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Fingerprinting sources of orogenic plutonic rocks from Variscan belt with lithium isotopes and possible link to subduction-related origin of some A-type granites

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ABSTRACT

Lithium (Li) elemental and isotope data are presented for a suite of plutonic rocks (I-, S-, A- and S_S-type, coexisting mafic bodies) from the Western Carpathians, Slovakia, that were generated throughout the complete Variscan orogenic cycle. I-type granites show a limited range of δ^7Li, contrasting with the large variations found in S-type granites and orthogneisses of distinct ages. This contradicts previous reports of predominantly light-Li S-type granites, sourcing metasedimentary lithologies, and isotopically heavier I-type granites, derived from meta-igneous sources. An almost exclusively heavy Li isotope signature (δ^7Li >4.7‰) found in four out of five A-type granites rules out several commonly accepted petrogenetic scenarios such as remelting of granulitic residue after formation of I-type granitic melts, anatexis of calc-alkaline meta-igneous crust or extensive closed-system fractional crystallization of mantle-derived magmas. Instead, it most probably reflects a derivation of A-type granites from a mantle wedge modified by slab-derived fluids during an Early Variscan or Pan-African subduction episode. In contrast, the distinct and lower δ^7Li values (<1.2‰) in the contemporaneous S_S-type granites can be related to their likely pelitic parentage.
Mafic rocks (gabbros and diorites), associated with several occurrences of granites, are uniformly Li-rich and isotopically light (<−0.5‰), precluding a direct derivation from the mantle. These signatures testify to their cumulate origin whereby kinetic effects may be a viable explanation for the light Li isotope compositions, associated with diffusive redistribution of Li between mantle-derived mafic melts and acid magmas. This corroborates recent studies on faster diffusion of ^{6}Li compared with ^{7}Li in natural systems.
Taken together, the presumed dichotomy between the sources and processes leading to generation of S- and I-type granitic magmas does not seem to be reflected by Li isotope signatures in a simple and globally valid manner. Interpretation of Li isotope compositions thus needs to be paralleled by other available information on petrology, whole-rock geochemistry and the magmatic context.

1. Introduction

Lithium (Li) isotopes have proven useful for tracking geological processes in both low- and high-temperature environments, for example mantle processes (Aulbach and Rudnick, 2009; Halama et al., 2008; Ionov and Seitz, 2008; Jeffcoate et al., 2007; Magna et al., 2006a, 2008; Rudnick and Ionov, 2007; Seitz et al., 2004), subduction zone processes (Agostini et al., 2008; Chan et al., 2002b; Magna et al., 2006b; Marschall et al., 2007; Moriguti and Nakamura, 1998; Tomascak et al., 2000; Zack et al., 2003), seafloor alteration (Chan et al., 1992, 2002a) and continental weathering/erosion (Huh et al., 2001; Kısakűrek et al., 2004; Rudnick et al., 2004; Ushikubo et al., 2008). However, surprisingly little has been done so far to characterize Li isotope systematics in the continental crust (Bryant et al., 2004; Teng et al., 2004, 2008, 2009). Granitic rocks and their derivates represent a significant part of the upper continental crust (Rudnick and Gao, 2003; Taylor and McLennan, 1995). Lithium, being a moderately incompatible element, is substantially enriched in these chemically evolved rocks, which typically reflect several cycles of partial melting and fractional crystallization within the Earth's crust. It has been estimated that as much as ~5% of Li may reside in the continental crust (Teng et al., 2004) which represents ~0.5% of Bulk Silicate Earth (Rudnick and Gao, 2003; Taylor and McLennan, 1995). Thus, the continental crust, the composition and evolution of which

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still needs to be better constrained, represents an important repository for the global Li budget.

Given the complex interplay of processes potentially involved in granite genesis, the general lack of information on intra-crustal fractionation of Li isotopes as well as the scarcity of Li isotope data on granitoid rocks and their prospective sources severely limits our understanding of Li isotope systematics. Previous attempts to link Li isotope signatures of granites to their presumed protoliths have shown that there is considerable overlap for granitic rocks derived from meta-igneous sources and those of metasedimentary origin (I-type and S-type granitoids; Chappell and White, 1974, 1992, 2001). Bryant et al. (2004) and Teng et al. (2004) have suggested that the predominantly light isotope signature of S-type granites likely reflects Li fractionation during weathering, consistent with their sedimentary parentage. Indeed, the dominance of low positive to negative δ^7Li in the continental crust has been explained by magmatic reworking (recycling) of weathered terranes (Teng et al., 2004) that had lost their originally heavy Li component during extensive weathering (Kısakűrek et al., 2005; Pistiner and Henderson, 2003; Rudnick et al., 2004).

On the other hand, higher δ^7Li values recorded in a majority of I-type granites seem to point to metabasic igneous, mantle-derived sources (Bryant et al., 2004) or may reflect recycling of isotopically heavier Li from the oceans through subduction of altered ocean-floor basalts (Elliott et al., 2006). The tenuous boundaries between δ^7Li of diverse granitic types as well as the large number of possible processes that may further disturb the Li composition of the initial reservoirs (i.e., diffusive isotope fractionation, contact metamorphism, fluid percolation) result in rather nebulous Li isotope systematics. Huge variations in these upper crustal materials (Marks et al., 2007; Teng et al., 2006b, 2008, 2009) clearly speak for magmatic or post-magmatic perturbations imposed on compositionally diverse magmas derived from heterogeneous crustal sources.

An acceptable interpretation of the processes governing Li isotope compositions of granitic rocks has been hampered by the fact that in most studies the Li isotope data were discussed in isolation, without providing adequate information on petrology and ancillary geochemical data. A comprehensive approach is needed, integrating petrological, geochemical and isotope data from a well-studied terrain with a broad spectrum of granitic types related to distinct sources and/or geotectonic position.

A typical feature of the Alpine mobile belt in Europe is the presence of reworked huge and often well-preserved slices of Variscan crystalline basement within the deformed Mesozoic and Cenozoic sedimentary successions. Specifically, the Western Carpathians in Slovakia, which host a range of variable and well-studied granites generated throughout a complete Variscan orogenic cycle (Broska and Uher, 2001; Kohút, 2007; Petrík and Kohút, 1997), are exceptionally suitable for a case study testing the Li isotope system utility.

We have analyzed a series of West-Carpathian plutonic rocks in order to i) shed new light onto petrogenesis of individual granitoid suites and of associated, mafic rock types, ii) provide new data for so far insufficiently characterized A-type granites, and, in more general terms, iii) investigate the usefulness of Li isotopes as a tool for fingerprinting fundamental source characteristics of individual granitic associations.

2. Geological setting

2.1. Structure of the Western Carpathians

Most exposures of pre-Alpine (pre-Mesozoic) crystalline basement in the central Western Carpathians consist, from north to south, of three main super-units: Tatric, Veporic and Gemeric (Fig. 1) (Plašienka et al., 1997).

In the *Tatric Unit*, granites and pre-Mesozoic metamorphic rocks build an only little modified fundament (Krist et al., 1992) overlain by Mesozoic sedimentary rocks and/or nappes derived therefrom. The *Veporic Unit* is dominated by a large granodiorite–tonalite composite batholith with low- to high-grade metamorphic rocks, overlain by Upper Palaeozoic and Mesozoic sedimentary rocks. Due to complex Variscan and Alpine tectonic processes, the Veporic Unit has a very complicated imbricated structure with south-eastwards increasing penetrative brittle–ductile deformation. The *Gemeric Unit* is dominated by a large granitoid body that intruded the Ordovician to Late Carboniferous rocks, mostly low-grade flyschoid metasedimentary and metavolcanic rocks with remnants of an ophiolite complex (Kohút and Stein, 2005, and references therein).

2.2. Variscan granites and their presumed geotectonic setting

In the Western Carpathians, pre-Alpine igneous activity occurred episodically over a long time interval (Kohút, 2007; Petrík and Kohút, 1997). In brief, Ordovician to Devonian rifting was followed by Andean-type subduction and amalgamation of an oceanic lithosphere at the Devonian–Carboniferous boundary. The Palaeozoic orogenic cycle culminated in a Himalayan-type collision with crustal thickening between 350 and 330 Ma, succeeded by lithospheric mantle delamination or slab (?) break-off between 310 and 300 Ma and finally terminated in a phase of crustal relaxation/extension (Petrík and Kohút, 1997; Petrík et al., 2001). Such a development is in accord with generally accepted models for the granitic plutonism (e.g., Barbarin, 1990; Bonin, 1990; Kemp and Hawkesworth, 2003; Pearce, 1996; Pitcher, 1983). It seems that recycled continental crust with contributions from a subduction-modified mantle may have played a crucial role in the genesis of the West-Carpathian granites.

The temporally evolving processes have produced the following granitic suites: i) Late Cambrian–Ordovician felsic plutonites/volcanites, transformed into orthogneisses by Late Devonian shearing (hereafter referred to as *orthogneisses* or the *OG-suite*) and mainly identified within the polymetamorphosed basement rocks of the Veporic and Tatric units; ii) Early Carboniferous peraluminous S-type granites (hereafter referred to as *S-suite*) common in the Tatric Unit and the Veporic composite batholith; iii) Late Carboniferous calc-alkaline I-type granites (hereafter referred to as *I-suite*); iv) Permian post-orogenic mildly alkaline A-type granites (hereafter referred to as *A-suite*) within the Veporic Unit and at its contact with the Gemeric Unit and (v) strongly peraluminous, Sn–W mineralized Gemeric granites (hereafter referred to as S_S*-suite*).

The detailed overview of the individual suites, including selected petrological and geochemical parameters, is given in Appendix 1 and summarized in Table 1. Several diagrams portraying the whole-rock geochemical variability of the studied samples are given in Fig. 2.

2.3. Mafic rocks

In the Western Carpathians, mafic rocks of variable ages (hereafter referred to as *mafic suite*) are scarce. The oldest gabbros occur as lenses within the ~500 Myr old leptyno-amphibolitic complex of the Tatric and Veporic units, where they are thought to having been contemporaneous with the emplacement of the protolith to the orthogneisses (Putiš et al., 2009).

Recently, significantly younger gabbroic rocks were identified as a part of an ophiolitic sequence in Tatric Unit dated at ~371 Ma (Putiš et al., 2009) and ~373 Ma (Kohút et al., 2009). Younger gabbros–diorites occasionally form small independent bodies (usually <70 m, rarely 100–200 m across) and dioritic rocks occur as numerous mafic