

# Oxygen isotopes in Samoan lavas: Confirmation of continent recycling

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## ABSTRACT

Lavas from the Samoan volcanic chain show the most enriched geochemical signatures ever documented in oceanic lavas ( $^{87}\text{Sr}/^{86}\text{Sr}$  as high as 0.7205). In order to test the hypothesis that their source contains a component of recycled upper continental crust, we measured oxygen isotope compositions of olivine phenocrysts from these lavas. Correlations between  $\delta^{18}\text{O}$  of olivines (5.11‰–5.70‰) and  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{207}\text{Pb}/^{204}\text{Pb}$  of whole rocks, as well as Ce/Pb and Nb/Th ratios of whole rocks, indicate that (1) measured  $\delta^{18}\text{O}$  are primary, mantle-derived values, and (2) the enriched mantle source of these lavas contains continental crust or its derivative sediments. The observed trend between  $\delta^{18}\text{O}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  can be fit using either clastic marine sediment or continental crust values of  $\delta^{18}\text{O}$ , Sr concentration, and  $^{87}\text{Sr}/^{86}\text{Sr}$ , but only those for clastic marine sediments are compatible with trace element modeling. We conclude that the enriched source for Samoan basalts was created by sedimentation of continent-derived material into a marine environment, followed by subduction and mixing with ambient mantle.

**Keywords:** oxygen isotopes, Samoa, enriched mantle, EM2, ocean island basalts.

## INTRODUCTION

The Samoan hotspot chain, located in the central South Pacific Ocean basin, is particularly significant to mantle geochemistry, as basaltic lavas from there have the highest (most radiogenic)  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of all oceanic lavas documented to date (as high as 0.7205; Jackson et al., 2007a). These exceptionally high  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios define the Enriched Mantle 2 (EM2) chemical component of Earth's mantle (Zindler and Hart, 1986). Lavas having radiogenic isotope compositions approaching the EM2 end member are also relatively common in the Society Islands (White and Duncan, 1996) and are found more rarely and with less extreme  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in other island chains, including the Marquesas (Castillo et al., 2007) and the Azores (Widom and Farquhar, 2003). Hence, even though EM2 is found in its most extreme form in only one island chain (Samoa), it may be a widespread component of the mantle.

In this study we examine whether there is a component of recycled crust or sediment in the mantle source of Samoan lavas. This question, generalized to all enriched ocean island basalts (OIBs), has plagued mantle geochemists since the first geochemical studies on hotspot lavas (note that enriched is used here to describe OIBs with  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios higher than bulk silicate earth and incompatible trace element abundances much higher than in mid-oceanic ridge basalts [MORBs]). Trace element and isotopic enrichment in OIBs was originally interpreted as evidence for the presence of continental crust recycled to the mantle through subduction (cf. White, 1985). However, due to the inherent complexity of Earth's physical and chemical processes (resulting in many parameters in radiogenic isotope and trace element modeling), it is difficult to provide a unique solution to a given set of geochemical observations.

For example, Workman et al. (2004) suggested, as have others (cf. Zindler and Hart, 1986), that EM2 could form from intramantle metasomatism, based primarily on smooth trace element patterns of the enriched Samoan lavas. Jackson et al. (2007a), using newly acquired dredge samples of Samoan lavas that are even more enriched than those presented by Workman et al. (2004), successfully modeled the trace element compositions of end-member EM2 lavas (with  $^{87}\text{Sr}/^{86}\text{Sr}$  of 0.7205)

as melts from a mixture of slightly depleted mantle and 5%–6% upper continental crust. Here we test this newest crustal recycling model using oxygen isotope analyses of olivine phenocrysts from Samoan lavas.

Oxygen isotopes are useful as a tracer of crustal components in mantle-derived lavas because materials precipitated at or near Earth's surface, or that have undergone weathering at near-surface conditions, differ greatly in  $\delta^{18}\text{O}$  [ $\delta^{18}\text{O} = 1000 (^{18}\text{O}/^{16}\text{O}_{\text{sample}}/^{18}\text{O}/^{16}\text{O}_{\text{SMOW}}) - 1$ , where SMOW is standard mean ocean water at an  $^{18}\text{O}/^{16}\text{O}$  ratio of 0.0020052] from mantle materials as a result of low-temperature isotopic fractionations. Consequently, the range of  $\delta^{18}\text{O}$  in olivines from MORBs and upper mantle peridotites is small (5.2‰  $\pm$  0.2‰; Matthey et al., 1994; Eiler et al., 1997; Eiler, 2001), whereas  $\delta^{18}\text{O}$  values in various marine sediments and continental rocks can range from 0‰ to 40‰ (Eiler, 2001). Deviations from primary mantle values have been found in some OIBs and attributed to the addition of sedimentary or upper crustal material (in the case of high  $\delta^{18}\text{O}$ ) or lower oceanic crust (in the case of low  $\delta^{18}\text{O}$ ), either as magmatic contaminants sampled during passage through the crust or as recycled components in the mantle sources of these lavas (see review by Eiler, 2001).

A previous survey of the oxygen isotope compositions of OIBs, made using high-precision laser-fluorination techniques (Eiler et al., 1997), indicated that EM2 lavas are systematically higher in  $\delta^{18}\text{O}$  than other basalts and mantle peridotites. These data were interpreted as evidence for the presence of recycled crust in the sources of EM2 lavas, but the result is not conclusive because samples in that study are only moderately enriched in  $^{87}\text{Sr}/^{86}\text{Sr}$  (to 0.7066, far below the extreme EM2 composition). Furthermore, these samples are posterosional lavas of enigmatic origin. Here we reassess this observation by determining the  $\delta^{18}\text{O}$  values of a large, well-studied suite of Samoan shield-stage lavas having extremely enriched EM2 signatures.

## SAMPLES AND METHODS

Lavas analyzed in this study were previously characterized for major and trace element abundances and isotopic (Sr-Nd-Pb-He-Os) compositions (Workman et al., 2004; Jackson et al., 2007a, 2007b). Most are alkali basalts, with a few samples in the tholeiite field of MacDonald and Katsura (1964). Four samples are picrites with Mg#s [molar percent of  $\text{Mg}/(\text{Mg}+\text{Fe}^{2+})$ ] of  $\geq 80$  and MgO contents of  $\geq 23.9$  wt%. All lavas have Mg#s  $> 55$  with the exception of Upolu samples U14 and U19, which have Mg#s of 43 and 49, respectively. Olivine phenocryst abundances range from 3% to 40% by volume.

Sample names beginning with T, U, or the name "ofu" are subaerial lavas from the Samoan Islands; those beginning with numbers are dredged from Vailulu'u and Malumalu Seamounts or submarine portions of the islands Ta'u and Savai'i. The most enriched samples, in terms of  $^{87}\text{Sr}/^{86}\text{Sr}$ , were collected on a cruise in 2005 aboard the R/V *Kilo Moana* and are nicknamed SEUSS (superenriched underwater Savai'i samples; Jackson et al., 2007a).

Whole-rock samples were crushed, sieved to obtain a 300–660  $\mu\text{m}$  size fraction, then rinsed in acetone and deionized water. Olivine phenocrysts were hand picked under binocular microscope either dry or from a water bath, and chosen for the fewest visible melt inclusions, mineral inclusions, or surface discolorations.

Oxygen isotope compositions were determined by  $\text{CO}_2$  laser fluorination at the California Institute of Technology laboratory for stable isotope geochemistry, based on methods described by Valley et al. (1995) and Eiler et al. (2000). On each day, analyses of unknowns were accompanied by at least three analyses of the UWG-2 garnet standard (Valley et al.,

1995) and at least two analyses of the SCO-1 olivine standard (Eiler et al., 1995). On days when the UWG-2 standard averaged within 0.1‰ of the accepted value of 5.80‰, no systematic corrections were applied to the data; on days when the average standard value differed by 0.1‰–0.3‰ from its accepted value, a correction equal to the average offset was applied to the data. The 1 $\sigma$  standard error of samples run in duplicate to quadruplicate averages 0.05‰ and ranges from 0.01‰ to 0.09‰; repeat sample analyses were typically made over multiple days.

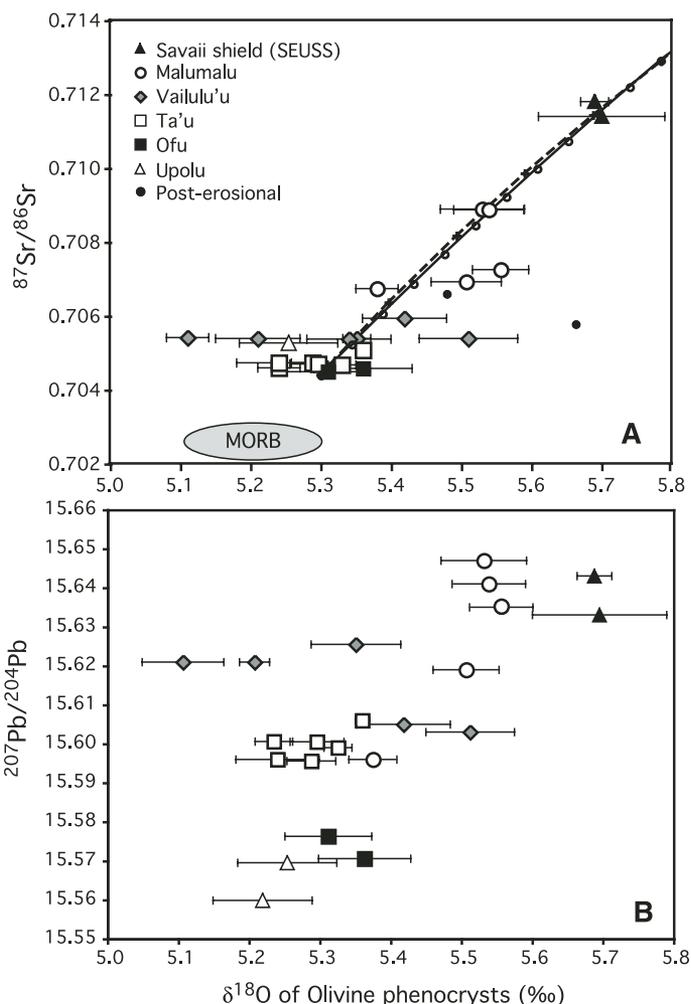
## RESULTS

Values of  $\delta^{18}\text{O}$  for olivine phenocrysts from 24 Samoan shield lavas range from 5.11‰ to 5.70‰ (GSA Data Repository Table DR1<sup>1</sup>). The lower end of this range overlaps with olivine from MORBs and upper mantle peridotites (5.2‰  $\pm$  0.2‰); the upper end of the range is comparable to the highest values observed in olivines from oceanic basalts (cf. Eiler, 2001).

Values of  $\delta^{18}\text{O}$  for olivines from Samoan shield-building lavas exhibit positive correlations with  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{207}\text{Pb}/^{204}\text{Pb}$  (Fig. 1) and an inverse correlation with  $^{143}\text{Nd}/^{144}\text{Nd}$  (not shown). The enrichments in radiogenic isotopes exhibited by these samples are primary, mantle-derived signatures and cannot be explained by assimilation of local oceanic crust or sediments into MORB-like lavas: the  $^{87}\text{Sr}/^{86}\text{Sr}$  of such local materials are all much too low to be the enriching component, and  $^{206}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$  of local marine sediments form a trend that diverges from the Samoan EM2 lavas (Jackson et al., 2007a). On this basis, we conclude that the correlated  $\delta^{18}\text{O}$  values are also primary mantle signatures. The SEUSS lavas define the most extreme EM2 basalts in the current global database, and therefore olivine from an end-member EM2-type basalt is established to have  $\delta^{18}\text{O}$  of at least 5.7‰; note that samples analyzed for  $\delta^{18}\text{O}$  only range as high as  $^{87}\text{Sr}/^{86}\text{Sr} = 0.7118$  because the most extreme lavas (at 0.7205) lack olivine phenocrysts.

Lavas with the lowest  $^{87}\text{Sr}/^{86}\text{Sr}$  (and highest  $^{143}\text{Nd}/^{144}\text{Nd}$ ), primarily from Ofu, Ta'u, and Upolu, are consistently lower in  $\delta^{18}\text{O}$ , La/Sm, Rb/Sr, Nb/Zr, and K/Na (i.e., show a lesser degree of incompatible element enrichment) than Malumalu and SEUSS lavas (Workman et al., 2004; Jackson et al., 2007a). Olivines from Vailulu'u lavas have a larger range in  $\delta^{18}\text{O}$  (~0.4‰) than do olivines from any other single volcano examined in this study, despite the fact that radiogenic isotope ratios and incompatible trace element ratios are nearly identical for the six Vailulu'u samples. We speculate that some factor other than source composition led to second-order variations in  $\delta^{18}\text{O}$  among olivines from Vailulu'u lavas; we suggest these lavas may contain relatively low  $\delta^{18}\text{O}$ , xenocrystic olivine entrained from the lithosphere. However,  $^3\text{He}/^4\text{He}$  ratios of olivine from the two lowest  $\delta^{18}\text{O}$  lavas are slightly elevated (9.3 and 10.0  $R_A$ ; Workman et al., 2004) compared to normal MORB values (~8  $R_A$ ), suggesting that either lithospheric olivine does not constitute 100% of the phenocrysts present, or the olivine and melt have diffusively exchanged He. Alternatively, there may be a component of HIMU (high U/Pb mantle reservoir) in the Vailulu'u lavas (Workman et al., 2004) that is lowering  $\delta^{18}\text{O}$  (olivines from HIMU lavas typically have  $\delta^{18}\text{O}$  values similar to MORBs; Eiler et al., 1997).

Correlations in Figure 1 indicate that  $\delta^{18}\text{O}$  variations of Samoan shield-building lavas primarily reflect variations in  $\delta^{18}\text{O}$  among their sources, as opposed to isotopic fractionations accompanying partial melting or crystal differentiation. The following observations further support this conclusion: (1)  $\delta^{18}\text{O}$  values of olivine do not correlate with Mg# of their host whole rocks, even when excluding the picrites (which we presume are cumulates), so  $\delta^{18}\text{O}$  values are not demonstrably controlled by crystal fractionation; (2) 20% or less fractional crystallization of olivine (typical for magmas presented here) will increase a melt's  $\delta^{18}\text{O}$  composi-



**Figure 1.** Plots showing correlations between  $\delta^{18}\text{O}$  of olivine phenocrysts (this study) and whole-rock  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{207}\text{Pb}/^{204}\text{Pb}$  (data from Workman et al., 2004; Jackson et al., 2007a, 2007b). Two posterosional data points are from Eiler et al. (1997). Correlations such as these are strong evidence that the  $\delta^{18}\text{O}$  values are representative of mantle sources. Error bars on  $\delta^{18}\text{O}$  represent 1 $\sigma$  standard error, except for Upolu samples, which were run only once and have error bars representing 1 $\sigma$  reproducibility of UWG-2 garnet standard (0.07‰). Ticks along mixing lines in panel A are for every 1% upper continental crust (UCC) addition (curve with circles; UCC values are  $\delta^{18}\text{O} = 10\text{‰}$ , Sr = 52 ppm,  $^{87}\text{Sr}/^{86}\text{Sr} = 0.75$ ) or 0.5% sediment addition (dashed curve with crosses; sediment values are  $\delta^{18}\text{O} = 25\text{‰}$ , Sr = 250 ppm,  $^{87}\text{Sr}/^{86}\text{Sr} = 0.75$ ) to the least enriched Samoan mantle component (represented by Ta'u lavas; details in text). MORB—mid-oceanic ridge basalt; SEUSS—superenriched underwater Savai'i samples.

tion by 0.1‰ or less (Eiler, 2001); and (3) variations in the degree of partial melting to produce the Samoan lavas could only lead to small variations in  $\delta^{18}\text{O}$  (~0.1‰; Eiler, 2001). Thus, crystallization and partial melting may cause slight scatter in the trends shown in Figure 1, but cannot account in any plausible way for the observed 0.6‰ variation in  $\delta^{18}\text{O}$ .

Of the two posterosional lavas from Savai'i documented by Eiler et al. (1997), one fits along the  $^{87}\text{Sr}/^{86}\text{Sr}$  versus  $\delta^{18}\text{O}$  trend in Figure 1 and one does not. A more in-depth evaluation of this issue is beyond the scope of this paper.

## DISCUSSION

EM2 lavas with elevated  $\delta^{18}\text{O}$  values require their source to contain a component of altered upper oceanic crust, marine sediments, and/or upper continental crust, as these are the most common, abundant (and frequently

<sup>1</sup>GSA Data Repository item 2008133, Table DR1, supplementary geochemical data, and Figures DR1 and DR2, is available online at [www.geosociety.org/pubs/ft2008.htm](http://www.geosociety.org/pubs/ft2008.htm), or on request from [editing@geosociety.org](mailto:editing@geosociety.org) or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

subducted) materials having  $\delta^{18}\text{O}$  values higher than typical mantle. The upper ~500 m of oceanic crust, which is altered at low temperature, typically varies in  $\delta^{18}\text{O}$  from 8‰ to 25‰, decreasing with depth and having a weighted average of ~8‰–10‰ depending on geographic location (Muehlenbachs, 1986; Staudigel et al., 1995; Alt, 2003). Clastic (i.e., continent derived) marine sediments and pelagic clays typically have  $\delta^{18}\text{O}$  values in the range of 10‰–25‰ throughout all documented geologic time, the most common values being 15‰–20‰ (Land and Lynch, 1996). The average  $\delta^{18}\text{O}$  value of upper continental crust has recently been estimated to be  $9.6\text{‰} \pm 1.3\text{‰}$  (Simon and Lécuyer, 2005).

Key trace element ratios can help distinguish which of these three high- $\delta^{18}\text{O}$  components may be present in the EM2 mantle source; in particular, ratios of elements that have similar compatibility during peridotite melting (thus melt ratios are similar to source ratios), and that differ markedly between primitive to slightly depleted mantle compared to altered oceanic crust, upper continental crust, and marine sediments. Ce/Pb and Nb/Th ratios satisfy both of these criteria, and are plotted versus the  $\delta^{18}\text{O}$  of olivine in Figure 2.

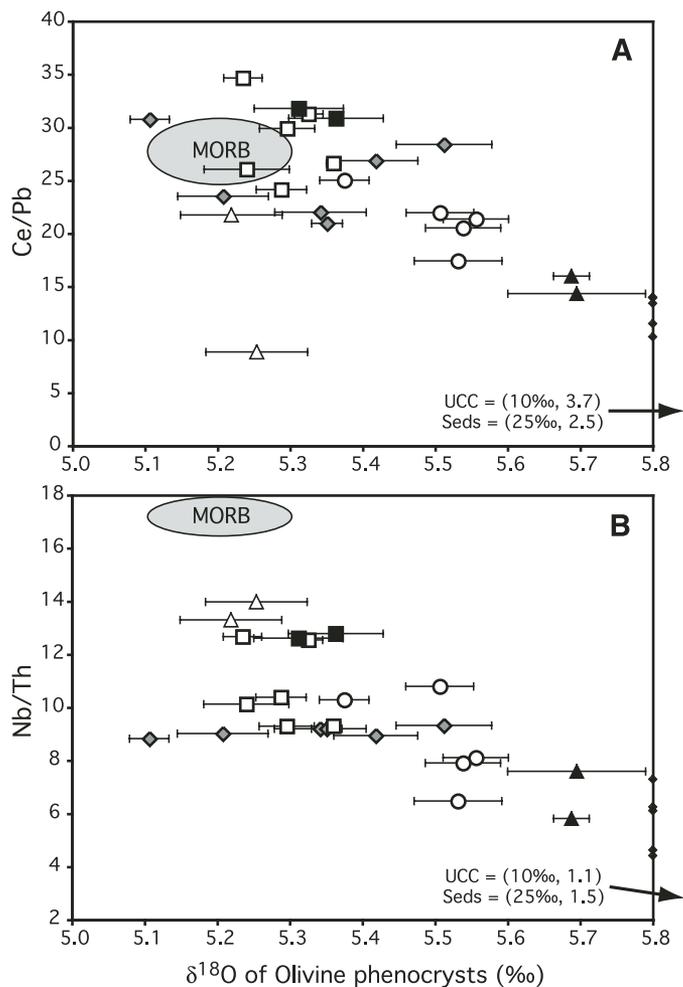
Based on Figure 2, can any of the three components be ruled out? There is substantial evidence that alteration of oceanic crust does not affect either Nb or Th (Staudigel et al., 1996; Kelley et al., 2003), so altered oceanic crust will have a Nb/Th ratio very similar to fresh MORBs (~18). Hence, altered oceanic crust could not account for the low Nb/Th (<5) of the SEUSS lavas, making it unlikely to be the dominant enriched component in EM2. However, it is difficult to completely rule out mantle metasomatism by a melt of subducted altered oceanic crust generated in the presence of residual rutile, in which Nb is compatible (Rudnick et al., 2000).

On the other hand, upper continental crust has low Ce/Pb and Nb/Th, which could easily explain the SEUSS lavas (Fig. 2). In fact, the overall pattern of trace element abundances for upper continental crust closely resembles that for the SEUSS lavas (Fig. 3), and is the main reason Jackson et al. (2007a) identified upper continental crust as the enriching component. Figure 3 shows that both upper continental crust and the SEUSS lavas are characterized by elevated Rb, Ba, Th, U, K, and Pb, with low Nb, Ta, and Ti.

There is very little difference in trace element composition between upper continental crust and clastic marine sediments (Figs. 2 and 3) because they are genetically related; the dominant source for clastic marine sediments is the upper continental crust. Therefore both upper continental crust and clastic marine sediments are suitable recycled, enriching components in EM2. However, the distinction between the two is geodynamically meaningful: if the recycled component responsible for the EM2 source is unmodified upper continental crust, this implies that enriched continental material is added to the mantle by mechanical erosion of continents by subducting slabs, a process that may be very difficult to quantify. However, if the crustal component is recycled clastic marine sediment, then it is produced by subduction of an observable (and possibly quantifiable) feature.

Although upper continental crust and clastic marine sediments are difficult to distinguish from each other by their trace element compositions, they have distinctly different  $\delta^{18}\text{O}$  values, leading to the different model mixing curves shown in Figure 1. In the following calculations, the low- $\delta^{18}\text{O}$  source is fixed at Sr = 28 ppm (from Jackson et al., 2007a),  $^{87}\text{Sr}/^{86}\text{Sr} = 0.7044$ , and  $\delta^{18}\text{O}_{\text{olivine}} = 5.3\text{‰}$  (representing lavas from Ta'u). With an enriching component having  $\delta^{18}\text{O}$  equal to average upper continental crust (10‰), a wide range of coordinates in Sr- $^{87}\text{Sr}/^{86}\text{Sr}$  space (1) results in a good fit to the Sr-O trend and (2) overlaps with values reported for continental cratons (Fig. DR1). However, all successful mixtures have much lower Sr ( $\leq 200$  ppm) than average upper continental crust (320 ppm; Rudnick and Gao, 1995) and require >20% upper continental crust in order to reach the most extreme EM2 lava at  $^{87}\text{Sr}/^{86}\text{Sr} = 0.7205$  (Figs. DR1 and DR2).

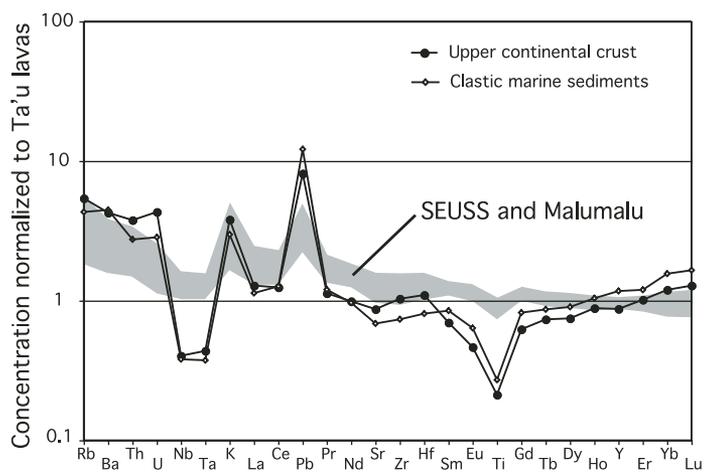
On the other hand, with an enriching component having  $\delta^{18}\text{O}$  in the upper range of clastic marine sediments (20‰–25‰), only 5%–8% sedi-



**Figure 2.** Plots showing systematic trends of decreasing Ce/Pb (A) and Nb/Th (B) of whole rocks with increasing  $\delta^{18}\text{O}$  values of olivine phenocrysts in Samoan lavas. Upper continental crust (UCC) values are from Rudnick and Gao (2003), and average clastic marine sediment values are from Plank and Langmuir (1998). SEUSS (super-enriched underwater Savai'i samples) lavas without  $\delta^{18}\text{O}$  data are plotted on vertical axis on right for reference (data from Jackson et al., 2007a). MORB—mid-oceanic ridge basalt.

ment is required to reach the most extreme EM2 lava (Fig. DR2). The forward modeling of Jackson et al. (2007a) indicates that misfits to the trace element pattern of the most enriched SEUSS lavas increase rapidly when >6% crustal component is used in the mixture, even when maximizing the goodness of fit by tuning the amount of garnet versus spinel facies melting, the degree of melting, and the estimate for the Ta'u source composition (the low  $\delta^{18}\text{O}$  component). Because of the greater misfit at greater amounts of crust, we prefer a model in which the enriching component in the SEUSS lavas is recycled, continent-derived, clastic marine sediments. Assuming such, we calculate there is 2%–3% sediment in the highest  $\delta^{18}\text{O}$  samples (5.7‰); this predicts a  $\delta^{18}\text{O}$  of 6.2‰–7.0‰, depending on the Sr concentration and isotopic composition of the sediment component, in the most enriched SEUSS lava (having an estimated 5%–6% sediment).

The above calculations imply a history of marine sedimentation of upper continental crust (during which sediments become strongly  $^{18}\text{O}$  enriched) prior to subduction recycling and formation of the EM2 mantle component. However, it is nearly impossible to rule out an upper continental crust reservoir that may have had elevated  $\delta^{18}\text{O}$  (i.e., >15‰) prior to its subduction; in any case, the enriching component in EM2 is in no way average upper continental crust.



**Figure 3.** Trace element abundance patterns in ultraenriched Samoan lavas compared to upper continental crust of Rudnick and Gao (2003) and modern, clastic marine sediments from Plank and Langmuir (1998). Concentrations are normalized to the least enriched Samoan lavas (average of 19 lavas from Ta'u; Workman et al., 2004) in order to visualize what elements are added or subtracted to make the ultra-enriched samples. Samples in gray field are 78-1 and 78-3 from Workman et al. (2004); 115-03, 115-18, 115-28, and 128-21 are from Jackson et al. (2007a). All lava samples have been fractionation corrected by incremental equilibrium olivine fractionation to obtain a whole-rock Mg# of 73. SEUSS—superenriched underwater Savai'i samples.

## SUMMARY AND CONCLUSIONS

1. Oxygen isotope compositions of olivine phenocrysts from Samoan basalts range from normal upper mantle values of 5.1‰ to some of the highest values in all oceanic lavas at 5.7‰; the amplitude of this signal is ~10 times analytical precision.

2. We interpret the  $\delta^{18}\text{O}$  values of olivines from Samoan shield building lavas as primary, mantle-derived signatures based on correlations with radiogenic Sr and Pb isotopes.

3. Correlations between  $\delta^{18}\text{O}$  and key trace element ratios (Ce/Pb, Nb/Th) are consistent with the finding of Jackson et al. (2007a) that EM2 contains a component of recycled continental material.

4. The observed trend between  $\delta^{18}\text{O}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  can be successfully fit with a wide range of  $\delta^{18}\text{O}$ -Sr- $^{87}\text{Sr}/^{86}\text{Sr}$  coordinates for the enriching component, but only those for clastic marine sediments are compatible with the trace element modeling of Jackson et al. (2007a). Therefore, we conclude that the creation of the EM2 mantle component was through a process we recognize in the modern world, i.e., sedimentation of upper continental crust material into a marine environment, followed by subduction and mixing with ambient mantle.

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