

A brief review of isotopically light Li – a feature of the enriched mantle?

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Lithium isotope geochemistry is increasingly being used to trace deep-earth processes, reflecting the observed large variation of Li isotope ratios in mantle-derived rocks, including peridotite xenoliths associated with ancient continents. We briefly review the Li isotopic compositions of major geochemical reservoirs, the assumed mechanisms of Li isotopic fractionation, and, in particular, the origins of isotopically light Li in mantle-derived rocks based on the latest developments in Li isotope geochemistry. Comparison of Li isotope data with existing Sr-Nd isotope ratios reflects the subduction-recycling of ancient oceanic crust and the reappearance of Li in volcanic rocks. This circulation may play an important role in generating the isotopically light-Li component in the mantle – perhaps the enriched mantle end member defined by the Sr-Nd isotopic compositions of oceanic basalts.

Keywords: lithium; isotope geochemistry; fractionation mechanisms; peridotite xenoliths; review of Li isotopic evolution

Introduction

Li isotope geochemistry has developed rapidly in recent years (Ushikubo *et al.* 2008; Vigier *et al.* 2008; Chan *et al.* 2009; Richter *et al.* 2009) because Li has many favourable characteristics as a geochemical tracer (Tomascak 2004; Tang *et al.* 2007b). Inasmuch as Li is a mildly incompatible element with $D_{solid/melt}$ values between 0.1 and 0.5 for most mantle minerals (Ryan and Langmuir 1987; Brenan *et al.* 1998a; Seitz and Woodland 2000; Ottolini *et al.* 2009), it can be concentrated in crust materials compared to the mantle. In addition, Li is highly mobile and tends to partition preferentially into fluid phase in the processes of near-surface weathering, sea-floor alteration, and subduction-zone metamorphism (Seyfried *et al.* 1984; You *et al.* 1996; Brenan *et al.* 1998b); thus Li can be strongly enriched in the oceanic crust by seawater alteration and then released in the subduction zone.

The main reason that the Li isotope system is a powerful tracer reflects the large relative mass difference between ⁶Li and ⁷Li, which favours the great range of δ^7 Li [Li isotopic composition relative to the L-SVEC (lithium carbonate prepared by H. Svec; Flesch *et al.* 1973) standard supplied by the National Institute of Standards and Technology: [(⁷Li/⁶Li)_{sample}/(⁷Li/⁶Li)_{L-SVEC}-1]×1000], from -34 to +50‰ in terrestrial samples (Hoefs and

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Sywall 1997; Košler *et al.* 2001; Tomascak 2004; Zhang *et al.*, personal communication). The fractionation of Li isotopes appears to be especially strong in low-temperature systems and highly variable in altered oceanic crust (Chan and Edmond 1988; Chan *et al.* 1994), hydrated mantle rocks (Decitre *et al.* 2002; Agostini *et al.* 2008), and eclogitic slabs (Zack *et al.* 2003; Marschall *et al.* 2007). In contrast, extremely small fractionation at higher temperatures (>350°C) has been recognized (Chan *et al.* 1994; Tomascak *et al.* 1999b; Chan and Frey 2003; Wunder *et al.* 2006) and thus Li isotopic fractionation is negligible in magmatic processes, implying that lavas have the potential of recording the isotopic compositions of their sources (Moriguti *et al.* 2004; Elliott *et al.* 2006; Agostini *et al.* 2008; Chan *et al.* 2009; Košler *et al.* 2009).

Mechanisms producing large fractionations of Li isotopes and origins of the extremely low δ^7 Li observed in mantle-derived rocks are still poorly understood despite major advances in the study of Li isotope geochemistry. In this article we briefly review the latest developments in this field and the assumed mechanisms of Li isotopic fractionation. Our main purpose is to more fully constrain the origins of low δ^7 Li in mantle-derived rocks and their implications for crust/mantle recycling by comparison of Li isotopic results with existing Sr-Nd isotopic evidence.

Li isotopic compositions of major reservoirs

A rapidly growing database allows us to put new constraints on the Li isotopic compositions of major geochemical reservoirs, most of which were summarized previously (Tomascak 2004; Tang *et al.* 2007b). The variable ranges of δ^7 Li in major reservoirs are shown in Figure 1. Seawater has a relatively homogeneous Li isotopic composition and is isotopically quite heavy, with an average of δ^7 Li ~ +32‰, while marine biogenic carbonates, marine pore water, and marine sediments show wide ranges of δ^7 Li. Marine biogenic carbonates have the highest δ^7 Li of +50‰ among the reservoirs (Košler *et al.* 2001).

Another relatively homogeneous reservoir is the normal mantle, represented by fresh mid-ocean ridge basalts (MORB), with δ^7 Li of +1 to +6‰ (Figure 1). However, the mantle is actually heterogeneous as reflected by the variable Li isotopic compositions of oceanic island basalts (OIB) with δ^7 Li of about +2 to +11‰ and mantle-derived peridotites showing a large δ^7 Li range, from +13 to -34‰ (Zhang *et al.*, personal communication).

The upper continental crust is isotopically lighter than the normal mantle, with an average of $\delta^7 \text{Li} \sim 0\%$ (Teng *et al.* 2004), which is inferred to reflect the influence of weathering, with heavy Li partitioned into surface waters, leaving light Li in the weathered residua. This inference is supported by the high $\delta^7 \text{Li}$ values of river waters (Huh *et al.* 1998, 2001) and ground waters (Hogan and Blum 2003), as well as the experimental data (Pistiner and Henderson 2003). Most altered oceanic basalts and marine sediments display $\delta^7 \text{Li}$ intermediate between that of the mantle and seawater, due to the uptake of heavier seawater Li.

Orogenic eclogites, with the lowest δ^7 Li of -35% (Cheng *et al.* personal communication), are isotopically lighter than the upper crust, reflecting loss of heavy Li during metamorphic dehydration of subducting slab (Zack *et al.* 2003; Wunder *et al.* 2007; Agostini *et al.* 2008) and/or diffusive influx of Li into the eclogites from the country rocks during exhumation (Marschall *et al.* 2007).

As illustrated in Figure 2, surface weathering produces the high $\delta^7 \text{Li}$ of river waters that feed the oceans. Low-temperature alteration of oceanic crust then makes seawater heavier than river water. Li isotopic fractionation during the dehydration of altered oceanic crust could produce isotopically heavy-Li fluids and light-Li slab residue, which



Figure 1. Li isotopic compositions of major geochemical reservoirs. Data sources: chondrites (McDonough and Sun 1995; Tomascak 2004; Magna *et al.* 2006; Seitz *et al.* 2006, 2007), seawater (Chan and Edmond 1988; You and Chan 1996; Moriguti and Nakamura 1998; Tomascak *et al.* 1999a; James and Palmer 2000; Rudnick *et al.* 2004b; Tomascak 2004), river waters (Huh *et al.* 1998; 2001), high-temperature vent fluids (Chan *et al.* 1993; Foustoukos *et al.* 2004; Kisakürek *et al.* 2004), arc lavas (Moriguti and Nakamura 1998; Tomascak *et al.* 2002b; Agostini *et al.* 2008; Košler *et al.* 2009), OIB (Tomascak *et al.* 2000, 2002; Chan *et al.* 2002a, 2002b; Moriguti and Nakamura 1998; Tomascak *et al.* 2009), fresh MORB (Chan *et al.* 1992, 2002a, 2002b; Moriguti and Nakamura 1998; Tomascak and Langmuir 1999; Elliott *et al.* 2006; Nishio *et al.* 2007; Tomascak *et al.* 2008), altered MORB (Chan *et al.* 1992, 2002a), marine sediments (Chan *et al.* 1994, 2006; Zhang *et al.* 1998; James *et al.* 1999; Chan and Kastner 2000; Bouman *et al.* 2004), loess, shales, and upper continental crust (Teng *et al.* 2004), Eclogite data (Zack *et al.* 2003; Marschall *et al.* 2007; Cheng *et al.* 2007b; Zhang *et al.* 2004), Eclogite data (Zack *et al.* 2003; Marschall *et al.* 2007; Cheng *et al.* 2007b; Zhang *et al.*, personal communication), granites (Tomascak 2004; Teng *et al.* 2009), other reservoirs (Tomascak 2004).

metasomatized the overlying mantle wedge, and thus the arc lavas derived from the mantle wedge show considerable variation of $\delta^7 \text{Li}$, from +12 to -6‰.

Li isotopic fractionation mechanisms

Differential partitioning of ⁷Li and ⁶Li between solid and aqueous phases is the principal mechanism invoked to explain the Li isotopic variations in major near-surface reservoirs because ⁷Li is preferentially partitioned into the fluid while ⁶Li is incorporated into the solid (Taylor and Urey 1938; Chan *et al.* 1999, 2002b; Yamaji *et al.* 2001; Wunder *et al.* 2006). Since a large fractionation can occur at low temperatures while absent at high temperatures, temperature should be a key factor of Li isotopic fractionation (Seyfried *et al.* 1998; Tomascak *et al.* 1999b; Coogan *et al.* 2005).

Li isotope compositions of mantle material may be affected by diffusion processes due to the exceptionally high diffusivity of Li (Nakamura and Kushiro 1998; Coogan *et al.* 2005) and faster diffusion of ⁶Li than ⁷Li (Richter *et al.* 2003). Thus high-temperature diffusive fractionation has been invoked to account for striking δ^7 Li variation in country



Figure 2. Schematic illustration of Li isotopic systematics in different settings, modified from Zack *et al.* (2003), Elliott *et al.* (2004), and Tang *et al.* (2007b). The numbers in the figure represent the δ^7 Li values. Weathering of continental rocks results in the heavy Li isotopic compositions of river water which feeds the oceans. Low-temperature alteration of oceanic crust then makes seawater heavier than river water. High δ^7 Li in decollement fluids and serpentinite diapirs indicate Li isotopic fractionation during dehydration of altered oceanic crust. The high δ^7 Li of fluids escaping the slab at low temperatures likely enrich the overlying mantle wedge in ⁷Li, which may be the source of arc lavas. Thus, variable δ^7 Li in arc lavas might be explained by incorporation of the mantle wedge. Data sources are the same as those identified in Figure 1.

rocks of pegmatite (Teng *et al.* 2006), within individual phenocrysts (Beck *et al.* 2006; Parkinson *et al.* 2007), and intra-granular, inter-granular, and inter-sample scales in mantle-derived xenoliths (Lundstrom *et al.* 2005; Jeffcoate *et al.* 2007; Rudnick and Ionov 2007; Tang *et al.* 2007a; Ionov and Seitz 2008; Kaliwoda *et al.* 2008; Magna *et al.* 2008; Aulbach and Rudnick 2009).

One recent study proposed that Li isotopic zoning can occur as a natural result of cooling magmatic systems based on parameterizations of the temperature dependence of Li partitioning and diffusivity in clinopyroxene (cpx) (Gallagher and Elliott 2009). This calculation model can produce an asymmetric isotope profile similar to those documented in some olivine and cpx phenocrysts with isotopically normal cores but light rims; hence temperature dependence of diffusivity and partition coefficient of Li isotopes are considered as key factors in producing the diffusion profile (Gallagher and Elliott 2009). This conclusion supports the speculation that significant δ^7 Li variations in lava-hosted peridotite xenoliths may be related to different cooling times for thicker and thinner flows (Ionov and Seitz 2008).

In brief, differential partitioning of ⁷Li and ⁶Li between solid and aqueous phases is the primary mechanism for Li isotopic fractionation in major near-surface reservoirs, and diffusive fractionation of Li isotopes is very prevalent in deep-seated rocks (such as eclogites, pegmatites, granites, granulite and peridotite xenoliths), which could be strongly affected by temperature.

Origins of low δ^7 Li in mantle-derived rocks

As mentioned above, diffusion-driven isotopic fractionation has been accepted as an important mechanism for extremely large variation of δ^7 Li in mantle peridotites (Richter *et al.* 2009). However, the boundary conditions for the diffusion processes, such as cooling of magmatic systems (Beck *et al.* 2006; Ionov and Seitz 2008; Kaliwoda *et al.* 2008; Gallagher and Elliott 2009), alkali diffusion and melt extraction (Lundstrom *et al.* 2005),

melting of peridotites promoted by metasomatism (Magna *et al.* 2008), interactions of xenoliths with host rocks (Jeffcoate *et al.* 2007; Parkinson *et al.* 2007; Rudnick and Ionov 2007), asthenospheric melt (Aulbach *et al.* 2008; Aulbach and Rudnick 2009), and/or recycled crustal melt (Nishio *et al.* 2004; Tang *et al.* 2007a), are currently disputed. Among these controversies, Li isotopic composition of recycled, dehydrated oceanic slab is a hot topic.

Progressive metamorphic dehydration of a highly altered oceanic crust (basaltic protoliths) during its subduction, with concomitant loss of heavy Li into expelled aqueous fluids, could produce low δ^7 Li in the slab residual by an approximate process of Rayleigh distillation, which is documented for exhumed orogenic eclogites from Trescolmen, Switzerland, whose δ^7 Li ranges from -11 to +5‰ (Zack *et al.* 2003). This scenario is modelled for Li in cpx according to experiments involving Li isotopic fractionation between cpx and Cl- and OH-bearing aqueous fluids between 500 and 900°C at 2.0 GPa (Wunder *et al.* 2006), which is further supported by the observation that continuous dehydration of altered oceanic crust could produce fluids enriched in Li and ⁷Li and rock residual with low δ^7 Li based on the experiments of Li isotope fractionation between synthetic Li-staurolite and aqueous fluids containing excess LiCl or LiOH at 3.5 GPa and 670 to 880°C and between Li-mica and similar fluids at 2.0 GPa and 300–500°C (Wunder *et al.* 2007).

However, Marschall *et al.* (2007) proposed a contrary prediction that the entire prograde metamorphic process, up to anhydrous eclogite, can account for a decrease in δ^7 Li of only $\leq 3\%$ in subducting oceanic crust during dehydration based on modelling calculations. They ascribed the low δ^7 Li of eclogites (as low as -21.9%) to kinetic fractionation of the Li isotopes during diffusive influx of Li from the country rocks into the exhuming eclogites and thus they predicted the deeply subducted eclogites to have a Li isotopic signature heavier than the mantle. This inference is likely supported by the heavy δ^7 Li values (up to 6.2‰) in olivine from the high μ (HIMU: $\mu = ^{238}U/^{204}Pb$) lavas from the Cook-Austral volcanic chain, indicating that the source for the HIMU lavas contains dehydrated recycled oceanic crust whose 'heavy' Li-isotope signature is partially preserved during passage to the mantle through the subduction factory (Chan *et al.* 2009).

Based on the Li, B, Sr, and Nd isotope variations in the lavas spanning approximately 10 Ma of subduction-related volcanism in western Anatolia, Agostini *et al.* (2008) suggest that the presumptions of Marschall *et al.* (2007) may be not valid in a case where subduction is stopping and a nearly 'hung' downgoing slab is being comprehensively devolatilized and that solid–fluid exchanges of Li as subduction slows could lower the δ^7 Li of the slab more extensively at shallow depths than is observed in normal arc settings. Thus, the very dehydrated slab will have a lower δ^7 Li than could develop during active subduction. High-temperature, Li-rich outfluxes from the slab at depth will perverse this lighter δ^7 Li signature (Agostini *et al.* 2008). This study may provide a necessary complement to the above models and shed light on the origins of low δ^7 Li in mantle-derived rocks.

Recently, we observe a rough trend between the δ^7 Li and Sr-Nd isotopic ratios of cpx in peridotite xenoliths from the Hannuoba and Fanshi Cenozoic basalts on the North China Craton. The δ^7 Li values increase with the increase of 143 Nd/ 144 Nd and the decrease of 87 Sr/ 86 Sr ratios for most samples (Figure 3). In one word, the δ^7 Li values increase with the depletion of Sr-Nd isotopic compositions in the peridotites, showing a mixing trend between the depleted mantle (DM) and enriched mantle (EM1). Some samples are plotted below the mixing fields due to their low δ^7 Li relative to a certain Sr or Nd isotope ratio, which may be the consequence of recent ingress of Li from infiltrated melt into the peridotites. Similarly, the cpx data from Nishio *et al.* (2004) also show a DM–EM1 mixing



Figure 3. Sr and Nd isotope ratios against δ^7 Li in cpx from peridotite xenoliths. The grey fields represent the assumed mixing of DM and EM1. The cpx plotted below the mixing fields are low in δ^7 Li due to recent Li ingress. Some samples from the Labait, Tanzania, have high δ^7 Li, possibly indicating the isotopically heavy Li of HIMU (Aulbach *et al.* 2008). Data sources: δ^7 Li for DM (Chan *et al.* 1992, 2002a; Moriguti and Nakamura 1998; Elliott *et al.* 2006; Nishio *et al.* 2007; Tomascak *et al.* 2008); for HIMU estimated from Nishio *et al.* (2004, 2005), Aulbach *et al.* (2008) and Chan *et al.* (2009); and for EM1 estimated from Nishio *et al.* (2004), Wunder *et al.* (2006), Tang *et al.* (2007a), Agostini *et al.* (2008), Košler *et al.* (2009) and Tang *et al.* (2009). Sr and Nd isotope data sources: Fanshi (Fan *et al.* 2000; Tang *et al.* 2008; Xu *et al.* 2000; Rudnick *et al.* 2004a; Tang *et al.* unpublished data), Yangyuan (Ma and Xu 2006; Xu *et al.* 2008), DM, HIMU and EM1 end members (Zindler and Hart 1986); other data: (Nishio *et al.* 2004; Aulbach *et al.* 2008).

trend, while the Labait peridotites from Tanzania may reflect a signature of isotopically heavy Li of HIMU (Aulbach *et al.* 2008).

Large variations of Sr-Nd isotopic ratios of cpx (Figure 4) – coupled with the petrology, mineralogy, and elemental and Re-Os isotopic geochemistry previously documented for peridotite xenoliths from the Fanshi (Tang *et al.* 2008; Xu *et al.* 2008) and two adjacent localities, Hannuoba (Song and Frey 1989; Tatsumoto *et al.* 1992; Fan *et al.* 2000; Rudnick *et al.* 2004a; Zhang *et al.* 2009) and Yangyuan (Ma and Xu 2006; Xu *et al.* 2008) – indicate that the lithospheric mantle beneath the North China Craton could have been previously modified by recycled materials that changed the Rb/Sr and Sm/Nd ratios in the old lithosphere, and then the modified lithosphere experienced a secular evolution. Furthermore, relatively depleted Sr-Nd isotopic compositions in some peridotites could be the products of recent asthenosphere is depleted in Sr-Nd isotopic compositions, the reaction products should be depleted in Sr-Nd isotopic compositions relative to their earlier counterparts (old lithospheric mantle) with evolved Sr-Nd isotopic ratios. Thus, the Sr-Nd isotope compositions of cpx from the peridotites (Figure 4) provide clear evidence for the DM–EM1 mixing.

Figure 5 shows the Li element and isotope data published recently and illustrates the behaviours of Li isotopes during the processes of fluids/melt-rock interactions. The normal mantle has δ^7 Li range similar to that of fresh MORB. Low-temperature alteration



Figure 4. Sr and Nd isotopic compositions in cpx from peridotite xenoliths. The grey field represents the mixing between DM and EM1. Data sources are the same as those in Figure 3.

can increase the Li contents and δ^7 Li in altered MORB (Chan *et al.* 1992, 2002a), while dehydration metamorphism can produce isotopically complementary heavy-Li fluids and light-Li slab residue, which hydrated the overlying mantle wedge, and thus arc lavas derived from the mantle wedge show variable δ^7 Li. Meanwhile, the low δ^7 Li in the slab residue could be further lowered (down to -10% or even lower?) by means of solid–fluid exchanges of Li (Agostini *et al.* 2008) and then be preserved in the mantle to form the EM1. Peridotite will acquire a low- δ^7 Li signature if initially metasomatized by a low- δ^7 Li agent from the EM1. Later interaction between the lower- δ^7 Li peridotite and higher- δ^7 Li asthenospheric melt will elevate the bulk δ^7 Li of peridotite. This inference is consistent with the linear correlation between the bulk δ^7 Li and 1/Li for these peridotites (Figure 5), showing a DM–EM1 mixing trend. Some peridotites and their constituent minerals are plotted below the mixing field due to their decreased δ^7 Li as a result of recent Li ingress.

Altogether, we believe in the presence of low $\delta^7 \text{Li}$ component in the mantle, which may form the EM1 end member according to the correlation between $\delta^7 \text{Li}$ and Sr-Nd isotopic compositions in the peridotites (Figure 3) and the growing data of low $\delta^7 \text{Li}$ in deep slab and mantle-derived rocks (Zack *et al.* 2003; Benton *et al.* 2004; Nishio *et al.* 2004, 2005, 2007; Nishio *et al.* 2007a; Agostini *et al.* 2008).

Implications for crust/mantle recycling

Since recycled crustal materials have evolved Sr-Nd isotopic ratios, interaction of a normal mantle peridotite with the recycled materials could lead to the enrichment of Sr-Nd isotopic composition in peridotite. However, such a case is highly complicated in terms of Li isotopic system due to the great variability of δ^7 Li in recycled crust materials. It has been proposed that δ^7 Li variation is closely related to the mineral composition (Wunder *et al.* 2007) and physical state (Marschall *et al.* 2007; Agostini *et al.* 2008) of the subducted slab. For example, mantle modified by subducted material will have negative δ^7 Li values if the major mineralogical Li reservoirs in the subducted slab have octahedral coordination of Li (such as micas and pyroxenes), as opposed to positive δ^7 Li values if Li



Figure 5. Variation of δ^7 Li with Li content for minerals and bulk rocks of peridotite xenoliths compared with published data. Dashed black arrow shows the trend for alteration of oceanic crust and the grey arrow for the dehydration trend of subducted slab. The dashed grey line represents the linear correlation of δ^7 Li and 1/Li for the bulk peridotites from the Fanshi and Hannuoba, showing a mixing trend of EM1 and DM. Some minerals with higher Li contents and lower δ^7 Li are plotted below the mixing field due to recent Li ingress into the minerals. Data sources are the same as those identified in Figures 1 and 3.

in the slab is tetrahedrally coordinated (such as staurolite). Besides, the behaviour of minerals might also have some influence on the isotopic fractionation (Wunder *et al.* 2007). Therefore, recycling of crustal materials could produce different components in the mantle: (1) ⁷Li-enriched fluids, which hydrated and elevated the δ^7 Li of mantle wedge; (2) isotopically light-Li slab residue, which possibly formed the EM1 member; and (3) isotopically heavy-Li components under certain circumstances, which might contribute high δ^7 Li to the HIMU member.

Conclusions

Lithium isotope geochemistry has developed very rapidly in recent years and been used to trace many geological processes related to fluids/melts. The large variations of δ^7 Li observed in newly studied OIB, MORB, granites, eclogites, arc lavas, lower crust-derived granulite, and mantle-derived peridotite xenoliths further enrich the database of major geochemical reservoirs.

Differential partitioning of ⁷Li and ⁶Li between solid and aqueous phases is the principal mechanism for the Li isotopic fractionation in major near-surface reservoirs. High-temperature diffusion fractionation of Li isotopes has recently been recognized as an important mechanism for the isotopic fractionation in crustal and mantle-derived rocks.

Many scientific achievements have been made in Li isotope geochemistry. However, the origins of low δ^7 Li in mantle-derived rocks are still poorly constrained.

The correlation between δ^7 Li and Sr-Nd isotope ratios of cpx in peridotite xenoliths, associated with previous findings, suggests that the recycling of ancient oceanic crust could produce a low- δ^7 Li component in the mantle and that isotopically light Li may be a feature of EM-1 end member. Meanwhile, dehydration of the subducted slab could result in the δ^7 Li in mantle wedge and HIMU member higher than that of the normal mantle. These processes may be dependent on the mineral compositions and physical state of subducted slab.

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