

# Osmium-isotope variations in Hawaiian lavas: evidence for recycled oceanic lithosphere in the Hawaiian plume

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## Abstract

Isotopic heterogeneity in Hawaiian shield lavas reflects the presence of two distinct recycled components in the Hawaiian plume, both from the same packet of recycled oceanic lithosphere. Radiogenic Os-isotopes and anomalously heavy oxygen-isotopes in Koolau lavas reflect melt generation from recycled oceanic crust plus pelagic sediment. In contrast, Kea lavas have unradiogenic Os-isotopes but anomalously light oxygen-isotopes. Oxygen–osmium–lead isotope correlations preclude generation of the Kea isotopic signature from asthenospheric upper mantle or the in situ lithospheric mantle or crust. Instead, melting of recycled, hydrothermally altered ultramafic lower crust or lithospheric mantle in the Hawaiian plume can produce Kea-type lavas. The preservation of both upper- and lower-crustal oxygen isotope signatures in plume-derived Hawaiian lavas indicates that chemical heterogeneities with length scales of only a few kilometers can be preserved in the convecting mantle for long periods of time, probably on the order of 1 Ga or more. © 1998 Elsevier Science B.V. All rights reserved.

*Keywords:* osmium; isotopes; oceanic crust; Hawaii; hot spots

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## 1. Introduction

Geochemical studies of mantle-derived basalts are a primary source of information on the composition and evolution of the mantle. Global compilations of isotopic data from oceanic basalts have identified several isotopic endmembers in the convecting mantle [1,2]. However, the origin, location and length scale of these components and their relationship to mantle convection remains a matter of considerable debate. Melt extraction, preservation of primitive mantle, intra-mantle differentiation or metasoma-

tism, and crustal recycling have all been advocated as mechanisms for producing isotopic heterogeneity in the mantle (e.g., [1,3,4]). Differentiation between these mechanisms is of great importance, because each has very different implications for the style and vigor of mantle convection throughout earth history.

Complicating our geochemical study of the convecting mantle is the fact that mantle-derived basalts undergo a variety of processes prior to eruption which have the potential to alter their composition, thereby obscuring the nature of their source. Several recent studies have suggested that much of the isotopic and chemical heterogeneity observed within individual ocean island basalt suites reflects different degrees of interaction between melts and the

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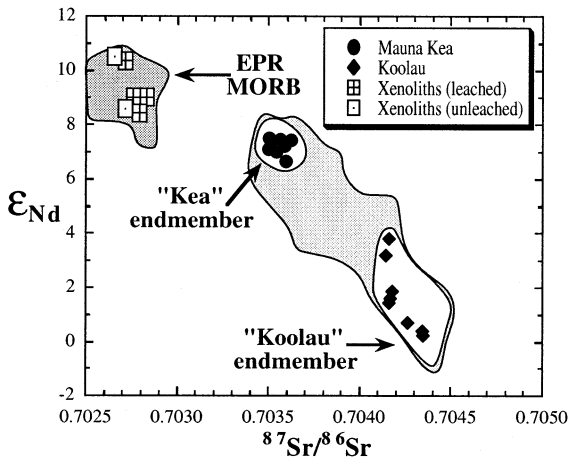


Fig. 1.  $^{87}\text{Sr}/^{86}\text{Sr}$  versus  $\epsilon_{\text{Nd}}$  in Hawaiian shield tholeiites. To first order, isotopic variations in Hawaiian shield lavas define a mixture between two principle components, an enriched component (the "Koolau" endmember) and a more depleted component (the "Kea" endmember). The MORB-related gabbro xenoliths from Hualalai plot within the field for East Pacific Rise (EPR) basalts. Data for these and subsequent plots are from this study and numerous literature sources, available on request from the corresponding author.

oceanic crust or lithospheric mantle [3,5,6]. It is thus important to evaluate possible magma/lithosphere interaction in order to distinguish chemical and isotopic signatures arising from such interaction rather than from heterogeneities in the convecting mantle.

Hawaiian lavas span a large range in isotopic composition over both long and short timescales [4,7]. Although at least three isotopically distinct components are required to produce the full range of isotopic compositions revealed in Hawaiian lavas (e.g., [8]), to first order the isotopic variations in shield lavas appear dominated by mixture of two components; a relatively 'depleted' or MORB-like component best observed in lavas from Kilauea and Mauna Kea volcanoes (henceforth the 'Kea' endmember), and an 'enriched' component that is most obvious in lavas from Koolau, Kahoolawe, and Lanai volcanoes (the 'Koolau' endmember) (Fig. 1). In this paper, we present new Os-isotope data for lavas from Mauna Kea and Koolau volcanoes. We evaluate possible crustal contamination of Hawaiian lavas through analyses of gabbroic xenoliths that provide a sample of the underlying Pacific crust. Isotopic data from compatible (Os) and major (O) elements

complement the considerable quantity of existing isotopic data for incompatible element tracers (e.g., Sr, Nd, and Pb), and provide important new constraints which preclude many earlier hypotheses for the identities of the Koolau and Kea endmembers. We propose a simple model whereby both endmembers are generated through the same physical process; alteration, subduction and recycling of ancient oceanic lithosphere.

## 2. Samples and results

We examined isotopic variations in a suite of 13 Mauna Kea lavas and 8 Koolau lavas. The Mauna Kea lavas are from well-characterized drill-core samples collected during the initial drilling phase of the Hawaii Scientific Drilling Project, which sampled approximately 200 kyr of Mauna Kea late-shield and early post-shield volcanism [9–14]. Olivine-rich Koolau samples were provided by Fred Frey. Chemical and Sr- and Nd-isotopic data for some of these samples have been previously reported [15,16]. Our new analyses are consistent with these prior results. Separation of Re and Os followed procedures similar to those of Hauri and Hart [17], except that sodium carbonate was eliminated from the flux mixture to reduce the procedural blank. Re and Os were analyzed using negative thermal ionization mass spectrometry as outlined by Pearson et al. [18] and references therein. Sr-, Nd-, and Pb-isotope analyses were performed on acid-leached samples, using procedures similar to those outlined in [19].

The results of our analyses are presented in Table 1. Mauna Kea tholeiites show limited variation in  $^{187}\text{Os}/^{188}\text{Os}$ , ranging from 0.1294 to 0.1328, and are only slightly more radiogenic than depleted upper mantle peridotite [20,21]. One alkalic lava (R177) has much more radiogenic Os (0.1522); however, the low Os concentration of this sample (20 ppt) makes it likely that its Os-isotope composition reflects minor assimilation of radiogenic crust or sediments. Similar contamination of low-[Os] lavas from Haleakala volcano has previously been proposed [22]. In contrast with Mauna Kea tholeiites, Koolau tholeiites have very radiogenic and more heterogeneous Os-isotopes. Initial  $^{187}\text{Os}/^{188}\text{Os}$  in Koolau lavas range from 0.1406 to 0.1477. Individually, neither Koolau

Table 1  
Isotopic compositions of Mauna Kea and Koolau lavas

	Re (ppt)	Os (ppt)	$^{187}\text{Os}/^{188}\text{Os}$	$^{87}\text{Sr}/^{86}\text{Sr}$	$\epsilon_{\text{Nd}}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$\delta^{18}\text{O}_{\text{Olivine}}$
<b>Mauna Kea Volcano</b>									
HSDP-R160-5.75A	175	219	0.1323	0.70350	7.5	18.444	15.490	38.049	4.87
R 160 duplicate	–	–	–	–	–	18.433	15.483	38.021	–
HSDP-R177-2.60A	180	20	0.1522	0.70356	–	18.426	15.462	37.967	–
HSDP-R189-8.50A	30	1.169	0.1312	0.70352	7.4	18.497	15.469	38.010	4.83
HSDP-R212-0.40A	96	1.172	0.1310	0.70356	7.5	18.418	15.453	37.939	4.76
HSDP-R243-8.40A	217	1.030	0.1308	0.70359	7.2	18.540	15.466	38.048	4.64
HSDP-R259-0.75A	–	1.066	0.1311	0.70362	7.4	18.533	15.466	38.048	4.82
HSDP-R303-3.00A	–	740	0.1294	0.70356	7.3	18.520	15.493	38.115	4.73
HSDP-R347-6.35A	–	1.236	0.1307	0.70356	7.2	18.520	15.464	38.071	4.95
HSDP-R365-0.05A	–	1.489	0.1297	0.70352	7.2	18.610	15.489	38.169	4.72
HSDP-R413-1.40A	241	642	0.1328	0.70350	7.1	18.521	15.470	38.089	4.99
HSDP-R446-2.40A	203	724	0.1301	0.70354	7.0	18.572	15.476	38.124	4.69
HSDP-R466-5.00A	287	242	0.1317	0.70360	6.7	18.553	15.465	38.112	4.83
<b>Koolau Volcano</b>									
KOO-8A	277	458	0.1469	0.70414	3.2	17.914	15.457	37.840	5.96
KOO-10 <sup>a</sup>	308	115	0.1457	0.70417	1.6	17.839	15.430	37.720	5.71
KOO-17A	248	1.377	0.1406	0.70435	0.2	17.877	15.447	37.894	–
KOO-19A	351	259	0.1424	0.70416	1.4	17.899	15.462	37.907	5.95
KOO-21	372	153	0.1477	0.70426	0.7	17.870	15.436	37.840	5.99
KOO-30A	429	254	0.1452	0.70434	0.4	17.861	15.440	37.933	5.98
KOO-49	382	244	0.1428	0.70421	1.9	17.853	15.423	37.774	5.90
KOO-49 duplicate	–	–	–	–	–	17.854	15.419	37.760	–
KOO-55	501	907	0.1420	0.70416	3.8	17.879	15.436	37.802	5.75

Os isotopes for Koolau samples are age corrected to 2.2 Ma. Age corrections are less than analytical uncertainty for most samples ( $<3\%$ ,  $2\sigma$ )  $^{87}\text{Sr}/^{86}\text{Sr}$  reported relative to a value of 0.71025 for NBS 987.  $\epsilon_{\text{Nd}}$  calculated using  $^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}} = 0.512636$ , and reported relative to La Jolla  $^{143}\text{Nd}/^{144}\text{Nd} = 0.511860$ . External precisions for Sr and Nd isotopes are  $\pm 0.00002$  and  $\pm 0.4\epsilon$  ( $2\sigma$ ). O-isotope data are from Eiler et al. [5,10]. Sr and Nd data for Mauna Kea lavas are from Ref. [11]; Pb isotopes were reanalyzed for these samples because of concerns about mass fractionation in the data from Ref. [11]. Pb-isotopes were corrected for mass fractionation using NBS 981 values of Todt et al. [61]. Reproducibility of measured ratios is better than  $0.05\%$  a.m.u<sup>-1</sup>. Average total procedural blanks were: Os, 10 pg; Re, 10 pg; Sr, 140 pg; Nd, 15 pg; Pb, 80 pg.

<sup>a</sup> Sr data for KOO-10 from Ref. [15].

nor Mauna Kea lavas show significant correlation between Os-isotopes and other isotopic tracers, except that Mauna Kea lavas show a statistically significant positive correlation between Os- and O-isotopes and a negative correlation between Os- and Pb-isotopes. However, Hawaiian lavas as a whole display very strong correlations between Os-isotopes and other isotopic tracers, as illustrated in Fig. 2.

Previous studies have revealed that a small subset of the gabbroic xenoliths contained within the 1801 Kaupulehu flow of Hualalai volcano represent accidental fragments of the pre-Hawaiian Pacific middle to lower crust [23,24]. We report isotopic compositions for 4 gabbroic xenoliths in Table 2.

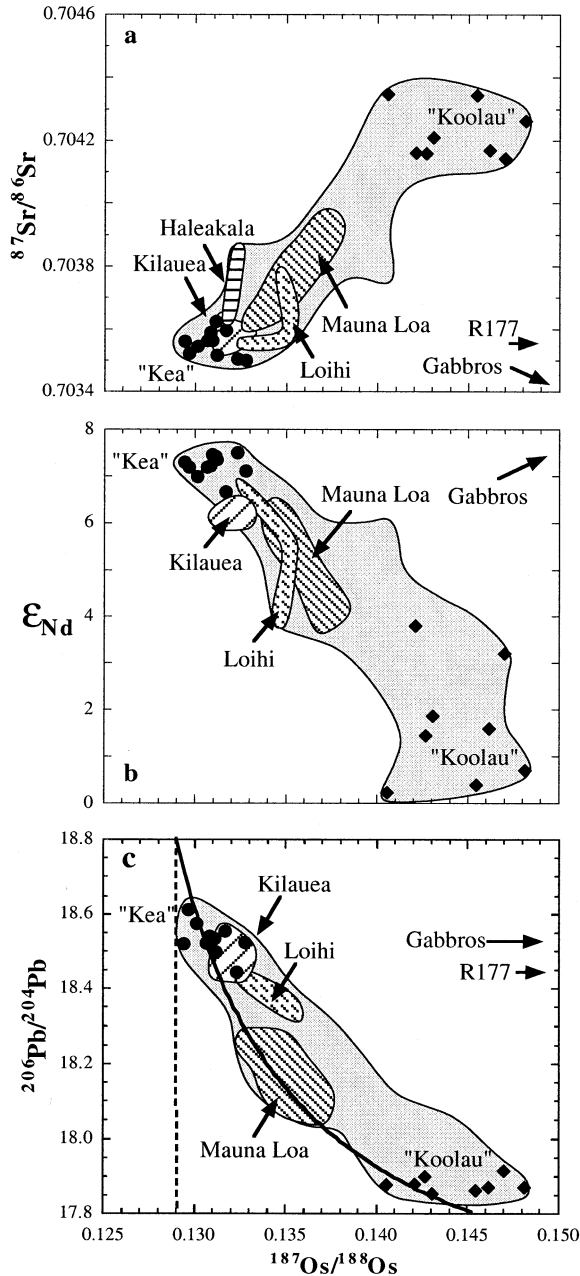
Acid-leached and unleached aliquots of the gabbroic xenoliths have similar MORB-like Sr-isotopes, with  $^{87}\text{Sr}/^{86}\text{Sr}$  ranging from 0.70271 to 0.70283 for leached samples and from 0.70266 to 0.70277 for unleached samples. Nd-isotopes are also MORB-like, with  $\epsilon_{\text{Nd}}$  ranging from 8.4 to 10.4. Pb-isotopes are somewhat more radiogenic in unleached samples than in leached samples, possibly reflecting the preferential removal of ingrown Pb by the leaching process.  $^{206}\text{Pb}/^{204}\text{Pb}$  ranges from 18.02 to 18.23 and from 18.04 to 18.62 in leached and unleached samples, respectively. Os-isotopes in the gabbros are very radiogenic, reflecting ingrowth of  $^{187}\text{Os}$  since formation of the Pacific crust  $\sim 100$  m.y. ago.

$^{187}\text{Os}/^{188}\text{Os}$  ranges from 0.2255 to 0.5388, much higher than any recent Hawaiian lavas.

Whole-rock oxygen isotopes were measured for four MORB gabbros by J. Fahrquar at the Geophysical Laboratory, Carnegie Institution. Samples were reacted with  $\text{BrF}_5$  using conventional, exter-

nally heated Ni-rod bombs [25]. Product oxygen was purified using liquid nitrogen traps and a Hg diffusion pump. Molecular oxygen was transferred to the mass spectrometer first by condensation at liquid nitrogen temperatures into sample tubes containing molecular sieve 5A and then by heating the sample tubes to  $80^\circ\text{C}$  prior to expansion into the bellows of the mass spectrometer. Oxygen isotope ratios were determined by dynamic gas source mass spectrometry (Finnigan MAT 252) measuring  $m/e$  32 and 34, and are reported relative to SMOW. Uncertainties on the oxygen isotope compositions are estimated to be  $\pm 0.2\text{‰}$  ( $1\sigma$ ). Wholerock  $\delta^{18}\text{O}$  ranges from 5.9 to 6.2‰, which is slightly heavier than fresh MORB glasses ( $\sim 5.7\text{‰}$ ; [26]).

The same components identified using incompatible element isotopic tracers (e.g., Fig. 1) are also apparent in the Os- and O-isotope systematics (e.g., Fig. 2); addition of these isotopic systems has not required the invention of new 'components' in Hawaiian isotope space. In the following discussion we evaluate the new constraints placed on models for the generation of the 'Kea' and 'Koolau' components by the O- and Os-isotope data.



### 3. Nature of the Koolau component

Koolau lavas possess the highest Sr- and lowest Nd- and Pb-isotopes of any Hawaiian shield lavas. Several models have been proposed to explain the isotopic signatures of the Koolau source. These include the entrainment of primitive lower mantle by the Hawaiian plume [1,8,27], involvement of a metasomatized mantle source (possibly recycled continental lithosphere) [28], and recycling of oceanic

Fig. 2.  $^{187}\text{Os}/^{188}\text{Os}$  in Hawaiian shield lavas versus (a)  $^{87}\text{Sr}/^{86}\text{Sr}$ ; (b)  $\epsilon_{\text{Nd}}$ ; and (c)  $^{206}\text{Pb}/^{204}\text{Pb}$ . Symbols are as in Fig. 1. The solid mixing curve is for  $\sim 1.8$  Ga crust and pelagic sediment (97:3) added to Kea-type mantle. Dashed curve is for pure sediment added to Kea-type mantle. Mixing parameters are as follows: Mantle [Os] = 3.1 ppb,  $^{187}\text{Os}/^{188}\text{Os}$  = 0.129, [Pb] = 50 ppb,  $^{206}\text{Pb}/^{204}\text{Pb}$  = 18.8; 1.8 Ga crust [Os] = 0.05 ppb,  $^{187}\text{Os}/^{188}\text{Os}$  = 2.74, [Pb] = 90 ppb (300 ppb before subduction),  $^{206}\text{Pb}/^{204}\text{Pb}$  = 21.5; 1.8 Ga pelagic sediment [Os] = 0.2 ppb,  $^{187}\text{Os}/^{188}\text{Os}$  = 0.64, [Pb] = 7500 ppb (25 ppm before subduction),  $^{206}\text{Pb}/^{204}\text{Pb}$  = 15.8.

Table 2

Isotopic compositions of MORB-related gabbroic xenoliths, Hualalai volcano, Hawaii

Sample	NMNH #	Os (ppt)	$^{187}\text{Os}/^{188}\text{Os}$	$^{87}\text{Sr}/^{86}\text{Sr}$	$\epsilon_{\text{Nd}}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$\sigma^{18}\text{O}_{\text{wholerock}}$
63KAP-13 leached	114358-14	–	–	0.70283	9.0	18.171	15.508	37.898	–
63KAP-13 unleached	114358-14	15	0.3507	0.70271	8.6	18.402	15.545	38.060	6.2
65KAP-13 leached	114370-13	–	–	0.70271	10.4	18.072	15.453	37.722	–
65KAP-13 unleached	114370-13	12	0.2901	0.70266	10.5	–	–	–	6.1
65KAP-14D leached	114370-15	–	–	0.70276	9.0	18.228	15.543	38.003	–
65KAP-14D unleached	114370-15	25	0.5388	0.70269	–	18.616	15.598	38.263	5.9
65-86-64 leached	114668-10	–	–	0.70279	8.4	18.019	15.463	37.798	–
65-86-64 unleached	114668-10	20	0.2255	0.70277	–	18.043	15.484	37.864	6.2

Samples were obtained from the Dale Jackson Collection at the National Museum of Natural History (NMNH).

crust and sediments [29,30]. Below, we evaluate each of these models in turn.

### 3.1. Primitive mantle

Koolau lavas have Sr- and Nd-isotope values that approach those inferred for the bulk silicate earth (BSE). Koolau lavas also have Pb-isotopes that approach the geochron, i.e. they have values similar to those of a silicate reservoir that has not experienced secondary changes in its U/Pb ratio since very early in earth history. This similarity between the isotopic compositions of Koolau lavas and that of the BSE has prompted repeated suggestions that primitive (presumably lower) mantle is involved in the generation of these lavas (e.g., [1,8,27]). However, many trace element ratios inferred for the Koolau source are non-primitive. For example, Rb/Sr ratios in fresh Koolau lavas ( $<0.017$ ; [15]), are lower than inferred for a BSE reservoir with a modern-day  $^{87}\text{Sr}/^{86}\text{Sr} \approx 0.7045$  ( $\text{Rb}/\text{Sr} \approx 0.030$ ; [31]). Because Rb is more incompatible than Sr during partial melting in the mantle, the Rb/Sr ratio of the Koolau source should be lower than that measured in the lavas, so that the measured ratios place an upper bound on the source ratio. Other trace element ratios that are not expected to significantly fractionate during partial melting (e.g., Nb/Th or Th/La) are also far removed from primitive mantle values [16,32,33].

Os- and O-isotope data further preclude the primitive mantle model. The Os-isotopic compositions of Koolau lavas ( $^{187}\text{Os}/^{188}\text{Os} = 0.140$  to  $0.148$ ) greatly exceed chondritic values or values commonly

observed in abyssal peridotites and other samples of the upper mantle ( $<0.127$  in ‘fresh’ peridotites; [20]), which appear to have evolved with a nearly chondritic or slightly sub-chondritic Re/Os [20,21]. Therefore, the Os-isotope values of Koolau lavas are inconsistent with derivation from a primitive mantle source, unless we further hypothesize some additional process for generating high  $^{187}\text{Os}/^{188}\text{Os}$  in ‘primitive’ lower mantle (e.g., [4,34]).

Furthermore, olivine phenocrysts in Koolau-type lavas possess systematically higher  $\delta^{18}\text{O}$  values than olivines from other Hawaiian volcanoes (e.g., Fig. 3) or from upper mantle peridotites. Koolau olivines have  $\delta^{18}\text{O} = 5.7\text{--}6.0\%$ , compared with  $5.2 \pm 0.1\%$  for olivines in equilibrium with upper mantle peridotites or fresh MORB [5,35]. The elevated  $\delta^{18}\text{O}$  values of Koolau olivines require a source with  $\delta^{18}\text{O}$  higher by nearly 1‰ than the upper mantle. However, mass-balance constraints indicate that primitive mantle should possess essentially the same oxygen-isotope composition as the depleted upper mantle.

In summary, many different features of Koolau lavas cannot derive from a primitive mantle source. The cumulative weight of these contradictions therefore makes any variant of the primitive mantle model untenable.

### 3.2. Intramantle differentiation (metasomatized mantle)

Intramantle differentiation processes (e.g., mantle metasomatism) can produce trace-element enriched regions in the mantle which over time may evolve

significant isotopic heterogeneities. Several studies have suggested that ‘enriched’ components in ocean island basalts may derive from metasomatized portions of the mantle (e.g., [3,28]). However, neither the radiogenic  $^{187}\text{Os}/^{188}\text{Os}$  nor the elevated  $\delta^{18}\text{O}$  values in Koolau lavas are likely to derive from metasomatized peridotite sources.

There is little evidence to suggest that mantle metasomatism significantly increases the Re/Os or  $^{187}\text{Os}/^{188}\text{Os}$  values in high-[Os] peridotites. Metasomatized trace-element enriched xenoliths from continental lithosphere typically have sub-chondritic rather than super-chondritic  $^{187}\text{Os}/^{188}\text{Os}$ , reflecting long-term Re-depletion due to melt removal [18,36]. To date, no peridotite xenoliths or massifs from either continental or oceanic settings with  $^{187}\text{Os}/^{188}\text{Os}$  as high as in the Koolau lavas have been found,

and the existence of such high  $^{187}\text{Os}/^{188}\text{Os}$  peridotite reservoirs in the mantle must be considered speculative at best.

Mantle enrichment via fluid or melt metasomatism is also unlikely to result in the  $^{18}\text{O}$ -enrichment characteristic of Koolau lavas. The relatively small equilibrium difference in the O-isotope composition between basaltic melt and mantle peridotite precludes significant shifts in O-isotope composition through the addition or removal of such melt. Given a  $\Delta_{\text{melt-ol}}$  of  $\sim 0.5\text{‰}$  [37], addition of 10% melt to peridotite will result in an increase in  $\delta^{18}\text{O}$  of less than  $0.05\text{‰}$ , far less than the increase required for the Koolau source. Several laser fluorination studies of O-isotope variations in mantle peridotites confirm that mantle metasomatism has little effect on O-isotope compositions. For example, no systematic O-isotope difference exists between hydrous and anhydrous mantle peridotites or between fertile and depleted peridotites [35,38]. Thus, neither the O- nor the Os-isotope enrichment of the Koolau source is consistent with a metasomatized mantle source.

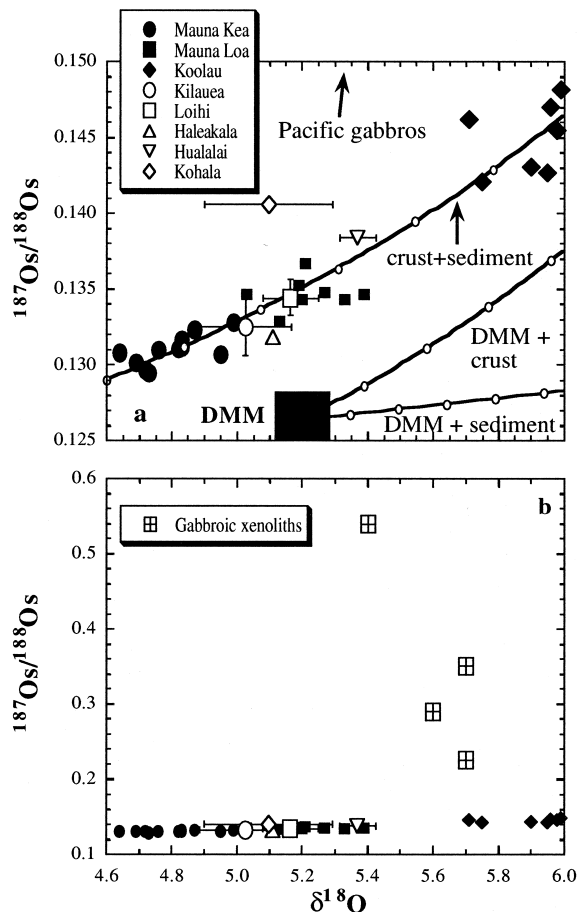


Fig. 3. (a)  $^{187}\text{Os}/^{188}\text{Os}$  versus  $\delta^{18}\text{O}$  in Hawaiian shield lavas. Olivine  $\delta^{18}\text{O}$  values are from Eiler et al. [5,10]. Os-isotope data for Mauna Loa lavas from Ref. [60]. Open symbols with error bars denote averages and standard deviations ( $1\sigma$ ) for volcanoes where O- and Os-isotope data do not exist for the same samples. No error bars for Os-isotopes are given for Kohala and Hualalai volcanoes because only one Os-isotope analysis for each of these volcanoes has been reported to date [4]. One Haleakala sample with  $>50$  ppt Os for which both Os- and O-isotope data exists is also plotted [5,22]. Haleakala samples with  $<50$  ppt Os are not plotted, but most follow the same general correlation defined by the rest of the data. The mixing curves show the result of mixing 1.8 Ga crust or sediment into depleted mantle or a mixture of crust and sediment (97:3) into mantle with a ‘Kea’-like isotopic composition. See text for details. Mixing parameters were as follows: Depleted mantle [Os] = 3.1 ppb,  $^{187}\text{Os}/^{188}\text{Os}$  = 0.1264,  $\delta^{18}\text{O}_{\text{olivine}}$  = 5.2‰; ‘Kea’-type mantle [Os] = 3.1 ppb,  $^{187}\text{Os}/^{188}\text{Os}$  = 0.129,  $\delta^{18}\text{O}_{\text{olivine}}$  = 4.6‰; basaltic crust [Os] = 50 ppt,  $^{187}\text{Os}/^{188}\text{Os}$  = 2.74,  $\delta^{18}\text{O}$  = 9‰; sediment [Os] = 200 ppt,  $^{187}\text{Os}/^{188}\text{Os}$  = 0.64,  $\delta^{18}\text{O}$  = 20‰. Tick marks denote 5% additions of crust or 1% additions of sediment to depleted mantle. (b)  $^{187}\text{Os}/^{188}\text{Os}$ – $\delta^{18}\text{O}$  in shield lavas and MORB-related gabbroic xenoliths from the 1801 Kaupulehu flow, Hualalai. Note change in scale for Os-isotopes.  $\delta^{18}\text{O}$  values for xenolith whole rock powders have been adjusted downward by  $0.5\text{‰}$  (reflecting an equilibrium  $\Delta_{\text{melt-ol}}$  of  $\sim 0.5\text{‰}$ ) to facilitate comparison with  $\delta^{18}\text{O}$  in olivine phenocrysts.

### 3.3. Recycled crust and sediments

In contrast with the models discussed above, recycling of oceanic crust and sediments can explain the EM-I type isotopic characteristics identified with the Koolau endmember (e.g., [30]). Although the effects of dehydration and melt extraction during subduction on incompatible element budgets and parent/daughter ratios (e.g., Rb/Sr) are poorly constrained, ancient recycled crust and sediments are likely to have higher  $^{87}\text{Sr}/^{86}\text{Sr}$  and lower  $\varepsilon_{\text{Nd}}$  than the depleted mantle, consistent with high  $^{87}\text{Sr}/^{86}\text{Sr}$  and low  $\varepsilon_{\text{Nd}}$  in Koolau lavas. Pelagic sediments commonly have high [Pb] and low U/Pb [30,39]. Although modern pelagic sediments are for the most part too radiogenic to explain the low  $^{206}\text{Pb}/^{204}\text{Pb}$  of Koolau lavas, ancient pelagic sediments should have much lower  $^{206}\text{Pb}/^{204}\text{Pb}$  as a result of their long-term evolution with low U/Pb [30]. Furthermore, in contrast with the primitive and metasomatized mantle models discussed above, crustal recycling provides a ready explanation for the Os- and O-isotope compositions of Koolau lavas.

Because oceanic crust possesses high Re abundances (average  $\sim 930$  ppt; [40]) but low [Os] (typically  $< 50$  ppt; see e.g., [41]), recycled oceanic crust will evolve very radiogenic Os over time. Pelagic sediments also typically have very radiogenic  $^{187}\text{Os}/^{188}\text{Os}$ , although the absolute concentrations of Re and Os are highly variable [42,43]. Recycling of ancient oceanic crust and pelagic sediments should therefore produce a mantle reservoir with elevated Re/Os and  $^{187}\text{Os}/^{188}\text{Os}$  (e.g., [17,40]). Using average Re and Os abundances in oceanic crust, sediment, and the mantle, we find that addition of  $\sim 30\%$  1.8 Ga crust containing a mixture of 97% basalt and 3% pelagic sediment into depleted mantle with modern day  $^{187}\text{Os}/^{188}\text{Os} = 0.126$  produces a hybrid source with  $^{187}\text{Os}/^{188}\text{Os} = 0.144$ , which is similar to the  $^{187}\text{Os}/^{188}\text{Os}$  values of Koolau basalts (0.140–0.148).

Recycling of oceanic crust and sediments is also the most viable mechanism for producing O-isotope heterogeneities in the mantle. Hydrothermal alteration of oceanic crust produces large O-isotope variations in altered crust. Numerous studies of ophiolites have revealed a consistent pattern of  $^{18}\text{O}$  enrichment in the upper oceanic crust and  $^{18}\text{O}$

depletion in lower oceanic crust and/or uppermost mantle [44,45]. Rejection of altered oceanic crust into the mantle therefore has the potential to produce reservoirs with both anomalously heavy and anomalously light oxygen isotopes. Most sediments are also strongly  $^{18}\text{O}$ -enriched, with  $\delta^{18}\text{O} \approx 10\text{--}30\%$  (e.g., [46]). Using the same proportions of basalt to sediment as in the previous example, and assuming a  $\delta^{18}\text{O}$  of 9‰ for altered basalt and 20‰ for sediment, we can generate a  $\delta^{18}\text{O}$  enrichment in the hybrid mantle of  $\sim 1.2\%$  by adding 30% recycled material to mantle peridotite, again very similar to the magnitude of enrichment observed in Koolau olivines. Although a mixture of normal upper mantle with recycled crust and sediment can produce the Koolau endmember, the overall Os–O isotope array is better reproduced if the ultramafic component has an isotopic composition similar to the source of Kea-type lavas rather than normal depleted mantle (Fig. 3). We discuss the identity of this Kea endmember in the following section.

Bennett et al. [4] argued against crustal recycling as an explanation for the Koolau component because the high Pb/Os of crust and sediments relative to depleted mantle should result in a sharply hyperbolic mixing array in Pb–Os isotope space. Such a mixing curve would appear to be inconsistent with the sub-linear array actually observed (Fig. 2). However, a significant proportion of the Pb (and other incompatible elements) in recycled crust and sediments is transported into the mantle wedge by melts or fluids generated during subduction (e.g., [39,47]). A mixing curve between depleted peridotite and subduction-modified crust and sediments therefore will be much less hyperbolic than suggested by Pb/Os ratios in un-subducted crust and sediments. The solid mixing curve in Fig. 2c shows the result of mixing depleted peridotite isotopically similar to the Kea source with the same mixture of crust and sediment as in the previous examples. We assume that approximately 70% of the Pb in the recycled components was removed during subduction, which is consistent with estimates based on the partitioning of Pb between fluids and eclogite [47]. In contrast with the results of Bennett et al. [4], we find that recycled crust and sediments can reproduce the observed correlation between Pb- and Os-isotopes provided that a substantial proportion of the Pb in the subducted

components is removed during the subduction process.

Given the large range in  $\delta^{18}\text{O}$  observed in altered oceanic crust and pelagic sediments and uncertainty regarding the age of the subducted crust or the amount of Pb that is removed during subduction, very different proportions of crust and sediment could produce similar enrichments as in the above examples. However, both the Os–O and the Os–Pb correlations in Hawaiian lavas suggest that the recycled Koolau component is predominantly recycled basaltic crust with only a small amount of sediment present. Mixing of pure sediment produces too great an increase in  $\delta^{18}\text{O}$  and decrease in  $^{206}\text{Pb}/^{204}\text{Pb}$  for a given increase in  $^{187}\text{Os}/^{188}\text{Os}$  in the hybrid source (Figs. 2 and 3). The amount of sediment in the Koolau source is therefore probably no more than  $\sim 5\%$  of the total recycled crust present.

#### 4. Nature of the Kea component

Shield lavas from Mauna Kea and Kilauea define a second endmember in the Hawaiian isotopic array (Fig. 1). In contrast to Koolau-type lavas, these lavas are characterized by the most ‘depleted’ or MORB-like isotopic signatures of any Hawaiian shield lavas (e.g., [8]). As with the Koolau source, several hypotheses have been proposed to explain the nature and origin of the Kea source. The relatively depleted Sr- and Nd-isotopic character of Kea lavas has led to suggestions that depleted upper mantle asthenosphere, perhaps entrained during ascent of the Hawaiian plume, is involved in the generation of these lavas [11,48]. However, Kea-endmember lavas possess anomalously high  $^{87}\text{Sr}/^{86}\text{Sr}$  at a given  $\epsilon_{\text{Nd}}$ , and are displaced to the right of the normal mantle array [49]. This has prompted suggestions that melting or assimilation of seawater-altered Pacific lithosphere (mantle or crust) is involved in generation or modification of Kea-type lavas [5,8,10,49]. In the following discussion, we reveal the shortcomings of both asthenospheric and in situ lithospheric sources for Kea lavas, and argue that, like the Koolau source, the Kea source represents a recycled component present in the Hawaiian plume itself.

##### 4.1. Depleted asthenosphere

Fluid dynamic models suggest that ascending mantle plumes should entrain upper mantle asthenosphere [50]. However, the anomalously high  $^{87}\text{Sr}/^{86}\text{Sr}$  in Kea-type lavas compared with MORB cannot be generated simply by melting of entrained MORB-source asthenosphere. The O-isotope results of Eiler et al. [5,10] further preclude simple generation of Kea-type lavas from depleted MORB-source asthenosphere. Olivine phenocrysts from Mauna Kea and other Kea-type lavas have  $\delta^{18}\text{O}$  values between 4.6 and 5.0‰, distinctly lower than most upper mantle peridotites. Furthermore, oxygen isotopes are correlated with radiogenic isotopes. These correlations suggest a common source for  $^{18}\text{O}$ -depleted material and Kea-type radiogenic isotope signatures. We therefore concur with Eiler et al. [5,10] that it is unlikely that Kea-type lavas are generated from normal asthenospheric mantle.

##### 4.2. Pacific crust

The elevated Sr-isotopes and especially the low  $\delta^{18}\text{O}$  values of Kea-type lavas strongly suggest their derivation from a component which has undergone high-temperature alteration from seawater, because hydrothermally altered oceanic crust (and in some cases serpentinized or otherwise altered lithospheric mantle) is the only known major reservoir of low- $\delta^{18}\text{O}$  material in the silicate earth. The general similarity of the Kea endmember with modern Pacific MORB led Eiler et al. [5,10] to argue that the Kea component is local hydrothermally altered Pacific crust.

Os–O isotope correlations in Hawaiian lavas preclude the large amounts of crustal assimilation ( $>10$ – $20$  wt.%) required to shift  $\delta^{18}\text{O}$  values in Mauna Kea olivines from ‘normal mantle’ values ( $\sim 5.2\%$ ) to observed values (4.6–5.0‰). Hawaiian lavas display a strong positive correlation between Os- and O-isotopes (Fig. 3). Even when only lavas from Mauna Kea are considered, the observed correlation is statistically significant at the 95% confidence level. Because MORB typically has very high Re/Os, the ca. 100 Ma Pacific crust beneath Hawaii should possess extremely radiogenic Os due to in situ decay of  $^{187}\text{Re}$ . The radiogenic nature of the Pacific

crust beneath Hawaii is confirmed by our analyses of MORB-related gabbroic xenoliths from Hualalai volcano. These xenoliths plot within the Sr–Nd isotopic field for modern MORB (Fig. 1). The gabbros have much higher  $^{187}\text{Os}/^{188}\text{Os}$  ( $^{187}\text{Os}/^{188}\text{Os} = 0.225$  to  $0.539$ ) than any Hawaiian lava. Therefore, if the low  $\delta^{18}\text{O}$  and ‘depleted’ Sr- and Nd-isotope values of Kea-type lavas derived from assimilation of hydrothermally altered Pacific lower crust, we would expect a trend towards increasing  $^{187}\text{Os}/^{188}\text{Os}$  with decreasing  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{18}\text{O}$ , the exact opposite of what we observe.

Pb- and Sr-isotopes in the gabbroic xenoliths also do not support derivation of Kea-type isotopic signatures from assimilation of in situ Pacific crust. The Kea endmember is characterized by the highest  $^{206}\text{Pb}/^{204}\text{Pb}$  values of any Hawaiian lavas. If in situ Pacific crust were the source of these signatures, we would expect the MORB-related gabbroic xenoliths to possess similar Pb-isotopes. In contrast, most of the gabbros we analyzed have lower  $^{206}\text{Pb}/^{204}\text{Pb}$  than observed in Kea-type lavas, with  $^{206}\text{Pb}/^{204}\text{Pb}$  ranging from 18.0–18.6. Furthermore, the only gabbros with elevated  $^{206}\text{Pb}/^{204}\text{Pb}$  have much higher  $^{207}\text{Pb}/^{204}\text{Pb}$  than Kea-type lavas. The MORB gabbros also have significantly lower  $^{87}\text{Sr}/^{86}\text{Sr}$  for a given  $^{206}\text{Pb}/^{204}\text{Pb}$  than do Mauna Kea lavas (Fig. 4). Assimilation of Pacific crust with a Pb- and Sr-isotope composition similar to that recorded in the Hualalai gabbros therefore cannot account for the high- $^{206}\text{Pb}/^{204}\text{Pb}$  Kea signature.

MORB recovered from ODP Site 843 approximately 400 km west of Hawaii have more radiogenic Pb-isotopes than the Hualalai gabbros ( $^{206}\text{Pb}/^{204}\text{Pb} = 18.2$ – $18.8$  versus  $18.0$ – $18.6$ ; [51]). However, as with the gabbros these basalts have systematically higher  $^{207}\text{Pb}/^{204}\text{Pb}$  for a given  $^{206}\text{Pb}/^{204}\text{Pb}$  than do Mauna Kea lavas ( $^{207}\text{Pb}/^{204}\text{Pb} = 15.51$ – $15.56$  in ODP Site 843 basalts versus  $15.45$ – $15.49$  in Mauna Kea lavas). Assimilation of MORB crust similar to the ODP Site 843 basalts therefore cannot account for the low  $^{207}\text{Pb}/^{204}\text{Pb}$  in Mauna Kea lavas any more than assimilation of the MORB gabbros.

Could deeper portions of the Pacific crust not sampled by either the gabbroic xenoliths or the ODP Site 843 basalts have systematically higher  $^{206}\text{Pb}/^{204}\text{Pb}$  and lower  $^{207}\text{Pb}/^{204}\text{Pb}$ , as required for any hypothetical assimilant? U-addition of Pacific

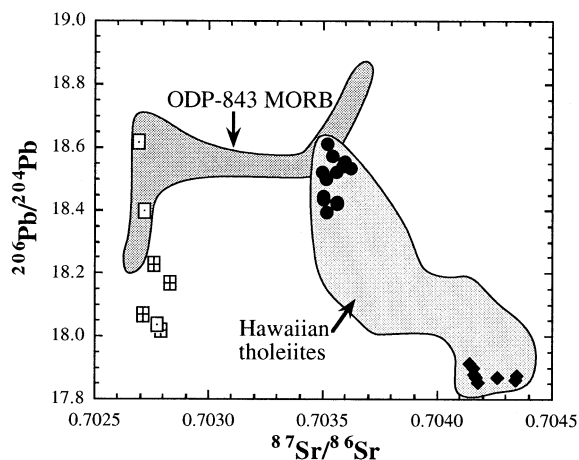


Fig. 4.  $^{206}\text{Pb}/^{204}\text{Pb}$  versus  $^{87}\text{Sr}/^{86}\text{Sr}$  for Hawaiian shield tholeiites. The ‘Kea’ endmember is characterized by high  $^{206}\text{Pb}/^{204}\text{Pb}$  ( $\sim 18.6$ ) but moderate  $^{87}\text{Sr}/^{86}\text{Sr}$  ( $\sim 0.7035$ ). Most samples of the Pacific crust from either ODP Site 843 or from the MORB-related gabbroic xenoliths do not have Sr–Pb isotopic values similar to that identified for the Kea endmember. Symbols are as in Fig. 1.

MORB during hydrothermal alteration could in principle have produced a crustal reservoir with high U/Pb which evolved high  $^{206}\text{Pb}/^{204}\text{Pb}$  over the past 100 Ma [5]. However, U-addition in MORB is usually a feature of low-T alteration in the upper oceanic crust [52]. As a result, altered MORB with high U/Pb will also tend to have high  $\delta^{18}\text{O}$  (as do the high- $\mu$ , high- $^{206}\text{Pb}/^{204}\text{Pb}$  MORB from ODP Site 843; [51]). In contrast, lower-crustal gabbros tend to have low U/Pb. For example, MORB-related gabbro xenoliths from Gran Canaria have  $\mu$  values between 3 and 6, much lower than in the overlying basalts [53]. Therefore, the combination of low- $\delta^{18}\text{O}$  and high- $\mu$  that is required for any hypothetical crustal assimilant is unlikely to be present in the local Pacific crust. In summary, the Os- and Pb-isotope composition of lower Pacific crust in the vicinity of Hawaii preclude generation of the low- $\delta^{18}\text{O}$  Kea endmember by assimilation of the local Pacific crust.

#### 4.3. Pacific lithospheric mantle

It is unlikely that any mafic portion of the lower Pacific crust would have sufficiently unradiogenic Os to produce the low  $^{187}\text{Os}/^{188}\text{Os}$  values in low  $\delta^{18}\text{O}$  Kea-type lavas. However, ultramafic components in

the lower crust and uppermost mantle are expected to have considerably lower  $^{187}\text{Os}/^{188}\text{Os}$ . In principle, it is possible that high-temperature hydrothermal alteration of the uppermost mantle could generate a low- $\delta^{18}\text{O}$  reservoir within the upper Pacific lithosphere while preserving the low  $^{187}\text{Os}/^{188}\text{Os}$  characteristic of mantle peridotite. Could either bulk assimilation of hydrothermally altered ultramafic material or partial melting of such material produce the low  $\delta^{18}\text{O}$ , isotopically ‘depleted’ Kea signature? Although the O- and Os-isotope data alone do not preclude this model, other features of the Kea-type lavas strongly discount such massive assimilation of shallow lithospheric mantle.

Trace element ratios in Hawaiian lavas require the presence of garnet as a residual phase during melt generation [54,55]. For example, Yb contents in fractionation-corrected Hawaiian lavas are nearly constant, and Sm/Yb ratios are high. Both of these features reflect the compatibility of Yb in garnet and the presence of garnet in the Hawaiian source. Because hydrothermal alteration of lithospheric mantle is restricted to shallow portions of the lithosphere at pressures below the stability field of garnet, partial melts from hydrothermally-altered Pacific lithosphere will necessarily lack a residual garnet signature. For example, plagioclase lherzolite xenoliths from Oahu are LREE-depleted ([56]; unpublished data), as expected for lithospheric mantle that is the residue from MORB generation. Partial melts of similar depleted lithospheric mantle will have much lower Sm/Yb than primary Hawaiian melts. Assimilation of either bulk lithospheric mantle or partial melts from the shallow mantle will decrease the Sm/Yb of the hybrid lavas. Therefore, if the isotopic signatures of Kea-type lavas derive from the lithospheric mantle, we should observe a weaker ‘garnet signature’ in these low- $\delta^{18}\text{O}$  lavas. However, Mauna Kea lavas have Sm/Yb ratios that extend to higher values than in lavas from either Loihi or Mauna Loa.

Fig. 5 shows the result of mixing a parental Hawaiian magma with either bulk depleted mantle or a small-degree partial melt of depleted mantle with a plagioclase lherzolite mineralogy [57]. The mixing trends fail to produce the necessary increase in  $\epsilon_{\text{Nd}}$  observed in low- $\delta^{18}\text{O}$  Kea-type lavas before lowering Sm/Yb to values well below those of the Mauna Kea lavas, even when the starting magma has a Sm/Yb

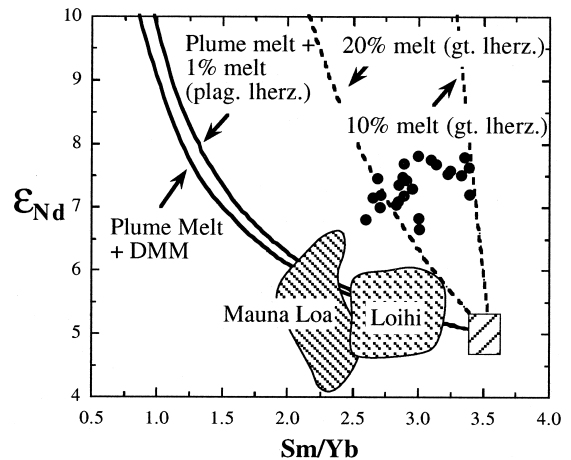


Fig. 5. Sm/Yb versus  $\epsilon_{\text{Nd}}$  for lavas from Mauna Kea, Mauna Loa, and Loihi. Mixing of depleted mantle or 1% partial melt of shallow depleted mantle (plagioclase lherzolite mineralogy) with Hawaiian melts results in nearly identical mixing curves, as shown. In the absence of garnet, partial melts of depleted mantle will have low Sm/Yb. Mixing of Hawaiian melts with melts from shallow depleted mantle therefore cannot generate Kea-type lavas unless the primary, uncontaminated melt is isotopically very similar to the final basalt. In contrast, partial melting of garnet lherzolite can produce the high Sm/Yb ratios in Hawaiian basalts (dashed mixing curves). Modal abundances and distribution coefficients for plagioclase and garnet lherzolite and Sm, Nd, and Yb abundance for typical depleted mantle from Ref. [57] and references therein. Minor variations in modal abundance or distribution coefficients will not significantly effect the results shown. ‘Primary’ Hawaiian melt [Nd] = 20.6 ppm, [Sm] = 6.4 ppm, [Yb] = 1.8 ppm. Data for Mauna Kea lavas from Refs. [11,13]. Symbols are as in Fig. 1.

ratio at the high extreme for Hawaiian tholeiites. The only way these mixing curves can intersect the bulk of the Mauna Kea data is if the parental magmas possess essentially identical Sr-, Nd- and Pb-isotope values as the final lavas, or in other words if assimilation does not affect these isotope values. However, this scenario would require a decoupling between major element tracers of assimilation (e.g.,  $\delta^{18}\text{O}$ ) and incompatible element tracers, and the strong correlations between  $\delta^{18}\text{O}$  and Sr-, Nd-, and Pb-isotopes [5] do not support this.

Previous authors have argued that Hawaiian melts may interact with the lithospheric mantle and yet retain their garnet signature (e.g., [58]). However, such considerations are largely irrelevant to the current discussion. It is true that many potential lithosphere-derived components have such low REE abundances

relative to Hawaiian lavas that assimilation of these components will have only modest effects on trace element ratios and abundances. However, the assimilation model of Eiler et al. [5] requires that a large fraction of the Sr, Nd, and Pb ( $\sim 40\%$  in the case of Nd) in Kea-type lavas derives from the assimilated material. It is extremely difficult to generate an assimilant from (garnet-free) shallow depleted mantle that can contribute sufficient Nd to a melt to shift its Nd-isotopes towards Pacific MORB values without simultaneously depressing the Sm/Yb of the resultant melts.

In summary, no component in the local Pacific crust or lithospheric mantle has been identified that can account for all of the isotopic and trace element characteristics of the Kea endmember. In contrast with Eiler et al. [5,10], we believe the isotopic and trace element constraints reviewed above preclude the generation of the Kea endmember isotopic signatures through shallow level assimilation processes. Instead, these signatures reflect the isotopic character of the sub-lithospheric (plume) source.

#### 4.4. Recycled oceanic lithosphere

The low  $\delta^{18}\text{O}$  of Kea-type lavas necessitates some hydrothermally altered source component, but neither the Pacific crust nor the upper lithospheric mantle possess the required mineralogy or isotopic characteristics of the Kea endmember. We have already reviewed several lines of evidence which indicate the presence of ancient recycled oceanic crust + sediment in the plume source of Koolau lavas. Given this evidence for recycled upper crust in the Hawaiian plume, it seems likely that recycled lower crust and lithospheric mantle from the same subducted packet is also present in the plume.

Several ophiolite sequences contain hydrothermally altered lower crust/upper mantle ultramafic sections with anomalously low  $\delta^{18}\text{O}$  (e.g., the MacQuarie ophiolite [45] and the Onverwacht Group [59]). Such ultramafic mantle/lower crust could provide a reservoir of low  $\delta^{18}\text{O}$ , low  $^{187}\text{Os}/^{188}\text{Os}$  material suitable for the Kea source. Furthermore, oceanic lithospheric mantle is expected to be incompatible-element depleted. As a result, even ancient recycled lithospheric mantle may possess relatively depleted Sr-, Nd-, and Pb-isotope values. Although the iso-

topic composition of recycled lithospheric mantle could qualitatively resemble the in situ Pacific lithosphere, the location of this component within the Hawaiian plume results in the stabilization of garnet in the source. This in turn produces melts with higher Sm/Yb than would be possible from partial melts of the shallow Pacific lithosphere (Fig. 5).

Fig. 6 schematically illustrates our model for generation of the Koolau and Kea endmembers. A

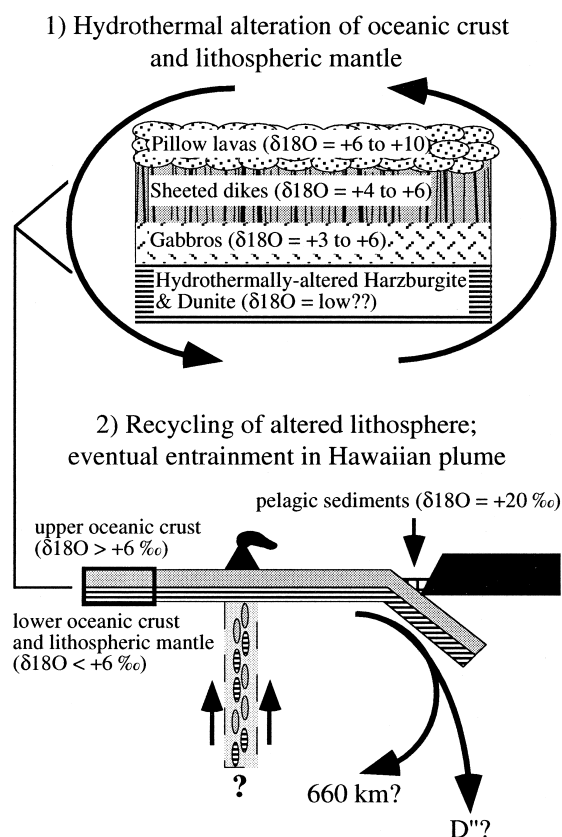


Fig. 6. Schematic illustration of our model for the origin of the Koolau and Kea isotopic components in the Hawaiian plume. Ocean crust cross-section reflects typical variations in  $\delta^{18}\text{O}$  preserved in ophiolite complexes. Recycling of oceanic lithosphere will typically introduce both  $^{18}\text{O}$  enriched and depleted components into the mantle. Preservation of one without the other would require physical separation of the upper crust and sediments from the lower crust/lithospheric mantle, and is therefore not generally expected. Mixing of the recycled lithospheric package will dampen O-isotope heterogeneities. The preservation of both light and heavy O-isotope components in the Hawaiian plume therefore suggests that mixing of the recycled lithospheric packet has been relatively inefficient.

distinct advantage of this model is that a single process (lithosphere subduction) generates both the Kea and Koolau endmembers. An important implication of this model is that the recycled lithospheric packet sampled by Hawaiian shield lavas has remained relatively intact and unmixed for a geologically long period of time. This ineffective mixing of recycled material into the convecting mantle may indicate that recycled lithosphere can behave as a lithologically distinct unit, perhaps because the presence of eclogite provides the recycled packet greater strength (higher viscosity) than the surrounding mantle. Alternatively, the preservation of short length scale heterogeneities associated with recycled lithosphere may reflect longer mixing times in the (high viscosity) lower mantle than in the upper mantle.

## 5. Conclusions

Os-isotopes in Hawaiian shield-stage lavas are strongly correlated with other isotopic tracers (Sr, Nd, Pb, and O), suggesting common sources for compatible, major, and incompatible elements in Hawaiian lavas. Isotopic variations in Hawaiian shield lavas can be explained to first order by simple two-component mixing.

Koolau lavas possess the most radiogenic Os-isotopes of any Hawaiian shield, and also possess anomalously heavy O-isotopes. Combined, these two observations place a formidable burden on models which equate the Koolau source with either primitive or metasomatized mantle. In contrast, these features are easily explained by the incorporation of ancient subducted oceanic crust into the Hawaiian plume.

Mauna Kea lavas possess the least radiogenic Os-isotopes of any Hawaiian shield, and anomalously light O-isotopes. Although Sr- and Nd-isotopes in Mauna Kea lavas approach (but do not reach) those of Pacific MORB, Os-, O- and Pb-isotope data for MORB-related gabbro xenoliths preclude generation of Mauna Kea lavas by assimilation of in situ Pacific crust. Assimilation of shallow, hydrothermally altered Pacific lithospheric mantle also cannot account for the Kea-component, because such assimilation would dilute the 'garnet signature' preserved in the trace element ratios of Mauna Kea lavas.

Given the failure of potential in situ components

to account for the Kea source, and also given the strong evidence for recycled material in the Koolau source, the best explanation for the low- $\delta^{18}\text{O}$  Kea component is recycled oceanic lithosphere in the Hawaiian plume. The chemical and isotopic differences between the Koolau and Kea endmembers reflect preferential sampling of upper oceanic crust and sediment in the former, whereas the latter contains a greater proportion of recycled ultramafic components from the hydrothermally altered lower crust and lithospheric mantle. In all probability, the upper crust + sediment present in the Koolau source and the lower crust + lithospheric mantle in the Kea source derive from the same packet of subducted lithosphere. The preservation of this stratigraphically-derived chemical and isotopic heterogeneity in the Hawaiian plume indicates that heterogeneities with length-scales of only a few kilometers can be preserved in the convecting mantle for geologically significant periods of time.

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