagioclase (Table 3), which may further limit enrichment during anatexis<sup>44</sup>.

**The partitioning of lithium between minerals and melts.** Lithium is moderately incompatible in staurolite, biotite, plagioclase, cordierite, garnet, and alkali feldspar ( $D_{Li}^{Mineral/Melt} = 0.2 - 0.5$ ), whereas it is highly incompatible in tourmaline, hercynite, magnetite, rutile, and quartz ( $D_{Li}^{Mineral/Melt} = 0.01 - 0.1$ ; Fig. 5). The  $D_{Li}^{Mineral/Melt}$  obtained in this study are consistent with previous experimental partition coefficients<sup>17-24</sup> and are largely consistent with  $D_{Li}^{Mineral/Melt}$  derived from migmatites and rhyolites<sup>13,44-48</sup> (Fig. 5).

Partition coefficients derived from granites and pegmatites<sup>49-51</sup> were not considered in this study because of several limitations that hinder their application to magmatic processes. Firstly, lithium-rich pegmatites form by disequilibrium crystallisation at a low temperature (<600°C<sup>23</sup>) compared to anatectic regimes in nature (>750°C), which likely influences the partitioning behaviour of trace elements<sup>52</sup>. Secondly, lithium behaves as a major element in micas in RMGPs; therefore, conventional partition coefficients derived from zinnwaldite or lepidolite are not representative due to the non-Henry's law behaviour of lithium<sup>53</sup>. Lastly, the redistribution of trace elements during post-magmatic metasomatic processes may overprint the geochemical signature of granites and pegmatites<sup>54</sup>, which likely results in significant variability in apparent partition coefficients. The high mobility of lithium during late-stage processes is also reflected by the range of partition coefficients calculated from rhyolites and migmatites<sup>46</sup> (Fig 5); therefore,  $D_{Li}^{Mineral/Melt}$  derived from natural samples should be used cautiously.

Experimental  $D_{Li}^{Mineral/Melt}$  may also vary significantly as a function of experimental conditions, and its effect on the composition of minerals and melts (Fig. 5). Most notably, the  $D_{Li}^{Pl/Melt}$  ( $0.48 \pm 0.07$ ) and  $D_{Li}^{Afs/Melt}$  ( $0.2 \pm 0.1$ ) obtained in this study are over an order of magnitude higher than the partition coefficients that were previously used to model lithium enrichment during crustal anatexis ( $0.01-0.02$ )<sup>16</sup>. Low  $D_{Li}^{Pl/Melt}$  and  $D_{Li}^{Afs/Melt}$  are typically derived from pegmatites or a single low-temperature crystallisation experimental study (600°C)<sup>24</sup>. The comparatively high  $D_{Li}^{Pl/Melt}$  from this study are consistent with the range of experimental partition coefficients obtained at magmatic conditions (>750°C; Fig. 6a)<sup>22</sup> and with  $D_{Li}^{Pl/Melt}$  derived from rhyolites or migmatites<sup>13,44,46-48</sup>. At first order, there is an observed temperature effect on the substitution of lithium into plagioclase in granitic systems. A similar temperature effect on the partitioning of lithium into alkali feldspar could not be

identified (Fig. 6b); therefore, the reasons for the variation remains unclear. The  $D_{Li}^{Afs/Melt}$  from this study are consistent with the partition coefficients calculated from the experiments of Maneta and Baker<sup>19</sup> and Sirbescu et al.<sup>21</sup>, as well as  $D_{Li}^{Afs/Melt}$  obtained from rhyolites and migmatites<sup>44,46-48</sup>. In contrast,  $D_{Li}^{Afs/Melt}$  derived from granites and pegmatites<sup>49,51</sup> is up to an order of magnitude lower than the  $D_{Li}^{Afs/Melt}$  observed in rhyolites and migmatites<sup>44,46-48</sup>.

The  $D_{Li}^{Bt/Melt}$  obtained in this study ( $0.5 \pm 0.1$ ) are consistent with previous experimental  $D_{Li}^{Bt/Melt}$  (Fig. 5); however, they are lower than the partition coefficients used by Knoll et al.<sup>14</sup> and Koopmans et al.<sup>16</sup> for trace element modelling (1.6-1.7). The effect of temperature or mineral and melt compositions on  $D_{Li}^{Bt/Melt}$  could not be identified due to the limited temperature range of biotite stability and the complex substitution mechanism of lithium into both lattice and interstitial sites in micas<sup>47</sup> (Fig. 6c). Nevertheless, the comparatively higher lithium content in staurolite relative to biotite is consistent with the partitioning behaviour of lithium between common metamorphic minerals. In micas, lithium substitutes into octahedral sites, whereas in staurolite, lithium partitions into energetically favourable tetrahedral sites<sup>35</sup>. Consequently, lithium preferentially partitions into staurolite over biotite<sup>35</sup>, which is reflected by the higher  $D_{Li}^{St/Melt}$  relative to  $D_{Li}^{Bt/Melt}$  in the experimental run products. In natural rhyolitic samples,  $D_{Li}^{Bt/Melt}$  is considered to be unusually high due to the entrapment of exsolved, lithium-rich fluids during the low-temperature crystallisation of biotite<sup>55</sup>. In contrast, unaltered biotite that crystallised at high-temperature (e.g., in phonolites) yields  $D_{Li}^{Bt/Melt}$  that are consistent with experimental partition coefficients<sup>55</sup>. The lithium content of residual biotite and plagioclase is comparable in the experiments from this study, which is consistent with previous analyses in unaltered migmatites<sup>44</sup>.

Experimental  $D_{Li}^{Crd/Melt}$  from this study and from Evensen and London<sup>18</sup> are considerably lower than the partition coefficients derived from migmatites<sup>13,44</sup> (Fig. 5). The lithium content of cordierite is governed by the substitution of  $Li^+$  for  $Mg^{2+}$  due to the similar ionic radii of the two cations<sup>56</sup>. Cordierite from migmatites is typically magnesium-poor (and iron-rich), which is consistent with the elevated concentration of lithium in the crystal lattice<sup>13,44</sup>. In comparison, newly crystallised cordierite from this study and from Evensen and London<sup>18</sup> is magnesium-rich and iron-poor. Consequently, the lithium content of experimental cordierite is relatively low compared to some natural samples. Similar to biotite, lithium preferentially partitions into staurolite over cordierite<sup>35</sup>, which is consistent with the calculated  $D_{Li}^{Mineral/Melt}$  in the experiments.

Partition coefficients of lithium in quartz also vary significantly between natural and experimental samples. The lithium content of quartz is controlled by the coupled substitution<sup>57</sup> of  $Li^+$  and  $Al^{3+}$  for  $Si^{4+}$ . The aluminium content of quartz in the experiments is considerably lower than in natural volcanic rocks<sup>46-48</sup>, which likely reflects the slow diffusivity of aluminium at magmatic conditions. The diffusive modification of aluminium in quartz is considered to be negligible at 750°C over several millions of years<sup>58</sup>. Therefore, residual crystals of quartz in anatectic regimes are depleted in aluminium and lithium, similar to their metamorphic source (Supplementary Tables 6 and 7). The  $D_{Li}^{Quartz/Melt}$  obtained in this study are comparable to the partition coefficients calculated from the experiments of Maneta and Baker<sup>19</sup> and Pichavant<sup>23</sup>; therefore, they are considered to be representative of natural systems.

The newly constrained partition coefficients from this study demonstrate that lithium is not as incompatible in minerals as previously interpreted from  $D_{Li}^{Mineral/Melt}$  derived from granitic rocks. Pegmatites form by low-temperature, disequilibrium crystallisation, during

which trace elements may behave differently compared to high-temperature magmatic processes<sup>52</sup>. To fully understand the variability of mineral-melt partition coefficients, it is imperative to further investigate the effect of experimental conditions and the composition of minerals and melts on the partitioning behaviour of lithium in felsic systems. At present, the existing and newly constrained  $D_{Li}^{Mineral/Melt}$  should only be applied to systems that crystallised at conditions that are comparable to the experimental conditions in this study.

## Discussion

The lithium content of partial melts is a complex function of the metasedimentary source composition, the melting reactions, the degree of melting, and the partitioning of rare-metals between minerals and melts. The enrichment of lithium is maximised during the low-degree partial melting of lithium-rich metasedimentary rocks and the complete breakdown of staurolite, which becomes unstable at >700-800°C<sup>14,34,59-62</sup>. To further constrain lithium enrichment in such anatectic regimes, the partial melting of crustal rocks was modelled.

The model was first used to quantify the enrichment of lithium in the experiments from this study, the partial melting experiments of Michaud et al.<sup>26</sup>, and the crystallisation experiments of Pichavant<sup>23</sup> (Figs. 7a and 7b). The calculations were constrained using the mean  $D_{Li}^{Mineral/Melt}$  from this study and the modal mineralogy of the run products (Supplementary Fig. 3; Supplementary Tables 3 and 12). Partition coefficients of corundum and sillimanite could not be determined due to the small size of these crystals (e.g., Fig. 3); therefore,  $D_{Li}^{Als/Melt}$  and  $D_{Li}^{Crn/Melt}$  were fixed to 0.01, based on the analyses of matrix glasses and phenocrysts in natural rhyolitic samples<sup>48</sup>. Bulk partition coefficients of lithium ( $\bar{D}_{Li}$ ) were determined according to the equation:

$$\bar{D}_{Li} = \sum_{i=0}^n D_{Li}^{Mineral/Melt} \times X_i^{Mineral}, \quad (6)$$

where  $X_i^{Mineral}$  is the modal proportion of a phase in the assemblage. Bulk partition coefficients are comparable between fluid-absent melting, fluid-present melting, and crystallisation experiments of all starting materials. The  $\bar{D}_{Li}$  of the experiments varies between 0.12 and 0.25 (Table 2 and Supplementary Table 8), which demonstrates that lithium is moderately incompatible in the minerals of the run products. Crustal anatexis was modelled using a modal batch melting model<sup>42</sup>:

$$C_{Melt} = \frac{C_{Bulk}}{F(1-\bar{D}_{Li}) + \bar{D}_{Li}}, \quad (7)$$

where  $C_{Melt}$  is the concentration of lithium in the melt,  $C_{Bulk}$  is the bulk lithium content in the metasediments, and  $F$  is the fraction of melt produced. The modelled and experimental results are in good agreement (Figs. 7a and 7b), which corroborates the applicability of the newly constrained partition coefficients and verifies that all lithium-bearing phases were accounted for in the experiments.

The model was subsequently used to simulate the partial melting of crustal rocks with variable lithium contents (Fig. 7c). The degree of melting was limited to 7-30%, comparable to previous numerical models<sup>13,14,16</sup>. Two crustal rocks were considered, including the staurolite-bearing micaschists from Knoll et al.<sup>14</sup> ('TK23'; 270 ppm lithium) and the anatectic granite from Koopmans et al.<sup>16</sup> ('LK23'; 538 ppm lithium). The concentration of lithium in the starting LK23 granite is comparable to the highest recorded lithium content in metapelites and gneisses (Fig. 1a). The partial melting of the mean (1000 ppm lithium) and most enriched units (2290 ppm lithium) of an unconventional metabauxite was also considered ('Mb')<sup>63</sup>. Such lithologies are representative of metasedimentary rocks that may form by the metamorphism of lithium-rich bauxites, clays, or volcano-sedimentary deposits<sup>27,28</sup>. The

model was constrained using the range of  $\bar{D}_{Li}$  determined from all fluid-absent melting experiments in this study, assuming a fixed modal mineralogy and the complete breakdown of residual staurolite and biotite ( $\bar{D}_{Li} = 0.09$  to  $0.21$ ; Supplementary Table 13). The partial breakdown of biotite was also investigated (Fig. 7d), which has a significant effect on the enrichment of lithium, as well as other rare-metals such as Rb, Cs, W, Nb, and Ta (Supplementary Discussion and Supplementary Fig. 5).

Lithium may become enriched in the partial melts by up to a factor of 5-6 during the low-degree (<10%) dehydration-melting of staurolite and biotite (Fig. 7c). The anatexis of the TK23 micaschists produces a partial melt with up to ~1500 ppm lithium (Fig. 7c), which is comparable to weakly enriched RMGPs (Fig. 1b). The low-degree partial melting of the LK23 granite forms a melt with a lithium content that is consistent with the mean composition of RMGPs and rare-metal rhyolites (~3000 ppm lithium). The partial melts produced by the anatexis of the micaschists and the granite must become further enriched by moderate fractional crystallisation<sup>48</sup> or the re-melting of granitic cumulates<sup>16</sup> to produce high-grade RMGP deposits (>5000 ppm lithium). Assuming that the partitioning behaviour of lithium is similar during anatexis and fractionation, it is estimated that the parental melts of highly fractionated RMGPs (>70% crystallisation) must contain at least 1000 ppm lithium (Fig. 7c). These modelled results are consistent with high-grade granite-related deposits being sourced from enriched crustal rocks.

An alternative hypothesis to this petrogenetic model is the formation of potential RMGPs by the anatexis of unconventional metasedimentary rocks (>1000 ppm lithium). The partial melting of the mean and most enriched units of the Mb metabauxite may produce a melt with up to ~6500 ppm and ~14000 ppm lithium, respectively. Other rare-metals (Rb, Cs, Ta) and major elements ( $\text{Na}_2\text{O}$ ,  $\text{P}_2\text{O}_5$ , and F) can become further enriched during the fractional crystallisation of extracted melts<sup>48</sup> or late-stage metasomatic processes<sup>54</sup>. To test

whether RMGPs are sourced from such unconventional deposits, it is important to constrain the occurrence and size of enriched metasedimentary rocks in nature. There are several known examples of bauxites or low-grade metabauxites at active continental margins, which may contain up to 8200 ppm lithium<sup>29,64</sup>. However, the high-grade equivalents of these deposits are poorly explored in orogenic settings. Lithium-rich metabauxites are typically found as thin lenses (~1-15 meters) within carbonate rocks that extend for up to a few hundred meters in length<sup>63,64</sup>. The anatexis of interlayered bauxite and carbonate units can produce weakly peraluminous felsic melts<sup>65</sup>; however, the Al<sub>2</sub>O<sub>3</sub> and K<sub>2</sub>O content of glasses produced by such experiments is significantly enriched relative to RMGPs or glasses produced by the anatexis of metapelites. Furthermore, the partial melting of thin lenses of metabauxites (<1 km<sup>3</sup>) would produce small-volume pegmatite-forming melts. However, in order to produce a Beauvoir-like granite (~1 km<sup>3</sup> and ~5800 ppm lithium) by low-degree (<10%) anatexis, approximately ~10 km<sup>3</sup> of a lithium-rich (~1000 ppm) metasedimentary sequence must be melted. The metamorphism of lithium-rich clays (up to 2400 ppm), tuffaceous sediments (up to 3300 ppm), or Jadar-type deposits (8400 ppm) may produce such voluminous metasedimentary sequences; however, these have not yet been identified in nature. Therefore, it is imperative to improve exploration prospects for such lithologies to test whether they can source RMGPs. The complex structural, temporal, and spatial variability of sediments, metamorphic rocks, and RMGPs in nature makes it challenging to establish a link between pegmatites and their crustal source. For example, lithium-rich sediments that are Cenozoic in age do not provide an accurate representation of the crustal rocks that source Precambrian or Palaeozoic RMGPs that are found in nature<sup>15</sup>. Therefore, it is critical to better constrain the redistribution of lithium during metamorphism in order to interpret the lithium content of metasedimentary rocks that are sourced from enriched sedimentary protoliths.

In conclusion, high-grade RMGP deposits (>5000 ppm lithium) may form by the low-degree (~10%) partial melting of enriched crustal rocks (~300-500 ppm lithium), followed by the moderate fractional crystallisation of the extracted melts or the re-melting of granitic cumulates. Alternatively, the anatexis of unconventional metasedimentary sources (>1000 ppm lithium) can produce enriched granitic melts, containing >5000 ppm lithium. Other rare-metals (Rb, Cs, and Ta) and major elements (Na<sub>2</sub>O, P<sub>2</sub>O<sub>5</sub>, and F) may become further enriched during fractional crystallisation of extracted melts or late-stage metasomatic processes. In both genetic models, the enrichment of lithium in RMGPs begins with the anatexis of an enriched crustal source (>300 ppm lithium). Such deposits are poorly explored in nature; therefore, it is important to further investigate their occurrence to better understand RMGP genetic models.

## Methods

**Experimental setup.** The experiments were conducted in either a piston-cylinder apparatus or an internally-heated pressure vessel at the Institute des Sciences de la Terre d'Orléans (ISTO), France (CNRS, BRGM, Université d'Orléans) (Table 1). To prepare the starting materials, the metasedimentary samples were powdered or melted at 1200°C. Partial melting and crystallisation experiments were performed on rock powders and glasses, respectively, at 750-800°C and 400-1000 MPa. The pressure-temperature conditions were chosen to investigate the dehydration melting of white micas and staurolite in orogenic settings<sup>26,34,36,59-62,66</sup>. Water was added to some of the charges to investigate the melting of biotite (Table 1).

The starting materials were crushed and ground using an agate mortar until the minerals were finely powdered (<10 µm). The powders were loaded in 5 mm outer diameter gold capsules for the partial melting experiments, and Au-Pd (80% gold, 20% palladium)

capsules for the crystallisation and high temperature experiments. Water was subsequently added to the fluid-present experiments (~10 wt%), and the capsules were welded shut.

The high temperature melting experiments were performed in an internally-heated pressure vessel at 1200°C ( $\pm 0.1^\circ\text{C}$ ) and 200-400 MPa ( $\pm 1$  bar)<sup>67</sup>. The vessel was pressurised with pure argon and 20 MPa of an Ar-H<sub>2</sub> gas mixture (2% volume hydrogen) in order to buffer the oxygen fugacity ( $f_{\text{O}_2}$ ) of the system, which is calculated to be ~QFM. The apparatus was heated to 1200°C, which was monitored using a K-type thermocouple. After 72 hours, the samples were quenched using a drop-quench mechanism. At the start of the experiments, the capsules were placed in an alumina basket, which was suspended near the hot end of the vessel with a platinum wire. At the end of the experiments, the wire was cut by a current, and the alumina basket with the capsules fell down to the cold end of the vessel. The Barr888 glass was extracted, crushed, and re-loaded into Au-Pd capsules for the crystallisation experiments (Table 1). The low-temperature partial melting and crystallisation experiments of Barr888 were subsequently performed in an internally-heated pressure vessel at 800°C, 400 MPa, and an  $f_{\text{O}_2}$  of ~QFM (20 MPa of Ar-H<sub>2</sub>). At the end of the experiments (up to 120 hours), the power to the vessels was cut, and the samples were quenched within ~5 minutes by the cooling system.

Low-temperature partial melting and crystallisation experiments of the three metasedimentary rocks were also performed in a Bristol type piston-cylinder apparatus<sup>37,68</sup> using 3/4 inch assemblies. The capsules were wrapped in an MgO sleeve and the empty space was filled with MgO powder. The sleeve was placed in a graphite-Pyrex-talc assemblage. The piston-cylinder was over-pressurised and heated to the desired temperature (monitored using a B-type thermocouple). The FM1 and ST19.03 partial melting and crystallisation experiments were conducted at 750°C ( $\pm 0.1^\circ\text{C}$ ) and 700 MPa ( $\pm 10$  bars). In the crystallisation experiments, the samples were first melted at 1150°C for ~24 hours, then the apparatus was

cooled down to 750°C. The Barr888 low-temperature partial melting and crystallisation experiments were performed at 750-800°C and 700-1000 MPa. High temperature melting experiments were also performed on the metabauxite at 1000-1200°C and 700-1000 MPa. The oxygen fugacity in the Barr888 experiments was buffered at CCO by the presence of carbon (ankerite) and sulfur (pyrite) in the metabauxite<sup>37</sup>. In contrast, the oxygen fugacity of the FM1 and ST19.03 experiments evolved from intrinsic (CCO)<sup>37</sup> to increasingly oxidising conditions following the dissociation of water to H<sub>2</sub>, and the diffusion of hydrogen through the capsule walls<sup>37</sup>. The piston-cylinder experiments lasted between 48 and 430 hours. At the end of the experiments, power to the apparatus was cut, and the samples were rapidly quenched within <1 minute by the cooling system. After the quenching, the capsules were cut in half, mounted in a 1-inch ring mount, and the samples were polished.

**Scanning electron microscopy.** All analyses of the starting materials and the run products were carried out at ISTO-CNRS-BRGM. The imaging of the samples and the identification of minerals was performed using a Merlin compact ZEISS scanning electron microscope<sup>67</sup>. Imaging was performed by backscattered electron (BSE) imaging and minerals were identified using a Bruker QUANTAX – Xflash6 energy dispersive spectrometer (EDS). The analyses were conducted using a focused beam with an accelerating voltage of 15 kV and a diaphragm diameter of 60 µm.

**Electron probe microanalysis.** Major element analyses were conducted using a CAMECA SX-Five or a JEOL IHP 200F electron probe microanalyser<sup>67</sup>. Analyses were performed using a defocused beam, with a diameter of 1-20 µm for minerals and 10-20 µm for glasses in order to limit the diffusion of alkalis. The accelerating voltage and intensity of the electron beam

was 15 kV and 5-6 nA, respectively. The concentration of major elements was quantified by wavelength dispersive spectroscopy (WDS). Sodium and potassium were analysed first in order to limit their diffusion under the electron beam. The conditions, standards, and crystals used for the analyses of both microprobes are summarised in Supplementary Table 1. All analyses are reported in Supplementary Data 3.

**Laser ablation inductively coupled plasma mass spectrometry.** Trace element analyses were performed using a RESolution-SE 193 nm Ar-F excimer laser ablation system with a Laurin Technique S155 ablation cell coupled with an Agilent 8900 QQQ quadrupole mass spectrometer<sup>67</sup>. A detailed description of the system parameters is summarised in Supplementary Table 2. The fluence and repetition rate of the beam was varied according to the phase analysed. Glasses were analysed at 2 J/cm<sup>2</sup> and 5 Hz, micas at 4-5 J/cm<sup>2</sup> and 5 Hz, other ferromagnesian minerals (staurolite, garnet, cordierite, and tourmaline) at 5 J/cm<sup>2</sup> and 5 Hz, oxides at 4 J/cm<sup>2</sup> and 3 Hz, feldspars at 4 J/cm<sup>2</sup> and 5 Hz, and quartz at 5-7 J/cm<sup>2</sup> and 5 Hz. All analyses were performed with a SQUID to smooth out signals at the low repetition rates. The beam diameter was kept constant at 10-20 µm. The trace element composition of phases could not be determined in some experiments due to the small size of melt pockets and crystals (<10 µm). The analysed isotopes include <sup>7</sup>Li, <sup>9</sup>Be, <sup>11</sup>B, <sup>23</sup>Na, <sup>24</sup>Mg, <sup>27</sup>Al, <sup>29</sup>Si, <sup>31</sup>P, <sup>35</sup>Cl, <sup>39</sup>K, <sup>44</sup>Ca, <sup>47</sup>Ti, <sup>51</sup>V, <sup>53</sup>Cr, <sup>55</sup>Mn, <sup>56</sup>Fe, <sup>59</sup>Co, <sup>60</sup>Ni, <sup>63</sup>Cu, <sup>66</sup>Zn, <sup>71</sup>Ga, <sup>72</sup>Ge, <sup>85</sup>Rb, <sup>88</sup>Sr, <sup>89</sup>Y, <sup>90</sup>Zr, <sup>93</sup>Nb, <sup>115</sup>In, <sup>118</sup>Sn, <sup>133</sup>Cs, <sup>137</sup>Ba, <sup>139</sup>La, <sup>140</sup>Ce, <sup>141</sup>Pr, <sup>146</sup>Nd, <sup>147</sup>Sm, <sup>153</sup>Eu, <sup>157</sup>Gd, <sup>172</sup>Yb, <sup>178</sup>Hf, <sup>181</sup>Ta, <sup>182</sup>W, <sup>208</sup>Pb, <sup>232</sup>Th, and <sup>238</sup>U. The isotopes were each measured at a dwell time of 10 ms, with a total sweep time of 0.4-0.7 seconds. The instrument settings were tuned following the ablation of a NIST-612 standard in order to minimise isotope fractionation (U/Th = 1.00 ± 0.02%), the production of oxides (ThO/Th < 0.1%), and the production of doubly-charged ions (<sup>42</sup>Ca<sup>2+</sup>/<sup>42</sup>Ca<sup>+</sup> < 0.2%). A NIST-610 glass was used as the external

standard and NIST-612 and BCR-2G glasses were used as standards for quality control in silicate minerals. For the oxides, a GSE-2G glass was used as the external standard, and GSD-2G as a standard for quality control. Three analyses were performed on the external standards and 1 analysis was performed on the quality control standards after every 10-15 analyses of the samples. The raw data were processed with Glitter<sup>69</sup> using a linear fit to correct for instrument drift. To quantify the trace elements, SiO<sub>2</sub> and FeO or TiO<sub>2</sub> were used as the internal standards for silicate minerals and Fe-Ti oxides, respectively. The signals were individually screened to identify contaminated analyses. Time-resolved signals that showed element spikes or irregular shapes resulting from the analyses of mixed phases were discarded (Supplementary Fig. 1). The precision and accuracy of lithium analyses in the quality control reference materials (NIST-612, BCR-2g, and GSD-2G) were mostly better than 2%, 5%, and 15%, respectively (see Supplementary Data 4).

**Bulk composition.** The bulk composition of the starting materials was determined by major and trace element analyses of the homogeneous glasses produced by the high-temperature (1200°C) melting experiments. Minor traces of sillimanite and corundum are present as evenly distributed residual phases in the Barr888 high temperature experiment (A1); therefore, major and trace element analyses were performed with a beam size of 100 µm on clusters of glass, corundum, and sillimanite.

**Modal mineralogy.** The modal proportion of phases in the run products were determined by mass balance calculations (Supplementary Fig. 3). A linear least-squares regression fit was applied to the normalised major element composition of the assemblages using the *lsqlin* function in MATLAB<sup>25</sup>. In the calculations, it is assumed that the bulk composition of the

sample remains constant before and after the experiments. The modal mineralogy of the run products and the uncertainty is reported in Supplementary Tables 3 and 4, respectively. Phase proportions could only be determined for some of the experiments because the minerals were too small to analyse in several samples.

**Trace element modelling.** The anatectic model was constrained using the experiments from this study, partial melting experiments on a paragneiss ('Pg') by Michaud et al.<sup>26</sup>, and the crystallisation experiments on the Beauvoir granite by Pichavant<sup>23</sup> (Figs. 7a and 7b). The modal mineralogy of the Pg experiment (6% biotite, 35% plagioclase, 9% alkali feldspar, 42 % quartz, 5% orthopyroxene, 4% spinel) was calculated using the modal abundance of minerals in the paragneiss and the stoichiometry of the melting reactions established in Michaud et al.<sup>26</sup>. Partition coefficients of orthopyroxene (0.2) were derived from Neukampf et al.<sup>46</sup>. The modal mineralogy of the Beauvoir crystallisation experiments was taken from Pichavant<sup>23</sup>. Michaud et al.<sup>26</sup> has also performed partial melting experiments on an orthogneiss ('Og'); however, these were not considered for the trace element modelling of lithium due to under-estimations of the bulk lithium and caesium contents by mass balance calculations<sup>26</sup>.

The anatexis of variably enriched crustal rocks in nature (Fig. 7c) was modelled using the mean  $D_{Li}^{Mineral/Melt}$  obtained from all experiments in this study. Using the variable  $D_{Li}^{Mineral/Melt}$  from previous experimental studies can yield a wide range of  $\bar{D}_{Li}$  values (0.02 to 0.31), which can lead to significantly different interpretations of lithium endowment during crustal anatexis. For example, at low melt fractions (~10%), the enrichment of lithium during the complete breakdown of biotite may vary between a factor of ~2.5 and ~8 as a function of  $\bar{D}_{Li}$  calculated from previous experiments (Fig. 7d).

574

## 575 **Data availability**

576 The authors declare that the data supporting the findings of this study are available within the  
577 paper and its supplementary files.

578

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## Author contributions

The manuscript was prepared by BH, a PhD candidate, who is supervised by FG, AMG, ÉG, AP, and JM. The project was conceptualized by JM, ÉG, and FG. Funding acquisition and

resource management was carried out by JM. The starting materials were obtained and characterized by ÉG and AP, respectively. The experiments were performed by BH, AMC, and JG. These authors were helped by IDC and SE for major and trace element analyses, respectively. Data curation and the writing of the manuscript was done by BH, with the support of AMG and FG. The final version of the manuscript was reviewed and approved by all authors.

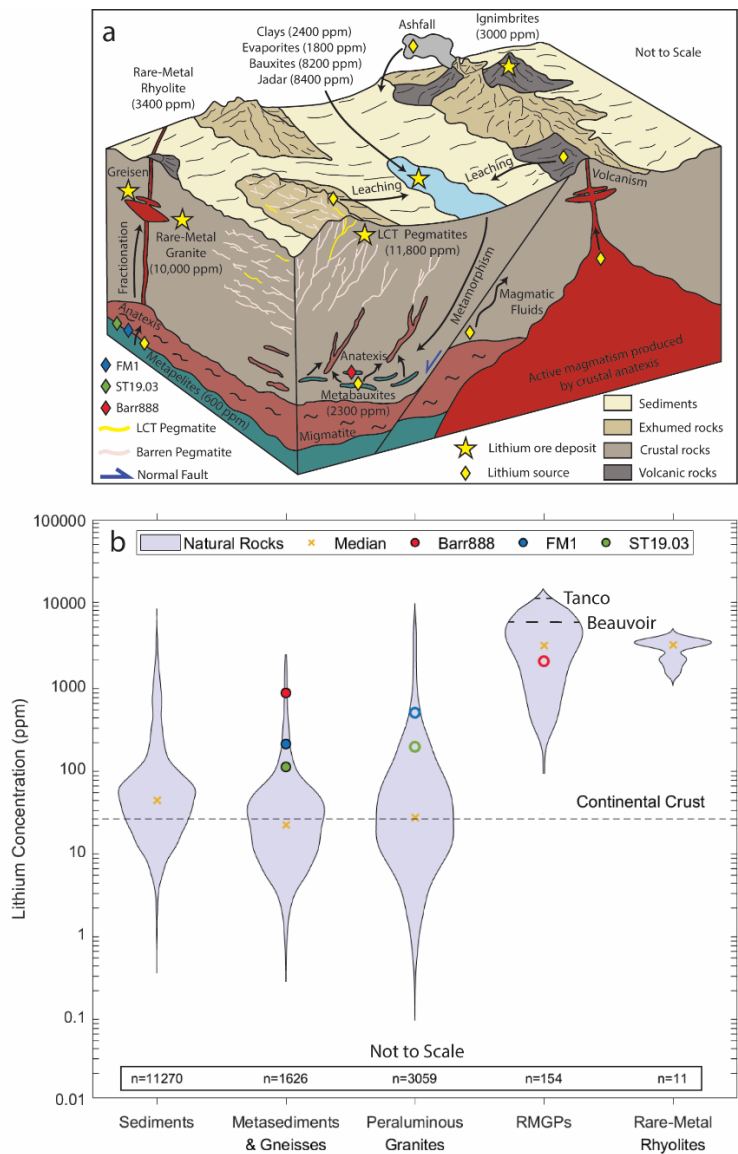
### **Competing interests**

The authors declare no competing interests.

### **Additional information**

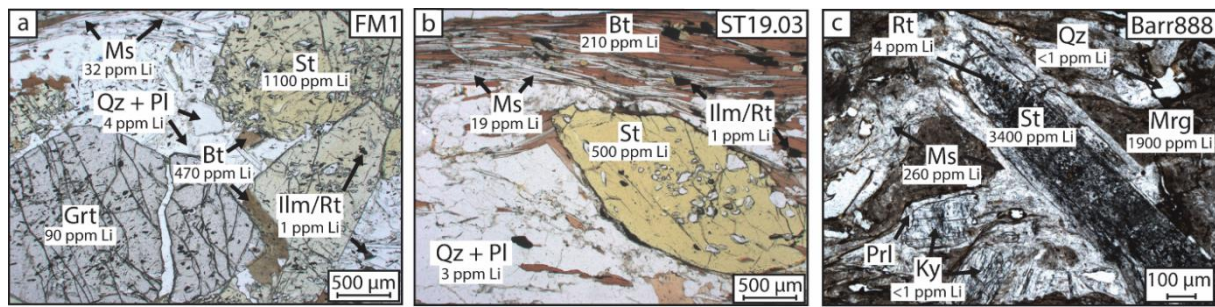
**Supplementary information.** The online version contains supplementary material, available at <https://doi.org/10.1038/XXXX>.

**Correspondence** and requests for materials should be addressed to Bence Horányi.



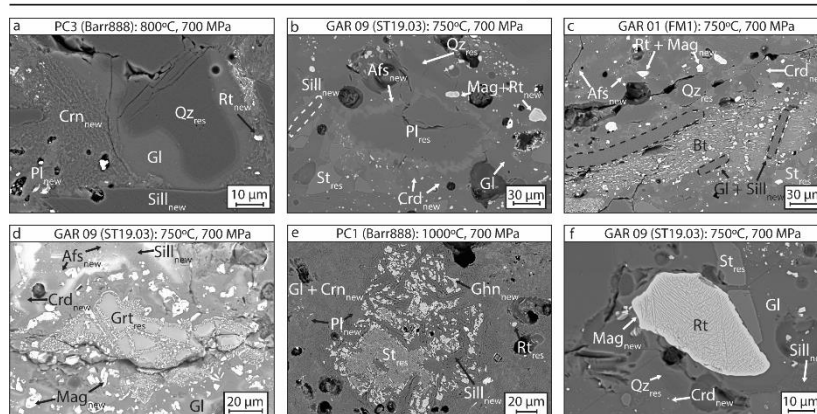
807

808 **Fig. 1 The lithium cycle in the continental crust. a.** The distribution and possible cycling of  
809 lithium between magmatic, sedimentary, and metamorphic reservoirs (modified from  
810 Gloaguen et al.<sup>30</sup> and Lefebvre and Tavignot<sup>70</sup>). Numbers in brackets indicate the highest  
811 recorded lithium content in each deposit. **b.** Violin plots of the lithium distribution in various  
812 crustal reservoirs (see Supplementary Data 2). Filled and open circles represent the  
813 composition of the starting materials and the mean composition of the glasses from the low-  
814 degree melting experiments, respectively.



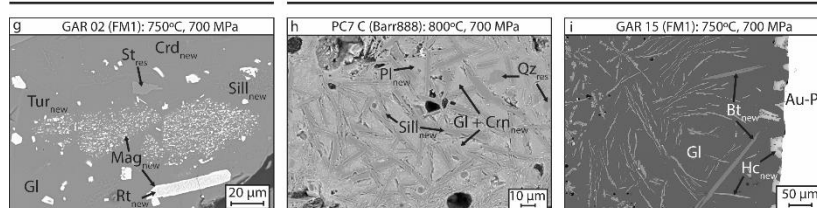
**Fig. 2 Representative thin section images of the starting materials and the distribution of lithium between minerals. a. FM1 micaschist. b. ST19.03 micaschist. c. Barr888 metabauxite.** Mineral abbreviations are: St = staurolite, Bt = biotite, Ms = muscovite, Mrg = margarite, Grt = garnet, Qz = quartz, Pl = plagioclase, Ilm = ilmenite, Rt = rutile, Prl = pyrophyllite, Ky = kyanite.

### Fluid-Absent Melting



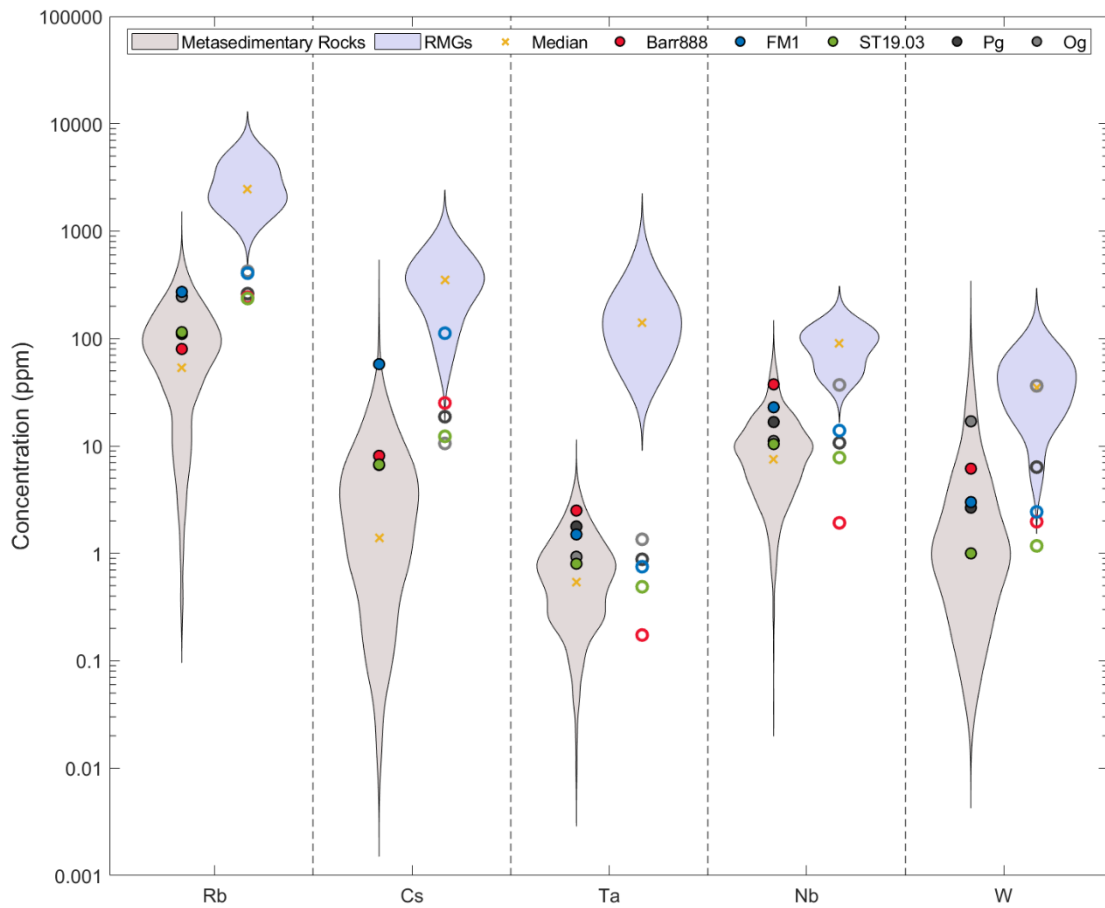
### Fluid-Present Melting

### Crystallization

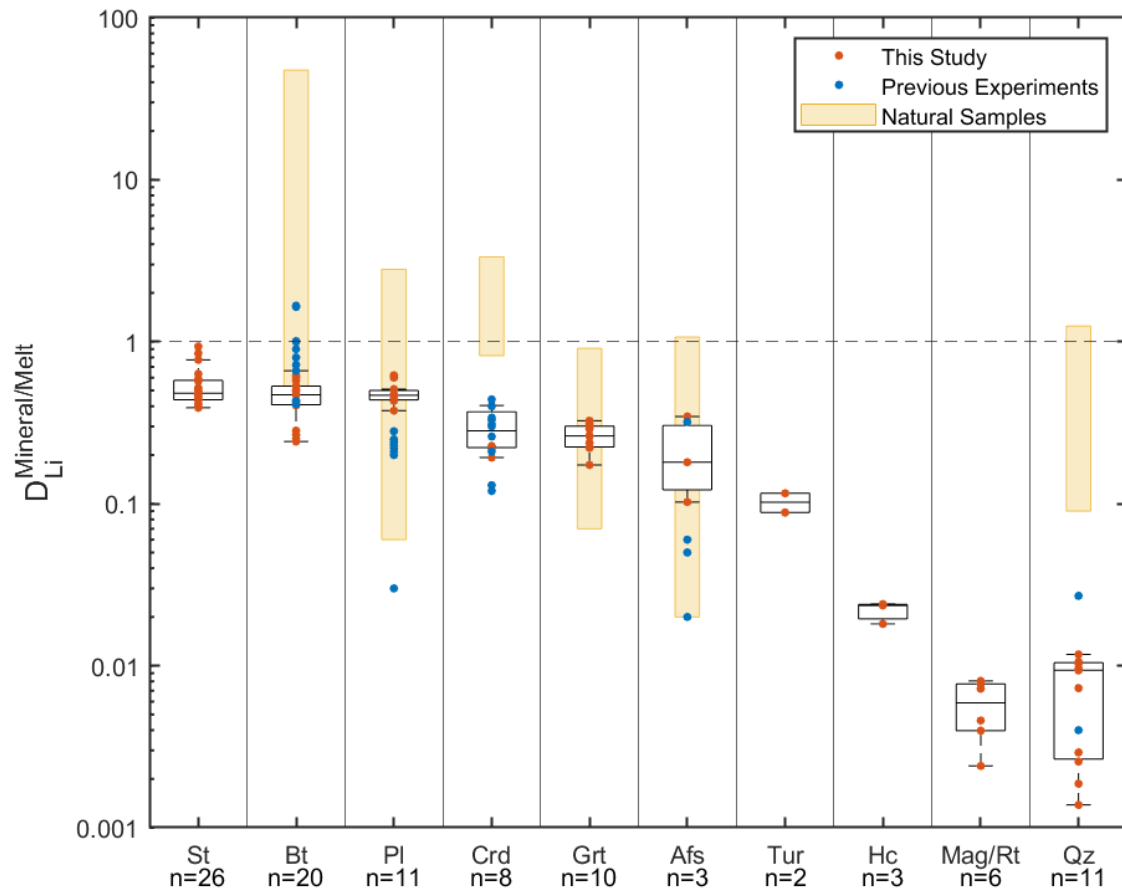


**Fig. 3 Representative SEM images of mineral textures in the experimental run products.**

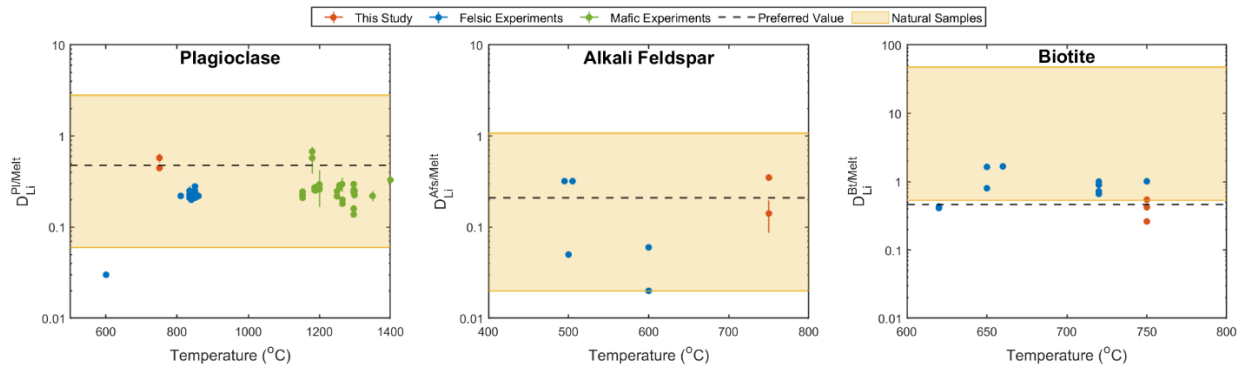
**a.** Melt pockets next to residual quartz phenocrysts. **b.** The replacement of residual plagioclase rims with newly crystallising alkali feldspar. **c.** The breakdown of muscovite to sillimanite and melt and the exsolution of magnetite from residual biotite. **d.** The incomplete breakdown of garnet to cordierite, magnetite, and melt. **e.** The partial breakdown of staurolite to gahnite, sillimanite, and melt. **f.** Rutile and magnetite intergrowths in pseudomorphs of ilmenite. **g.** The complete breakdown of biotite to garnet, tourmaline, magnetite, and melt in the fluid-present melting experiments. **h.** Representative mineralogy of Barr888 crystallisation experiments. **i.** Representative mineralogy of the FM1 crystallisation experiments. Phase abbreviations are: Gl = glass, Crn = corundum, Rt = rutile, Sill = sillimanite, Pl = plagioclase, Mag = magnetite, St = staurolite, Grt = garnet, Ghn = gahnite, Hc = hercynite, Qz = quartz, Afs = alkali feldspar, Bt = biotite, Crd = cordierite. 'res' denotes residual phases, whereas 'new' signifies newly crystallising minerals.



**Fig. 4** The rare-metal content of the starting metasedimentary rocks (filled circles) and glasses from the low-degree partial melting experiments (open circles). Violin plots in grey and blue represent the distribution of rare-metals in metasedimentary rocks (see Supplementary Data 2) and rare-metal granites<sup>6</sup>. The ‘Pg’ and ‘Og’ experiments were derived from Michaud et al.<sup>26</sup>.

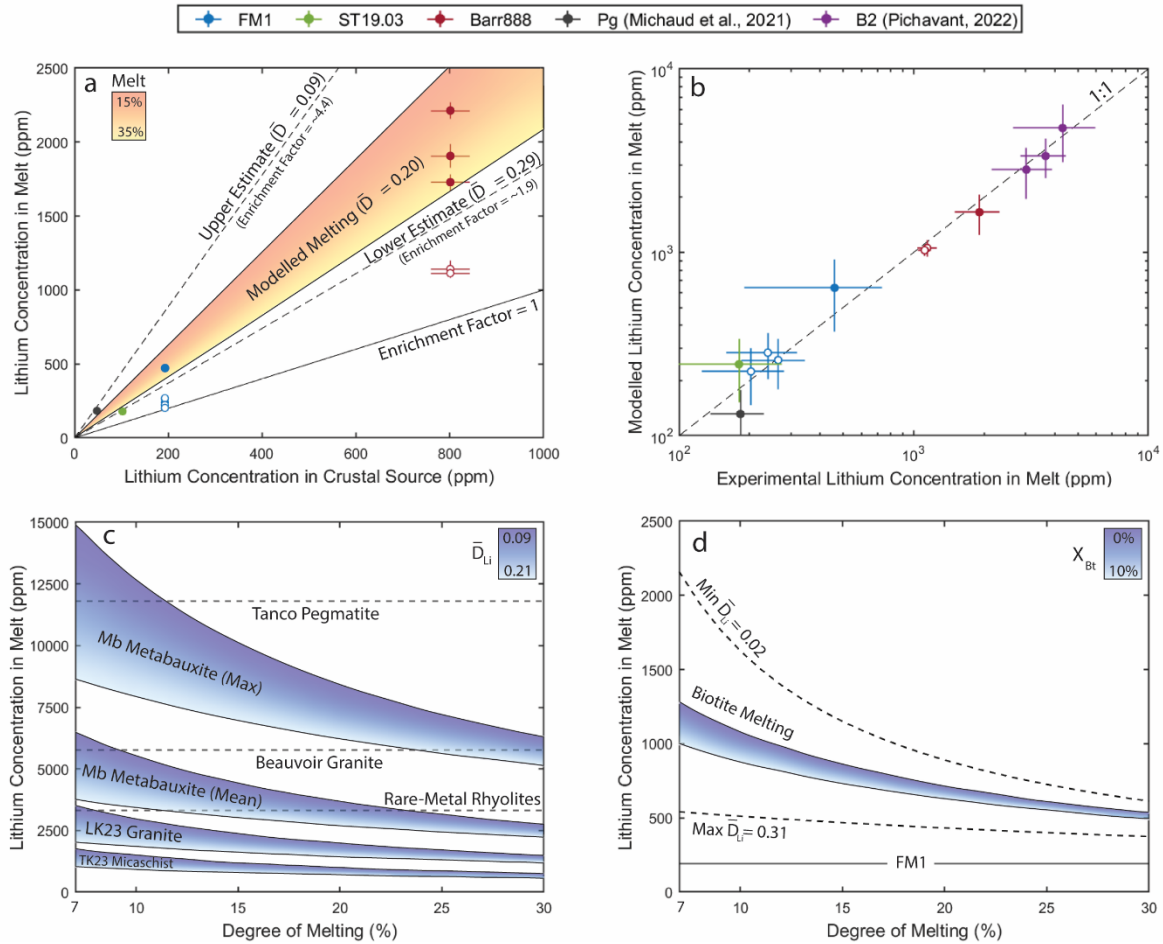


**Fig. 5 Mineral-melt partition coefficients of lithium obtained from the partial melting and crystallisation experiments (red circles and box plot).** The range of partition coefficients from natural samples<sup>13,44-48</sup> and from previous experimental studies in felsic systems<sup>17-24</sup> are indicated by the orange box and the blue circles, respectively. Mineral abbreviations are reported in Fig. 3.



847

848 **Fig. 6 The effect of temperature on the partitioning of lithium into residual and newly**  
849 **crystallising minerals. a. Plagioclase. b. Alkali Feldspar. c. Biotite.** Partition coefficients are  
850 presented from this study (red circle), felsic experiments in the literature<sup>17,19-24</sup> (blue circles),  
851 mafic experiments in the literature (green circles; Supplementary Table 14), and natural  
852 samples<sup>13,44-48</sup> (range shown by the orange box). The dashed line highlights the preferred  
853 values that were subsequently used for trace element modelling. The error bars represent the  
854 standard deviation of the mean ( $1\sigma_m$ ).



**Fig. 7 The enrichment of lithium during crustal anatexis. a.** The enrichment of lithium in the partial melts of the experiments from this study and the ‘Pg’ experiment of Michaud et al.<sup>26</sup>. Filled circles represent fluid-absent melting experiments, whereas open circles represent glasses from crystallisation, fluid-present, or high-temperature experiments. **b.** Comparison of the experimental and modelled lithium contents in partial melts. The error bars represent the standard deviation of the mean ( $1\sigma_m$ ). **c.** The enrichment of lithium during the partial melting of a diverse composition of crustal rocks, taken from Franceschelli et al.<sup>63</sup>, Knoll et al.<sup>14</sup>, and Koopmans et al.<sup>16</sup>. **d.** Sensitivity test of the trace element model to highlight the effect of biotite breakdown during crustal anatexis. The effect of using variable  $D_{Li}^{Mineral/Melt}$  from previous experimental studies is demonstrated by the dashed lines.

Table 1: Experimental conditions and run products.

Run	Starting Material	Apparatus*	Temperature (°C)	Pressure (MPa)	Duration (Hours)	H <sub>2</sub> O added (wt%)	Newly Formed Phases	Residual Phases
<b>High Temperature Melting</b>								
PC1	Barr888	PC	1000	700	96	0	Gl + Pl + Gnt + Sill + Crn + Rt	St
PC2	Barr888	PC	1200	1000	48	0	Gl + Sill + Crn	n.a.†
A1	Barr888	IHPV	1200	400	48	10		
BH_MSed1	FM1	IHPV	1200	200	72	0	Gl + Crn + Hc	n.a.
BH_MSed4	ST19.03	IHPV	1200	200	72	0		
<b>Fluid-absent Melting</b>								
GAR01	FM1	PC	750	700	330	0	Gl + Crd + Tur + Afs + Sill + Rt + Mag	St + Bt + Grt + Qz + Pl
GAR09	ST19.03	PC	750	700	69	0	Gl + Crd + Afs + Sill + Rt + Mag	St + Bt + Grt + Qz + Pl
PC5	Barr888	PC	750	700	144	0		
PC3	Barr888	PC	800	700	168	0	Gl + Pl + Afs + Sill + Crn + Aln + Rt + Fe-Ti Oxide <sup>§</sup>	St + Qz
PC7 F	Barr888	PC	800	700	168	0		
PC8 F	Barr888	PC	800	1000	168	0		
A2 F	Barr888	IHPV	800	400	120	0	Gl + Pl + Hc + Sill + Crn + Aln + Rt + Fe-Ti Oxide	St + Qz
A3	Barr888	IHPV	800	400	48	0		
<b>Fluid-present Melting</b>								
GAR02	FM1	PC	750	700	330	10	Gl + Crd + Tur + Sill + Crn + Rt + Mag	St + Qz
GAR03	FM1	PC	750	700	330	10		
<b>Fluid-absent Crystallisation</b>								
GAR05	FM1	PC	750	700	72	0	Gl + Bt + Hc + Sill + Crn	St + Qz
GAR08	FM1	PC	750	700	72	0		
GAR13	FM1	PC	750	700	430	0	Gl + Bt + Hc + Sill	n.a.
PC7 C	A1 Glass	PC	800	700	168	0	Gl + Pl + Afs + Hc + Sill + Crn + Fe-Ti Oxide	Qz
PC8 C	A1 glass	PC	800	1000	168	0	Gl + Pl + Bt + Hc + Sill + Crn + Fe-Ti Oxide	Qz
A2 C	A1 Glass	IHPV	800	400	120	0		
A4	A1 Glass	IHPV	800	400	168	0	Gl + Pl + Hc + Sill + Crn + Fe-Ti Oxide	Qz
<b>Fluid-present Crystallisation</b>								
GAR06	FM1	PC	750	700	72	10	Gl + Bt + Hc + Sill + Crn	St + Qz
GAR07	FM1	PC	750	700	72	10		
GAR15	FM1	PC	750	700	430	10	Gl + Bt + Hc	n.a.

Abbreviations are Gl = glass, St = staurolite, Bt = biotite, Grt = garnet, Crd = cordierite, Tur = tourmaline, Rt = rutile, Mag = magnetite, Gnt = gahnite, Hc = hercynite, Qz = quartz, Pl = plagioclase, Afs = alkali feldspar, Sill = sillimanite, Crn = corundum.

\*PC = piston-cylinder (oxygen fugacity = ~CCO), IHPV = internally-heated pressure vessel (oxygen fugacity = ~QFM)

†n.a. = not applicable.

<sup>§</sup>Fe-Ti oxides are too small to be precisely identified.

Table 2: Representative major and trace element compositions of the starting materials, experimental glasses, and RMGs.

Sample wt%	Starting Materials			Fluid-Absent Melting Experiments			RMGPs
	FM1	ST19.03	Barr888	GAR 01	GAR 09	A2 F	RMGs
	n = 10	n = 10	n = 12	n = 19	n = 11	n = 3	n = 127
SiO <sub>2</sub>	63.7 (4)	61.2 (3)	45.3 (9)	71 (1)	73.3 (7)	68.4 (3)	70 (2)
TiO <sub>2</sub>	0.82 (2)	0.94 (4)	1.99 (9)	0.14 (6)	0.13 (3)	0.29 (1)	b.d. to 0.02
Al <sub>2</sub> O <sub>3</sub>	17.1 (2)	18.6 (3)	35 (1)	12.7 (5)	12.7 (3)	14.7 (2)	16.4 (9)
FeO <sub>(t)</sub>	7.0 (2)	6.7 (1)	2.3 (2)	1.3 (4)	0.44 (6)	1.1 (1)	0.4 (2)
MgO	2.11 (6)	2.40 (6)	0.25 (3)	0.3 (1)	0.11 (3)	0.10 (1)	0.2 (2)
MnO	0.17 (3)	0.08 (3)	n.a.*	0.11 (3)	0.02 (2)	b.d.†	b.d. to 0.35
CaO	0.23 (2)	0.25 (2)	3.3 (1)	0.38 (6)	0.17 (1)	0.99 (2)	0.6 (6)
Na <sub>2</sub> O	1.01 (8)	2.05 (7)	1.29 (7)	1.9 (4)	2.8 (2)	1.2 (4)	4.3 (8)
K <sub>2</sub> O	3.89 (8)	3.67 (7)	2.3 (1)	5.6 (5)	5.2 (1)	6.2 (3)	3.3 (6)
ZnO	n.a.	n.a.	0.4 (1)	n.a.	0.04 (2)	0.0902 (6)	n.a.
P <sub>2</sub> O <sub>5</sub>	0.11 (4)	0.14 (3)	0.03 (2)	0.16 (8)	0.15 (5)	0.04 (2)	2 (1)
F	b.d	b.d.	b.d.	0.06 (5)	0.2 (1)	0.46 (6)	2 (1)
Cl	0.02 (1)	0.010 (6)	n.a.	0.22 (3)	0.006 (4)	n.a.	n.a.
-(F,Cl) = O	0.005 (3)	0.002 (1)	b.d.	b.d. to 0.02	0.08 (5)	0.20 (3)	0.7 (5)
Total	96.1 (5)	96.0 (4)	92 (1)	94 (2)	95.0 (6)	93.2 (6)	98 (1)
ppm	n = 10	n = 10	n = 16	n = 13	n = 7	n = 8	n = 127
Li	193 (3)	102 (1)	800 (40)	470 (30)	180 (10)	1900 (80)	3000 (2000)
Rb	224 (2)	138 (2)	80 (4)	432 (6)	240 (20)	235 (8)	3000 (1000)
Nb	14.4 (2)	10.9 (3)	37 (2)	13 (2)	8 (2)	2.1 (9)	90 (30)
Cs	49.5 (5)	6.9 (1)	8.1 (0.3)	130 (20)	12.3 (5)	25 (2)	300 (200)
Ta	0.91 (6)	0.70 (5)	2.5 (1)	0.8 (3)	0.5 (1)	0.2 (1)	200 (100)
W	1.5 (2)	0.9 (1)	6.2 (4)	2 (1)	1.2 (5)	3 (1)	40 (30)
Bulk D <sub>Li</sub> <sup>§</sup>	n.a.	n.a.	n.a.	0.15	0.22	0.22	n.a.
Bulk D <sub>Rb</sub>	n.a.	n.a.	n.a.	0.34	0.37	0.004	n.a.
Bulk D <sub>Nb</sub>	n.a.	n.a.	n.a.	1.84	1.95	2.29	n.a.
Bulk D <sub>Cs</sub>	n.a.	n.a.	n.a.	0.22	0.29	0.004	n.a.
Bulk D <sub>Ta</sub>	n.a.	n.a.	n.a.	2.05	2.30	4.01	n.a.
Bulk D <sub>W</sub>	n.a.	n.a.	n.a.	0.44	0.39	0.01	n.a.
ASI <sup>#</sup>	n.a.	n.a.	n.a.	1.3 (1)	1.21 (4)	1.4 (1)	1.5 (2)
Melt Fraction	n.a.	n.a.	n.a.	18%	25%	34%	n.a.

Detailed major and trace element compositions of glasses is reported in Supplementary Table 8. Composition of 'PHP' RMGs is taken from Linnen and Cuney<sup>6</sup>. Uncertainty is reported to the last digit(s) in the brackets as the standard deviation of the mean ( $1\sigma_m$ ).

\*n.a. = not analysed.

†b.d. = below detection.

<sup>§</sup>Bulk partition coefficients are calculated using equation (6).

<sup>#</sup>ASI is calculated as the molar ratio of  $Al_2O_3/(Na_2O + K_2O + (CaO - \frac{1}{2}P_2O_5))$ .

Table 3: Representative major and trace element compositions of minerals in the FM1 experimental run products.

	St <sub>(res)</sub>	Bt <sub>(res)</sub>	Grt <sub>(res)</sub>	Cr <sub>d</sub> ( <sub>new</sub> )	Tur( <sub>new</sub> )	Pl( <sub>res</sub> )	Afs( <sub>new</sub> )	Qz( <sub>res</sub> )	Hc( <sub>new</sub> )	Mag( <sub>new</sub> )
Sample	GAR 01	GAR 01	GAR 01	GAR 03	GAR 01	GAR 01	GAR 01	GAR 01	GAR 15	GAR 03
wt%	n = 16	n = 8	n = 10	n = 6	n = 2	n = 4	n = 6	n = 4	n = 1	n = 3
SiO <sub>2</sub>	27.4 (7)	38 (2)	36.0 (3)	48.9 (2)	36 (3)	67.6 (8)	64.6 (4)	100.7 (6)	0.25	0.13 (6)
TiO <sub>2</sub>	0.5 (1)	1.6 (3)	0.05 (3)	b.d.* to 0.2	0.6 (1)	b.d. to 0.02	0.02 (1)	0.006	0.65	1.4 (5)
Al <sub>2</sub> O <sub>3</sub>	53.9 (9)	19.9 (4)	20.7 (1)	34.7 (5)	32 (2)	20.7 (6)	19.1 (8)	0.04 (3)	48.17	2.7 (3)
FeO <sub>(t)</sub>	13 (1)	12 (1)	34 (2)	3.2 (6)	8 (2)	0.17 (6)	0.4 (1)	0.2 (1)	41.41	85.0 (6)
MgO	3 (1)	14.1 (9)	1.5 (3)	11.6 (1)	5.6 (2)	0.016 (7)	0.02 (1)	0.02 (2)	7.34	0.12 (3)
MnO	0.3 (2)	0.09 (6)	7 (2)	0.3 (1)	0.10 (3)	0.05 (3)	0.04 (2)	b.d.	1.50	0.03 (1)
CaO	0.04 (3)	b.d. to 0.02	1.1 (2)	0.09 (3)	0.8 (4)	1.6 (4)	0.4 (3)	b.d.	0.02	0.03 (1)
Na <sub>2</sub> O	b.d. to 0.01	0.48 (9)	0.04 (4)	0.13 (6)	1.8 (2)	10.1 (4)	2.1 (7)	0.03	0.02	0.01
K <sub>2</sub> O	0.06 (2)	9.3 (9)	0.012 (9)	0.15 (7)	b.d. to 0.04	0.5 (3)	13 (1)	0.013 (6)	0.05	0.11 (2)
ZnO	n.a.†	n.a.	0.06 (4)	0.04 (3)	n.a.	0.12	b.d.	0.06 (3)	n.a.	n.a.
P <sub>2</sub> O <sub>5</sub>	0.07 (6)	b.d. to 0.03	0.03 (3)	0.06 (4)	n.a.	0.13 (7)	0.06 (4)	0.03 (2)	0.04	0.03
F	b.d.	n.a.	b.d.	b.d.	n.a.	0.05 (4)	0.2 (1)	b.d.	n.a.	n.a.
Cl	0.02	n.a.	0.006 (5)	0.005 (2)	n.a.	0.007 (1)	0.007	0.0057 (9)	n.a.	n.a.
-(F,Cl) = O	0.004	n.a.	b.d. to 0.0005	b.d. to 0.0004	n.a.	b.d. to 0.02	0.09 (6)	0.0013 (2)	n.a.	n.a.
Total	99 (1)	95 (1)	100.3 (7)	99.3 (3)	85 (2)	101.0 (6)	100.0 (8)	101.0 (5)	99.45	89.5 (9)
ppm	n = 9	n = 5	n = 7	n = 8	n = 2	n = 3	n = 2	n = 3	n = 3	n = 1
Li	240 (40)	199 (6)	130 (20)	80 (20)	48 (9)	270 (30)	70 (30)	4.6 (3)	4.4 (7)	b.d.
Rb	5 (2)	790 (50)	0.24 (6)	20 (5)	n.a.	b.d. to 3.92	340 (30)	0.03 (2)	b.d.	b.d.
Nb	1.7 (3)	37 (2)	0.03 (2)	b.d. to 0.07	0.14 (7)	0.12	0.6 (6)	0.0005	0.38 (5)	135
Cs	1.1 (5)	260 (80)	b.d.	23 (4)	n.a.	b.d. to 0.59	10 (7)	0.011 (2)	0.1	b.d.
Ta	0.24 (6)	2.0 (2)	0.03 (1)	0.011 (3)	0.017 (6)	b.d.	0.02 (1)	0.0008	0.02 (1)	5.51
W	0.05 (5)	0.7 (3)	0.04 (2)	0.06	b.d.	b.d.	0.2 (1)	0.006	b.d.	43.13
D <sub>Li</sub> <sup>§</sup>	0.51 (9)	0.42 (1)	0.28 (5)	0.29 (8)	0.10 (2)	0.58 (6)	0.14 (6)	0.0098 (6)	0.022 (3)	b.d.
D <sub>Rb</sub>	0.010 (4)	1.8 (1)	0.0006 (1)	0.06 (2)	b.d.	b.d. to 0.009	0.79 (8)	0.00006 (4)	b.d.	b.d.
D <sub>Nb</sub>	0.12 (2)	2.8 (2)	0.002 (1)	b.d. to 0.009	0.011 (5)	0.009	0.04 (4)	0.00004	0.070 (9)	17.25
D <sub>Cs</sub>	0.009 (4)	2.0 (6)	b.d.	0.34 (6)	b.d.	b.d. to 0.004	0.08 (6)	0.00008 (2)	0.001	b.d.
D <sub>Ta</sub>	0.32 (8)	2.7 (2)	0.03 (1)	0.025 (6)	0.022 (8)	b.d.	0.03	0.001	0.04 (2)	12.33
D <sub>W</sub>	0.02 (2)	0.3 (1)	0.015 (7)	0.01	b.d.	b.d.	0.09	0.003	b.d.	6.57

Mineral abbreviations are given in Fig. 3. Detailed major and trace element compositions of minerals in the starting materials and experimental run products are reported in Supplementary Tables 6, 7, 8, 9, 10, and 11. Uncertainty is reported to the last digit(s) in the brackets as the standard deviation of the mean ( $1\sigma_m$ ).

\*b.d. = below detection.

†n.a. = not analysed.

<sup>§</sup>Partition coefficients are calculated using equation (5).



13 June 2025

Dear Dr. Evan Hastie

Enclosed is my review of manuscript COMMSENV-25-2064-T entitled “**Lithium enrichment in granites and pegmatites: a reflection of their crustal source**” by *B. Horányi, A. Gion, F. Gaillard, E. Gloaguen, A. Plunder, J. Melleton, A. Moradell-Casellas, J. Garde, S. Erdmann, and I. Di Carlo*.

I recommend the manuscript for publication with minor revisions.

The article brings much needed experimental data to answer the question as whether or not all rare-metal granitoids originate from fractionated crystallization from a sizeable peraluminous parental granite. The authors relied on an integrated study using experimental work to calculate partition coefficients that subsequently feed melt-production modelling. Note that as I reviewed this paper from a metamorphic petrologist perspective and not as an experimental petrologist. Nonetheless, my expertise is relevant since the authors experimented on metamorphic rocks as potential sources for rare-metal granitoids.

The demonstration is compelling and in line with recent studies and support the conclusion that the anatexis of rare-metal-enriched metapelites and other metasedimentary rocks of more exotic compositions such as bauxite is a suitable mechanism to produce rare-metal granitoids. For metabauxites, because of their rare occurrence in nature, the frequency at which this material would melt may be overstated.

The manuscript is well written and well organized. My comments are minor and mostly deal with wording and adding some clarification over a few points, particularly in the method section.

I hope my review will be helpful in reaching a decision.

Sincerely yours,

Dr. Manuel Duguet

### **General comments:**

Recent work (laboratory experiment and field studies) called into question whether rare-metal granites and pegmatites originated exclusively from parental peraluminous granites by fractionated crystallization. Age discrepancies between putative parental granites (when present) and the rare-metal granites and the level of fractionation required to reach lithium saturation for spodumene crystallization led researchers to consider anatexis of a suitable protolith (i.e., enriched) as an alternate viable mechanism. Although many field studies documented the possible relationships between anatexis and rare-metal granites, the feasibility of such a genetic link still needed to be demonstrated on the petrogenetic front. This was only addressed very recently by several laboratory experiment studies testing the partial melting of diverse enriched sources (e.g., granitoids). In addition of testing these sources, these studies aimed also at improving our knowledge of melt-minerals partitioning coefficients for rare metals that are indispensable for the accuracy of melt modelling.

This article explores the suitability of metasedimentary rocks moderately to highly enriched in rare metals as potential source for rare-metal granites and pegmatites. 3 samples were studied; two are staurolite-bearing micaschists enriched in rare metals compared to the average metapelites, the third one is a metabauxite. Chemical composition of these samples is compared to those in a voluminous database assembled by the authors and covering sediments (pelites, bauxites), sedimentary rocks and their metamorphic equivalents and rare-metal granitoids. This problematic is addressed in two consecutive steps. The authors present first results related to their partial melting experimental work on these samples which objective is to assess mineral-melt partition coefficients for the elements of interest (Li, Rb, Nb, Cs, Ta, W) with a clear emphasis on lithium. Both fluid-absent and fluid-present melting configurations are investigated for a range of pressures commonly encountered in the continental crust (200 MPa to 1000 MPa). Using newly acquired melt-minerals partitioning coefficients, the authors model the composition of melt generated by crustal anatexis of aluminum-rich metasedimentary rocks and granitoids. Without surprise, low-degree partial melting experiments (less than 10%) produce the most enriched melts. Passed that threshold, enrichment of the melt in these elements may drop in some cases by an order of magnitude. The most enriched protolith in elements such as lithium (i.e., bauxite) will yield melts that require little to no fractionation to reach the saturation point in lithium for spodumene to crystallize.

Results of mineral-melt partition coefficients presented in this paper do not deviate substantially from previous studies for some of the silicate phases. The novelty is clearly in the type of material, both rocks and mineral phases (e.g. staurolite), that is investigated. The silicate phases present in the starting material are ubiquitous in metamorphosed metapelites and the case of staurolite is particularly interesting because it can accommodate up to several 1000s ppm of lithium making it a primary source for lithium in the melt. However, only rocks displaying anomalous compositions in zinc, aluminum and lithium would stabilize staurolite at pressures and temperatures at which it would participate as a reactant to melt generation and not reacting out before the solidus at lower temperature (metastable staurolite). Another way to involve staurolite in melting-reaction would be to lower the solidus by having unusual high-fluid content. Very few studies have addressed the possibility of existence of suprasolidus staurolite, either being the product of melting reactions or participating to them (see for instance Spear

et al. 1999 and Carcía-Casco et al. 2003). This is a problematic that is not currently discussed or taken into account (regrettably so) in thermodynamic modelling for metamorphic rocks.

For this experiment, staurolite-melting reaction could only be texturally documented for the metabauxite sample (Barr888). Bauxite is not the most abundant nor is it the most survivable material in geological formations. Beside their very peculiar chemistry, their preservation before erosion and potential burial necessitates a specific set of circumstances that are rarely met. Erosion of such material and deposition in turbiditic sequences of highly aluminous metapelites is a viable alternative that was not mentioned. In addition, such sources would not be suitable to generate the numerous LCT-type pegmatites in the Archean because aluminum-rich pelites do not appear in significant amount before the Paleoproterozoic. There were other possible scenarios with less enriched metapelites such as metastable staurolite at P-T conditions at which partial melting place or melt infiltration (disequilibrium melting of staurolite) that could have extended the pool of fertile material. It is argued that the small volume of enriched sources such as metabauxites and staurolite-melting reactions may be under documented; especially for the former, I am not fully convinced by this. This is one of the very few criticisms I would have for this paper. However, it must be pointed out that the authors try to address this issue by assembling a voluminous rock geochemistry database (several 10000s of chemical analyses) to assess the representativeness in term of frequency and composition of the chosen samples. The other objective was to compare the chemical compositions of experimental melt products to those encountered in natural enriched rocks (i.e., rare-metal granitoids). Discussion on the experiment results is sound and compared to different sources (e.g., orthogneiss). For fluid-absent experiments, some elements (Li, Cs, Rb) in the produced melt are a good match with natural product composition whereas others (Nb, Ta, W) are not. These discrepancies are appropriately pondered and discussed in the light of other studies. It is particularly relevant because the peritectic phases crystallizing with the melt play a significant role in this. This can only be explored in the light a careful examination of the textures of the melting experiment from which melting reactions are deduced. This contribution executed this part of the study appropriately.

This experimental work and modelling study succeed at demonstrating the viability of anatexis of aluminous metasedimentary rocks enriched for producing melts enriched in rare metals. The geochemistry of the experimental melt falls within the range of those of natural rocks and are enriched enough in rare metals that lower amount of fractionation compared to the parental granite model is required to achieve observed ore-grade deposits. The behavior of lithium partitioning depends strongly on what phases crystallize during melting and this complexity is properly discussed by the authors. Similar discussions, although shorter, are conducted for the other elements. Melt modelling from the newly acquired partitions coefficients strengthens conclusions presented in this paper.

In conclusion, this article is a valuable and necessary contribution to our understanding of the petrogenesis of rare-metal granitoids. Length wise the article falls within the parameters allowed by Nature Communications with the following tally: abstract: 163 words, body of the text: 5873 words, methods: 2044 words.

### **Specific comments:**

Most of my specific comments are embedded in the manuscript and the supplementary note. I will address in this section corrections related to the tables of the supplementary material.

- Regarding the excel files of the supplementary material, I suggest that you add in the supplementary note a list detailing the names of the files and what they contain (i.e., sheet tabs). For instance, **In the excel file 12179\_0\_data\_set\_10715251\_svhfqm**, the names of the different sheet tabs are imprecise because excel is quite limited in the number of characters you can input in these. As an example, the second sheet is titled *Major elements- Metasediments* whereas *minerals chemistry-major elements-metasediments* would be more accurate. I believe it is a necessity considering the number of tables that you have. You need to make it easy for a reader to navigate through them.
- In some tables (body of the article and supplementary material alike): parameter “n” is present but is never explicated. I assume that you talk about the number of analyses here. This should be added to the relevant tables. It may sound like a mere technicality, but every acronym and abbreviation should be explained.
- **Supplementary Table 5:**  
For FM1 sample: longitude for a sample located in Spain should be 8.11W, therefore -8.11 according to your convention.  
for ST19.03 sample, Lat-Long coordinates were likely inverted. A latitude of -2.97 would place that sample close to the equator.
- **Supplementary Table 5:** 2 typos need to be corrected at the bottom line: ***Institut*** instead of ***Institute*** and ***Grenoble*** instead of ***Grénoble***
- **Supplementary Table 11:** Title is obviously incomplete and needs to be fixed.
- **In the excel file 12179\_0\_data\_set\_10715250\_svhfqm:** in the sediments sheet, please specify the unit of the “age” column. I guess they are in millions of years (Ma).