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Li isotope ratios of spring fluids as an effective tracer of slab-derived subducted sources across the Costa Rica forearc

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ABSTRACT

Spring waters from across the Costa Rica margin were analyzed for their Li and He isotope compositions to determine the utility of Li isotopes as a tracer of volatile sources in subduction zones. Li isotope ratios systematically decrease with increasing depth to the subducting slab: averaging +15.0‰ ± 9.2‰ in the outer forearc (<40 km to the slab), +9.3‰ ± 4.3‰ in the forearc (40–80 km to the slab), and +5.8‰ ± 2.8‰ in the arc (>80 km to the slab). In contrast, air-corrected ^{3}He/^{4}He values (reported relative to the ratio in air, R$_A$) range from 0.4 to 7.5 R$_A$ and increase from predominantly crustal values near the trench to mantle values in the arc. Together, these data support progressive devolatilization of the subducting plate with slab-derived Li components sourced from shallowly expelled pore fluids in the outer forearc, sedimentary and/or altered oceanic crust contributing to the forearc, and limited slab input beneath the arc.

INTRODUCTION

Recent developments in Li isotope geochemistry have raised concerns about the utility of Li isotopes as tracers of fluid sources in magmatic and metamorphic systems (e.g., Marschall and Tang, 2020). Initial excitement surrounding the use of Li isotopes to trace subduction of crustal components through the subduction cycle (e.g., Zack et al., 2003; Elliott et al., 2004) was dampened by the recognition of diffusive fractionation of Li isotopes at magmatic temperatures (e.g., Jeffcoate et al., 2007). Although Li may be subject to diffusive fractionation at high temperatures, we assert that Li isotopes can still be an effective means to trace fluid sources within subduction zones given the right parameters. Lithium isotopes remain advantageous in that they undergo minimal fractionation during liquid-vapor separation (Liebscher et al., 2007) and during dehydration of the subducting slab, thus preserving the isotopic composition of their source (e.g., Marschall et al., 2007; Penniston-Dorland et al., 2012).

In this study, we report δ^7Li values in cold (<40 °C) spring fluids to determine the change in slab-derived fluid components across the Costa Rica convergent margin. Cold fluids were targeted to limit secondary geochemical modification via fluid-rock interaction at high temperatures. For comparison, we analyzed higher-temperature fluids from adjacent springs. Costa Rica is one of few localities in which forearc springs are exposed subaerially due to several uplifted peninsulas and therefore easily accessible. We collected spring samples from the Santa Elena, Nicoya, and Burica Peninsulas as well as across the margin to the volcanic arc, conducting a transect covering depths to the subducting slab from ~20 to ~120 km.

GEOLOGIC SETTING AND SPRING SAMPLES

The subduction of the Cocos plate beneath the Caribbean plate at a convergence rate of 60–90 mm/yr at the Middle America Trench results in an arc of active volcanoes stretching from Guatemala to southern Costa Rica. Along-arc variations in the subduction parameters drive large changes in the chemistry of arc products. Steep subduction of hydrated oceanic crust formed at the fast-spreading East Pacific Rise in Nicaragua and northern Costa Rica is associated with strong slab-fluid signatures (e.g., Ba/La; Carr et al., 1990) in arc lavas. In central and southern Costa Rica, subduction of thicker oceanic crust formed at the Cocos-Nazca spreading center under the influence of the Galapagos plume results in shallower slab dip, less-pronounced slab-fluid signatures, and oceanic island basalt (OIB)–like lava geochemistry (e.g., Gazel et al., 2011). Subduction of seamounts and the Cocos Ridge result in uplift of the Nicoya and Osa Peninsulas and tectonic erosion of the outer forearc (e.g., Vannucchi et al., 2006).

A total of 43 springs (47 samples) along and across the Costa Rica margin were studied (Fig. 1; Table S1 in the Supplemental Material[1]). Nine samples (2017 collection dates; Table S1) were previously studied (Barry et al., 2019). Depth to the subducting slab interface (Lücke and Arroyo, 2015) was used to classify a spring as "outer forearc" (<40 km), "forearc" (40–80 km), or "volcanic arc" (>80 km). The lithology of the overriding plate varies, with sampled springs hosted in Quaternary alluvium, Quaternary volcanics, Tertiary volcanics, Cretaceous to Quaternary deep-water sediments, and Jurassic seafloor basalts. Several of the outer forearc springs on the Nicoya Peninsula (SI, RS, SR, SN, SG, SM, SV; spring abbreviations in Table S1) are located along the Falla Morote, a major

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[1] Supplemental Material. Detailed methods, Figures S1–S3, Tables S1–S4. Please visit https://doi.org/10.1130/GEOL.S.23571741 to access the supplemental material, and contact editing@geosociety.org with any questions.

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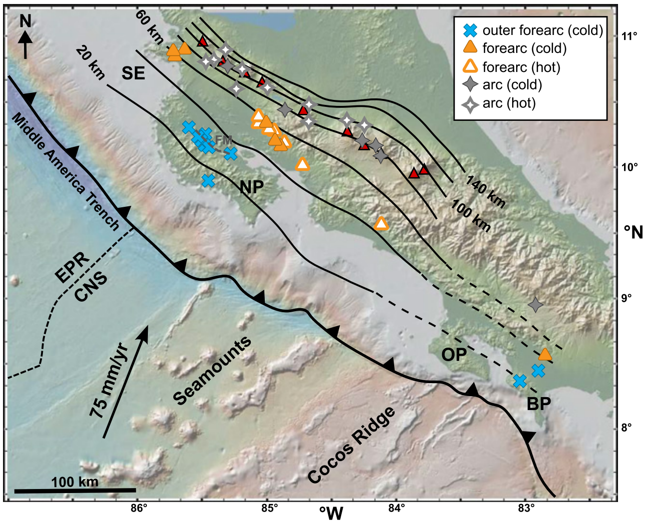

Figure 1. Map of the Costa Rica convergent margin showing sample sites. Solid symbols are cold fluids (<40 °C); open symbols are warm fluids (>40 °C). SE—Santa Elena Peninsula; NP—Nicoya Peninsula; OP—Osa Peninsula; BP—Burica Peninsula; FM—Falla Morote (dashed gray line); EPR—East Pacific Rise; CNS—Cocos-Nazca spreading center. Depth to subducting slab (solid black lines) is shown in 20 km contours after Lücke and Arroyo (2015, and references therein). Depth to subducting slab in southern Costa Rica is not well defined and shown as speculative dashed black lines. Volcanoes are shown as red triangles.

active northwest-southeast–trending reverse fault (Denyer et al., 2014). Two of the forearc springs (CP, LF) are in altered and mineralized volcanic rocks of the Aguacate Formation. Four springs were sampled twice (SL, LW, FA, GY) to investigate chemical variability. Laurel Well (LW) was sampled 454 days before and 8 days after a magnitude 6.3 earthquake occurred on the Burica Peninsula.

ANALYTICAL METHODS

Li concentrations and δ^7Li values were analyzed at the University of Texas at Austin (Austin, Texas, USA). Concentrations were determined via solution mode inductively coupled plasma mass spectrometry (ICP-MS) using an Agilent 7500ce quadrupole ICP-MS, and Li isotope ratios were measured using a Nu Plasma 3D multi-collector ICP-MS. In addition, ^{3}He/^{4}He values, reported relative to the ratio in air (R/R_A), were measured and air corrected (R_C) for either the fluid or free gas. Helium analyses were performed at Woods Hole Oceanographic Institution (Woods Hole, Massachusetts, USA). Major anions were analyzed at the National University of Costa Rica (Heredia, Costa Rica). See the Supplemental Material for details.

RESULTS

The spring waters show a dramatic range in geochemical parameters, with temperatures from 20.6 °C to 98.4 °C and pH from 1.7 to 11.3 (Table S1). Water types range broadly from sulfate dominated to bicarbonate and chloride dominated (Fig. S1; Table S2). Lithium concentrations range from 0.4 to 5173 µg/L and δ^7Li values from −2.4‰ to +27.3‰ (Fig. 2). Air-corrected ^{3}He/^{4}He (R_C/R_A) values range from 0.4 R_A to 7.5 R_A (Fig. 2). Overall, δ^7Li values decrease with increasing depth to the subducting slab from an average of +15.0‰ ± 9.2‰ in the outer forearc to +9.3‰ ± 4.3‰ in the forearc and +5.8‰ ± 2.8‰ in the arc (Fig. 2). The maximum [Li] increases with increasing depth to the slab. However, there remains a large range in [Li] at a given distance from the trench (e.g., arc samples range from <1 to >5000 µg/L Li), resulting in no correlation with δ^7Li values (Fig. 2; Fig. S2). In contrast, ^{3}He/^{4}He values increase with distance from the trench (averaging 1.3 ± 0.8 R_A in the outer forearc, 2.7 ± 1.5 R_A in the forearc, and 5.9 ± 1.6 R_A in the arc), correlating with decreasing δ^7Li values (Fig. 3). No clear correlations are observed between δ^7Li values and major anions (Fig. 4A).

Springs that were sampled twice (FA, SL, GY) show minimal change in [Li] and/or δ^7Li values over the course of a year and up to four years. LW shows minimal to no change in [Li] and δ^7Li values from pre- and post-earthquake samples. Samples from different springs at Quepos Hot Springs have δ^7Li and ^{3}He/^{4}He values within error and similar [Li].

DISCUSSION

Li Isotopes as a Tracer of Slab-Derived Fluids in Spring Fluids

The δ^7Li values of the spring fluids are bounded by the end members of seawater (+31‰) and mid-ocean-ridge basalt (MORB) (~+3‰ to ~+5‰) (Tomascak et al., 2016, and references therein). With increasing distance to the subducting slab, the δ^7Li values steadily decrease to mantle values (Fig. 2). Values from samples from the outer forearc, except for outliers along the Falla Morote (see discussion below), largely overlap with those

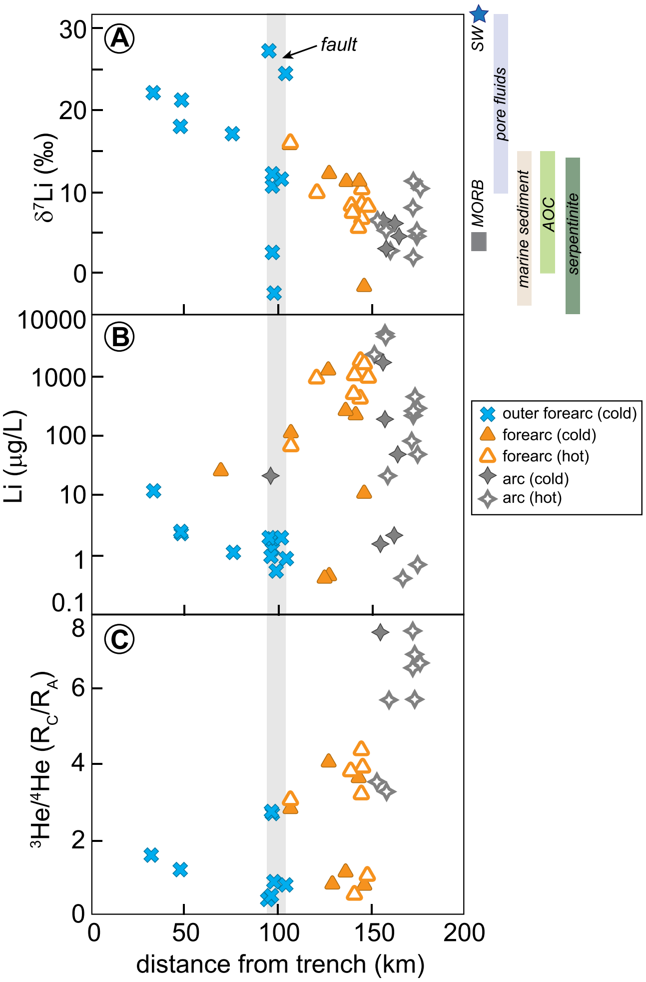

Figure 2. Li isotope composition (A), Li concentration (B), and ^{3}He/^{4}He values (reported as air corrected and relative to the ratio in air, R_C/R_A) (C) of spring waters with distance from the trench. Symbols are larger than the error. Range in δ^7Li values is given for various reservoirs (references in text). MORB—mid-ocean-ridge basalt; SW—seawater; AOC—altered oceanic crust. Location of Falla Morote is highlighted by gray bar. Río Cotón and Río Colorado samples plot closer to the trench for forearc and arc given the shallower dip of the subducting slab in southern Costa Rica; δ^7Li and ^{3}He/^{4}He values were not analyzed for these two samples.

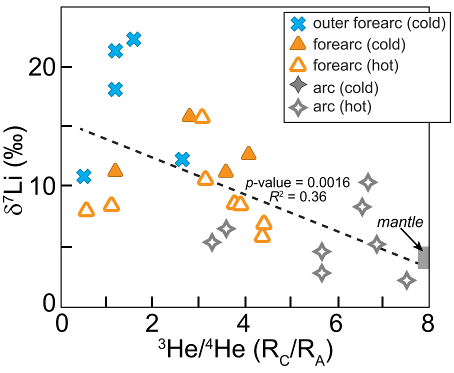

Figure 3. Li isotope ratios of spring fluids decrease with increasing ^{3}He/^{4}He values (reported as air corrected and relative to the ratio in air, R_C/R_A) ($R^2 = 0.36$; *p*-value = 0.0016). Black dashed line is the best fit line. Samples that experienced modification of δ^7Li values along the Falla Morote (springs SI, SV, SM, SG) or contribution from surficial mining drainage (spring CP) are not included. Gray box highlights mantle values.

of sedimentary pore fluids, which range globally from $\sim$+10‰ to +45‰ (Tomascak et al., 2016, and references therein) and $\sim$+20‰ to +40‰ in Central America (Chan and Kastner, 2000). Values in fluids from the forearc overlap with those of altered oceanic crust (AOC; $\sim$0‰ to +15‰) (e.g., Chan et al., 1992, 2002) as well as marine sediments (globally between $\sim$−4‰ and +15‰ with samples offshore Costa Rica $\sim$+2‰ to $\sim$+13‰) (Chan et al., 2006) (Fig. 2). However, values from seafloor serpentinites also overlap, ranging from $\sim$−28‰ to $\sim$+14‰ (Decitre et al., 2002; Vils et al., 2009). Values from arc springs are consistent with MORB values, likely reflecting the ambient mantle wedge, but overlap with those of other potential deeply subducted sources (Fig. 2). Collectively, the decrease in δ^7Li values suggests an evolving slab component due to progressive devolatilization of the subducting slab from shallow loss of pore fluids in the outer forearc, devolatilization of sediments and/or AOC below the forearc, and minimal input from the slab beneath the arc.

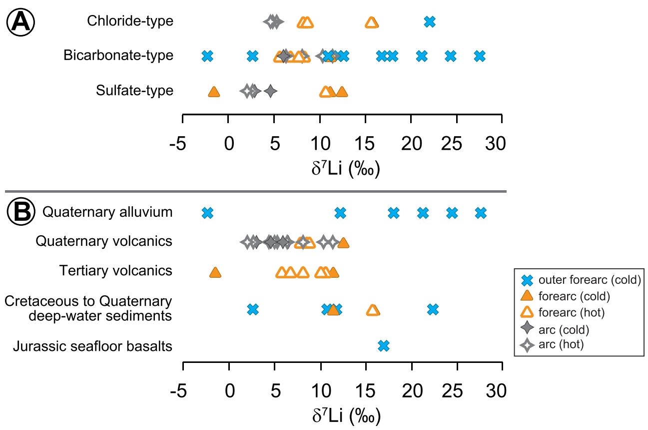

Figure 4. Li isotope composition of springs grouped by water classification (Fig. S1 [see text footnote 1]) (A) and lithology of the upper-plate host rock (B). Lithology from Schruben (1996).

With decreasing distance to the arc, ^{3}He/^{4}He values increase, approaching upper mantle values ($\sim$8 R_A) (Fig. 2) (Barry et al., 2019; this study). Lower ^{3}He/^{4}He values are recorded in the outer forearc and forearc but still indicate a mantle component (i.e., samples with R_C/R_A >1) consistent with a deep volatile source mixed with shallow crustal fluids (Barry et al., 2019). The correlation between δ^7Li and ^{3}He/^{4}He values (Fig. 3) supports the interpretation of slab-derived Li components to the outer forearc and forearc springs and limited slab input beneath the arc.

Potential of Li Isotope Fractionation

As with any isotopic system, isotopic fractionation during secondary processes (e.g., fluid-rock interaction) must be evaluated. In general, ^{7}Li preferentially partitions into aqueous fluids relative to minerals (Tomascak et al., 2016, and references therein). Therefore, the formation or dissolution of Li-bearing minerals may result in isotopic fractionation. Li is a highly fluid-mobile element thought to undergo limited interaction with the host rock and thus considered to be a conservative tracer (e.g., Giggenbach, 1991). However, some work has shown that Li can be both incorporated in secondary minerals and lost to solution via mineral breakdown, resulting in isotopic fractionation typically at higher temperatures (>150 °C) (e.g., James et al., 2003; Reyes and Trompetter, 2012). Despite numerous studies on Li in hydrothermal systems, a complete understanding of Li behavior is lacking, with studies commonly showing contradictory results. For example, some studies report δ^7Li values of hydrothermal fluids varying with temperature (e.g., Millot and Négrel, 2007), whereas others show minimal temperature dependence (Bernal et al., 2014; Cullen et al., 2019).

To avoid complications due to shallow fluid-rock hydrothermal interaction, we have focused on cold springs. Cold and hot springs with similar distances from the trench result in similar δ^7Li values, suggesting minimal temperature effects. Calculated subsurface spring temperatures using silica thermometry (Fournier, 1977) show a weak correlation ($R^2 = 0.2$) with δ^7Li values (Table S4; Fig. S3). Previous work on hydrothermally altered basalts has shown correlations between [Li] and δ^7Li values due to incorporation of Li into secondary minerals (Chan et al., 1992, 2002). Importantly, in the Costa Rica springs, there is no correlation between [Li] and δ^7Li values (Fig. 2; Fig. S2). Dilution with meteoric water likely results in variation of [Li] without affecting δ^7Li values, as is exemplified by arc sample VB200923, which has low temperature and very low [Li] yet preserves a typical arc δ^7Li value. In addition, δ^7Li values do not correlate with the lithology of the upper-plate host rock (Fig. 4B). For example, high δ^7Li values (>15‰) are recorded in three different lithologies: Quaternary alluvium, Cretaceous to Quaternary sediments, and Jurassic seafloor basalts. Furthermore, the observation that δ^7Li values do not correlate with Cl-SO_4-HCO_3 chemistry of spring waters (Fig. 4A) argues against hydrothermal processing (e.g., Henley and Ellis, 1983; Giggenbach and Corrales Soto, 1992) as the primary driver of δ^7Li variations. The steady decrease in δ^7Li values toward mantle values with distance from the trench and correlation with He isotope ratios strongly supports the interpretation that Li isotopes in the forearc are predominately recording slab-derived components to the springs.

Despite the decreasing trend in δ^7Li values with depth to the subducting slab, there are some distinct outliers. Several springs along the Falla Morote have δ^7Li values significantly above (springs SI, SV) or below (SM, SG) the overall trend line. These values may result from diffusion due to highly variable degrees of fluid-rock interaction along this fluid pathway in the crust. Li concentrations in these samples are very low, making these fluids highly sensitive to modification by shallow processes within the fault zone. In contrast, primary fluids with high [Li] would be relatively impervious to secondary processing in the shallow crust. The CP sample has a much lower δ^7Li value (−1.6‰) than other forearc samples and is from a mineralized region where field observations suggest contribution from associated acid mine drainage. The low He isotope ratio (0.8 R_A) also suggests a shallow, crustal component. These observations demonstrate that although Li isotopes can be an effective tracer of deeper fluid sources, which feed thermal spring systems, care must be taken with samples showing modification by shallow processes.

Comparison with Previous Work

There has been extensive previous work focusing on the cycling of volatile and fluid-mobile elements through the Central American subduction zone; however, most of the work has focused on the volcanic arc. Based on various elemental and stable isotope ratios, slab contributions to the Costa Rica arc volcanic products are estimated to be minimal (e.g., Carr et al., 1990; Patino et al., 2000; Fischer et al., 2002). Work focused on volatiles in the Costa Rica outer forearc and forearc is limited. Carbon, O, and H isotope compositions of offshore cold seeps have been interpreted as recording a sedimentary-derived fluid component from the subducting slab (Hensen et al., 2004; Füri et al., 2010). Recent carbon isotope work on outer forearc and forearc springs also suggests a deeply derived (mantle + slab) component (Barry et al., 2019). Nitrogen isotope work on gases from forearc springs records δ^{15}N values of −4.4‰ to +1.6‰, similar to mantle-like values from arc springs (−3.0‰ to +1.9‰), which are interpreted to reflect subduction erosion of accreted ophiolites from the upper plate with minimal input from subducted sediments (Lee et al., 2017).

Overall, prior work is consistent with the Li isotope geochemistry across the Costa Rica margin. Outer forearc springs have a component of Li sourced from sedimentary pore fluids. With increasing subduction, dehydration of sediments and/or AOC contributes to forearc fluids, consistent with carbon and nitrogen isotope work. The δ^7Li values of the arc springs predominantly record a mantle signature with minimal slab contribution, consistent with the compositions of arc lavas and gas emissions.

CONCLUSIONS

Li isotope ratios vary systematically across the Costa Rica convergent margin and correlate with ^{3}He/^{4}He values. Poor correlation of δ^7Li values with host lithology, temperature, or water chemistry argues against a major fractionation control. Li in outer forearc and forearc fluids is instead derived from dewatering of evolving subducted slab components, and little slab contribution is preserved in arc samples, which are dominated by Li from the mantle wedge. Li isotopes are thus an effective tracer of slab fluid source in appropriate samples. Additionally, forearc spring emissions are a potentially significant output of volatiles fluxes through the subduction zone, which is commonly overlooked.

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