Terra Nova

doi: 10.1111/j.1365-3121.2012.01058.x

Silicic magma petrogenesis in Iceland by remelting of hydrothermally altered crust based on oxygen isotope diversity and disequilibria between zircon and magma with implications for MORB

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ABSTRACT

Petrogenesis of silicic magmas in Iceland has fundamental significance for understanding the relative importance of fractional crystallization of mantle-derived basalt and partial melting of hydrothermally altered basaltic crust in formation of the earliest continental crust. First results of *in situ* oxygen isotope investigation of zircons in large-volume silicic eruptive products of three volcanoes in Iceland (Askja, Torfajökull, and Hekla) demonstrate isotope diversity and disequilibria and long U–Th zircon pre-eruptive residence of 10^3–10^4 year. This suggests that zircons did not grow from their host melts but instead were inherited from older magma batches and leftover cumulates with generally low and variable $\delta^{18}O$ values. This study demonstrates that segregation of cubic kilometres of silicic magma is faster than mineral-diffusive or recrystallization time-scales (estimated at $\sim 10^3$ years), and it suggests that partial melting of hydrothermally altered and oxidized oceanic crust is the mechanism that best explains silicic rocks in Iceland and early earth environments.

Terra Nova, 00, 1–6, 2012

Introduction

Petrogenesis of silicic magmas at mid-ocean ridges has fundamental significance in understanding the origin of the first continental crust (e.g. McBirney, 2006). This topic is the subject of a century-long debate between proponents of origins by fractional crystallization of basaltic magma (Bowen, 1928; Marsh, 2006; Namur *et al*., 2011) and partial melting of basaltic rocks (e.g. Gillis and Coogan, 2002; Jonasson, 2007; Wanless *et al*., 2010).

Iceland represents the only subaerial portion of the mid-ocean ridge that is also influenced by a mantle plume, resulting in abnormally thick crust. The differentiation of the early silicic (continental) crust on Earth possibly happened in environments similar to Iceland, with thickened mafic crust and high heat fluxes and magma production rates (e.g. Rapp *et al*., 1991; Condie and Pease, 2008; Martin *et al*., 2008). The proportion of silicic rocks in Iceland is 5–10% (Walker, 1966; Jonasson, 2007) and this proportion is higher by approximately a factor of two than is possible to generate and extract by pure fractional crystallization of tholeiitic basalt (e.g. McBirney, 2006).

An important feature of Icelandic crust, especially in its rift zones, is that it is pervasively hydrothermally altered by rain, snow and glacial waters and thus has low-$\delta^{18}O$ values of −12 to +4‰ (Hattori and Muehlenbachs, 1982; Gautason and Muehlenbachs, 1998). This hydrothermal metamorphism has led to the widespread abundance of low-$\delta^{18}O$ hydrothermally altered rocks and magmas (Muehlenbachs *et al*., 1974; Bindeman *et al*., 2008). As oxygen is the most abundant element in the crust, mantle and hydrosphere, variation of its isotopic composition provides robust constraints on mass balances in magma genesis.

If silicic rocks in Iceland were solely derived by fractional crystallization of basalt, then low-$\delta^{18}O$ values in them must reflect low-$\delta^{18}O$ values of the parental basalt and the Iceland plume. Although primitive magnesian olivines in basalts from some mantle plumes display moderately low-$\delta^{18}O$ values (~+4.7‰; Eiler *et al*.,1997), we do not share the opinion of Thirlwall *et al*. (2006) that Icelandic plume is depleted to a greater extent to explain lower $\delta^{18}O$ values, and large isotopic diversity in both basaltic and silicic magmas. A critical test in advocating for shallow, crustal origin of low-$\delta^{18}O$ values in the majority of Icelandic magmas comes from investigation of their crystalline cargo.

Single-crystal oxygen isotope analysis, including ion microprobe analysis of zircons, has resulted in discoveries of tremendous diversity in many volcanic units around the world (e.g. Bindeman, 2008; Watts *et al*., 2011 and references therein). This is particularly true for environments that are fingerprinted by low-$\delta^{18}O$ values such as Iceland or Kamchatka, which make recognition of the zircon heritage resolvable. These low-$\delta^{18}O$ magmas are produced by partial or complete melting or assimilation of hydrothermally preconditioned low-$\delta^{18}O$ crust, providing clear test of crustal melting vs. fractional crystallization.

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Zircon, a prime mineral for U–Pb geochronology, will retain $\delta^{18}O$ values of its host magmas (e.g. Valley *et al.*, 2005). Often, zircons with diverse $\delta^{18}O$ are found in ordinary-looking units showing equilibrium $\Delta^{18}O$ isotope fractionation values between other phenocrysts and their host melt (Watts *et al.*, 2011). Zircons thus serve as excellent probes into the partial melting process of hydrothermally altered rocks (variable in $\delta^{18}O$) and subsequent magma assembly processes. The presence of heterogeneous zircon populations can also serve as a chronometer of magma assembly time-scales, provided that the rates of diffusion or solution-reprecipitation are established (e.g. Bindeman, 2008). Finally, fractionation of oxygen isotopes between melt and zircon, and melt and olivine, are very similar to each other. Equilibrium mantle-derived zircons and olivines have nearly identical 5.3 ± 0.3‰ and 5.1 ± 0.3‰ values (Valley *et al.*, 2005), higher and lower values define a different source: high-$\delta^{18}O$ zircon indicates a 'supracrustal', metasedimentary source, or a source that interacted with seawater at low temperature (Cavosie *et al.*, 2011), whereas low-$\delta^{18}O$ zircon indicates derivation from a hydrothermally altered source altered by meteoric (rain and snow) or seawater at high temperature (Bindeman, 2008).

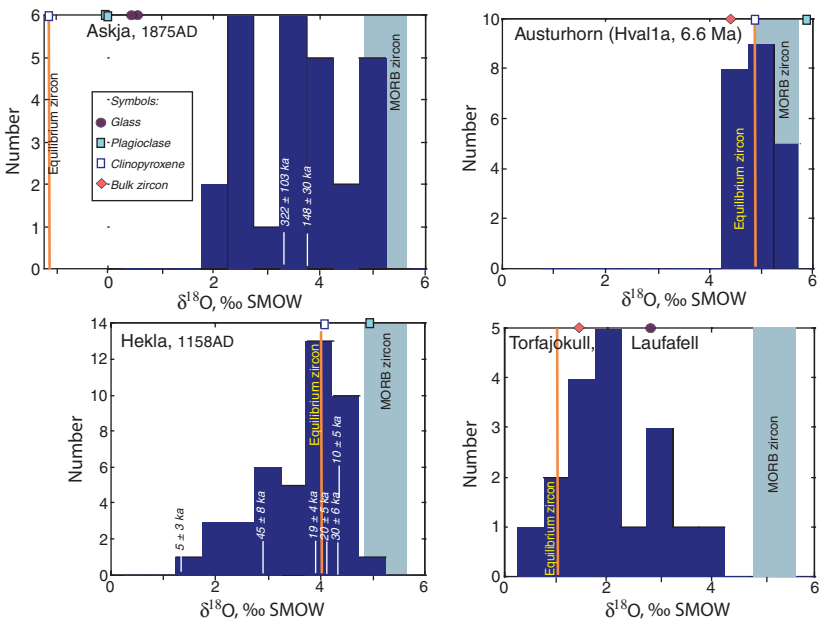

Fig. 1 Histograms of $\delta^{18}O$ values of zircon analyses for four investigated silicic units. Vertical line denotes estimated equilibrium $\delta^{18}O$ zircon value, based on its exchange equilibria with silicic melt ($\Delta^{18}O_{rhyolite\text{-}zircon}$ = 1.8 ± 0.3‰ at 850 ± 50 °C, see Bindeman, 2008; Trail *et al.*, 2009). The melt value is estimated from glass or plagioclase phenocrysts in equilibrium with it. Notice that diverse zircons plot between MORB and equilibrium zircon values displaying different kurtosis (skewness) of the histogram relative to the expected equilibrium zircon values: left lateral (Hekla) suggesting inheritance of a few low $\delta^{18}O$ zircons by a moderately $\delta^{18}O$ depleted magma, right-lateral (Torfajokull) suggesting that a majority of zircons are near equilibrium with several higher $\delta^{18}O$ zircons remaining, and diverse, poly-modal and significantly higher than equilibrium (Askja) suggesting that melt transformation to low-$\delta^{18}O$ was rapid enough, leaving behind many zircons, and/or that assimilated low-$\delta^{18}O$ rocks of $\sim<2$ permil contributed much oxygen by mass, but did not supply zircons (e.g. they were zircon-undersaturated). Vertical lines are U–Th disequilibria ages for specific zircons; see Carley *et al.* (2011) for ages for rocks from different units.

Samples and methods

In this work we investigated in detail four well-known, zircon-bearing, large-volume silicic units of Iceland (Figs 1 and 2; Table S1): 1875 AD eruption of Askja (MacDonald *et al.*, 1987), 1158 AD eruption of Hekla (Sigmarsson *et al.*, 1992), sub-glacial Pleistocene Laufafell eruption of Torfajökull (Gunnarson *et al.*, 1998; Martin and Sigmarsson, 2007, 2010), and 6.6 Ma silicic Austurhorn intrusion (sample Hval1a, Walker, 1966, Furman *et al.*, 1992; Martin and Sigmarsson, 2010; Martin *et al.*, 2011). Descriptions of these units are provided in the supplementary electronic file and respective papers. Analysis of oxygen isotopes by ims 1270 ion microprobe used a 25 μm beam, targeting zircon cores to find inheritance. Zircon rims were too thin (~5–15 μm) to be analysed individually by the ~25 μm beam used in this study, but some analyses approach the $\delta^{18}O_{rim}$ value based on estimated equilibrium values. Errors on standards, mounted and analysed in blocks with the unknowns, range from 0.1 to 0.35‰ (1 SD). Analytical details and a data table are provided in the Supporting Information. We have also analysed glasses and major phenocryst phases by laser fluorination to determine the $\delta^{18}O$ value of the host glass, and also to check if these other phenocrysts are in isotope disequilibrium with zircons at magmatic temperatures. The results of these analyses are also presented in the Table S1 and are shown on Figs 1 and 2.

Results

The main result of this study demonstrates that zircons from the three studied volcanic units have $\delta^{18}O$ values that are both higher and lower than the expected equilibrium $\delta^{18}O_{zircon}$ values and contain zircon cores that span >2–3‰ in $\delta^{18}O$ (Fig. 1). In contrast, a plutonic granophyre unit from Austurhorn has a homogeneous zircon population that plots tightly around MORB-like zircon values, in equilibrium with the estimated melt.

The degree and the character of disequilibrium ranges dramatically: cores of Askja zircons are diverse (2.2–5.1‰) and much higher in $\delta^{18}O$ than the estimated −1‰ equilibrium zircon rim values. Zircons in Torfajökull have cores mostly higher in $\delta^{18}O$ (up to 4‰) than the equilibrium, +1‰ $\delta^{18}O$ zircon value, but have a few cores plotting at equilibrium to slightly lower values, at ~+0.5‰. Hekla zircons mostly cluster around equilibrium $\delta^{18}O$ values at 4‰, with

a prominent low-$\delta^{18}O$ zircon tail, reaching +1.5‰ values likely indicating inheritance of low-$\delta^{18}O$ zircons from hydrothermally altered carapace. No zircons with $\delta^{18}O$ higher than MORB were found among 111 analyses, but all units contain zircons that approach MORB-like $\delta^{18}O$ values (5.3‰; Grimes *et al.*, 2011).

The U–Th disequilibria ages of zircon from historic and recent Icelandic eruptions investigated in this work, including Askja and Hekla samples, were recently reported by Carley *et al.* (2011). These authors demonstrated that (1) Icelandic zircons predate the age of the eruption by 10^3–10^4 years, with a few analyses approaching 10^5 years; (2) zircons often have lower Ti-in-zircon temperatures than the estimated temperature of the host magma; (3) trace elemental concentrations and ratios, as well as morphology and cathodoluminescence zoning patterns, often indicate entrained crystal cargo rather than an equilibrium crystallizing assemblage. Askja and Hekla zircons were particularly U and Th poor (tens of p.p.m.) and we were able to obtain meaningful U–Th ages for only eight zircons studied herein, shown by arrows on Fig. 2. Not only do these zircons significantly predate the age of the eruption, they also display a range in $\delta^{18}O$ values, suggesting that the source rocks varied in both age and level of $\delta^{18}O$ depletion. No old (> *c.* 0.35 Ma) zircons in secular equilibrium were found here, nor in the dataset of Carley *et al.* (2011), suggesting that magmatic zircon recycling taps young crust in rift zones, subject of further investigation.

Discussion

Diversity of zircon and olivine in Iceland

The *in situ* investigation of $\delta^{18}O$ of Icelandic zircons stems from our earlier *in situ* effort that demonstrated widespread oxygen isotope disequilibrium of olivine in nearly all large-volume basaltic eruptions of Iceland (Bindeman *et al.*, 2008). Due to very sluggish oxygen diffusion, both minerals will retain initial $\delta^{18}O$ values during inheritance and recycling processes. The persistent disequilibrium indicates that time-scales since inheritance were not sufficient to equilibrate oxygen isotopes in zircons by diffusion and/or solution–reprecipitation, but sufficiently long to equilibrate in all other phenocrysts, which appear to be in high-*T* isotope equilibrium (Fig. 1). Given temperatures of their parental magmas (~800–900 °C) estimated using Zr saturation and Ti-in-zircon thermometries (Carley *et al.*, 2011), isotope equilibrating would take ~10^3–10^4 years by analogy with Yellowstone and other rhyolitic units with similar temperatures (e.g. Bindeman, 2008, see Appendix S1 for further discussion of estimating zircon residence time-scales). Preservation of $\delta^{18}O$ diversity in zircons while other magma products are more or less homogenous also means that magma segregation, assembly, mixing and eruption time-scale are faster than zircon isotope equilibration, making zircon a potential chronometer to investigate these processes.

It now appears that Icelandic zircons add to a rapidly growing dataset for zircons analysed *in situ* from a variety of magmatic systems worldwide that demonstrate zircon diversity and disequilibrium: Yellowstone hot spot track, Timber Mt Oasis Valley Caldera complex, Nevada, and Kamchatka (Bindeman, 2008; Watts *et al.*, 2011; I. Bindeman, T. Carley and C. Miller, unpublished data). This zircon diversity is due to remelting of crust that is heterogeneous in $\delta^{18}O$, followed by batch assembly of isotopically diverse bulk or partial melts.

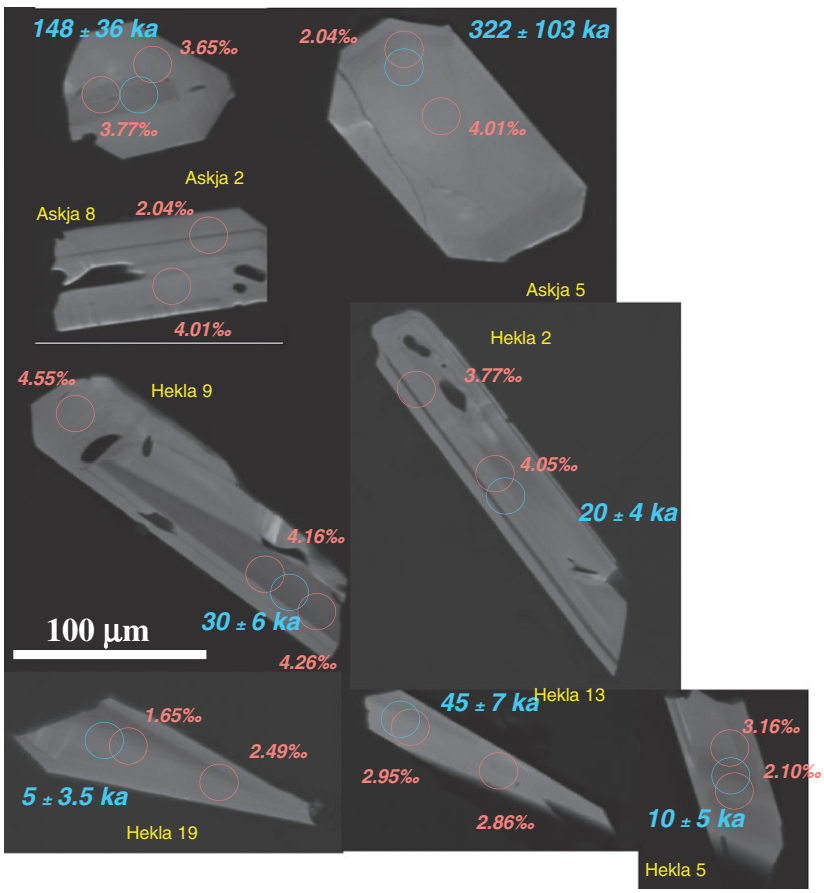

Fig. 2 Cathodoluminescence images of analysed zircons showing dull indistinct patterns for zircons of different age and $\delta^{18}O$ values.

Iceland zircons vs. MORB zircon

Zircons in Iceland display low-$\delta^{18}O$ values and heterogeneity, in contrast to the majority of submarine MORB zircons, which are more homogeneous with mantle-like $\delta^{18}O$ values (Grimes *et al.*, 2011). This result may not reflect differences in petrogenesis, but rather differences between isotopically fingerprinted low-$\delta^{18}O$ crust, and

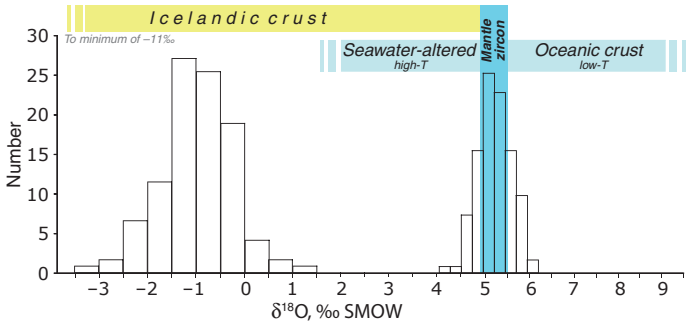

Fig. 3 The $\delta^{18}O$ of zircons crystallized from a hybrid melt after a normal-$\delta^{18}O$ mantle-derived MORB magma (5.7‰, in equilibrium with 5.3 ± 0.3‰ zircons; Valley *et al.*, 2005) digested hydrothermally altered crust. The envisioned process involves small increment addition calculated using: $xM + (1 - x)A$, where M, A, and x are the $\delta^{18}O$ values of magma and assimilant, and % of assimilant added. This equation was put in a Monte-Carlo simulator and yielded histograms shown. It is assumed that the normal $\delta^{18}O$ magma is only capable of assimilating a maximum of 50% of the crust held at near solidus temperature, before becoming fully crystalline. Silicic magmas generated by this process average out large volumes of the crust and mix with mafic magma differentiates that caused melting in the first place. It is also possible, although less likely, that oxygen exchange occurred by a magmatic flow in sub-Icelandic magma plumbing systems. Again, such flow will cross-cut variable in $\delta^{18}O$ rocks and is random in nature. The dominant range for submarine-altered oceanic crust is taken here as from +2 to +9‰, based on survey of available DSDP core data, see also Fig. 4; more extreme values from ~0‰ to +20‰ are characteristic for vanishingly small amounts of rocks. The lower $\delta^{18}O$ range in subaerially altered Icelandic crust is shown based on 100% exchange with −14‰ waters. Such low values are observed in drill-cores (e.g. Hattori and Muehlenbachs, 1982), and in our analyses of 12 hydrothermally altered xenoliths from deposits of the Askja 1875 caldera forming eruption. The graph demonstrates that up to 50% of bulk assimilation of oceanic crust does not lead to significant 'heterogenization' nor lowering of $\delta^{18}O$ in the silicic partial melt, although similar processes in isotopically depleted Icelandic crust lead to a significant diversity of $\delta^{18}O_{magma}$ and $\delta^{18}O_{zircon}$ values.

crust altered by seawater (or not altered at all), with much less extreme range in $\delta^{18}O$ values in assimilants, both higher and lower than the mantle-like values (Fig. 3).

The majority of oceanic crust altered by seawater has ~7–9‰ range of $\delta^{18}O_{WR}$ values (+2 to +9 – +11‰, e.g. Alt *et al.*, 1986; Grimes *et al.*, 2011), plotting on both sides of the canonical mantle 5.7‰ value (Figs 3 and 4). Submarine oceanic crust is high in $\delta^{18}O$ (~+6 to +9‰) in its upper portions (pillow basalts, sheeted dike complexes) due to low-T (< 300 °C) exchange with seawater, and low-$\delta^{18}O$ (~+2 to +4‰) in its middle and lower part, where high-temperature hydrothermal alteration leaches out ^{18}O from rocks (Gregory and Taylor, 1981). Because of the 0 ± 1‰ $\Delta^{18}O_{rock\text{-}water}$ isotope fractionation at high, > 500 °C temperature, oceanic crust is never altered to values lower than 0 to +2‰ SMOW. A much larger, ~17‰ range of $\delta^{18}O$ values of the Icelandic crust (5.7 to −11‰; Figs 3 and 4) is exclusively lower than MORB values, meaning that any assimilation process will shift mantle magma to more negative values. Normal $\delta^{18}O$ silicic rock in Iceland is almost certainly a product of normal closed system fractional crystallization of a normal $\delta^{18}O$ mantle-derived basalt (e.g. Martin and Sigmarsson, 2007), but some authors (e.g. Thirlwall *et al.*, 2006) favour depleted (~4.5–4.8‰) values for the Icelandic plume. However, these minor variations have no significant effect on our conclusions below.

The ~1‰ variation around mantle-like-$\delta^{18}O$ values recorded by MORB zircons is interpreted by Grimes *et al.* (2011) to represent predominance of fractional crystallization (with variability due to effects of differences in temperature and extent of crystallization). We suggest that partial melting may also explain these results, and demonstrate on Fig. 3 that the mutually cancelling effects of both low and high-$\delta^{18}O$ assimilants result in scattering around mantle-like values. As zircon saturates only in evolved melts, and relatively later in the basaltic crystallization/assimilation processes (cf. Watson and Harrison, 1983), the $\delta^{18}O_{zircon}$ values of final melt reflect time- and $\delta^{18}O$-averaged prior assimilant additions to the original basaltic magma. Figure 3 compares distribution of $\delta^{18}O_{zircon}$ values for Icelandic crust and submarine MORB, demonstrating that the $\delta^{18}O$ values of Icelandic $\delta^{18}O$ zircons are low and the variance is large, while MORB zircons form a normal distribution around canonical mantle mean with only 1‰ standard deviation variations. Likewise, mantle-like $\delta^{18}O$ values of the best global examples of adakites (Bindeman *et al.*, 2005), which represent partial melts of subducted crust that is diverse in $\delta^{18}O$ (high-$\delta^{18}O$ top, and low-$\delta^{18}O$ bottom; Fig. 4), are interpreted to represent mixing of incremental melts that averages out the differences. Published analyses of $\delta^{18}O$ in zircon from Archaean TTG suites also demonstrate scatter around normal-$\delta^{18}O$ MORB field (Bindeman *et al.*, 2005; Valley *et al.*, 2005, and references therein) and these zircons were clearly not solely derived by fractional crystallization of mantle magmas. Remelting of the hydrothermally altered (and thus diverse in $\delta^{18}O$) mafic crust was perhaps the leading process of silicic magma generation in the Archaean (Martin and Moyen 2002), the partial melt averaging process shown on Fig. 3 is likely responsible for this $\delta^{18}O$ homogeneity.

Partial melting of hydrated crust in Iceland, MORB and on early Earth

This first *in situ* $\delta^{18}O$ dataset for 111 spot analyses of Icelandic zircons found no zircons with values higher than MORB, suggesting lack of high-$\delta^{18}O$ 'supracrustal' slivers of old continental crust contributed to the petrogenesis of the studied silicic rocks of Iceland. We note the persistent presence of zircons with mantle-like $\delta^{18}O$ in almost all studied units. Along with persistent presence of olivines with mantle-like $\delta^{18}O$ in low-$\delta^{18}O$ large volume basalts of Iceland (Bindeman *et al.*, 2008), these provide

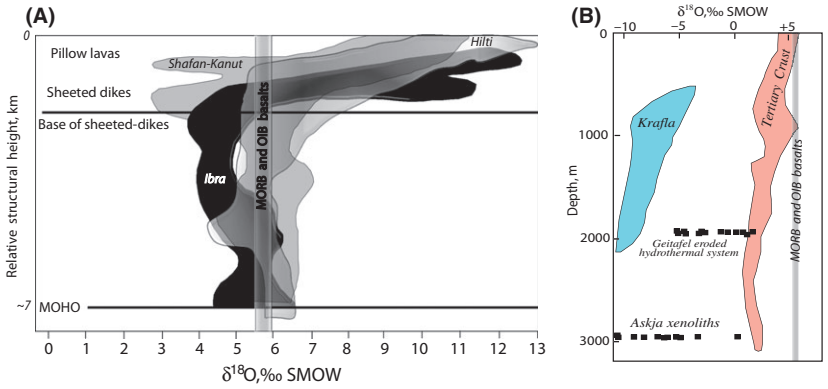

Fig. 4 (A) The $\delta^{18}O$ profile through the oceanic crust based on three profiles from Gregory and Taylor (1981) and Stakes and Taylor (1992) measured in Oman ophiolite; shown relative stratigraphic height of the base of sheeted dike complex is adjusted based on VanTongeren *et al.* (2008) and assuming realistic 1 : 3 extrusive/intrusive ratio. Notice that each individual profile is different and diverse with respect to $\delta^{18}O$, including high and low $\delta^{18}O$ portions, and lower 75% of profiles are not universally low in $\delta^{18}O$ as is commonly assumed. The $\delta^{18}O$ value of the magma generated by partial melting or exchange in the lower 75% of the oceanic crust is expected to be random around MORB, as is shown in Fig. 3. (B) Oxygen isotope profile through the top portion of Icelandic crust based on drillcore data (Hattori and Muehlenbachs, 1982) and our shallow xenoliths data from Askja shown at ~3 km depth based on the inferred depth of magma chamber (Pagli *et al.* 2006), and our data on hydrothermally altered rocks from 5 to 6 Ma Geitafell volcano in eastern Iceland, shown at ~2 km erosion level (Fridleifsson, 1983). See Appendix S1 for analyses.

material evidence that the Icelandic plume is not universally low in $\delta^{18}O$.

Our data lend new support to interpretation that the genesis of silicic rocks predominantly involves partial melting of crust that is diverse in $\delta^{18}O$, age, relatively evolved and zircon-bearing, hydrothermally altered in shallow (rift) environments, thus generally low-$\delta^{18}O$ (Fig. 4B), hydrated and oxidized. Because of the hydration and oxidation during hydrothermal metamorphism, we envision that a partial melting process will proceed with significant retention of Fe-oxides in the residue, thus making a greater proportion of silicic rocks (Fig. A2 in Appendix S1). This result is qualitatively similar to 'calc-alkaline' trend vs. tholeiitic trends, explaining the greater proportion of silicic rocks in arcs, as compared to normal mid-ocean ridges (e.g. McBirney, 2006). Thus, we consider hydrothermal metamorphism as a key to understanding the genesis of silicic rocks in Iceland and on early earth.

Acknowledgements

Supported by NSF EAR grants EAR-CAREER 0844772, EAR0911093, CNRS-CRPG (contribution 2157), and Icelandic Science Foundation. We thank Gerhard Woerner, Alfred Kroener, and two anonymous reviewers, for excellent comments, which helped us to improve the manuscript.

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Received 5 October 2011; revised version accepted 9 January 2012

Supporting Information

Additional Supporting Information may be found in the online version of this article:

Appendix S1. Supporting Material.

Table S1. Oxygen isotope analyses of zircons from Iceland by ion microprobe and laser fluorination analyses of glass and major phenocryst phases by laser fluorination.

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