GEOCHEMISTRY

Origin of potassic postcollisional volcanic rocks in young, shallow, blueschist-rich lithosphere

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Potassium-rich volcanism occurring throughout the Alpine-Himalayan belt from Spain to Tibet is characterized by unusually high Th/La ratios, for which several hypotheses have brought no convincing solution. Here, we combine geochemical datasets from potassic postcollisional volcanic rocks and lawsonite blueschists to explain the high Th/La. Source regions of the volcanic melts consist of imbricated packages of blueschist facies mélanges and depleted peridotites, constituting a new mantle lithosphere formed only 20 to 50 million years earlier during the accretionary convergence of small continental blocks and oceans. This takes place entirely at shallow depths (<80 km) without any deep subduction of continental materials. High Th/La in potassic rocks may indicate shallow sources in accretionary settings even where later obscured by continental collision as in Tibet. This mechanism is consistent with a temporal trend in Th/La in potassic postcollisional magmas: The high Th/La signature first becomes prominent in the Phanerozoic, when blueschists became widespread.

INTRODUCTION

Potassic volcanism commonly occurs in the late stages of orogenes— the process of collision of plates—between a long-lasting period of subduction and the end of postcollisional collapse. The transformation of an arc system into a collisional regime is usually accompanied by volcanism that geochemically resembles the older arc lavas and yet has important additional ingredients derived from the continental lithosphere. The syn-collisional episode evolves into a postcollisional phase during which volcanism may be extremely variable due to a combination of slab detachment, delamination, slab rollback, and extension, and each of these processes may involve different sources and melting regimes (1, 2). These processes are reflected in the geochemistry of the erupted orogenic magmas, which represent the end result of complex, multistage processes. Furthermore, in some regions such as Tibet, where massive continental collision occurred later, the tectonic regime in which potassic volcanism occurs may be obfuscated by later tectonic processes. Here, the potassic volcanism may have originated in an environment similar to the modern eastern Mediterranean, which predated the continental collision (3).

The tectonic regime that triggered widespread orogenic volcanism in the Alpine-Himalayan chain is controversial (4, 5). There is now abundant evidence from trace elements and isotopes for the involvement of continental material in the source (5, 6), but the style and mechanism of this crustal involvement remain unclear. Two competing scenarios invoke (i) direct melting of continental crust during deep intercontinental subduction (7) and (ii) delamination of heavily metasomatized mantle lithosphere into the convecting mantle where melting of its most fusible parts occurs (8, 9). However, direct evidence to distinguish between these two models has been lacking.

The Th/La ratio has been used to assess whether certain chemical features shared by arc magmas and continental crust are induced by subduction processes or are derived from recycled subducted sediment (10). This study suggested that in normal circumstances, the Th/La ratio of mantle-derived magmas should be relatively constant and no greater than 0.5. However, postcollisional lamproites of the “Tethyan-realm,” which later developed into the Alpine-Himalayan orogenic belt (AHOB), were reported to have high Th/La ratios of up to 2.2 (8). Given their exceptional enrichment in potassium and other strongly incompatible elements, lamproites have been considered the optimal probe to characterize strongly metasomatized mantle to constrain the source of the Alpine-Himalayan orogenic magmatism (3, 11). The unusual geochemical signature of extremely high Th/La coupled with relatively low Sm/La is not restricted to lamproites in the AHOB, as described by Tommasini et al. (8), but also occurs in many other K-rich volcanic rocks in the same belt (Fig. 1). Two main mechanisms have been put forward to explain this anomalously high Th/La: (i) preferential uptake of La relative to Th in mafic minerals during direct melting of continental crust (10, 12) and (ii) melting of mélanges that include lawsonite/zoisite-bearing blueschists accreted to the colliding continental plates (8, 9, 13, 14).

Here, we propose a comprehensive model that is a variant of the second hypothesis. This model explains the source regions of the AHOB potassic volcanic rocks as consisting of blueschist facies mélanges (including oceanic crust, oceanic, and continental sediments) imbricated together with extremely depleted forearc peridotites. Together, these make up a mantle lithosphere that was newly formed during the convergence of small continental blocks and oceans. The imbrication process took place entirely at shallow depths (<80 km) and did not require any deep subduction of continental materials. Lawsonite blueschists provide notable new petrological and geochemical evidence to explain the Th/La fractionation widely seen in AHOB K-rich lavas.

RESULTS

Geological background: Tavşanlı zone, Turkey

During the Cretaceous, the Alpine-Himalayan chain was dominated by the convergence of continents that eliminated the northern
In situ 40 Ar/39 Ar dating study on phengite showed much wider (fig. S1B) and lawsonite garnet-lawsonite blueschist age of 83.3 Ma (18), whereas more recent studies of Lu-Hf dating yielded a lawsonite eclogite age of 91.1 Ma, (17) and the other three samples are more enriched in K, and especially in Th, U, and REE (LREE), and show variably radiogenic $^87$Sr/$^{86}$Sr and mostly unradiogenic $^{143}$Nd/$^{144}$Nd. The most unradiogenic $^{143}$Nd/$^{144}$Nd indicates the presence of an enriched, probably continental crust-like, component. These samples have more radiogenic $^{206}$Pb/$^{204}$Pb, $^{207}$Pb/$^{204}$Pb, and $^{208}$Pb/$^{204}$Pb than the MORB-like samples, although they mostly plot within the mantle array.

**Lawsonite compositions**

Calcium aluminosilicate phases such as epidote group minerals and lawsonite may contain significant amounts of trace elements, with lawsonite known to be a potentially important repository for Th, Sr, U, and REE (17, 22–25). The concentrations of Th, large-ion lithophile element (LILE), and LREE in lawsonite were found to be highly variable within individual Tavşanlı samples. This strong heterogeneity in trace element concentrations is not due to zoning or contamination from inclusions and probably results from differing origins of the lawsonite (23, 24). Sample 10tav07 is distinctive in showing a positive correlation between extremely high Th/La (up to 1.1) and relatively high Sm/La ratios. This is consistent with several previous studies, in which high Th/La (up to 4.88) has been found in lawsonites from nearby Sivrihisar (Fig. 2) (23). Mass balance calculations indicate that ~40% of Th is held in lawsonite (25 modal %) in sample 10tav07 (17), underscoring the significance of lawsonite in governing the Th budget of blueschists. This enrichment is coupled with REE fractionation in lawsonite and low $^{87}$Sr/$^{86}$Sr and $^{143}$Nd/$^{144}$Nd in the whole rock, which suggests the involvement of an enriched component similar to continental crust. In contrast, the MORB-like samples exhibit low Th/La but markedly high Sm/La ratios (Fig. 2).

Minor amounts of inclusions of minerals such as monazite and epidote group minerals, especially allanite, have the potential to falsify the trace element concentrations of lawsonite markedly (12). This has been discounted for the data used here by thoroughly investigating lawsonites by three-dimensional confocal micro-Raman spectroscopic mapping: only glauconephane, quartz, and titanite were found, which cannot greatly affect trace element concentrations in lawsonite (17).

**Fingerprinting the geochemistry of Alpine-Himalayan orogenic magmatism**

Previous studies of the isotopic and chemical heterogeneities of mantle-derived K-rich lavas from the AHOB, mostly of lamproitic
subdivide them into two major groups using K2O/Na2O as a geochemical discriminator: (i) low-K lavas or “normal” arc lavas (K2O/Na2O < 1; e.g., South Aegean active volcanic arc), with potassium contents that are low in the most primitive members of the suite but increase with fractionation; (ii) high-K lavas (K2O/Na2O > 1), which have high K2O content also in the most Mg-rich samples (Fig. 3, A and B). Despite some overlap between the two populations on some plots, high-K lavas are systematically more enriched in Th, with many showing elevated Th/La and Th/Yb (Fig. 3, C and E). Moreover, they have more radiogenic strontium and less radiogenic neodymium isotopes (Fig. 3D), interpreted to suggest that their source contains a terrigenous crustal component. The isotopic and chemical differences between low- and high-K lavas indicate the existence of distinct mantle sources.

DISCUSSION

The crust-like signature and Th enrichment identified previously in lamproites may be extended to the entire K-rich group of AHOB lavas. Because Th enrichment in the high-K lavas (high Th/La and Th/Yb) rises as K2O and 87Sr/86Sr increase and as 143Nd/144Nd decreases, we may conclude that recycled continental crust plays a significant role in the source. However, neither continental and oceanic crust nor their derivatives show enrichment in Th and elevated Th/La and Th/Yb ratios (8, 10), implying that this signature does not simply arise by crustal recycling. In other words, the high Th/La signature must be newly created during the orogenic cycle without losing the isotopic affinities.

Lawsonite blueschists in the source of potassic magmas

Th/La ratios of both the blueschist whole rocks and the lawsonites yield heterogeneous values, whereby the most elevated Th/La ratios are in the terrigenous samples with the continent-derived geochemical component characterized by unradiogenic Nd isotopes (17). The most enriched blueschist sample (10tav07) plots at values typical for continental crust, implying that blueschist from mélanges may play a substantial role in the origin of the AHOB lavas, providing the geochemical ingredients responsible for the continental crust-like signature and elevated Th/La ratios. Lawsonites from this sample show the extremely high Th/La values as well as a positive correlation between Th/La and Sm/La ratios (Fig. 2), a similar correlation to that seen in the orogenic lavas (Fig. 1). This contrasts with melts of the upper crust, which fail to appreciably fractionate Th from La at any degree of melting (14). This supports the hypothesis that lawsonite blueschist in mélanges can host the high Th/La signature in the postcollisional potassic volcanic rocks of the Alpine-Himalayan belt. The high Th/La ratios of lawsonites can be conveyed to the AHOB lavas by a series of progressive geological processes, not only by prograde metamorphic recrystallization into new mineral phases, which would redistribute trace elements differently (22).

Allanite, monazite, and epidote group minerals are known to host considerable amounts of Th and LREE (28, 29), so these minerals are often assumed to be the most likely candidates to account for the elevated Th/La ratios. Although a few experimental studies have reported that allanite is able to fractionate Th from La (12, 30), existing data on natural minerals show that they are not likely to be responsible for the high Th/La ratios, because none of these minerals fractionate Th from La as effectively as lawsonite (all have Th/La < 0.5; fig. S2). Moreover, even if allanite does have the potential to account for the high Th/La, it is also a common lawsonite blueschist facies mineral (22, 23, 31) and so does not contradict our proposal of the significance of blueschist facies mélange in conveying the Th/La characteristics to AHOB lavas.
A key question that needs to be addressed pertains to the petrological mechanism of blueschist involvement in the origin of the melts, particularly why they are potassium rich and have unusually high Th/La ratios. It is clear from our data that only the terrigenous blueschists can provide the coupled elevated Th/La and crust-like trace element signature to the source of the AHOB lavas (17). Because phlogopite is often assumed to be an essential contributor to strongly potassic magmatism (32), it is easy to generalize the origin of K-rich magmatism as being derived from deep mantle levels. However, reaction experiments between quartz phyllite and depleted peridotite produce potassic, silicic melts without residual phlogopite (33), indicating that the assumption of phlogopite in the source may not apply in all cases. Although our samples are not very phengite-rich, we envisage that some portions of the blueschist facies mélangé are extremely enriched in potassium and trace elements typical for the continental crust [e.g., (34)]. Experimental studies of the melting of mélangé show that melt compositions depend critically on the exact rock type among the heterogeneous mélangé that melts. Some indicate that granodioritic to tonalitic melts would be produced (35), whereas melting of chlorite-omphacite-dominated rocks produces melts with the major and trace element characteristics of postcollisional alkaline lavas (36). However, because of the lack of terrigenous lawsonite blueschist in the starting material, their study did not produce melts with elevated Th/La ratios.

Multistage evolution of elevated Th/La in Alpine-Himalayan volcanic rocks

The history of the collision, lithosphere formation, and remelting in the AHOB might be exclusively a shallow level process in which mélange recycling takes place within the relatively cold fore-arc region at low pressure (path a in Fig. 4A) (14). This contrasts with conventional deep Andean-style subduction (path c in Fig. 4A) in which a succession of additional reactions at higher pressures would dilute the Th/La signature, as shown in the quantitatively modeled figure (Fig. 4D).

Lawsonite blueschists may have an array of trace element ratios depending on their origins: terrigenous blueschists will contain the high Th/La feature while others (e.g., MORB-like blueschists) do not (17). As tectonic imbrication occurs, a variety of rock types can be stored for considerable time in the blueschist facies in the newly formed lithosphere, unlike Andean-style subduction in which metamorphism would progress relatively quickly to the eclogite facies (path c in Fig. 4A, A and B). Blueschists are initially stable in the
newly formed lithosphere but will slowly heat up during postcollisional orogenic collapse. Because the thickened crust slowly relaxes and thins in postcollisional conditions (37), it takes 10 to 30 Ma for the lower block to achieve thermal equilibrium (38), which agrees well with the timing of postcollisional volcanism, which first occurs after this period. However, the P-T-t path consists mostly of heating with a slight reduction in pressure due to extension and to erosion of the continent above it during postcollisional relaxation. The effect is to slowly move from the blueschist to epidote amphibolite then to amphibolite facies but does not pass through the blueschist facies. (B and C) Full pressure-temperature (P-T) pseudosection diagrams for the Tavşanlı blueschist 10tav07 at 600° to 1000°C, 0.8 to 1.6 GPa and 2.2 to 3.0 GPa following paths a and c in (A), respectively. Also shown are calculated modal proportions for melting along apparent pressure of 1.2 and 2.6 GPa. Plots of modal proportions versus temperature (also known as modebox diagrams) illustrate the changing abundance of phases, modeled along linear pressure of 1.2 and 2.6 GPa (orange arrow lines). (D) Comparison between modeled Th/La ratios for melts produced at 1.2 GPa (shallow imbrication scenario) and melts produced at 2.6 GPa (conventional deep subduction scenario) at given degree of melting. Starting Th/La in blueschist 10tav07 is 0.22 (17), many blueschists have higher ratios. Note the notable difference of Th/La ratios between the two types of modeled melts. Grt, garnet; Omp, omphacite; Gl, glauconaphane; Ph, phenelite; Coe, coesite; Qz, quartz; Rt, rutile; Kfs, K-feldspar; Pl, plagioclase; Liq, melt; Cpx, clinopyroxene; Opx, orthopyroxene; Hbl, hornblende; Ttn, titanite; Bt, biotite; Ilm, ilmenite; Ms, muscovite; Ab, albite.

Fig. 4. Deep subduction versus shallow imbrication. (A) Three contrasting prograde P-T-t paths for (a) shallow subduction and tectonic imbrication discussed for the Alpine-Himalayan belt (blue line), (b) normal regional (orogenic) metamorphic scenario [after Çetinkaplan et al. (57) and Plunder et al. (16)] (green line), and (c) conventional deep Andean-style subduction (beige shaded field after Syracuse et al. (58)]. During subduction, path a reaches blueschist facies, and tectonic imbrication may store abundant blueschists in the lithosphere. This material is now removed from the subduction environment and is isolated from further movements. During postcollisional processes, these rocks slowly heat up path a and move slowly from blueschist, through epidote amphibolite to upper amphibolite facies. In contrast, path b follows a path from greenschist to epidote amphibolite then to amphibolite but does not pass through the blueschist facies. (B and C) Full pressure-temperature (P-T) pseudosection diagrams for the Tavşanlı blueschist 10tav07 at 600° to 1000°C, 0.8 to 1.6 GPa and 2.2 to 3.0 GPa following paths a and c in (A), respectively. Also shown are calculated modal proportions for melting along apparent pressure of 1.2 and 2.6 GPa. Plots of modal proportions versus temperature (also known as modebox diagrams) illustrate the changing abundance of phases, modeled along linear pressure of 1.2 and 2.6 GPa (orange arrow lines). (D) Comparison between modeled Th/La ratios for melts produced at 1.2 GPa (shallow imbrication scenario) and melts produced at 2.6 GPa (conventional deep subduction scenario) at given degree of melting. Starting Th/La in blueschist 10tav07 is 0.22 (17), many blueschists have higher ratios. Note the notable difference of Th/La ratios between the two types of modeled melts. Grt, garnet; Omp, omphacite; Gl, glauconaphane; Ph, phenelite; Coe, coesite; Qz, quartz; Rt, rutile; Kfs, K-feldspar; Pl, plagioclase; Liq, melt; Cpx, clinopyroxene; Opx, orthopyroxene; Hbl, hornblende; Ttn, titanite; Bt, biotite; Ilm, ilmenite; Ms, muscovite; Ab, albite.
they contribute a relatively high SiO$_{2}$ end-member to the cocktail of source components. This is consistent with the positive correlation of Th/La with SiO$_{2}$ contents in the postcollisional volcanics.

The uniquely high Th/La signature in AHOB volcanic rocks may, therefore, result from three consecutive processes that progressively increase the Th/La ratio: (i) initial incorporation of high Th/La in lawsonite; (ii) loss of a hydroxyl fluid (breakdown of lawsonite); and (iii) melting of originally continental crustal rocks in the amphibolite facies. It should be noted that amphibolites need not be restricted to metabasites; a metapelite may have distinct mineralogy, containing quartz, orthoclase, muscovite, biotite, garnet, etc. (44, 45). Melts of this amphibolite would be potassic and could give rise to the potassium enrichment seen widely in AHOB volcanic rocks.

The P-T-t path usually considered for the stability of many accessory minerals (such as allanite, monazite, and epidote) is different to that in this new geodynamic scenario. A normal regional (orogenic) metamorphic field gradient moves through greenschist to amphibolite and not via the blueschist facies (path b in Fig. 4A), so that the prograde mineral assemblages differ. For an Andean subduction (path c in Fig. 4A), blueschists cannot escape the fate of being transformed to eclogite as the slab proceeds to deeper levels. Therefore, experiments that simply displace a continental crustal rock to high PT conditions and analyze trace elements in accessory minerals do not account for the correct history of mineral assemblage changes relevant to shallow accretionary processes.

**Temporal trends of Th/La in orogenic magmatism**

The oldest glaucohane schists preserved on Earth are Neoproterozoic and lawsonite-bearing blueschists appear much later (~Ordovician), so that blueschist-facies metamorphism is practically restricted to the Phanerozoic (46). If lawsonite blueschists are essential for the production of K-rich orogenic magmas with high Th/La ratios, then a logical consequence of our model is that K-rich volcanic rocks in older orogenic belts should show progressively lower and more confined Th/La ratios as we go back in time. To test this prediction, we plot published Th/La data in K-rich orogenic magmas from geochronologically diverse orogens in Fig. 5. As we go back in time from <80 Ma through 200 to 400 Ma and from 410 to 800 Ma through 980 to 1800 Ma, a secular trend from high to low Th/La ratio is indeed clearly visible, with no elevated Th/La present for Grenvillian and Svecofennian times (980 to 1800 Ma; Fig. 5). This further justifies the shallow subduction model and the significance of lawsonite blueschists in the production of K-rich orogenic magmas with high Th/La ratios.

**Implications and outlook**

Convergence along the Alpine-Himalayan belt varied in style, involving accretion of small continental slivers and numerous oceanic island arcs in the west, culminating in the world's most comprehensive continental collision in the east. Most of the convergence between Gondwana and Eurasia since the Cretaceous has been accommodated by nappe stacking and lithospheric slab underthrusting. In many regions, oceanic subduction waned during the Cretaceous, shortly after which postcollisional tectonics dominated during a period of large-scale extension (47). Magmatism is closely related to tectonics: It postdates the final accretionary events that formed the Alpine-Himalayan chain, with the most voluminous and widely distributed episode(s) beginning in the late Cretaceous. Magma genesis is controlled by a combination of rollback of the underthrusting lithospheric slab that initiated postcollisional extension and collapse of the orogenic belts, coupled with the initiation and progression of slab tear (5).

Complex geodynamic settings are the key to activating and melting a variety of mantle and crustal sources: Slab rollback triggers the suction of hot convecting mantle toward shallow levels in the mantle wedge, and hot, fresh asthenospheric mantle may also penetrate through slab tears, causing melting of previously enriched domains in the lithospheric mantle. In this process, rollback plays a specific role in the fate of lawsonite, which exerts control of critical trace element ratios in relatively shallow subduction zone conditions along cold geotherms (450° to 650°C and 2 to 3 GPa) (23). As the subducting slab descends into the mantle, continental sediments experience prograde metamorphism during which trace elements are redistributed from precursor minerals (most likely epidote group minerals or Mn-garnet) to lawsonite, glaucophane, jadeite, and white mica. Lawsonite takes up much of the Th, Sr, and REE, whereas Nb and Ta are preferentially incorporated in rutile and titanite, Zr and Hf in zircon, LILE in phengite, and heavy REE in garnet. Lawsonites that grow in locally different components of a lithologically heterogeneous subduction mélangé inherit the geochemical characteristics of their oceanic or continental protoliths. It is now uncertain whether the high Th/La signature is derived from mixed continental and oceanic sedimentary protoliths or from metasomatic reactions caused by fluids transporting more La than Th from continental blocks when they infiltrate oceanic mélangé.
The Alpine-Himalayan orogenic volcanics thus owe their unusual trace element geochemistry, especially the extremely high Th/La ratios, to the involvement of lawsonite blueschists in the source region. Their incorporation in newly formed lower lithosphere and the 30- to 50-Ma time span before magmatism are consistent with petrological-thermomechanical modeling (48). The extremely high Th/La signature in lawsonite can be transferred to the postcollisional lavas by multistage accumulation in an entirely shallow level process (Fig. 4), consistent with P-T estimates for the Tavşanlı zone of ~300° to 500°C, 12 to 24 kbar, which correspond to lithospheric depths of 40 to 80 km. This highlights an unrecognized bias in past studies, which have unconsciously supposed deep, Andean-style subduction where the slab proceeds to lower mantle depths.

These scenarios are specifically relevant to accretionary orogens in which the limited size of small subducting oceanic slabs and continental blocks prevents deep subduction processes from dominating. For places where the accretionary stages could be largely obscured by later major collision, e.g., in Tibet (3, 49), the proposed scenario is especially applicable and could provide additional geodynamic perspectives complementary to existing knowledge. Although this tectonic scenario has been recognized in Turkey and Tibet because the continental sediment signature is observed in the Pb isotope compositions of post-collisional lavas (5, 8), we predict that there will often be abundant ancient sediments integrated into the lithosphere. All these components are reactivated and melted during mechanical relaxation millions of years after the collision. These processes are significant for arcs and orogenic magmatism on the modern Earth but will be particularly pertinent to the late Archean, during which crustal formation by the amalgamation of arcs may have been widespread (50).

MATERIALS AND METHODS
Thermodynamic modeling
To constrain melt crystallization at high-pressure and low-pressure conditions, P-T pseudosection calculations have been applied for a representative terrigenous blueschist composition (sample 10tav07) (17). Phase equilibria were calculated using PerpleX version 6.8.1 software package (51) with the upgraded thermodynamic database DS6.22 from Holland and Powell (52). Pseudosections for composition of blueschist 10tav07 were calculated in the MnO–Na2O–CaO–K2O–FeO–MgO–Al2O3–SiO2–H2O–TiO2–O (MnNCKFMASHTO) system. The activity-solution models used were taken from literature and tabulated in data file S4. The composition of CaO of blueschist 10tav07 has been adjusted using the Rock Maker software (53) to account for the presence of 6 mol% of apatite that has not been included in the pseudosection calculations. The O content has been determined using calculated T–X(O) pseudosection at 1.2 GPa to constrain the oxidation state able to predict stability of the rock mineral assemblage. The H2O contents at high- and low-pressure conditions have been determined using two calculated T–X(H2O) pseudosections to constrain the minimum content in H2O necessary for the rock to cross the solidus after potential subsolidus water loss at 2.6 and 1.2 GPa, respectively. The results for high-pressure melting are shown in a P–T pseudosection calculated with H2O = 1.53 mol% and O = 0.17 mol% between 2.2 and 2.6 GPa and 600° and 1000°C. The results for low-pressure melting are shown in a P–T pseudosection calculated with H2O = 3.94 mol% and O = 0.17 mol% between 8 and 16 GPa and 600° and 1000°C. Assemblage fields are labeled with stable phases while pure phases include quartz, rutile, sphene (titane), and aqueous fluid (H2O). The thick labeled black line represents the liquid-in reaction. The depth of shading reflects increasing variance of phase assemblage fields. The bulk-rock composition used for pseudosection calculations at high- and low-pressure conditions is given in data file S4. The software and data files used to calculate the pseudosections can be downloaded from http://www.perplex.ethz.ch.

Trace-element modeling
For Th/La ratio modeling (Fig. 4D), we used blueschist sample 10tav07 as a starting composition (Th/La = 0.22) (17) and the calculated abundance of phases at 600° to 1000°C, 1.2 and 2.6 GPa, respectively, at various melt fractions (Fig. 4, B and C, and data file S4). Mineral/melt partition coefficients (D) were taken from the literature and tabulated in data file S4.

SUPPLEMENTARY MATERIALS
Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/7/29/eabc0291/DC1

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2021; 7 : eabc0291 14 July 2021
Origin of potassic postcollisional volcanic rocks in young, shallow, blueschist-rich lithosphere
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Sci Adv 7 (29), eabc0291
DOI: 10.1126/sciadv.abc0291

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