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Survival of lithium isotopic heterogeneities in the mantle supported by HIMU-lavas from Rurutu Island, Austral Chain

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ABSTRACT

Thirty years ago, Hofmann and Hart [Hofmann, A.W., Hart, S.R., 1978. An assessment of local and regional isotopic equilibrium in the mantle, Earth Planet Sci. Lett. 38, 44–62] showed that local disequilibria of slowly diffusing radiogenic tracers (e.g. Sr) are preserved in the mantle over 1–2 Ga time scales. Recently, it was suggested that this is not the case for fast diffusing elements such as lithium [Halama, R., McDonough, W.F., Rudnick, R.L., Bell, K., 2008. Tracking the lithium isotopic evolution of the mantle using carbonatites, Earth Planet Sci. Lett. 265, 726–742], thus questioning the ability of lithium isotopes to constrain long-term effects of recycling of material with crustal signatures. A key issue in this debate is the identification in hotspot volcanism of Li isotopic fingerprint consistent with recycling. Previous studies proposed that HIMU type volcanism, which is thought to sample mantle domains that include subducted altered oceanic crust, has $^{7}Li/^{6}Li$ distinctively higher than the fresh mid-ocean ridge basalts.

This work focuses on Rurutu island, where both HIMU ($20.88<^{206}Pb/^{204}Pb<21.42$) and non-HIMU ($19.11<^{206}Pb/^{204}Pb<20.45$) lavas occur. When considering only the freshest and most primitive lavas, the lithium isotopic signatures of HIMU ($+5.4<\delta^{7}Li<+7.9‰$) and non-HIMU ($+2.9<\delta^{7}Li<+4.8‰$) lavas do not overlap, thus supporting the idea that HIMU mantle has distinctly elevated $\delta^{7}Li$. This result suggests that Li isotopic heterogeneities could survive diffusion over 1–2 billion years, the amount of time required to develop the highly radiogenic Pb signature of HIMU-lavas.

Modeling lithium diffusion out of a lithium-rich, isotopically heavy altered oceanic crust reveals that isotopic disequilibrium persists over long time periods in comparison with the rapid decrease of chemical disequilibrium. For instance, a kilometer-thick altered oceanic crust loses most of its Li excess in a matter of a few tens of millions of years but could preserve $^{7}Li/^{6}Li$ distinctively higher than the ambient mantle over a time period in excess of 1.5 Ga. After 1.5 Ga, a kilometer-thick altered oceanic crust has heavy but uniform $^{7}Li/^{6}Li$, while large isotopic variations persist in the nearby mantle (<20 km). Thus, decoupling between Li and slowly diffusing radiogenic isotopes can be predicted in the mantle nearby subducted crust.

1. Introduction

Until very recently, lithium stable isotopes were considered as an ideal tracer of recycling processes in the mantle, because of the large variations (tens of permil) in their relative abundance in surface environments (e.g. Elliott et al., 2004), but this view was recently challenged for a number of reasons. First, the precise mantle signature is accessible only after accounting for the modification processes during magma storage and segregation, such as contamination (Tomascak et al., 2008; Chan et al., 2009), and diffusion-driven fractionation (Richter et al., 2003; Seitz et al., 2004; Lundstrom et al., 2005; Jeffcoate et al., 2007; Parkinson et al., 2007; Ionov and Seitz, 2008). Second, Li isotopes rarely correlate with long-lived radiogenic isotopes that are commonly used to identify mantle heterogeneities. Third, the range of isotopic variations thought to reflect mantle heterogeneity is unexpectingly narrow (a few permil) (e.g. Tomascak et al., 2008). The cause of these small variations has been ascribed to a) small compositional contrast between the average input at subduction zones and mantle (Marschall et al., 2007), b) isotopic homogenization during melting (Ryan and Kyle, 2004) and/or c) fast diffusion of lithium in the asthenospheric mantle (Halama et al., 2008).

On the other hand, some observations support the existence of small, but real Li isotopic variations in the mantle. Along mid-oceanic ridges,

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Li–Sr–Nd isotope co-variations were identified locally (Elliott et al., 2006; Simons et al., 2008). Among the ocean islands with a strong HIMU affinity (St Helena, Mangaia, Rurutu, Raivavae and Tubuai), lithium compositions are distinctly heavier ($+5.0>\delta^7Li>+7.4‰$) than those of the fresh mid-ocean ridge basalts ($+2.5<\delta^7Li<+5.5‰$). This observation possibly reflects the existence of mantle domains with distinctly heavy lithium compositions (Ryan and Kyle, 2004; Nishio et al., 2005; Chan et al., 2009). However, it is difficult to evaluate whether the heavy signatures represent genuine mantle source values or values modified by near surface petrogenetic processes, primarily because of the small number of samples analyzed for each locality.

The HIMU mantle end-member is an ideal target to test if lithium isotopes can constrain recycling processes. HIMU is a relatively pure mantle component probably derived from recycling of altered oceanic crust free of sediments known to have a highly heterogeneous Li signature (Chan et al., 2006). Thus, if the subduction processes do not significantly modify the Li isotopic composition of altered oceanic crust (Marschall et al., 2007), heavy lithium composition is expected in the HIMU component. In addition, the highly radiogenic Pb signature of HIMU requires storage in the mantle over a period of time exceeding 1.5 Ga (Chauvel et al., 1992). The preservation of heavy Li signature in HIMU mantle would indicate that isotopic heterogeneities in Li could survive diffusion over time scales much longer than those proposed by Halama et al. (2008).

Chan et al. (2009), who recently investigated in detail Li isotopic variations in Cook–Austral islands (Raivavae, Rapa, Mangai, and Tubuai), concluded that the isotopic composition of the HIMU mantle is lighter than the previously proposed composition, but still distinctly heavier than the non-HIMU mantle. The present study focuses on Rurutu Island, where both moderately radiogenic Pb and typical HIMU signatures have been identified (Chauvel et al., 1997). Having distinguished secondary processes from the mantle signature, we confirm that high $^7Li/^6Li$ is a robust feature of the HIMU mantle component and, based on diffusion modeling, we discuss how Li isotopic heterogeneities can be preserved in the mantle over billion-year time scales.

2. Geologic, petrologic and geochemical background

Rurutu island belongs to the Austral–Cook volcanic chain, which extends over 2000 km in the south Pacific Ocean, from the Macdonald seamount to the Palmerson atoll (Fig. 1a). As first shown by Duncan and McDougall (1976), Rurutu was built during two main volcanic stages. The old lavas erupted ca. 12 Ma ago, first in submarine then in subaerial environments. The young lavas emplaced about 11 Ma later on top of the carbonate sediments that were deposited during the long period of quiescence. The old volcanism is generally associated with the hotspot track linking the active Mcdonald seamount to the 19 Ma old Mangaia island (Diraison, 1991; Chauvel et al., 1997), although a connection with a northern track extending from Raivavae (6.5 Ma) to Tubuai (9 Ma) and Rurutu islands has been recently suggested (Bonneville et al., 2006). The young Rurutu volcanism belongs to the hotspot track that extends from the 8 Ma old Atiu Island to the recently discovered, 0.2 Ma old Arago seamount, 130 km southeast of Rurutu island (Bonneville et al., 2002).

Rurutu is made of aphyric (young volcanism) to olivine–clinopyroxene–plagioclase phyric lavas (old volcanism) (Guille et al., 1998). The old lavas consist mainly of alkali basalts whereas the young lavas are more differentiated, with composition ranging from basanite to hawaiite. As Mangaia and Tubuai volcanics, the old Rurutu lavas have a typical HIMU isotopic fingerprint ($^{206}Pb/^{204}Pb$ from 20.88 to 21.42; $^{87}Sr/^{86}Sr$ from 0.702761 to 0.703003 and $^{143}Nd/^{144}Nd$ from 0.512852 to 0.512906) (Chauvel et al., 1997). The young lavas show much less HIMU affinity (with $^{206}Pb/^{204}Pb$ ranging from 19.11 to 20.45), possibly reflecting contamination of plume melts within the lithosphere (Chauvel et al., 1997) or at shallower level. Previous Li isotopic analyses of Rurutu include one sample (RRT-C10)from the old lava group with a composition ($\delta^7Li=+5.4‰$) similar to Tubuai ($+4.9<\delta^7Li<+5.5‰$), but lighter than Mangaia ($+5.5<\delta^7Li<+7.8‰$) compositions (Nishio et al., 2005; Chan et al., 2009).

3. Samples

Samples listed on Table 1 were selected on the basis of freshness by previous geochemical and geochronological studies after inspection of thin sections and lost on ignition (LOI) measurements (Duncan and McDougall, 1976; Dupuy et al., 1988, 1989; Diraison, 1991; Chauvel et al., 1997; Guille et al., 1998).With the exception of samples from Dupuy et al. (1988, 1989), sampling locations are known (Fig. 1b). Miocene samples have slightly higher LOI (0.8 to 2.2% except for one sample with 2.9%) compared to the Pleistocene samples (LOI from −0.2 to 0.7% except for one sample with 1.6%). The negative LOI of sample 74-388, which is ascribed to oxidation of iron, is indicative of freshness. Guille et al. (1998) described traces of palagonitic alteration in the oldest basaltic samples, consistently with their emplacement in submarine environment. Using X-ray diffraction and scanning electron

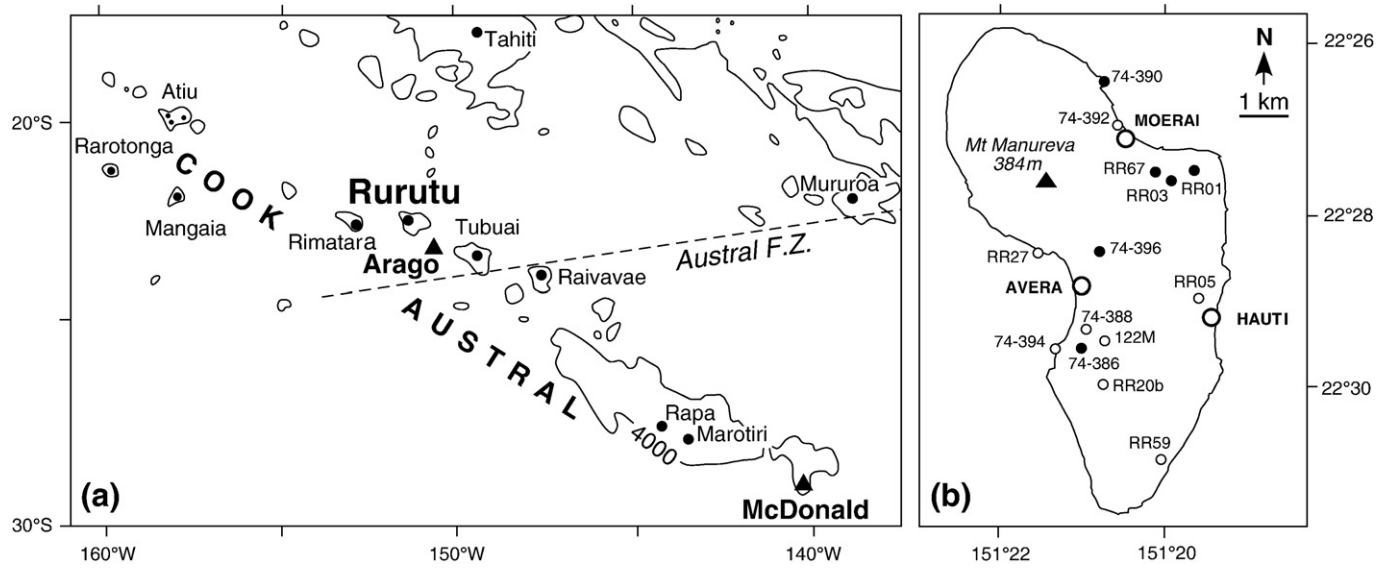

Fig. 1. Location and map of Rurutu island. (a) The 4000-m depth contour as well as major islands (circles) and seamounts cited in text (triangles) are shown. (b) Location of samples is shown, when known (Duncan, 1975; Duncan and McDougall, 1976; Diraison, 1991; Guille et al., 1998).