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# Re–Os isotope systematics in Samoan shield lavas and the use of Os-isotopes in olivine phenocrysts to determine primary magmatic compositions

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ABSTRACT

Samoan shield-stage lavas (from the islands of Ta'u, Savai'i, and Ofu and the seamounts of Vailulu'u and Malumalu) with Os concentrations >30 ppt have $^{187}$Os/$^{188}$Os ratios that exhibit a narrow range of values between 0.128 and 0.132. Lavas with ≤30 ppt Os show more radiogenic $^{187}$Os/$^{188}$Os ratios, in some cases as high as 0.191, suggesting that the $^{187}$Os/$^{188}$Os ratios of the extreme Samoan EM2 (enriched mantle 2) lavas likely have been compromised by assimilation of altered oceanic crust. The $^{187}$Os/$^{188}$Os ratios for rejuvenated-stage lavas from Savai'i are lower than shield lavas, and they exhibit some of the lowest $^{187}$Os/$^{188}$Os ratios in the global ocean island basalt database (Hauri and Hart, 1993). The difference may owe to contamination of the rejuvenated lavas with unradiogenic Os from disaggregated xenoliths from the mantle lithosphere, and their low Os isotopic composition does not reflect the EM2 mantle source of magmas. The limited range in $^{187}$Os/$^{188}$Os ratios of the higher Os-abundance shield lavas (0.128–0.132), coupled with a tremendous range of $^{87}$Sr/$^{86}$Sr (0.7045–0.7114), are characteristics of the EM2 source that can be explained by mixing a continental crustal sediment characterized by a high Sr/Os (~$10^7$) with a mantle peridotite that has low Sr/Os (~$10^4$).

Os abundances for nine whole rocks and their olivine phenocrysts show opposite trends with respect to whole rock MgO content: olivines have less than 0.1 ppb Os at 15 wt.% MgO and increase to 1–2 ppb Os in lavas with 8 wt.% MgO. Even though the Os isotopic composition of an evolved lava is susceptible to crustal assimilation, olivine phenocrysts with high Os concentrations preserve the pre-assimilation magmatic $^{187}$Os/$^{188}$Os ratio. For example, the range of $^{187}$Os/$^{188}$Os ratios measured on magmatic olivines from Samoan shield lavas (0.127 to 0.130) is lower (and narrower than) the range estimated using Os-rich (>30 ppt) whole rock lavas (0.128–0.132).

## 1. Introduction

Oceanic crust and terrigenous marine sediment enter the mantle at subduction zones (Hofmann and White, 1982; White and Hofmann, 1982), but their fate in the mantle afterwards is not well known. As a result of incorporation of subducted crust over geologic time, the Earth's mantle is chemically and isotopically heterogeneous (Hofmann, 1997; Zindler and Hart, 1986). However, the Earth's mantle mixes and stirs chaotically on geologic timescales, and any surviving portions of subducted crust are likely attenuated and their associated geochemical signatures diluted. Therefore, if oceanic and continental crust is returned to the surface in mantle upwellings, or plumes, and melted beneath hotspots, their geochemical signatures prove difficult to detect.

Samoa is a hotspot that has long been considered to host a component of recycled continental material (Farley et al., 1992; Jackson et al., 2007a; White and Hofmann, 1982; Workman et al., 2008; Wright and White, 1987). It has not been as well-studied as Hawaii and Iceland, and yet it is known to have unique compositional variability (e.g., Farley et al., 1992; Jackson et al., 2007a,b; Workman et al., 2004; Wright and White, 1987). Samoa is located on ~100 myr old altered Pacific lithosphere and displays a petrologically-diverse suite of geochemically well-characterized lavas (e.g., Hawkins and Natland, 1975; Natland, 1980; Workman et al., 2004). Therefore, Samoan lavas are ideal for probing questions of the petrogenesis of OIB during eruption and emplacement, such as how assimilation of oceanic lithospheric materials occurs in intraplate settings, and how phenocrysts can be used in OIB suites to establish primary magma compositions. They are also ideal for questions that have global significance such as identifying the recycled signatures that can better establish the end-member EM2 (enriched mantle II) composition.

The Re–Os isotopic system is a powerful geochemical tool that helps to detect crustal assimilation and to resolve the compositional

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imprint of ambient mantle peridotite from recycled crust. The Re–Os radiogenic isotope system – where $^{187}$Re undergoes $\beta^-$ decay to $^{187}$Os, $t_{1/2} = 41.6$ Ga (Shen et al., 1996; Smoliar et al., 1996) – is well suited for detecting recycled crustal signatures in hotspot lavas (e.g., Shirey and Walker, 1998). During mantle melting, Re is a moderately incompatible element and Os behaves compatibly. The disparate behavior on melting results in extreme fractionations of parent and daughter isotopes, which generates high time-integrated Re/Os (and high $^{187}$Os/$^{188}$Os) ratios in crustal reservoirs and correspondingly low Re/Os (and low $^{187}$Os/$^{188}$Os) ratios in the depleted mantle. This is in contrast to much smaller parent–daughter fractionation associated with melting in the more commonly measured radiogenic systems such as Sr, Nd or Pb isotopes, and highlights the potential of the Re–Os isotope system to resolve recycled crustal material in the mantle.

Os isotopes were previously reported in Samoan rejuvenated lavas from Savai'i (Hauri and Hart, 1993) and Samoan shield lavas (Jackson et al., 2008a; Workman et al., 2004). However, the data obtained on the Samoan shield lavas were acquired using an ICP-MS (inductively coupled plasma mass spectrometer) method (Jackson et al., 2008a; Workman et al., 2004), and the corresponding analytical errors were large (typically 1–2% for basalts). Such large analytical uncertainties make it problematic to resolve the small variations in $^{187}$Os/$^{188}$Os ratios that we identify in Samoan shield lavas and to see how truly uniform they are. Additionally, Re concentrations were never determined for Samoan shield lavas and the Re concentration for only a single Samoan rejuvenated lava was reported (Hauri and Hart, 1993). The sample suite examined in this study for Re and Os abundances and Os isotopic compositions includes some unusual Samoan lavas, including lavas with the highest $^3$He/$^4$He from Ofu Island (up to 34 Ra, or ratio to atmospheric; Jackson et al. 2007b) and the highest $^{87}$Sr/$^{86}$Sr from Savai'i Island (up to 0.7208 in magmatic clinopyroxene [cpx] separates; Jackson et al. 2007b).

Complementing the new whole rock Os-isotopic data, Os isotopes and Os concentrations on olivines hosted in the same lavas were measured. The new paired whole rock-olivine data provide important clues about the behavior of Os in magmas as they evolve. Owing to their lower Os abundances, evolved lavas can be more susceptible to having their Os-isotopic compositions modified by assimilation of oceanic crustal materials and seawater. However, the elevated Os concentrations that we report in olivines from evolved lavas may better shield olivines from the effects of crustal assimilation. As a result, Os-isotopic measurements in olivines may allow us to "see through" the effects of assimilation, particularly in evolved (and therefore low-Os) lavas that are susceptible to having their primary magmatic $^{187}$Os/$^{188}$Os ratios overprinted by assimilation.

## 2. Samples and methods

All samples for which we report new Re and Os abundances and $^{187}$Os/$^{188}$Os ratios are Samoan shield lavas. These occur on Savai'i, Ta'u and Ofu Islands, as well as Malumalu and Vailulu'u seamounts (Jackson et al., 2007a,b; Workman et al. 2004). Re and Os abundances and $^{187}$Os/$^{188}$Os ratios for Samoan rejuvenated lavas from Savai'i Island were reported in Hauri and Hart (1993). With relatively few exceptions (see Table 1) most of the samples analyzed here are new aliquots of the powders characterized geochemically in earlier studies (Jackson et al., 2007a,b, Workman et al., 2004). Powder preparation was as follows. Whole rocks were cut on a rock saw and crushed by hammer in plastic bags. The freshest chips were selected under binocular microscope with care to avoid surfaces that came into contact with the saw blade. The chips were subsequently cleaned by sonication in Milli-Q water. After drying, sample chips were powdered in an agate mortar. Additional chips were crushed in a jaw crusher, sieved and separated magnetically to concentrate the olivine fraction. The freshest olivines were selected, regardless of inclusion content, and sonicated in 6 N HCl for 30 min at room temperature, followed by rinsing and 10 min of sonication in Milli-Q water. The olivines were then re-examined to ensure that no visible groundmass or alteration phases remained. Olivines were then powdered by hand in an agate mortar that was precleaned by grinding with silica followed by thorough rinsing in Milli-Q water.

The Re–Os method employed in this study was most recently described in Class et al. (2009) and Heinonen et al. (2010). Os extraction followed the Carius tube method (Shirey and Walker, 1995), which was followed by solvent extraction (Cohen and Waters, 1996) with carbon tetrachloride. The Os was then purified using micro distillation (Roy-Barman and Allègre, 1995). Re was separated by anion exchange chromatography (Morgan and Walker, 1989; Walker, 1988). Isotopic measurements of Os were made by negative thermal ionization mass spectrometry (N-TIMS) on the Thermofinnigan Triton at the Department of Terrestrial Magnetism (DTM) on Pt ribbon filaments as an $OsO_3^-$ species; all measurements were made at low intensity on a single-collector secondary electron multiplier. All runs were corrected for their oxide compositions to their metal composition and mass bias corrected to a $^{192}$Os/$^{188}$Os ratio of 3.082614 (Creaser et al., 1991). From December 2008 to May 2010, twenty-nine separate 100 pg loads of the DTM solution standard "J-M Os" were analyzed as part of this study, and they yielded an average $^{187}$Os/$^{188}$Os ratio of 0.17366 ($\pm 0.00045$, $2\sigma$ std. dev., $n = 29$). However, variation in the $^{187}$Os/$^{188}$Os ratio of the J-M Os standard runs was generally lower during individual analytical sessions. Runs of basalts and olivine samples were corrected for the offset between the measured and accepted (0.17399) standard values for each analytical session. In-run precision for basalts and olivine samples was 0.03 to 0.32% ($2\sigma$) for $^{187}$Os/$^{188}$Os ratios (except for AVON3-76-9ol, which was 0.78%). Re concentrations were determined by isotope dilution, and measurements were made on either the P54 or the NU plasma machines hosted at DTM. The average proportion of blank Os to sample Os was 1.2% (ranging from 0.03 to 5.2%) for all whole rock samples and 1.8% (ranging from 0.04 to 4.5%) for olivine samples for which $^{187}$Os/$^{188}$Os ratios are reported. AVON3-78-3ol has a higher blank proportion – 18.1% – so Os isotopic ratios are not reported. The average proportion

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**Note to Table 1**

Abbreviations: whole rock is wr, olivine is ol, clinopyroxene is cpx, standard error (absolute) is s.e. (abs), replicate is rep., submarine is mar, subaerial is aer. With three exceptions, all whole rocks have had Re and Os measured on the same aliquot of powder. Os and Re were measured on different aliquots of the same batch of powder for samples ALIA-115-18 (wr), ALIA-115-03 (wr), and Ofu-04-15 (wr). In order to estimate the $^{187}$Re/$^{188}$Os ratios of the samples with "missing" Re, the Re and Os data obtained on different aliquots of powder were combined: Re concentrations for sample ALIA-115-18 were taken to be the averages of rep1 and rep2; Re concentrations for ALIA-115-03 were taken from the replicate run; Re concentrations for Ofu-04-15 were taken from the rep. Ofu-04-14 is the only whole rock sample for which Re concentrations are not reported.All lavas with the prefix "ALIA" have been age corrected using ages from Koppers et al. (2008): ALIA-115-18 is 5.29 Ma; ALIA-115-28 is 5.04 Ma; ALIA-115-03 is 5.06 Ma; ALIA-128-21 is 4.80 Ma. Lavas from all other locations are thought to have much younger ages (<1 Ma) and have not been age-corrected.The $^{87}$Sr/$^{86}$Sr of ALIA-115-18 cpx is the average of two measurements, where one measurement was made on green cpx (0.720830) and the other on black cpx (0.720232) (Jackson et al., 2007a). The Sr-isotopic composition of Ofu-04-06 cpx is reported in Jackson et al. (2009b). Whole rock Sr and Pb isotopic data are from Jackson et al. (2007a,b) and Workman et al. (2004). Owing to small sample sizes combined with relatively high blank Re to sample Re ratios, it is difficult to determine Re concentrations in most of the olivine samples examined here. Therefore, Re data are not reported for most olivine separates.All osmium standard and unknown runs with $^{185}ReO_3/^{188}OsO_3 > 0.00035$ were discarded.AVON3-78-3 (ol) was overspiked, and the Os-isotopic composition is not reported; Os concentrations were measured precisely for this sample and and are reported.Each replicate involves a new aliquot of powder that was processed independently through all steps of the wet chemistry procedure. Whole rock Sr-isotope and MgO compositions are published for all whole rock lavas (Jackson et al. 2007a,b; Workman et al., 2004), except for T54, 63-11, 63-2 and MgO for all Ofu lavas. Sr-isotopic ratios were measured on the WHOI Neptune following the procedures outlined in Workman et al. (2004). Whole rock MgO was measured in the same way as reported in Workman et al. (2004).