

# Generation of Hawaiian post-erosional lavas by melting of a mixed lherzolite/pyroxenite source

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## Abstract

Melting of mafic veins in a marble-cake mantle may play an important role in generating isotopic and chemical heterogeneities in mid-ocean ridge and ocean island basalts. Mafic veins have lower solidi than mantle peridotite and will be preferentially sampled during partial melting, particularly at low melt fractions. However, the abundance of mafic components in the mantle or their role during melt generation has been difficult to quantify because most isotopic systems (e.g. Rb–Sr, Sm–Nd, U–Th–Pb) are not diagnostic of the presence or absence of mafic components. The compatible behavior of Os during mantle melting combined with the incompatible behavior of Re makes the Re–Os isotopic system uniquely well suited for distinguishing mafic and ultramafic contributions to melt generation. Almost all peridotites have low  $^{187}\text{Os}/^{188}\text{Os}$  (e.g. chondritic to subchondritic). In contrast, mafic rocks have much higher Re/Os than peridotites, which results in the rapid ingrowth of  $^{187}\text{Os}$  and the development of large isotopic contrasts between mafic and ultramafic components within the mantle. In this paper, we show that Os-isotopes in Hawaiian post-erosional lavas extend to more radiogenic values than are found in Hawaiian lherzolites, abyssal peridotites or most other ultramafic samples. Os-isotopes are not correlated with other isotopic tracers, in contrast with plume-derived Hawaiian shield-stage lavas. The lack of correlation between Os-isotopes and Sr-, Nd- or Pb-isotopes and the more ‘depleted’ or MORB-like Sr–Nd isotopic signature of the post-erosional lavas relative to other Hawaiian lavas precludes significant melt input from the Hawaiian plume. However, Os-isotopes are correlated with major and trace elements. Lavas with more radiogenic Os-isotope compositions have higher silica and alumina and lower calcium and incompatible trace element abundances than lavas with less radiogenic Os-isotopes. These correlations result from mixing of pyroxenite- and peridotite-derived melts, both likely derived from the ~100 Ma Pacific lithospheric mantle. © 2000 Elsevier Science B.V. All rights reserved.

**Keywords:** partial melting; mafic composition; isotope ratios; ocean-island basalts; mid-ocean ridge basalts

## 1. Introduction

Melting of mafic veins in the mantle is a potential source of chemical and isotopic heterogeneity in mantle-derived melts [1–5]. Because mafic components in the mantle (e.g. pyroxenites or eclo-

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gites) have lower solidi than peridotite [6,7], low-degree partial melts will preferentially sample such components. Previous studies have suggested that melting of garnet pyroxenite may in part explain the ‘garnet signature’ inferred from trace element ratios and U–Th isotopic disequilibrium observed in many mid-ocean ridge basalts (MORB) and ocean island basalts (OIB) [7–9]. However, the chemical and isotopic signatures of pyroxenite melting at the relatively high degrees of melting responsible for the generation of MORB are likely to be subtle, because pyroxenite-derived melts will be greatly diluted by peridotite-derived melts.

In contrast, highly alkalic, incompatible element-enriched basalts found in many ocean island volcanic suites are generated by much smaller extents of partial melting than MORB [10] and may possess a more pronounced signature of pyroxenite melting. Os-isotopes are particularly sensitive to pyroxenite melting because of the high Re/Os and  $^{187}\text{Os}/^{188}\text{Os}$  ratios present in most pyroxenites [11–13]. This contrasts with the unradiogenic  $^{187}\text{Os}/^{188}\text{Os}$  ( $< 0.130$ ) found in almost all peridotites (e.g. [14–17]). We have examined the Os-isotope systematics of Hawaiian post-erosional lavas from Kauai and Oahu to evaluate the role of pyroxenite melting in their generation. Highly radiogenic Os-isotopes in these lavas and correlations between Os-isotopes and major elements suggest that a substantial fraction of post-erosional melts derives from mafic rather than peridotitic mantle components.

## 2. Post-erosional lavas

Hawaiian post-erosional or rejuvenated-stage lavas have erupted on several volcanoes after a period of up to 2.5 Myr quiescence following cessation of shield- and post-shield-stage volcanism [18]. Similar rejuvenated-stage lavas are found on many other ocean islands, although the age gap between the shield-stage volcanism and rejuvenated-stage volcanism in some instances is much longer than observed for the Hawaiian Islands [19]. At Hawaii, these lavas are typically more silica-undersaturated and incompatible element-

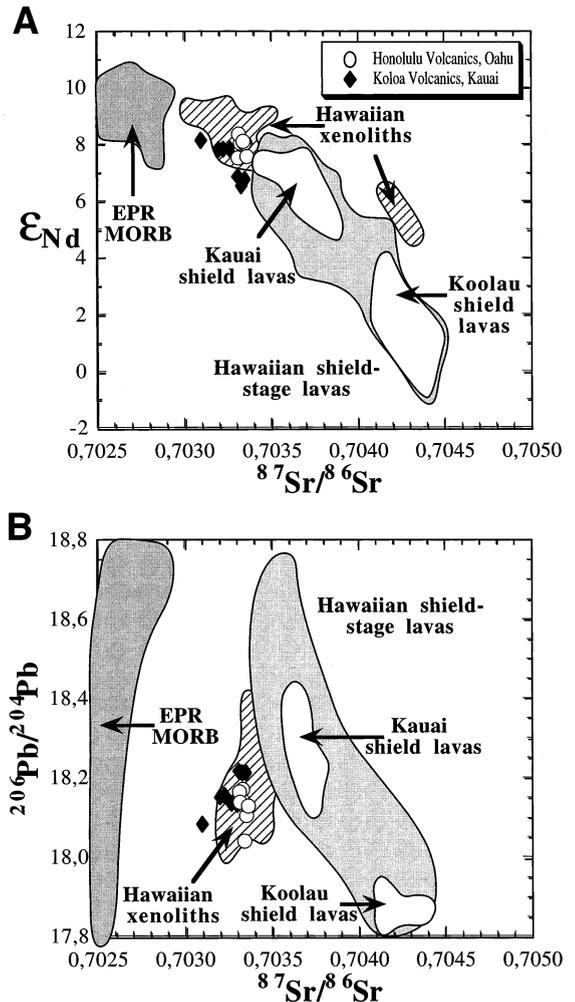


Fig. 1. Sr–Nd and Sr–Pb correlations in Hawaiian shield-stage and post-erosional lavas. The post-erosional lavas are isotopically distinct from shield lavas, and plot closer to the field for East Pacific Rise MORB. Lherzolite and pyroxenite xenoliths have Sr–Nd–Pb isotopes similar to post-erosional lavas. Fields are from numerous literature sources, available from the GEOROC database (<http://georoc.mpch-mainz.gwdg.de>). Separate fields for pyroxenite and lherzolite xenoliths are not shown because the Sr-, Nd- and Pb-isotope compositions of pyroxenite and lherzolite xenoliths completely overlap.

enriched than shield- and post-shield-stage lavas, and have distinctly more ‘depleted’ or MORB-like Sr- and Nd-isotopes than their earlier shield counterparts (Fig. 1) [20–22]. Lavas compositionally and isotopically similar to the post-erosional lavas

have also been found north of Oahu along the Hawaiian Arch, a flexural arch that results from the lithospheric loading of the Hawaiian Islands [23–25].

The origin of the post-erosional and North Arch lavas remains one of the more puzzling questions regarding Hawaiian volcanism. The depleted isotopic character of post-erosional lavas, the age gap between them and earlier shield lavas, and trace element evidence for the presence of hydrous phases in the source of some post-erosional lava suites (e.g. the Honolulu Volcanic Series, or HV) [26,27], suggest they may form by partial melting of lithospheric mantle heated from below by the Hawaiian plume [28,29]. Melting of entrained asthenosphere at the plume periphery might also explain the age and spatial distribution of the post-erosional and North Arch lavas and the isotopic differences between them and earlier (presumably plume-derived) shield-stage lavas [30]. Ribe and Christensen [31] recently proposed that post-erosional lavas derive from a secondary melting region within the ascending Hawaiian plume that results from the lateral spreading of buoyant plume material beneath the lithosphere. Although this model can account for both the time gap between shield-stage and post-erosional volcanism and the relative volumes of erupted material, it does not explain the isotopic differences between shield- and post-erosional lavas or the origin of the North Arch lavas.

### 3. Samples and results

Isotopic data for post-erosional lavas from Oahu and Kauai and for pyroxenite and lherzolite xenoliths from Oahu are presented in Table 1. Sr-, Nd- and Pb-isotope analyses of acid-leached samples were obtained using procedures similar to those outlined in [32]. Re and Os were measured by negative thermal ionization mass spectrometry. For the lavas and pyroxenite xenoliths, spike-sample equilibration and Re and Os separation and purification were achieved using a modified Carius tube technique [33], and extraction of Os and Re via solvent extraction [34] and anion resin column exchange [35] techniques. The spinel lher-

zolite xenoliths, however, were measured as part of an earlier project, at which time Os was pre-concentrated via NiS fusion [36]. High Re blank associated with the NiS fusion technique precludes direct measurement of Re on the same sample aliquots for which Os was measured. Re concentrations are therefore not reported for the lherzolite xenoliths.

We selected samples from the Honolulu Volcanics (HV), Oahu and the Koloa Volcanics (KV), Kauai that span a wide range in major and trace element compositions (M.G., unpublished data; [37]). The samples analyzed are all near-primitive lavas with high MgO (>10.7 wt%) and Ni (>220 ppm) content but with widely varying SiO<sub>2</sub> content (36.7–45.4 wt%). Most HV lavas are more silica-undersaturated than the KV lavas, and have greater enrichments in incompatible trace elements. These differences reflect lower degrees of melting and/or greater incompatible element and volatile (especially CO<sub>2</sub>) enrichment of the source of the HV lavas [25,26] relative to the source of the KV lavas. Os-isotopes in both suites span a large range, with <sup>187</sup>Os/<sup>188</sup>Os ranging from 0.135 to 0.160. Os concentrations also span a wide range, from ~20 to 90 ppt in the KV lavas, and from ~50 to 280 ppt in the HV lavas. In contrast with Hawaiian shield lavas and most other basaltic lava suites, Os concentration is not clearly correlated with either MgO or Ni content. Correlations between MgO, Ni and Os in many basalt suites [38–40] have been interpreted as reflecting the compatibility of Os in olivine or olivine-hosted phases (e.g. Cr-spinel) during fractional crystallization [39,40]. The lack of such a correlation in the present lavas is consistent with previous interpretations that these lavas are near-primary mantle melts that have experienced minimal fractional crystallization [26,37].

Many HV and KV lavas contain mantle xenoliths [26,41]. We attempted to analyze samples from xenolith-free or xenolith-poor flows to avoid Os contamination from disaggregated xenoliths [38]. Duplicate Os-isotope analyses of separate powder splits were conducted for several samples to assess the magnitude of within-sample Os-isotope heterogeneity. In addition, two samples from separate outcrops of the Kamaikai flow were

Table 1  
Isotopic compositions of Hawaiian post-erosional lavas and hosted xenoliths

	[Re]	[Os]	$^{187}\text{Os}/^{188}\text{Os}$	$^{87}\text{Sr}/^{86}\text{Sr}$	$\epsilon_{\text{Nd}}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$
<i>Honolulu Volcanics (HV), Oahu</i>								
Kaupo	341	189	0.1503	0.70335	8.0	18.10	15.46	37.78
Kaupo dup.	307	90	0.1507	–	–	–	–	–
Kamanaiki-A	569	279	0.1465	0.70332	7.9	18.14	15.44	37.76
Kamanaiki-B	495	277	0.1424	0.70333	7.8	18.17	15.45	37.80
Kamanaiki-B dup.	–	232	0.1449	–	–	–	–	–
Ainoni	401	101	0.1598	0.70331	7.5	18.16	15.44	37.79
BP-F1	110	64	0.1432	0.70333	8.0	18.13	15.45	37.79
Rocky Hill	374	59	0.1419	0.70335	8.0	–	–	–
Makuku	145	47	0.1517	0.70332	8.3	18.14	15.44	37.74
Kalama	162	78	0.1538	0.70336	7.6	18.13	15.45	37.78
Molili	150	108	0.1351	0.70334	8.1	18.04	15.44	37.70
<i>Koaloa Volcanics (KV), Kauai<sup>a</sup></i>								
94-PRKP-4	66	56	0.1500	0.70326	7.9	18.14	15.43	37.79
94-PRKP-6	80	74	0.1438	0.70322	7.9	18.16	15.45	37.87
94-PRKP-12	44	73	0.1587	0.70329	7.8	18.14	15.42	37.79
94-PRKP-12 dup.	52	69	0.1578	–	–	–	–	–
94-PRKP-14	16	69	0.1410	0.70320	7.9	18.15	15.44	37.84
95HTZ425	30	74	0.1380	0.70333	6.6	18.22	15.43	37.91
95HTZ460	–	54	0.1387	0.70331	6.9	18.22	15.44	37.94
95HTZ525	19	89	0.1366	0.70335	6.8	18.21	15.44	37.95
95HTZ1000	127	21	0.1430	0.70309	8.2	18.09	15.44	37.73
<i>Pyroxenite xenoliths<sup>b</sup></i>								
114884-11	793	167	0.1513	0.70333	8.7	18.18	–	37.67
114884-27	998	6587	0.1534	0.70324	9.3	18.11	–	37.89
114923-29	992	266	0.1588	0.70329	8.4	18.21	–	37.98
114923-141	708	53	0.1750	0.70323	9.0	18.09	–	37.86
114923-147	248	1258	0.1339	–	–	–	–	–
114923-192	118	370	0.1386	–	–	–	–	–
<i>Lherzolite xenoliths<sup>b</sup></i>								
114744-1	–	1021	0.1257	0.70338	8.6	–	–	–
114744-2B	–	1348	0.1269	0.70303	9.2	–	–	–
114745-2	–	431	0.1258	0.70326	8.8	–	–	–
114745-3	–	441	0.1297	0.70327	8.7	18.19	–	37.98
114886-5	–	1417	0.1244	–	–	–	–	–
114908-7	–	3970	0.1252	–	–	–	–	–
114908-20	–	1204	0.1238	0.70336	7.5	–	–	–
114908-22	–	1601	0.1259	–	–	–	–	–

Re and Os concentrations in ppt.  $^{187}\text{Os}/^{188}\text{Os}$  reported as measured values. Age corrections for all samples are insignificant ( $< 5\%$ ).  $^{87}\text{Sr}/^{86}\text{Sr}$  reported relative to a value of 0.71025 for NBS 987.  $\epsilon_{\text{Nd}}$  calculated using  $^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}} = 0.512638$ , 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  $\pm 0.4\epsilon$  ( $2\sigma$ ). Pb-isotopes were corrected for mass fractionation using NBS 981 values of Todt et al. [70]. Reproducibility of measured ratios is better than 0.05% a.m.u.<sup>-1</sup>.  $^{187}\text{Os}/^{188}\text{Os}$  measured on the DTM J-M shelf standard during the course of this study was 0.1742 ( $\pm 3\%$   $2\sigma$ ).  $^{207}\text{Pb}/^{204}\text{Pb}$  data from [44] are not included because of reported large uncertainty in mass fractionation. Xenoliths are from the Dale Jackson Collection at the National Museum of Natural History. Total procedural blanks are  $< 1$  pg for Os,  $< 2$  pg for Re,  $< 140$  pg for Sr,  $< 15$  pg for Nd and  $< 80$  pg for Pb.

<sup>a</sup>Sr- and Nd-isotope data from [37].

<sup>b</sup>Italicized Sr-, Nd- and Pb-isotope data from [44]. Other data are from this study.

analyzed to assess larger-scale flow heterogeneities. The measured isotopic and concentration variations for duplicate analyses are considerably larger than the analytical uncertainties or typical sample reproducibility based on previous studies. This apparently real within-flow heterogeneity, which may reflect variable amounts of xenocrystic olivines within the flows, is a source of scatter in the isotope composition trends discussed below. However, the scatter observed within an individual flow is small compared to the total observed range in Os-isotopes.

Mantle xenoliths present in HV lavas include lherzolites, dunites and pyroxenites [42]. Pyroxenite and lherzolite xenoliths span identical ranges in Sr-, Nd- and Pb-isotopes. The majority of both the lherzolites and pyroxenites have Sr-, Nd- and Pb-isotope values similar to the post-erosional lavas [43,44], which suggests a genetic relationship between the lavas and xenoliths. Major and trace element and mineral compositions of HV xenoliths have been previously reported (e.g. [42,45–48]). The lherzolite xenoliths have widely varying REE patterns and abundances, ranging from LREE-depleted to strongly LREE-enriched. These xenoliths have been interpreted as fragments of the ~100 Ma Pacific lithosphere that have been variably metasomatized by LREE-enriched melts [9,48] subsequent to MORB melt extraction. Estimated equilibration temperatures and pressures for the spinel lherzolite xenoliths range from 900 to 1150°C and ~1.2 to 2.6 GPa [9,46].

Pyroxenite xenoliths from the HV lavas include clinopyroxenites (with and without garnet present), websterites and wehrlites [46]. Sen [46] estimated pressures and temperatures of equilibration for the pyroxenite xenoliths between 1.6 and 2.5 GPa (50–75 km) and 1050 and 1120°C, similar to estimates for the spinel lherzolites. Several studies have proposed that the pyroxenite xenoliths represent frozen melts or cumulates formed from earlier alkali-rich magmas passing through the lithosphere, although whether the pyroxenites formed during earlier Hawaiian volcanism (e.g. [45,46]) or near the mid-ocean ridge [44] is unclear. The pyroxenite xenoliths are highly heterogeneous in both phase modal abundances and

major element compositions. Fe-rich garnet lherzolites in the HV xenolith suite appear compositionally intermediate between the spinel lherzolite and garnet pyroxenite xenoliths, and may represent mixtures of lherzolite wallrocks with pyroxenite veins [47]. In contrast with the highly variable REE patterns displayed by the lherzolite xenoliths, the pyroxenite xenoliths have more uniform convex upward REE patterns consistent with clinopyroxene precipitation from an LREE-enriched melt (e.g. [45]). The uniformity of REE patterns in the pyroxenite xenoliths suggests that the pyroxenites were not metasomatically enriched subsequent to their formation. Instead, the melt from which they formed was likely the same metasomatic agent responsible for variable LREE enrichment in the lherzolite xenoliths.

Os-isotopes in the lherzolite xenoliths are much less radiogenic than in the post-erosional lavas.  $^{187}\text{Os}/^{188}\text{Os}$  values for the lherzolite xenoliths (0.124–0.130) are identical to the range reported for abyssal peridotites [14–17]. Os concentrations span a wide range, from 430 ppt to nearly 4 ppb. Although the Hawaiian lherzolites show evidence for variable degrees of metasomatic enrichment of incompatible elements (e.g. La/Yb<sub>N</sub> in clinopyroxenes ranges from <0.5 to >4; [48]), there is no correlation between the degree of LREE enrichment and Os-isotopes (Fig. 2).

In contrast with the lherzolite xenoliths, the pyroxenite xenoliths possess more variable and more radiogenic Os-isotopes ( $^{187}\text{Os}/^{188}\text{Os}$  = 0.134–0.175), and extend to higher values than observed in the host lavas. Os concentrations show extreme variation, ranging from 50 ppt to 6.6 ppb. Os concentration does not obviously correlate with the modal phase abundances observed in the handsamples. However, sulfides are common in the pyroxenite xenoliths [46], and heterogeneous distribution of Os-rich sulfides could account for the large scatter in Os abundances. In particular, the high Os concentration in sample 114884-27 seems unreasonable given the MgO content of the bulk xenolith (16.9 wt%; [45]), and probably results from such a ‘nugget effect’.

The pyroxenite xenoliths possess higher average Re concentrations than either the post-erosional lavas or most other Hawaiian or OIB lavas. Four

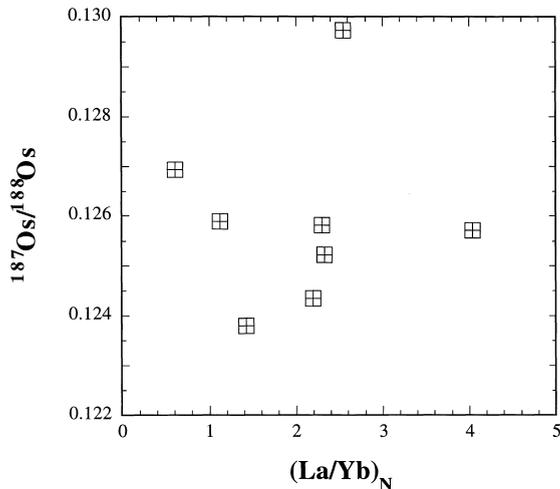


Fig. 2.  $^{187}\text{Os}/^{188}\text{Os}$  in Hawaiian lherzolite xenoliths versus chondrite-normalized La/Yb in clinopyroxene cores (unpublished data). Hawaiian lherzolites have highly variable REE patterns, ranging from LREE-depleted to spoon- or V-shaped to LREE-enriched (cf. [48]). This variability reflects interaction of the lherzolites with LREE-enriched melts or fluids isotopically similar to Hawaiian post-erosional lavas. However, no correlation between Os-isotopes and indices of metasomatic enrichment (e.g. La/Yb or total REE content) is observed. Mantle metasomatism of the sort experienced by the Hawaiian lherzolites did not significantly effect Os-isotopes.

of the six pyroxenites analyzed have Re concentrations between 700 and 900 ppt. Re/Os ratios also span a large range (0.15–13.5) and extend to higher values than in the post-erosional lavas (0.2–6.4). Two pyroxenite xenoliths with lower Re abundances are garnet wehrlites with high modal olivine, and are similar in appearance to the high-Fe garnet lherzolites described by Sen and Leeman [47]. These two xenoliths also have lower  $^{187}\text{Os}/^{188}\text{Os}$  than other pyroxenite xenoliths. Furthermore, one of the two xenoliths with low Re content, 114923-147, is a composite pyroxenite/lherzolite xenolith and was sampled near the lherzolite contact. The unradiogenic Os in these samples and the low Re abundances (relative to the olivine-poor pyroxenites) probably reflect mixing with Os-rich, Re-poor lherzolite.

The high Re concentrations and Re/Os ratios in the olivine-poor pyroxenites suggest that these pyroxenites are unlikely to have formed directly from earlier Hawaiian magmas. Shield and post-

shield lavas from Oahu average 358 and 305 ppt Re, respectively ([49], this study). Lavas from Kauai have much lower Re concentrations, but it is possible that these low concentrations reflect Re loss during weathering. Re behaves as a moderately incompatible element in most mantle silicate phases, so the concentration of Re in the pyroxenites should be less than or equal to the Re concentration in the melts from which they originated. Re is compatible in garnet [50]. However, the garnet present in Hawaiian pyroxenites is rarely of primary magmatic origin, but instead formed during subsolidus cooling [46]. Therefore, the high Re content in the pyroxenites cannot reflect garnet fractionation from a melt with lower Re content.

Sulfides are also present in the pyroxenites [46], and sulfide precipitation could increase Re content in some samples (e.g. 114884-27). However, Os is much more compatible in magmatic sulfides than Re (e.g. [51]). Therefore, if the high Re abundances in the pyroxenites reflected local accumulation of sulfides, we should observe a trend of increasing Os abundance and decreasing Re/Os with increasing Re abundance. However, with the exception of xenolith 114884-27, the high-Re xenoliths have lower Os abundances than the low-Re xenoliths. Therefore, we conclude that sulfide precipitation cannot explain the combined overall high Re abundance and high Re/Os present in most olivine-poor pyroxenites. Instead, the high Re abundance and high Re/Os of the pyroxenite xenoliths probably reflects derivation from melts with Re abundances significantly higher than commonly observed in Hawaiian lavas.

#### 4. Source of the post-erosional lavas

Os-isotopes in Hawaiian post-erosional lavas are considerably more radiogenic than in Hawaiian lherzolite xenoliths or in abyssal peridotites worldwide. Although abyssal peridotites span a large range in Os-isotopes, all but the most highly altered samples have  $^{187}\text{Os}/^{188}\text{Os} < 0.130$  [14–17]. Melts generated solely by melting of upper mantle peridotite should possess similarly unradiogenic Os-isotopes. The much higher  $^{187}\text{Os}/^{188}\text{Os}$  values

in the post-erosional lavas require an additional contribution from a more radiogenic component. Below, we evaluate whether this radiogenic Os-isotope signature derives from interaction with the  $\sim 100$  Ma Pacific crust, from an enriched plume component, from metasomatically enriched asthenospheric or lithospheric peridotite, or from mafic veins (hereafter loosely termed pyroxenite) within the upper mantle lithosphere or asthenosphere.

#### 4.1. Crustal contamination

The primitive compositions of the lavas analyzed and the presence of mantle xenoliths in many KV and HV lavas indicate that these lavas were not stored at crustal depths for a significant period prior to eruption. Os concentrations in the post-erosional lavas (average  $\approx 100$  ppt) are much higher than in typical MORB (average  $< 10$  ppt; [52]) or lower crustal Pacific gabbros [49]. Therefore, moderate amounts of crustal contamination will produce a relatively small effect on the Os-isotopic composition of erupted lavas. For example, 100 Ma average MORB with 930 ppt Re [53] and 10 ppt Os will have  $^{187}\text{Os}/^{188}\text{Os} \approx 0.85$ . 10% bulk assimilation of MORB crust by a post-erosional lava with an initial Os concentration of 100 ppt and  $^{187}\text{Os}/^{188}\text{Os} = 0.126$  (the approximate average of Hawaiian lherzolites and abyssal peridotites) will produce a hybrid magma with  $^{187}\text{Os}/^{188}\text{Os} \approx 0.134$ . Although this value is higher than the hypothetical initial composition, it is much lower than the average isotopic composition of the post-erosional lavas. To produce the range of Os-isotopes observed in the post-erosional lavas, much more than 10% crustal contamination is required. Such large amounts of crustal contamination are inconsistent with the primitive major element compositions of the erupted lavas.

Furthermore, crustal contamination typically produces an inverse correlation between Os concentrations and isotopic values, because lavas with low Os abundance are more susceptible to contamination than higher-abundance lavas. Correlations between Os abundance and isotopic composition are observed in many suites of

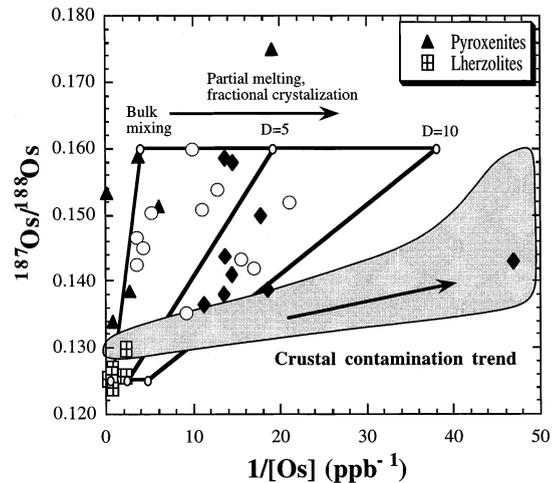


Fig. 3. Os-isotopes versus inverse Os concentration. Strong near-linear correlations are observed in evolved, crustally contaminated lavas from Haleakala, Kilauea and Mauna Kea, as well as in other ocean islands and MORB. Contamination trend defined by data from [39,54,55]. No correlation between Os-isotopes and Os concentration is observed for the post-erosional lavas, and most of the post-erosional lavas have higher Os concentrations than typically observed in crustally contaminated OIBs. Pyroxenite xenoliths have highly variable Os concentrations and isotopic compositions, but on average have higher Os concentrations than the post-erosional lavas. Tie lines reflect either bulk mixing of representative pyroxenite and peridotite or mixing of partial melts from these sources, assuming a bulk distribution coefficient for Os between five and 10. More complex mixing scenarios (e.g. melt/wallrock reaction) are also possible. Symbols for HV and KV lavas as in Fig. 1.

evolved, low-MgO lavas, including lavas from Haleakala Volcano, Maui [39,54,55] (Fig. 3). Such correlations are a clear indication of contamination accompanying or subsequent to fractional crystallization. Because Os abundances in the post-erosional lavas vary over an order of magnitude (20–280 ppt), we would expect a correlation between Os concentration and isotopic composition to exist if crustal contamination was responsible for the high  $^{187}\text{Os}/^{188}\text{Os}$  values present in these lavas. However, no such correlation is observed in either the KV or the HV lavas (Fig. 3).

In addition, KV and HV lavas with high incompatible element abundances have less radiogenic Os-isotopes than lavas with lower abundan-

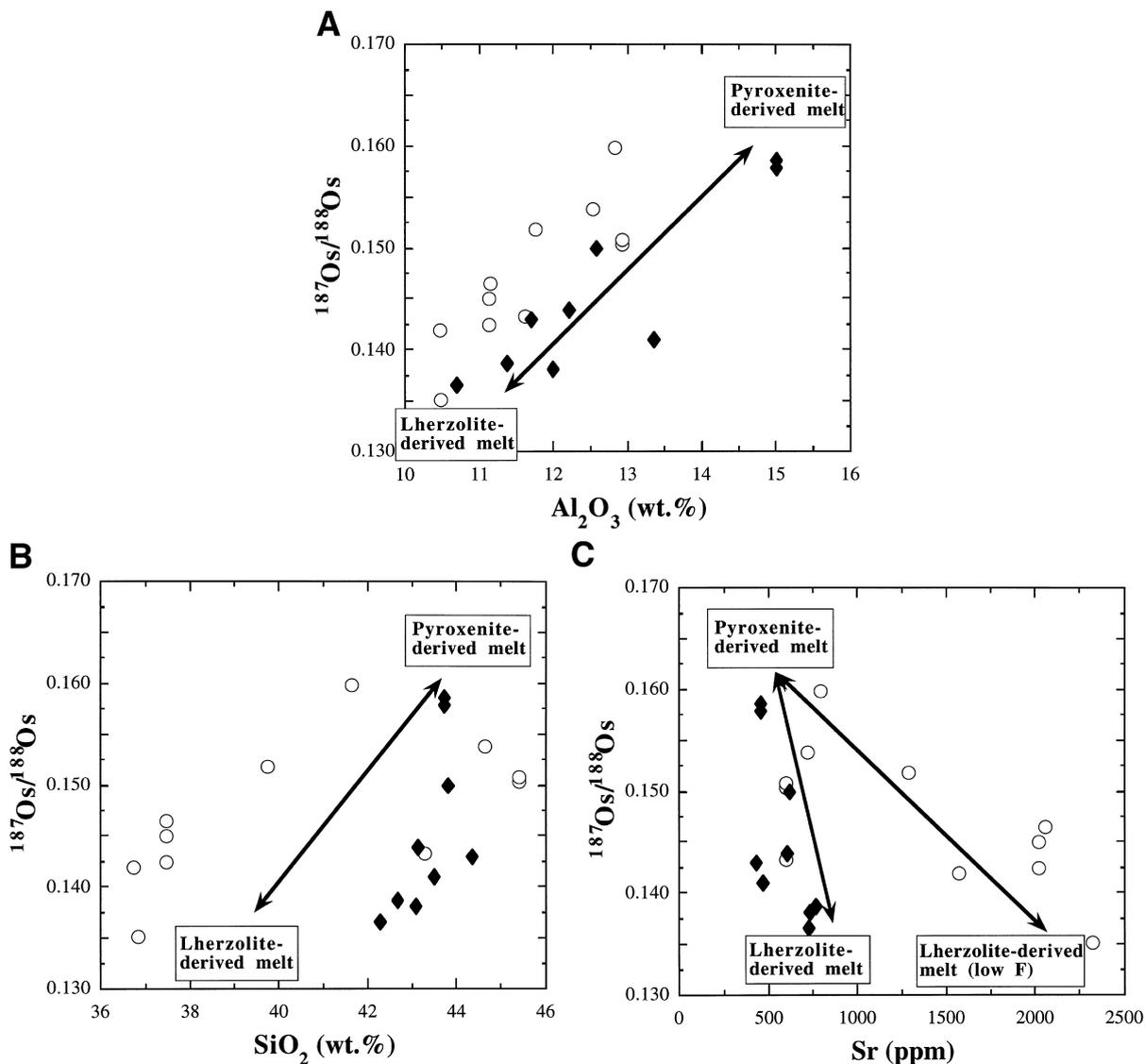


Fig. 4. Os-isotope correlations with  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$  and Sr. Post-erosional lavas from Kauai and Oahu define distinct trends that reflect differences in the melting processes that generated these two suites of lavas. However, both suites define separate positive correlations between Os-isotopes and  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  and a negative correlation with Sr. Three high- $\text{SiO}_2$  lavas from Oahu fall within the Kauai trends for both Os–Si and Os–Sr. Symbols as in Fig. 3. Arrows qualitatively denote effects of mixing high-degree pyroxenite melts with low-degree peridotite melts (see discussion).

ces (e.g. Fig. 4). Although the correlations between incompatible element abundance and Os-isotopes display considerable scatter, the correlations are observed for a large number of elements, including Sr, Th and the LREE. Elements that are incompatible during fractional crystallization are not removed from the magma by crystal fraction-

ation. Therefore, the concentrations of these elements can only be reduced during magma evolution through dilution by the addition of a component with lower incompatible element abundances. Assimilation of high- $^{187}\text{Os}/^{188}\text{Os}$ , incompatible element-poor MORB crust could in principle produce a negative correlation between

Os-isotopes and incompatible elements. However, extremely large amounts of crustal contamination would be required to explain the observed correlations. For example, Sr abundances in HV lavas vary by a factor of four, from 600 to > 2300 ppm (M.G., unpublished data). Bulk assimilation of more than 75 wt% of depleted crust would therefore be necessary to produce the observed range in concentrations via dilution. Such large amounts of assimilation are unreasonable given the primitive nature of the samples we examined. Therefore, the large variations in incompatible element abundances observed in the post-erosional lavas must be a primary feature, reflecting changes in the degree of partial melting or the composition of the mantle source(s). Because Os-isotopes correlate with incompatible element abundance, we again conclude that the Os-isotope values themselves must also be a primary feature unrelated to crustal interaction.

#### 4.2. Plume contribution

Several models involving mixing of plume and depleted mantle components have been proposed to explain the isotopic and trace element characteristics of Hawaiian post-erosional lavas [20,21,26,37]. These models involve either the addition of incompatible element-enriched plume-derived melts or fluids to a depleted mantle source prior to generation of the post-erosional lavas, or alternatively, addition of incompatible element-enriched depleted mantle-derived melts or fluids to a plume source. Metasomatic addition of volatiles or incompatible elements to the source of post-erosional lavas prior to melt generation is not precluded by the Os-isotope data, and may be necessary to explain the extreme incompatible element enrichments and low SiO<sub>2</sub> of some HV lavas [26]. Because some Hawaiian shield-stage lavas possess elevated <sup>187</sup>Os/<sup>188</sup>Os values, we must consider whether the Hawaiian plume could be the (direct or indirect) source of radiogenic Os-isotopes in the post-erosional lavas.

Os concentrations in peridotites are much higher than in most melts or fluids [36,40], in contrast with incompatible elements such as Sr or Nd. As a result, addition of plume-derived melts or fluids

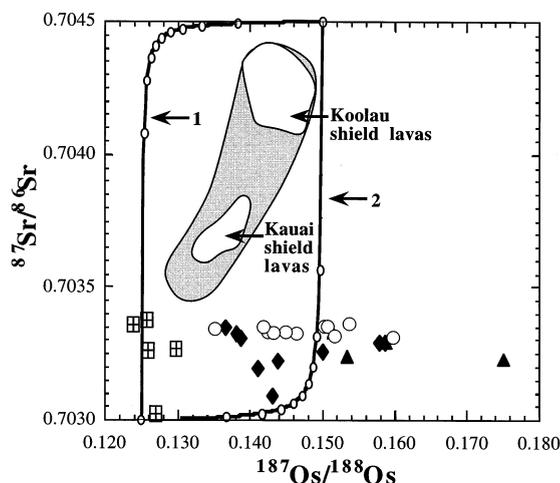


Fig. 5. Post-erosional lavas from both Kauai and Oahu have Os-isotopes that extend to more radiogenic values than are observed in Hawaiian shield lavas (stippled field), and Os-isotopes show no clear correlations with other isotopic tracers. Mixing curves reflect mixing of plume-derived melts with depleted mantle (DM) (curve 1) or depleted mantle-derived melts with the Hawaiian plume (PM) (curve 2). Mixtures of melts with mantle peridotite, whether from the depleted upper mantle or the Hawaiian plume, will produce strongly hyperbolic mixing arrays, so that only very large additions of melt will significantly affect the Os-isotopes of mantle peridotite. Small open circles indicate 10% increments of melt addition. Mixing parameters are as follows: DM [Sr]=13 ppm, <sup>87</sup>Sr/<sup>86</sup>Sr=0.7030, [Os]=2 ppb, <sup>187</sup>Os/<sup>188</sup>Os=0.125; PM [Sr]=20 ppm, <sup>87</sup>Sr/<sup>86</sup>Sr=0.7045, [Os]=2 ppb, <sup>187</sup>Os/<sup>188</sup>Os=0.150; DM-derived melt [Sr]=250 ppm, <sup>87</sup>Sr/<sup>86</sup>Sr=0.7030, [Os]=250 ppt, <sup>187</sup>Os/<sup>188</sup>Os=0.125; PM-derived melt [Sr]=250 ppm, <sup>87</sup>Sr/<sup>86</sup>Sr=0.7045, [Os]=2 ppb, <sup>187</sup>Os/<sup>188</sup>Os=0.150. However, the shape of mixing curves is insensitive to minor changes in mixing parameters provided (Sr/Os)<sub>melt</sub> ≫ (Sr/Os)<sub>mantle</sub>. Fields for Hawaiian shield, Koolau and Kauai shield lavas from literature sources and unpublished data. Symbols as in Fig. 3.

to depleted mantle will have a much more pronounced effect on Sr- or Nd-isotopes than on Os-isotopes. Os-isotopes are strongly correlated with Sr-, Nd- and Pb-isotopes in Hawaiian shield lavas (e.g. Fig. 5) and by inference in the Hawaiian plume itself [49]. Therefore, plume-derived melts with high <sup>187</sup>Os/<sup>188</sup>Os will also have correspondingly high <sup>87</sup>Sr/<sup>86</sup>Sr and low ε<sub>Nd</sub>. Fig. 5 illustrates the effect of adding a plume-derived melt to depleted mantle lithosphere or asthenosphere (mixing curve 1). The strongly hyperbolic shape of this

mixing curve is insensitive to the extent of partial melting involved, because most melts or fluids will have significantly higher Sr/Os than depleted mantle peridotite. Such mixing cannot produce the elevated  $^{187}\text{Os}/^{188}\text{Os}$  values in the source of post-erosional lavas without also increasing  $^{87}\text{Sr}/^{86}\text{Sr}$  values well beyond what is observed in the lavas.

Addition of low-degree melts from depleted mantle to the Hawaiian plume [10] could in principle produce the relatively unradiogenic Sr-isotopes of the post-erosional source without overprinting the radiogenic Os-isotopes present in some plume components (Fig. 5, mixing curve 2). However, at both Kauai and Oahu, post-erosional lavas extend to more radiogenic Os-isotope values than earlier shield-stage lavas. If the radiogenic Os-isotopes in the post-erosional lavas derived from a metasomatized Hawaiian plume component, it would therefore necessarily be a plume component that was not sampled by any of the earlier shield lavas, a proposition we feel is unlikely.

Finally, a plume source is also inconsistent with the inferred mineralogy of the source of HV lavas. Trace element enrichment ratios (e.g. K/Th or Ba/Th) in HV lavas indicate the presence of a residual potassium-bearing phase such as phlogopite or amphibole in the source [26,27]. Class and Goldstein [27] and Clague and Frey [26] both modeled trace element abundances and enrichments in HV lavas to infer the presence of minor quantities of phlogopite and/or amphibole in the source region. However, neither phlogopite nor amphibole are stable at the elevated temperatures present in mantle plumes [27], and are unlikely to be stable in the asthenospheric mantle under most conditions. The maximum temperature of stability for amphibole is dependent on source composition, especially total alkali content [56]. However, no experimental studies have demonstrated amphibole stability at asthenospheric temperatures ( $T_p \approx 1300^\circ\text{C}$ ) even in alkali-rich peridotites. Phlogopite could be stable in the asthenosphere at depths greater than  $\sim 160$  km, but not at shallower depths [57,58]. Given a maximum probable thickness for the oceanic lithosphere beneath Hawaii of  $\sim 90$  km [18], it seems unlikely that a

phlogopite signature in HV lavas could reflect melting at depths greater than 160 km. Therefore, the evidence for phlogopite or amphibole in the source of the HV lavas suggests that melt generation occurred at least partly within the cooler lithospheric mantle where such phases are stable [27]. In summary, although influx of plume-derived melts or fluids may be an important catalyst for generation of post-erosional lavas, the Hawaiian plume is not the source of the radiogenic Os-isotopes in these lavas.

#### 4.3. Melt generation from metasomatized asthenosphere or lithosphere peridotite

The  $\sim 100$  Ma oceanic lithosphere beneath Hawaii could have been previously metasomatized by melts or fluids unrelated to Hawaiian volcanism [25,60]. The low silica content of many post-erosional lavas suggests a source enriched in volatiles (especially  $\text{CO}_2$ ) [26,37]. Partial melts of dry mantle peridotite typically have significantly higher  $\text{SiO}_2$  than Hawaiian nephelinites and melilitites (e.g.  $> 44$  wt%  $\text{SiO}_2$  vs.  $\sim 36$ – $44$  wt%  $\text{SiO}_2$ ; cf. [59] and references therein). However, the addition of carbonate to peridotite results in melts with much lower  $\text{SiO}_2$  [59]. Further evidence for a volatile-rich source for the post-erosional lavas comes from the high  $\text{CO}_2$  and  $\text{H}_2\text{O}$  content of quenched glasses from the Hawaiian North Arch [24].

Could (hydrous) metasomatized lithosphere or asthenosphere peridotites provide a source for the radiogenic Os in the post-erosional lavas? Several lines of evidence suggest this is not the case. First,  $\text{SiO}_2$  and incompatible element content are negatively correlated in the post-erosional lavas, so that it are the low- $\text{SiO}_2$  nephelinites and melilitites that show the greatest evidence for involvement of a metasomatized ( $\text{CO}_2$ -rich) peridotite source. However, post-erosional lavas with low  $\text{SiO}_2$  and high incompatible element abundance have less radiogenic Os-isotopes than lavas with higher  $\text{SiO}_2$  (Fig. 4). This is opposite the trend we would expect if the high  $^{187}\text{Os}/^{188}\text{Os}$  derived from a metasomatized peridotite source. Thus, to the extent that a  $\text{CO}_2$ -rich metasomatized source is necessary to produce the low- $\text{SiO}_2$  nephelinites (e.g.

[25,26]), this source does not appear to be associated with the radiogenic Os-isotope signature of the higher-SiO<sub>2</sub> lavas.

Furthermore, Os-isotopes in Hawaiian lherzolite xenoliths do not correlate with the degree of LREE-enrichment (Fig. 2). Instead, metasomatized Hawaiian lherzolites have <sup>187</sup>Os/<sup>188</sup>Os values indistinguishable from abyssal peridotites (<sup>187</sup>Os/<sup>188</sup>Os < 0.130; [14–17]). Other studies of metasomatized xenoliths have found similar results. Handler et al. [61] reported that modally metasomatized phlogopite- and apatite-bearing wehrlites and peridotites from southeastern Australia have similar Re and Os abundances and isotopic compositions as non-metasomatized xenoliths from the same localities. Similarly, xenoliths from Tubuai and Savai'i with trace element patterns indicative of carbonatite metasomatism have Os-isotopic compositions (<sup>187</sup>Os/<sup>188</sup>Os ≈ 0.129) only slightly higher than average abyssal peridotites [62], although at Tubuai, the host lavas are considerably more radiogenic [36]. Only peridotite xenoliths from arc environments appear to have higher average <sup>187</sup>Os/<sup>188</sup>Os values than abyssal peridotites, and here as well the large majority of peridotites have <sup>187</sup>Os/<sup>188</sup>Os values < 0.133 [63]. Although we cannot categorically exclude the possibility that some unusual form of metasomatism increased <sup>187</sup>Os/<sup>188</sup>Os values and/or Re/Os ratios in some sub-Hawaiian mantle peridotites, evidence supporting this hypothesis is lacking. As a plume origin for the Os-isotope signature is also precluded (see above), we must consider whether melt generation from non-peridotite sources could produce the isotopic and compositional variations we observe.

#### 4.4. Melt generation from pyroxenite

Pyroxenites may contribute to melt generation to a disproportionate extent relative to their abundance in the mantle because of their lower solidi and greater melt productivity relative to most peridotites [7–9]. Pyroxenite-rich veins are observed in many ophiolites and xenolith suites (e.g. [11–13,45,46]). Although the abundance of pyroxenites varies considerably, they generally make up 2–5% of all mantle samples for many

localities (e.g. [7] and references therein). Pyroxenites typically have variable but high Re/Os and, as a result of radiogenic ingrowth of <sup>187</sup>Os, extremely radiogenic Os-isotopes [11–13]. Below, we evaluate whether the elevated Os-isotope values in the Hawaiian post-erosional lavas reflect melting of pyroxenite veins in the Pacific lithosphere or entrained upper mantle asthenosphere.

Pyroxenite xenoliths from Salt Lake Crater, Oahu, provide direct evidence that at least portions of the Pacific lithosphere contain pyroxenitic material. These xenoliths have high <sup>187</sup>Os/<sup>188</sup>Os values that extend beyond the range defined by the post-erosional lavas (Table 1). Several studies have proposed that the Hawaiian pyroxenites formed during earlier stages of Hawaiian magmatism [45,46]. However, the pyroxenites may also have formed prior to Hawaiian volcanism, possibly as trapped melts or cumulates near the mid-ocean ridge at ~100 Ma [44]. The high Re content in olivine-poor pyroxenites is consistent with their formation from MORB-type magmas, because MORBs have a higher average Re abundance than OIBs (930 vs. 350 ppt; [53]). Re/Os and <sup>187</sup>Os/<sup>188</sup>Os are roughly correlated in pyroxenite xenoliths. As previously discussed, one xenolith, 114884-27, has an anomalously high Os concentration, possibly due to the presence of sulfide in the sample aliquot analyzed. The other five xenoliths define a broad positive Re/Os–<sup>187</sup>Os/<sup>188</sup>Os correlation. This trend could reflect either mixing of radiogenic and unradiogenic Os, e.g., via melt/lherzolite reaction or radiogenic ingrowth of <sup>187</sup>Os since the formation of the pyroxenites. If the positive Re/Os–<sup>187</sup>Os/<sup>188</sup>Os trend defined by the pyroxenites reflects radiogenic ingrowth, the pyroxenites must be several 10s of Myr old, i.e. they must predate the arrival of the Hawaiian plume beneath the islands of Kauai and Oahu. However, regardless of the age and origin of these pyroxenite xenoliths, it is likely that pyroxenites are present in the sub-Hawaiian lithosphere and asthenosphere, and that these pyroxenites possess higher <sup>187</sup>Os/<sup>188</sup>Os than upper mantle peridotites.

If mixing of pyroxenite- and peridotite-derived melts produced the variable Os-isotopes in Hawaiian post-erosional lavas, we should observe correlations between Os-isotopes and major ele-

ments. Because the post-erosional lavas contain xenoliths that equilibrated at pressures up to 2.5 GPa (75 km) [46], they must have derived from greater depth. High-pressure phase relations of high-alumina basalts indicate that garnet and clinopyroxene are liquidus phases of such basalts at pressures greater than  $\sim 2.5$  GPa [64,65]. Pertermann and Hirschman [66] also reported that partial melting of a pyroxenite with a MORB-like bulk composition at 3.0 GPa produces basaltic andesite melts high in  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  but low in CaO, MgO and FeO. Furthermore, rare hydrous alkaline melts are observed in some Hawaiian pyroxenite xenoliths and along pyroxenite/lherzolite contacts in composite xenoliths. Compared with the host post-erosional lavas, these melt pockets have higher  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  and lower CaO [67,68]. Although these melts cannot be generated solely by melting of the host pyroxenite, they may reflect a combination of pyroxenite melting and reaction with the surrounding lherzolite [68]. Based on these results, we infer that melts derived from pyroxenite veins should have higher  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  and lower CaO than low-degree lherzolite melts.

As predicted from our model, Os-isotopes are positively correlated with  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  (Fig. 4) and negatively correlated with CaO in both the KV and HV lavas. Os-isotopes are also negatively correlated with concentrations of incompatible elements such as Sr (Fig. 4). The degree of melting of pyroxenite should be substantially higher than in surrounding lherzolite [7–9]. As a result, high-degree melts of pyroxenite may have lower concentrations of many incompatible elements than coexisting low-degree, lherzolite-derived melts [37]. Mixing of pyroxenite- and peridotite-derived melts would then produce the observed negative correlations between incompatible element abundances and  $^{187}\text{Os}/^{188}\text{Os}$ .

Os-isotopes are not correlated with other isotopic tracers (e.g. Sr or Nd) in the post-erosional lavas, in contrast with Hawaiian shield-stage lavas (Fig. 5; [49]). However, pyroxenite- and peridotite-derived melts should possess very different Sr/Os and Nd/Os ratios, so mixing of melts will produce strongly hyperbolic mixing curves if the pyroxenite and peridotite sources have significantly

different Sr- and Nd-isotope compositions. The lack of Sr–Os or Nd–Os isotope correlation therefore indicates that the low- $^{187}\text{Os}/^{188}\text{Os}$  (peridotite) and high- $^{187}\text{Os}/^{188}\text{Os}$  (pyroxenite) components have nearly identical Sr- and Nd-isotope compositions. This in turn suggests that the pyroxenite component is relatively young. Although an asthenospheric source for the post-erosional lavas cannot be precluded, a lithospheric source appears more likely. The age of the Pacific lithosphere beneath Hawaii ( $\sim 100$  Ma) is sufficient for pyroxenite veins produced near the mid-ocean ridge to develop high  $^{187}\text{Os}/^{188}\text{Os}$ , but insufficient for pyroxenites and peridotites to evolve significantly different Sr- or Nd-isotope signatures.

We note that several recent studies have argued both for [49] and against [69] the presence of ancient ( $\geq 6$  Ga) recycled eclogite or pyroxenite in the Hawaiian plume. The mafic component that is involved in the generation of Hawaiian post-erosional lavas is distinct from any such component in the plume (see Section 4.2), is much younger ( $< 100$  Ma if it resides in the lithosphere) and is therefore unlikely to differ significantly from surrounding peridotite in its Sr-, Nd- or Pb-isotope composition. Melts from such young mafic components are difficult to distinguish from peridotite-derived melts by examining potential covariations between these radiogenic isotopes and major or trace elements. It is the extreme fractionation of Re from Os and the resulting rapid ingrowth of  $^{187}\text{Os}$  in many natural pyroxenites that allows us to distinguish these melts on the basis of their Os-isotope signatures.

## 5. Implications for evolution of oceanic lithosphere

The  $P$ – $T$  conditions recorded in the Hawaiian xenoliths are hotter than predicted for 100 Ma conductively cooled oceanic lithosphere [46]. Deeper portions of the lithosphere (75–90 km) that are not sampled by the xenolith suite are likely to depart still further from a 100 Ma conductive geotherm. Conductive heating of the base of the lithosphere by the underlying Hawaiian plume [28] or by the passage of earlier shield-stage magmas [46] should raise the temperature of the

lower lithosphere above the solidus for pyroxenite or hydrous peridotite [28] and is therefore a reasonable mechanism for generation of the low-volume post-erosional lavas. If the pyroxenite component implicated by the Os-isotope systematics in Hawaiian post-erosional lavas derives from the Pacific lithosphere, it most likely reflects melts trapped in the lithosphere near the mid-ocean ridge during waning MORB production  $\sim 100$  Myr ago. We suggest that this pyroxenite component formed near the mid-ocean ridge as E-MORB-type melts generated from the limbs of the MORB melting region became trapped in the upper mantle. Although the xenoliths themselves cannot be the actual source of Hawaiian post-erosional lavas (which must derive from deeper depths), similar material near the base of the lithosphere could produce the isotopic and chemical compositions of the post-erosional lavas. Such veining of the shallow upper mantle near mid-ocean ridges is inevitable if melt extraction is not 100% efficient.

Melts generated from mafic veins within oceanic lithosphere in response to thermal perturbations or volatile fluxes from mantle plumes may be a significant source of chemical and isotopic heterogeneity in other OIB suites, particularly during the waning phases of volcanism when melt flux from the sub-lithospheric (plume) mantle is low. Furthermore, where oceanic lithosphere is not reheated by mantle plumes, mafic veins within the lithosphere will be returned to the convecting mantle through subduction. Recycled lithospheric mantle may therefore be an important source of isotopic heterogeneity in the mantle [60].

## 6. Conclusions

Hawaiian post-erosional lavas have highly variable Os-isotopes that extend to more radiogenic values than observed in Hawaiian shield lavas. This contrasts with the relatively uniform and depleted Sr-, Nd- and Pb-isotope compositions of these lavas. The primitive compositions of the lavas, the presence of mantle xenoliths in many post-erosional vents and the high Os abundances

in several samples all indicate that the radiogenic Os-isotopes do not derive from crustal assimilation or other contamination processes, but are instead a source feature. Because the Os-isotopes in the post-erosional lavas are more radiogenic than essentially all peridotite samples worldwide (including both metasomatized and non-metasomatized xenoliths), the Os-isotope signature of the post-erosional lavas cannot be generated by partial melting of upper mantle peridotite. The lack of correlation between Os-isotopes and other isotopic tracers (e.g. Sr, Nd and Pb) contrasts with the strong correlations observed in Hawaiian shield lavas, and precludes derivation of the radiogenic Os-isotope signature from contribution of plume-derived melts or fluids. Thus, pyroxenitic veins in the source of the post-erosional lavas appear the most likely source of the radiogenic Os-isotope signature. This hypothesis is supported (1) by the presence of highly radiogenic pyroxenite xenoliths in some post-erosional lavas, and (2) by correlations observed between Os-isotopes and both major and trace elements in the post-erosional lavas, with the most radiogenic samples possessing higher  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  and lower trace element abundances than the less radiogenic samples. These trends are consistent with the expected compositions of moderate-degree pyroxenite melts.

Although the Os-isotope trends in the post-erosional lavas suggest the involvement of pyroxenite in the generation of these lavas, the source of this pyroxenite is less clear. Both lithospheric and asthenospheric mantle are permissible sources for the post-erosional lavas, although trace element evidence for phlogopite or amphibole in the source of post-erosional lavas from Oahu suggests that melt generation within the lithosphere is more likely. If Hawaiian post-erosional lavas derive from the  $\sim 100$  Ma Pacific lithosphere, our data suggest that veining of oceanic lithosphere near mid-ocean ridges is an important mechanism for producing chemical and isotopic heterogeneity in the mantle. Alternatively, if the post-erosional lavas derive from entrained upper mantle asthenosphere near the plume periphery, the evidence for pyroxenite in the source of these lavas would suggest that such pyroxenite may be a ubiquitous,

albeit volumetrically minor, component in the Earth's convecting mantle.

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