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Barium isotope evidence for a magmatic fluid-dominated petrogenesis of LCT-type pegmatites

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Abstract

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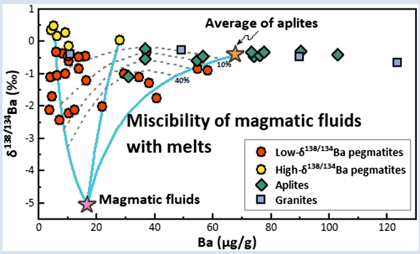

Understanding the petrogenesis of granitic pegmatites associated with Li mineralisation is fundamental in constraining rare metal enrichment mechanisms. However, there is still significant controversy surrounding the central issue of the nature of pegmatite-forming liquids. Here, we report Ba isotope data for a 3000 m borehole in the Jiajika pegmatite field that hosts the largest hard rock-type Li deposit in Asia. The pegmatites exhibit markedly lower $\delta^{138/134}$Ba than the wall rocks and the average upper continental crust, which cannot be attributed to low degree anatexis of metasedimentary or crystal fractionation of highly evolved granitic magmas. Instead, modelling suggests that their low $\delta^{138/134}$Ba most likely results from substantial involvement (10–40 %) of isotopically light hydrothermal fluids exsolved from the underlying magmatic reservoirs. In the context of transcrustal magmatic system, these magmatic fluid components may be a key medium in extracting, concentrating, and transporting Li to provide a material source for mineralisation in granitic pegmatites.

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Introduction

Granitic pegmatites are granitoids with characteristic textures and constitute one of the most important sources of rare metal elements (London, 2018). The Li-Cs-Ta (LCT) pegmatites, as a type of Li deposit with high grade and wide distribution (Fig. 1a), supply approximately half of global Li production and are the main target of future exploration (Benson *et al.*, 2017). Exploring the petrogenesis of LCT-type granitic pegmatites is thus crucial for understanding the enrichment mechanisms of Li and other rare metals.

Most LCT-type pegmatites are considered to be the product of extreme differentiation of granitic magmas (Thomas and Davidson, 2016; London, 2018). However, the nature of the pegmatite-forming liquids, especially with regard to the role of H_2O and fluxing elements (*e.g.*, B, F, and P), is still highly controversial. The constitutional zone refining model, based mainly on experimental petrology, large scale whole rock analyses, and simulations, suggests that the initial pegmatite-forming liquids are essentially granitic melts inherited from the parental magmas (London, 2018). Incompatible components, including H_2O, fluxing elements, and rare metals, are continuously enriched in a boundary layer liquid adjacent to the crystallisation front (London, 2014). In this scenario, H_2O and fluxing elements only play a role in locally lowering viscosity and increasing elemental diffusivity, with aqueous fluid considered unnecessary or even unlikely. In contrast, an alternative model, based mainly on the studies of melt and fluid inclusions, proposes that the formation of LCT-type pegmatites begins with single phase supercritical fluids resulting from miscibility of magmatic fluids and silicate melts (Thomas and Davidson, 2016). These supercritical fluids can acquire very high initial water and rare metal contents from granitic source magmas, followed by phase separation after emplacement that further enriches Li in the H_2O-rich melt/fluid phase (Thomas *et al.*, 2019). Numerous attempts have been made to distinguish between the two models (Zhang *et al.*, 2021; Troch *et al.*, 2022), yet a consensus remains elusive.

Barium (Ba) is fluid-mobile and compatible during the crystallisation of granitic magmas. Recent studies highlight the great potential of Ba isotopes in distinguishing between crystal fractionation and magmatic fluid effect. The near eutectic composition of granitic pegmatites implies that K-feldspar controls the Ba budget of the crystalline mineral assemblage, which could result in significantly higher $\delta^{138/134}$Ba (defined as $[(^{138}\text{Ba}/^{134}\text{Ba})_{\text{sample}} / (^{138}\text{Ba}/^{134}\text{Ba})_{\text{NIST3104a}} - 1] \times 1000$ (‰)) in the residual melts (Deng *et al.*, 2021). Conversely, as magmatic fluids are experimentally demonstrated to preferentially incorporate light Ba isotopes relative to silicate melts (Guo *et al.*, 2020), their substantial involvement would dramatically reduce $\delta^{138/134}$Ba of the pegmatite systems (Huang *et al.*, 2021; G. Deng *et al.*, 2022). Hence, Ba isotopes offer a powerful tool for deciphering the nature of pegmatite-forming liquids.

Here, we present high precision Ba isotope data for a suite of borehole samples from the Jiajika granitic pegmatite field (eastern Tibetan Plateau). This pegmatite field is formed within the Jiajika gneiss dome that has a two-mica granite core, mantled

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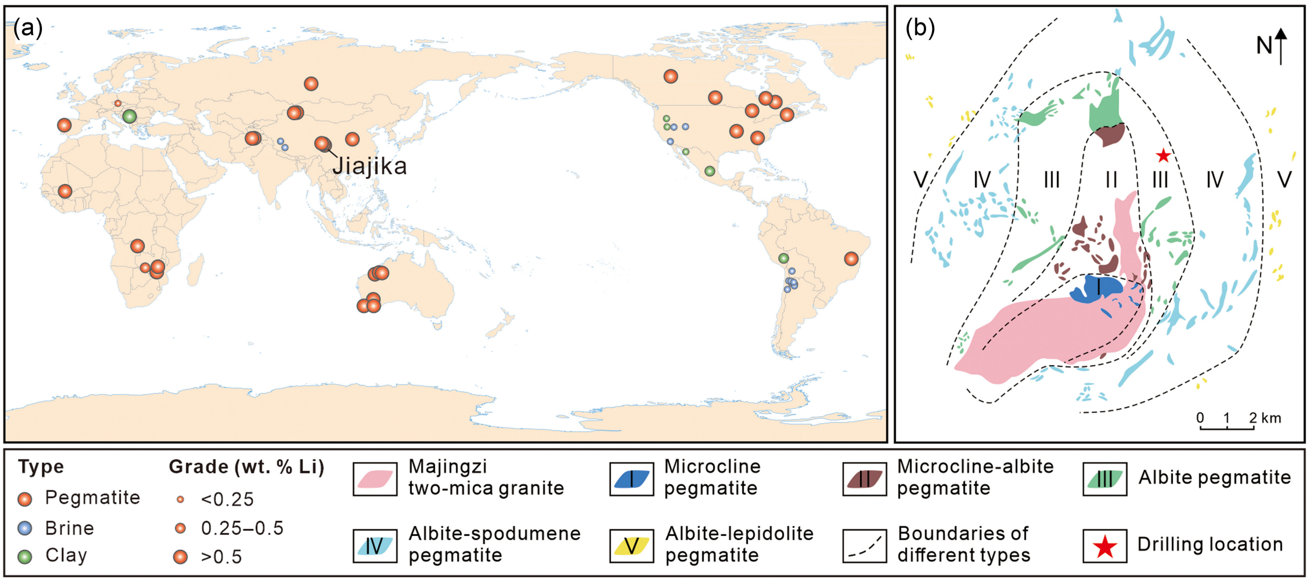

Figure 1 **(a)** Location and grades of representative pegmatite, brine, and clay Li deposits worldwide. See data sources in Table S-1. **(b)** Simplified geological map of the Jiajika pegmatite field (modified after Zhao *et al.*, 2022).

by a succession of metasedimentary rocks. More than 500 LCT-type pegmatite veins are spatially zoned around the two-mica granite, in the order from the pluton outward: microcline pegmatite (I), microcline-albite pegmatite (II), albite pegmatite (III), albite-spodumene pegmatite (IV), and albite-lepidolite pegmatite (V) (Fig. 1b). The Li mineralisation is mainly developed within zones IV and V, as well as some veins in zone III, with total estimated Li_2O reserves of up to 3.0 Mt, ranking the first in Asia (Huang *et al.*, 2020). The studied samples were collected from different depths of a borehole of 3211.21 m, which consists of 35 % metasediments, 14 % granite-aplites, and 51 % pegmatites (Fig. 2a) (Xu *et al.*, 2023). Spodumene-bearing pegmatites are only observed at depths between 0 and 100 m, while deeper pegmatites correspond to surface zones I and II pegmatites without Li mineralisation (Wang *et al.*, 2023). Detailed geological background and sample description are available in the Supplementary Information. Due to their proximity to the buried main part of the parental granite compared to samples collected from the surface, these borehole samples may provide a strong documentation of the initial compositional characteristics of the pegmatite system. Our results show extremely low $\delta^{138/134}$Ba of pegmatites, supporting a comprehensive influence of magmatic fluids on their compositions, and thus a possible key role in rare metal enrichment.

Results

The Ba isotope analytical method is detailed in the Supplementary Information. The long-term external precision for $\delta^{138/134}$Ba is better than 0.05 ‰ (2 s.d.). The $\delta^{138/134}$B of rock standards agree with published values, and the results of replicates are consistent within error (Table S-2), demonstrating the reliability of our data.

There are significant variations in $\delta^{138/134}$Ba of the borehole samples (Fig. 2). Overall, pegmatites have the lowest Ba contents (3.7–130.2 μg/g, mostly <50 μg/g) and the largest range of $\delta^{138/134}$Ba (−2.44 to +0.47 ‰) among the four sample types. Notably, their $\delta^{138/134}$Ba does not show a systematic variation with depth. Granitic aplites and granites have moderate and less variable Ba contents (30.9–102.9 μg/g and 11.0–123.8 μg/g, respectively) and narrower $\delta^{138/134}$Ba ranges (−1.08 to −0.22 ‰ and −0.67 to −0.28 ‰, respectively). The Ba contents of metasedimentary wall rocks (125–1238 μg/g) are significantly higher and more variable than the other samples, but their $\delta^{138/134}$Ba shows a rather restricted range (−0.31 to +0.40 ‰) around a mean of 0.00 ‰, which matches the upper continental crust (UCC, 0.00 ± 0.04 ‰; Nan *et al.*, 2018).

Discussion

Controls on Ba isotope variation. The possible mechanisms driving Ba isotope fractionation in granitoids include chemical alteration, wall rock assimilation, anatexis, and magma differentiation. The first scenario can be excluded as all igneous samples are fresh. Their loss on ignition (LOI) is generally very low (≤1 wt. %, Table S-2), the chemical index of alteration (CIA) mostly falls within the range of fresh granitoids, and neither parameter shows any correlation with $\delta^{138/134}$Ba (Fig. S-1). Secondly, because the wall rocks have a limited $\delta^{138/134}$Ba range that is higher than granites, aplites, and most pegmatites, wall rock assimilation could not produce the extremely low $\delta^{138/134}$Ba of most pegmatites. Although some pegmatite samples have similar $\delta^{138/134}$Ba as the wall rocks, they do not show significantly elevated Ba contents except for sample J084317 (higher than all aplites and granites) (Fig. 3a). Therefore, wall rock assimilation cannot explain Ba isotope fractionation between the Jiajika igneous samples.

Some studies suggest that LCT-type pegmatites may result from low degree muscovite dehydration melting of a metasedimentary source containing Li-rich rocks, rather than as products of extreme magma differentiation (Zhao *et al.*, 2022). However, our Ba isotope data do not support this model. According to the dehydration melting reaction Muscovite + Plagioclase + Quartz = K-feldspar + Sillimanite + melt, K-feldspar should govern the Ba budget of the restite. Because K-feldspar is enriched in light Ba isotopes relative to melts (Deng *et al.*, 2021), the resulting melts should acquire higher $\delta^{138/134}$Ba than the source. In this case, the extremely low $\delta^{138/134}$Ba of most pegmatites implies even lower $\delta^{138/134}$Ba for their source rocks, which is not consistent with the observation of (meta-)sedimentary rocks (Nan *et al.*, 2018). For the pegmatite samples with high $\delta^{138/134}$Ba, their low Rb/Sr does not support muscovite

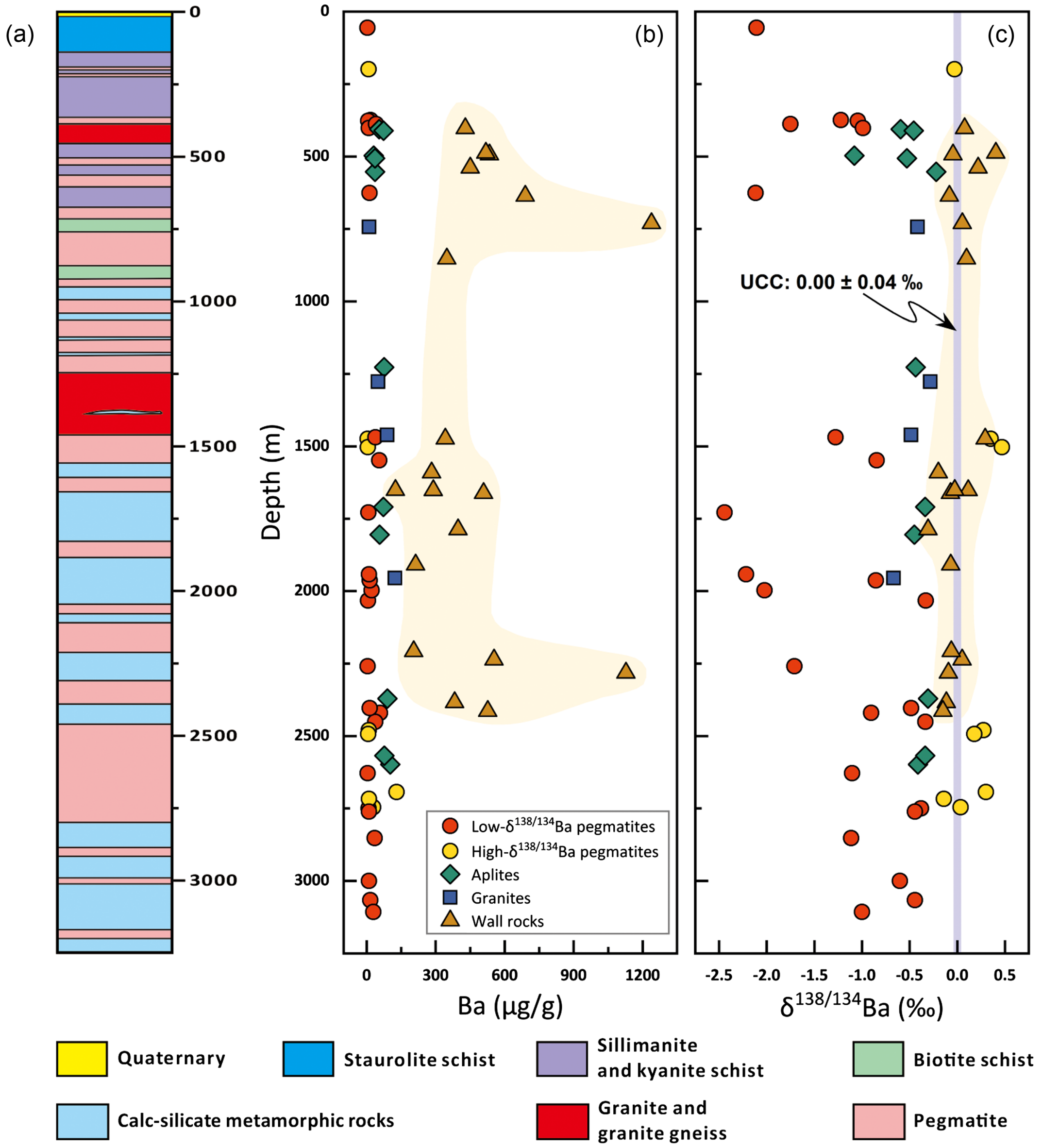

Figure 2 **(a)** Schematic cross section of the studied drill hole (modified after Xu *et al.*, 2023). **(b)** Ba and **(c)** $\delta^{138/134}$Ba *vs.* depth. The purple band represents the weighted average $\delta^{138/134}$Ba of the upper continental crust (UCC, 0.00 ± 0.04 ‰; Nan *et al.*, 2018). Error bar is smaller than the symbols.

dehydration melting of a metasedimentary source either (Gao *et al.*, 2017) (Fig. S-2a).

As the vast majority of studies support the Jiajika granite-pegmatite association (Huang *et al.*, 2020; Zhang *et al.*, 2021), the Ba isotope variation among them should be linked to the magma evolution toward pegmatite-forming liquids. Due to the highly evolved nature of the studied granitoids, major element indicators, such as SiO_2 and MgO contents, cannot discern the degree of differentiation (Fig. S-2b,c), while trace element ratios, such as Ba/Rb, can help (Zhang *et al.*, 2021). With the exception of sample J007522, which has abnormally high Ba/Rb likely due to sampling bias, Ba/Rb decreases continuously from granites through aplites to pegmatites (Fig. 3b). This reflects the fractional crystallisation of Ba-rich minerals, which, based on the variation in mineral composition of the borehole samples (Xu *et al.*, 2023), first involves both K-feldspar and mica, and then is dominated by K-feldspar. Because K-feldspar and mica preferentially incorporate light and heavy Ba isotopes, respectively (Deng *et al.*, 2021), such fractional crystallisation can explain the overall consistent $\delta^{138/134}$Ba of granites and aplites, as well as the elevated $\delta^{138/134}$Ba of some pegmatites (Fig. 3b). However, it cannot account for the significantly lower $\delta^{138/134}$Ba and higher K_2O contents of most pegmatites than those of granites and aplites (Fig. S-2d).

The low $\delta^{138/134}$Ba and high K_2O contents of most pegmatites cannot be explained by crystallisation within pegmatite veins. According to the constitutional zone refining model, light Ba isotopes with higher diffusivity can preferentially feed mineral crystallisation through the boundary layer liquid. Meanwhile, the variation in mineral assemblage of the Jiajika pegmatites indicates that the early crystalline minerals are dominated by Ba- and K-rich microcline that preferentially incorporates light Ba isotopes. Thus, $\delta^{138/134}$Ba of most pegmatites is expected to gradually increase with decreasing Ba and K contents, which is not observed (Figs. 3d, S-2d). Furthermore, although Fe isotope fractionation among these pegmatite samples was suggested to result from fractional crystallisation (Luo *et al.*, 2024), no significant correlation between $\delta^{138/134}$Ba and δ^{56}Fe is seen (Fig. S-2e).

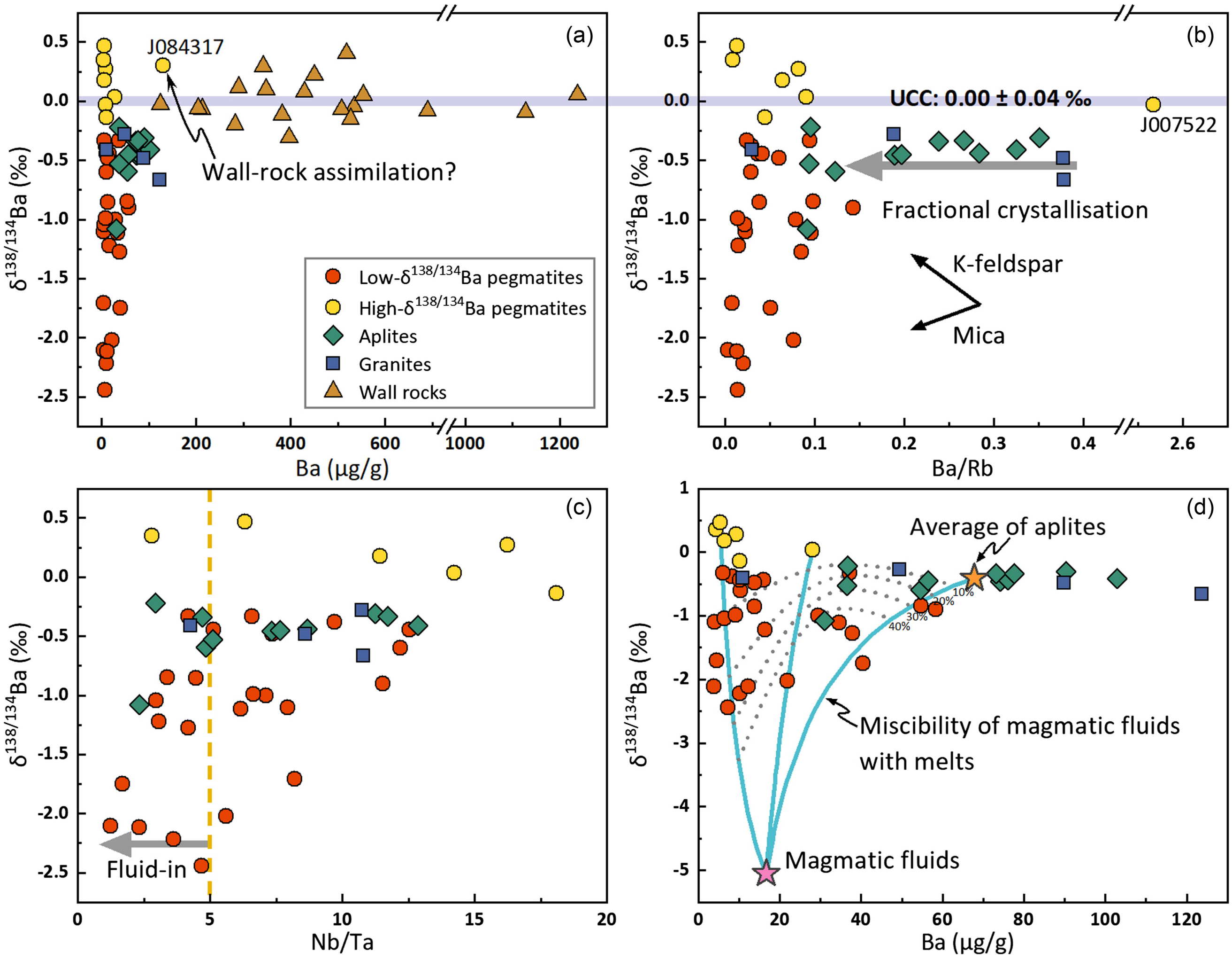

Figure 3 $\delta^{138/134}$Ba *vs.* **(a)** Ba, **(b)** Ba/Rb, and **(c)** Nb/Ta. Arrows in **(b)** indicate evolution of melts during fractional crystallisation of K-feldspar and mica. **(d)** Simulation of magmatic fluids mixing with highly differentiated melts and comparison with observations. Numbers on the line of mixing denote the fraction of magmatic fluids. The purple band represents the weighted average $\delta^{138/134}$Ba of the UCC. Error bar is smaller than the symbols.

The most likely process responsible for the low $\delta^{138/134}$Ba of pegmatites is modification by magmatic fluids. According to a previous experimental study by Guo *et al.* (2020), the apparent Ba isotope fractionation factor between exsolved fluids and residual melts can be up to −4.6 to −5.1 ‰ under the near solidus cold storage conditions (650–600 °C) of the shallow "mushy" magma reservoirs (Rubin *et al.*, 2017). Considering the generally low Ba contents of most pegmatites (<50 μg/g with an average of 17.7 μg/g), their extremely low $\delta^{138/134}$Ba could well be the result of the incorporation of magmatic fluids into the pegmatite-forming liquids. This interpretation is in line with their tendency towards low Nb/Ta (<5) and Zr/Hf (<26) (Figs. 3c, S-2f), which are often interpreted as results from magmatic-hydrothermal interaction (Ballouard *et al.*, 2016). The slightly higher Nb/Ta and Zr/Hf of some low-$\delta^{138/134}$Ba pegmatites are likely influenced by the crystallisation of accessory minerals, such as columbite-tantalite and zircon, indicating that Ba isotopes might be more sensitive to magmatic fluid activity than these elemental indicators. Consistent with our conclusion, the B and Li isotope compositions (also fluid-mobile elements) of the Jiajika pegmatites have been demonstrated to be significantly modified by magmatic fluids (Zhang *et al.*, 2021; Huan *et al.*, 2023). Metamorphic dehydrated fluids from the wall rocks are enriched in heavy Ba isotopes (Wu *et al.*, 2020), thus cannot provide the light Ba isotope compositions for most pegmatites.

To further decipher the exact mechanism of magmatic fluid participation in pegmatite formation, a model of magmatic fluid mixing with highly differentiated melt is developed in a $\delta^{138/134}$Ba-Ba diagram. Because of continuous decompression and crystallisation, fluid exsolution can occur early in magma evolution and throughout the magma ascent path (Edmonds and Woods, 2018). Thus, when calculating the composition of the magmatic fluid using the Rayleigh fractionation equations, the initial melt is represented by the average of all granites and aplites from various depths. The average of aplite samples and two pegmatites with elevated $\delta^{138/134}$Ba and different Ba/Rb ratios covering the main Ba/Rb range of Jiajika pegmatites are selected to represent highly differentiated melts. The calculations and results are detailed in the Supplementary Information.

As shown in Figure 3d, the variations in Ba elemental and isotope compositions of most pegmatite samples can be explained by the mixing of 10–40 % magmatic fluids with melts of different degrees of evolution. This range of fluid proportions is much wider than that of typical granitic magmas, but it is consistent with the supercritical fluids model of the pegmatite-forming liquids. The studies of melt and fluid inclusions show that

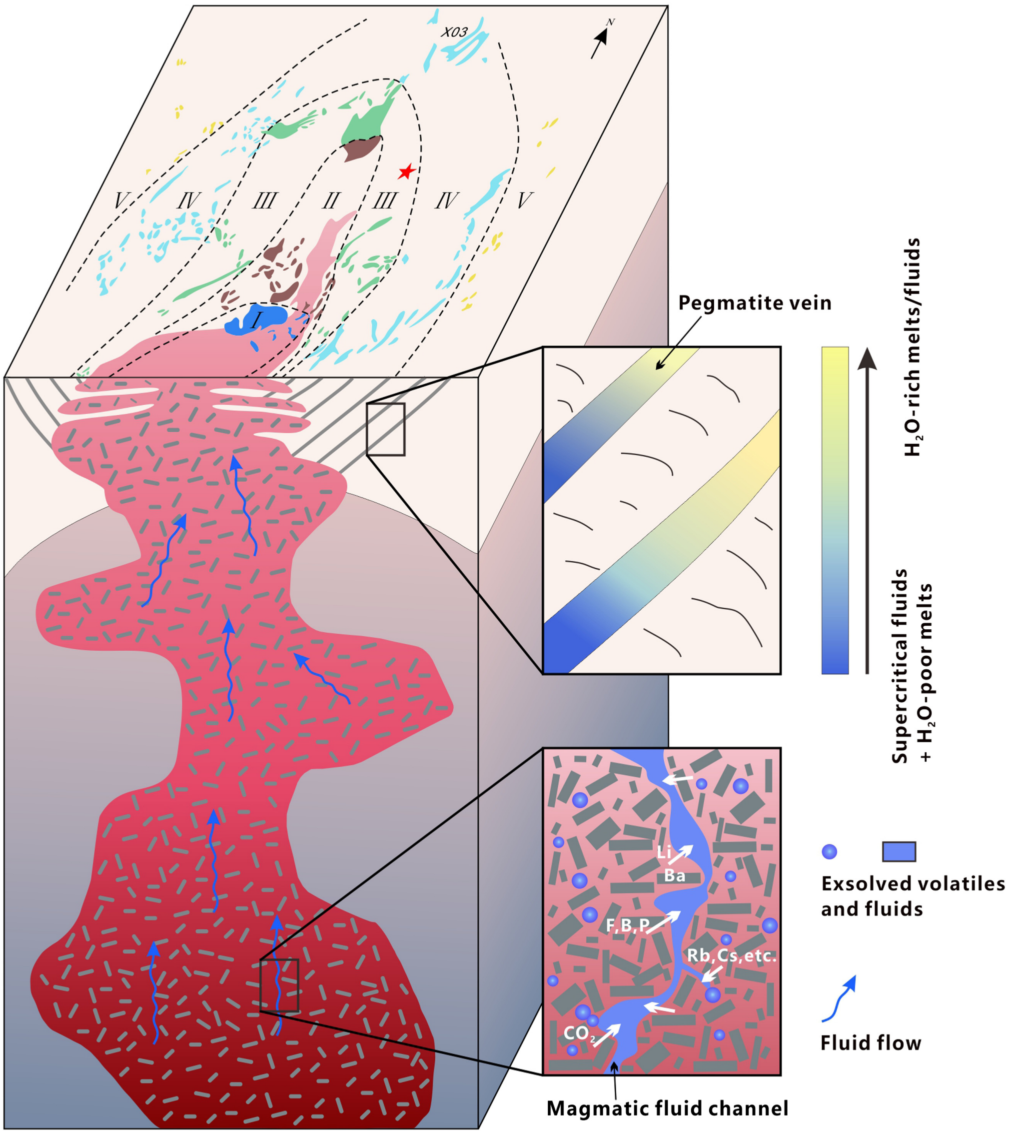

Figure 4 Schematic model for the role of magmatic fluids in intra-crustal material transport and petrogenesis and metallogenesis of granitic pegmatites.

supercritical fluids have initial water contents ranging from 20 to 33.3 % (Thomas and Davidson, 2016). After intrusion, they separate into conjugated H_2O-poor melts and H_2O-rich melts/fluids by melt-melt-fluid immiscibility. The former become the pegmatites with low fluid proportions, while the latter can continue to migrate away from the parental granite due to low viscosities and high mobilities (Troch *et al.*, 2022). Potential supercritical fluid activity in the Jiajika pegmatite field is also supported by previous observations of melt/fluid inclusions. Firstly, representative primary inclusions are commonly enriched in H_2O, fluxing components, alkaline elements, CO_2, and carbonates, but depleted in alkaline earth elements (Li and Chou, 2017; J. Deng *et al.*, 2022). These features are similar to those found in other pegmatites showing supercritical fluid activity (Thomas *et al.*, 2019; Wang *et al.*, 2023), and could explain the higher K_2O contents of most pegmatites compared to granites and aplites. Secondly, the trapping conditions of these inclusions (500–720 °C, 3–5 kbar) are consistent with the critical conditions for the complete miscibility of silicate melts with fluids obtained from experiments with addition of B, F, and Na (600 °C, 4 kbar) (Sowerby and Keppler, 2002). In this case, the multiple magmatic-hydrothermal events experienced by the Jiajika pegmatites (Xu *et al.*, 2023) could result from multiple injections of supercritical fluids, thereby leading to the lack of systematic variation in their $\delta^{138/134}$Ba with depth. Overall, the Ba isotope signatures of our studied deep pegmatites unequivocally demonstrate that magmatic fluids should overwhelmingly influence the composition and nature of the system from the early stages of pegmatite formation. Barium isotopes can therefore be used in studies of supercritical fluid activity associated with pegmatite systems.

Implications for Li enrichment in granitic pegmatites. Despite the importance of granitic pegmatite Li deposits in securing future lithium supply, the mechanism behind Li enrichment, exceeding thousands of times the average continental crust in the formation of such deposits, remains enigmatic. The substantial involvement of magmatic fluids in the petrogenesis of LCT-type pegmatites revealed by our study implies that they should be a key medium in extracting, concentrating, and transporting Li to provide a material source for mineralisation in granitic pegmatites (Fig. 4). The large and super-large pegmatite Li deposits are mainly developed within collisional orogenic belts related to supercontinent assembly

(London, 2018), where thickened crust implies more extensive intra-crustal magmatism. Available geophysical studies indicate the presence of vertically extended (up to 15 km) magma reservoirs beneath a number of LCT-type pegmatites like the Jiajika (Huang *et al.*, 2020). This is consistent with the recent concept of crystal mush-dominated transcrustal magmatic system (Cashman *et al.*, 2017), wherein pegmatites are regarded as the last drops of magmatic distillates at the top (Troch *et al.*, 2022). In such a scenario, hydrothermal fluids stored in shallow magma reservoirs represent the time-integrated products of magmatic differentiation and fluid exsolution operating over a significant vertical extent (Blundy *et al.*, 2021). Because ascending magmatic fluids can be decoupled from residual melts, even the fluid-mobile rare metals and fluxing components enriched in the deep crystal mush can be efficiently extracted and transported to aggregate in shallow magma reservoirs through fluid channels among the crystals (Parmigiani *et al.*, 2016). The elevated contents of fluxing components in shallow magma reservoirs not only effectively reduce the polymerisation of the melts and promote their miscibility with magmatic fluids, but also provide sufficient ligands, thereby enhancing solubility of incompatible rare metals in the resulting supercritical fluids (Thomas and Davidson, 2016). Theoretically, the supercritical fluids can efficiently extract Li and other rare metal elements with enrichment factors of up to 10^4 (Thomas *et al.*, 2019). Meanwhile, the accumulated magmatic fluids in shallow magma reservoirs are capable of driving overpressure and causing episodic hydraulic fracturing for supercritical fluids intrusion (Holtzman *et al.*, 2012). Finally, as phase separation occurs to supercritical fluids, Li can be further transported by H_2O-rich melt/fluid phase to mineralisation away from the parental granite, as shown by the Jiajika and other LCT-type granitic pegmatite fields worldwide (London, 2018; Troch *et al.*, 2022). In short, fluid exsolution within the parental magmatic system provides an additional material source for pegmatite petrogenesis and mineralisation. Liquid immiscibility within pegmatite veins determines the spatial distribution of Li mineralised pegmatites. Therefore, after observing highly differentiated granites with high Li content in the collisional orogenic belts, potential Li-rich pegmatites in wall rocks could occur vertically higher and/or horizontally distant (up to several kilometres), which can be an effective exploration strategy for pegmatite Li deposits.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2426.

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