



Assessing the Presence of Garnet-Pyroxenite in the Mantle Sources of Basalts Through Combined Hafnium-Neodymium-Thorium Isotope Systematics

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Abstract. [1] The existence of an enriched component in the mantle with a pyroxenitic or eclogitic composition and its importance for basalt genesis has been discussed for over two decades. Inferences about the depth of melting as well as the dynamics of melting based on the presence of garnet and the location of the spinel-garnet transition are different if garnet-pyroxenite is present in the peridotitic mantle. Trace element partition coefficients are dependent on composition, and the differences between garnet-pyroxenite and peridotite are large enough to produce significant differences in trace element fractionation between melts derived from these different lithologies. Melts derived from garnet-pyroxenite or eclogite-bearing sources will have small or no ²³⁰Th excesses, which are largely independent of melting and upwelling rate. Melts derived from garnet-peridotite will have significant ²³⁰Th excesses, which are dependent on melting and upwelling rate. We show that the combined hafnium-neodymium-thorium (Hf-Nd-Th) isotope and trace element data can distinguish between melts derived from peridotitic and pyroxenitic or eclogitic sources. We also present new Hf isotope data for Hawaiian basalts and use the combined Hf-Nd-Th isotope and trace element systematics to argue against the existence of garnet-pyroxenite or eclogite in the source of Hawaiian basalts. It is especially the large variation in degree of melting for relatively constant isotopic composition that allows us to rule out garnet-pyroxenite in the source of the Hawaiian basalts.

Keywords: Hawaii; garnet-pyroxenite; partition coefficients; U series disequilibrium; hafnium; thorium.

Index terms: Composition of the mantle; isotopic composition/chemistry; geochemistry cycles; igneous petrology.

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1. Introduction

[2] The existence of pyroxenite or eclogite veins and layers in parts of the peridotitic mantle has long been discussed [Zindler et al., 1979; 1984; Allègre and Turcotte, 1986; Hirschmann and Stolper, 1996] because pyroxenite veins are observed in peridotite massifs as well as in mantle xenoliths. Although a pyroxenitic or eclogitic component has often been suggested as the source of chemically and isotopically enriched isotopic signatures in oceanic basalts [Zindler et

al., 1979, 1984; Allègre and Turcotte, 1986; Prinzhofer et al., 1989; Hauri, 1996; Hauri et al., 1996; Hirschmann and Stolper, 1996; Lassiter and Hauri, 1998; Lundstrom et al., 1999; Niu et al., 1999], its role in causing these variations remains controversial. This controversy is partly due to the fact that the isotopic compositions of pyroxenite veins in peridotite massifs show large variations, and these pyroxenites do not have appropriate chemical characteristics to be sources for oceanic basalts [Pearson et al., 1991; Reisberg et al., 1991;

Blichert-Toft *et al.*, 1999a]. Nevertheless, isotope and trace element enrichments in mid-ocean ridge basalts, as well as ocean island basalt signatures such as high μ ($\mu = {}^{238}\text{U}/{}^{204}\text{Pb}$) (HIMU) [Zindler and Hart, 1986] have been associated with the presence of pyroxenite in a peridotitic host. Pyroxenite veins and layers in the suboceanic mantle can have several origins, and most often it is assumed that they represent either trapped melts or remnants of recycled oceanic crust. If pyroxenite layers are assumed to be remnants of recycled oceanic crust, they must have kept their integrity during incorporation into the mantle sources of oceanic basalts. Such pyroxenites would be more restricted in their composition than pyroxenites in peridotite massifs, and an assessment of their major and trace element compositions is possible. Pyroxenites derived from ancient oceanic crust are more enriched in fertile components (clinopyroxene and garnet) and trace elements than the chondritic mantle [Prinzhofer *et al.*, 1989; Hirschmann and Stolper, 1996] and are similar to basalts in terms of their major element composition. The lower Lu/Hf and Sm/Nd and the higher Rb/Sr and Re/Os ratios in pyroxenites compared to peridotites will, in time, lead to ingrowth of isotopic signatures distinct from the ambient mantle, and their melts will show enriched isotopic signatures similar to ancient mid-ocean ridge basalts (low ${}^{143}\text{Nd}/{}^{144}\text{Nd}$ and ${}^{176}\text{Hf}/{}^{177}\text{Hf}$ and high ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ and ${}^{187}\text{Os}/{}^{188}\text{Os}$).

[3] Recently, the possible existence of garnet-pyroxenite or eclogite in the source of oceanic basalts gained renewed attention, as the trace element and isotopic signatures of basalts from various tectonic settings indicate melting in the presence of garnet [Bender *et al.*, 1984; Salters and Hart, 1989; Johnson *et al.*, 1990; Frey *et al.*, 1993; LaTourrette *et al.*, 1993; Beattie, 1993b; Shen and Forsyth, 1995; Bourdon *et al.*, 1996]. The U-Th, Lu-Hf, and Sm-Nd systematics are strongly influenced by melting in the presence of residual garnet [Salters and Hart, 1989; Bourdon *et al.*, 1996] and thus place important constraints on melting in the presence of garnet-

pyroxenite or eclogite (for simplicity, pyroxenite in the remainder of the text is synonymous with garnet-pyroxenite or eclogite, unless garnet-pyroxenite or eclogite is referred to specifically). In the absence of pyroxenite the garnet signature is derived from garnet-peridotite and is the result of melting in the garnet stability field and an indicator of the depth of melting. In the presence of pyroxenite the garnet signature is additionally dependent on the proportion of pyroxenite derived melt in the extracted melt, and information about the depth of melting is hard to extract. Physical parameters of melting and melt transport such as the residual porosity, melting, and upwelling rate are commonly calculated from U series disequilibrium data, on the basis of the assumption of melting a garnet-peridotite source [McKenzie, 1985; Spiegelman and Elliott, 1993; Elliott, 1997; Sims *et al.*, 1999]. Since these estimates can be made only in the absence of pyroxenite, it is of paramount importance to distinguish between garnet signatures from garnet-peridotites and garnet-signatures from pyroxenite. We will show that the combined U-Th, Sm-Nd, and Lu-Hf isotope systematics are able to do just that.

[4] The presence of garnet-pyroxenite in a peridotitic mantle has important consequences for the trace element fractionations during melting. First, we discuss the compositional dependence of the partition coefficients. Second, we discuss trace element fractionations during melting of mixed peridotite-pyroxenite sources with both constant amount of pyroxenite as well as varying amount of pyroxenite. Third, we investigate whether the trace element and isotopic systematics of Hawaiian basalts permit the presence of pyroxenite in their sources.

2. Melting of a Pyroxenite-Bearing Peridotite

2.1. Effects of the Compositional Dependence of Partition Coefficients on Melt Composition

[5] In a recent study, Salters and Longhi [1999] determined partition coefficients for the

peridotite melt system at high pressures and in concurrence with other studies found that partition coefficients are compositionally dependent [Nielsen, 1985; Gaetani and Grove, 1995; Blundy et al., 1998; van Westerenen et al., 1999]. It is these variations in partition coefficients as a function of composition that allow us to distinguish between garnet-peridotite and garnet-pyroxenite (eclogite) derived melts. At high pressure (>2.5 GPa) the clinopyroxene (cpx) in a peridotite has a lower calcium (Ca) content than a cpx at lower pressures. Ca contents in cpx similar to the high Ca content in cpx in peridotites at low pressure are found in systems with a more basaltic bulk composition at high pressures. Similarly, peridotitic garnets (gt) have a lower grossular content than gt's in equilibrium with a more basaltic bulk composition. *Salters and Longhi* [1999] argue, on the basis of the major element composition of the bulk system, that partition coefficients determined in previous studies [*LaTourrette et al.*, 1993; *Hauri et al.*, 1994; *Lundstrom et al.*, 1994] are most applicable to basaltic systems (i.e., garnet-pyroxenite and eclogite). We use the *Salters and Longhi* [1999] partition coefficient data for peridotitic compositions at high pressure and use several of the previously published studies for gt-pyroxenite or eclogite melting [*Hart and Dunn*, 1993; *LaTourrette et al.*, 1993; *Hauri et al.*, 1994; *Lundstrom et al.*, 1994; *Johnson*, 1998]. Enough partitioning data are now available to formulate partitioning models and to parameterize the systematic variations in partitioning as a function of pressure, temperature, and composition [*Gaetani and Grove*, 1995; *Blundy et al.*, 1996; *Salters et al.*, 1999; *van Westerenen et al.*, 1999]. These parameterizations indicate that although the details of our calculations might change, the general trends in our melting calculations are expected to be robust.

[6] The compositional effect of the partition coefficient is illustrated for U and Th. To a first approximation, we can assume that for all bulk compositions considered here, $D_{Th} < D_U$ for gt, while $D_{Th} > D_U$ for cpx. However, peridotitic gt

has higher absolute D_U and D_{Th} than pyroxenitic gt, while in gt-peridotite, D_U and D_{Th} for cpx are lower than for cpx from a pyroxenite. Therefore, in gt-peridotite the leverage of gt on bulk D_U and D_{Th} is larger than in gt-pyroxenite, and U-Th fractionation is larger for gt-peridotites than for gt-pyroxenite. A simple way to assess trace element fractionations during melting is through bulk partition coefficient ratios. Bulk partition coefficient ratios for gt-pyroxenite (70% cpx and 30% gt) and eclogite (50% cpx and 50% gt) are $D_U/D_{Th} = 1.04$, $D_{Lu}/D_{Hf} = 9.7$, and $D_{Sm}/D_{Nd} = 1.9$ for gt-pyroxenite and $D_U/D_{Th} = 1.21$, $D_{Lu}/D_{Hf} = 15.2$, and $D_{Sm}/D_{Nd} = 2.26$ for eclogite (U-Th D for gt and cpx from *Hauri et al.* [1994], rare earth element (REE) and Hf D values for cpx from *Hart and Dunn* [1993] and for gt from *Johnson* [1998]). As a consequence, melts derived from gt-pyroxenite or eclogite (hereinafter called Py melts) are substantially enriched in Hf relative to Lu and moderately enriched in Nd relative to Sm. U and Th, however, are not significantly fractionated during melting of gt-pyroxenite or eclogite but are equally enriched in the melt ($D_U/D_{Th} \approx 1$). With partition coefficients most appropriate for peridotite melting at high pressure [*Salters and Longhi*, 1999], gt-peridotites (50% olivine, 20% orthopyroxene, 20% cpx, 10% gt) have $D_U/D_{Th} \approx 2$, $D_{Lu}/D_{Hf} \approx 7$, and $D_{Sm}/D_{Nd} \approx 2.6$. For similar degrees of melting, gt-peridotite derived melts (hereinafter called Per melts) have significantly larger ^{230}Th excesses and show smaller Lu-Hf and larger Sm-Nd fractionations compared to Py melts. Combining gt liquid and cpx liquid partition coefficients for U and Th from a number of different studies results in ranges of D_U/D_{Th} for gt-pyroxenite from 0.66 to 1.3 and of D_U/D_{Th} for eclogite from 0.91 to 1.76 [*LaTourrette et al.*, 1993; *Hauri et al.*, 1994; *Lundstrom et al.*, 1994], dependent on which combination of published D is used. A websteritic composition [*Hirschmann and Stolper*, 1996] results in a bulk D_U/D_{Th} almost identical those of the pyroxenitic composition used above. Only if cpx liquid partition coefficients for U and Th from *Beattie* [1993b] are used, can $D_U/D_{Th} > 1$ be generated

for gt-pyroxenitic and eclogitic compositions because the cpx liquid D_U and D_{Th} from this study are approximately a factor of 10 lower than those of any other study [LaTourrette *et al.*, 1993; Hauri *et al.*, 1994; Lundstrom *et al.*, 1994]. In this case, the gt liquid D_U and D_{Th} dominate the bulk D_U and D_{Th} of pyroxenitic and eclogitic compositions and result in bulk $D_U/D_{Th} > 1$. We decided not to use the Beattie [1993a, b] studies, as both D_U and D_{Th} are constant over a large range of composition, pressure, and temperature. This is contrary to numerous other studies on cpx partitioning and casts doubts on the accuracy of Beattie's partition coefficients. All other data indicate that even small-degree Py melts can only develop small ^{230}Th excesses owing to the small Th-U fractionation of gt-pyroxenite and eclogite. This is contrary to qualitative estimates in previous studies, where gt-pyroxenitic or eclogitic compositions were thought to be capable of generating large ^{230}Th excesses [Sigmarsson *et al.*, 1998; Lundstrom *et al.*, 1999]. It follows from the above discussion that accurately determined partition coefficients as a function of the chemical composition are crucial in assessing the partitioning behavior during melting of mineralogically different sources.

2.2. Trace Element Fractionation During Melting of Peridotite and Pyroxenite

[7] The $\alpha_{(Sm/Nd)}$, $\delta_{(Sm/Nd)}$, and $\delta_{(Lu/Hf)}$ parameters defined by DePaolo [1988] and Salters and Hart [1989] are a measure of Sm-Nd and Lu-Hf fractionation during melting (see Figure 1). The $\delta_{(Lu/Hf)}$ parameter is strongly affected by the amount of melting in the presence of residual gt, whereas $\delta_{(Sm/Nd)}$ is less sensitive to gt and is a good indicator of the relative degree of melting [Salters and Hart, 1989; Salters, 1996]. Contrary to simple trace element concentration ratios, the $\alpha_{(Sm/Nd)}$, $\delta_{(Sm/Nd)}$, and $\delta_{(Lu/Hf)}$ parameters have the advantage of measuring the Sm-Nd and Lu-Hf fractionation of basalts independent of their source composition. The $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ parameters use the isotopic composition of the basalt to calculate the trace element ratios in the source of

the basalts for a given model age of the source (2 Ga). A younger age for the pyroxenite (or peridotite) would yield higher $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ values; an older age would lead to lower $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ values. The relative variations of $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$, however, are relatively independent of the exact age of the source [Salters, 1996]. Although the translation of $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ parameters in absolute degrees of melting and the amount of melting in the presence of residual gt might be difficult, the relative difference between basalts is a robust estimate. Assuming secular equilibrium in the source, the ($^{230}\text{Th}/^{238}\text{U}$) ratios (parentheses denote activities) are, as $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ values, independent of source composition.

[8] Py melts are clearly distinguishable from Per melts in terms of $\delta_{(Lu/Hf)}$, $\delta_{(Sm/Nd)}$, ($^{230}\text{Th}/^{238}\text{U}$), $^{176}\text{Hf}/^{177}\text{Hf}$, and $^{143}\text{Nd}/^{144}\text{Nd}$. Although there can be substantial variations in the enrichment in incompatible trace elements, pyroxenites or eclogites derived from ancient oceanic crust are expected to have enriched radiogenic isotope signatures (low $^{176}\text{Hf}/^{177}\text{Hf}$ and $^{143}\text{Nd}/^{144}\text{Nd}$) relative to the peridotitic mantle. Figure 1 shows the calculated variations in $\delta_{(Lu/Hf)}$ and $\delta_{(Sm/Nd)}$ of Py and Per melts as a function of degree of melting. As we will apply the U-Th-Sm-Nd-Lu-Hf systematics to the Hawaiian hotspot, we have chosen the isotopic compositions for the Py melts to be very similar to those proposed for the Koolau end-member. On the basis of bulk partition coefficient ratios, Py melts are more enriched in Hf relative to Lu and more enriched in Nd relative to Sm than Per melts. This leads to higher $\delta_{(Lu/Hf)}$ values in Py melts than in Per melts for a given degree of melting. In Py and Per melts, $\delta_{(Sm/Nd)}$ shows large variations with the degree of melting, but owing to the higher D_{Sm}/D_{Nd} ratio in peridotite the variation in $\delta_{(Sm/Nd)}$ is larger in Per melts than in Py melts. Since Per melts are significantly more enriched in Th compared to U than Py melts, Per melts develop larger ^{230}Th excesses than Py melts (Figure 2). Unlike the $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ parameters, however, the Th isotopic composition is influenced by the dynam-

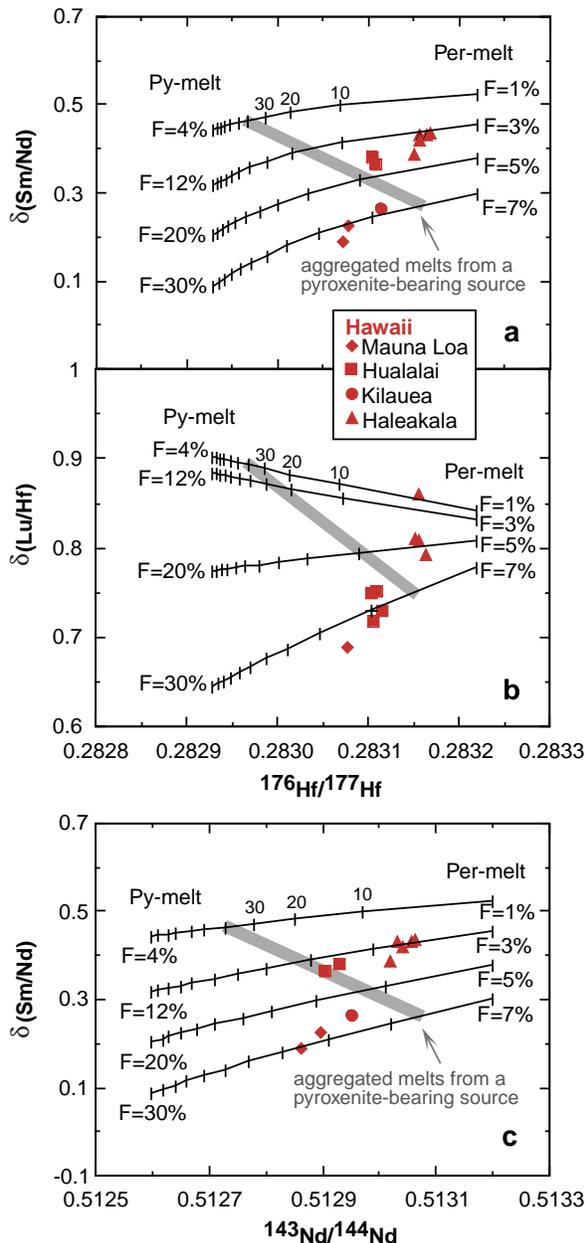


Figure 1. (a and b) Diagram of $^{176}\text{Hf}/^{177}\text{Hf}$ versus $\delta_{(\text{Sm}/\text{Nd})}$ and $\delta_{(\text{Lu}/\text{Hf})}$; (c) $^{143}\text{Nd}/^{144}\text{Nd}$ versus $\delta_{(\text{Sm}/\text{Nd})}$. Our Hawaiian data are shown as solid symbols. Solid lines are mixing lines between high-degree Py melts and low-degree Per melts. The $\delta_{(\text{Lu}/\text{Hf})}$ parameter is an indicator of the amount of melting in the presence of garnet, whereas $\delta_{(\text{Sm}/\text{Nd})}$ is an indicator of the degree of melting [Salters, 1996]. Py melts have enriched isotopic signatures as well as higher $\delta_{(\text{Sm}/\text{Nd})}$ and $\delta_{(\text{Lu}/\text{Hf})}$ than Per melts. The degree of melting (F) for each end-member is indicated at the end of the solid lines; tick marks are for 10% mixing intervals. The sense of the variations in aggregated melt compositions

due to variations in average degree of melting is indicated by the shaded bar, assuming sources with similar amounts of garnet-pyroxenite in the peridotitic source. Decreasing melt productivity for pyroxenite results in higher $\delta_{(\text{Sm}/\text{Nd})}$, $\delta_{(\text{Lu}/\text{Hf})}$, and ($^{230}\text{Th}/^{238}\text{U}$) than shown, which is inconsistent with the Hawaiian data. All melting calculations are done by incremental batch melting with small melt increments (0.1%) and similar porosity (0.1%), i.e., dynamic melting [Salters and Longhi, 1999]. The $\delta_{(\text{Sm}/\text{Nd})}$ and $\delta_{(\text{Lu}/\text{Hf})}$ parameters use the isotopic composition of the basalt to calculate the trace element ratios in the source of the basalts for a given model age of the source (2 Ga): $\delta_{(\text{Sm}/\text{Nd})} = [(^{147}\text{Sm}/^{144}\text{Nd})_{2\text{Ga}} - (^{147}\text{Sm}/^{144}\text{Nd})_m] / (^{147}\text{Sm}/^{144}\text{Nd})_{2\text{Ga}}$, where $(^{147}\text{Sm}/^{144}\text{Nd})_{2\text{Ga}}$ is the calculated $^{147}\text{Sm}/^{144}\text{Nd}$ of the source based on the present-day $^{143}\text{Nd}/^{144}\text{Nd}$ and an assumed source age of 2 Ga and $(^{147}\text{Sm}/^{144}\text{Nd})_m$ is the measured $^{147}\text{Sm}/^{144}\text{Nd}$ of the basalt. The $\delta_{(\text{Lu}/\text{Hf})}$ parameter is defined similarly. Solid symbols are the Hawaiian data from this study. The isotopic composition of the pyroxenite end-member is chosen to be similar to that of the Koolau basalts [Stille et al., 1983; Roden et al., 1994; Blichert-Toft et al., 1999b]. The trace element composition for the garnet-pyroxenite component is similar to depleted mid ocean ridge basalts (NMORB Hofmann, 1988). The isotopic and the trace element composition of the garnet-peridotite are similar to the MORB-source mantle (according to the geochemical Earth reference model (GERM, 1999, available at <http://www-ep.es.lnl.gov/germ>). U-Th partition coefficients for the garnet-pyroxenite component are from Hauri et al. [1994], REE and Hf partition coefficients are from Hart and Dunn [1993] for cpx and from Johnson [1998] for garnet. Peridotite partition coefficients are from Salters and Longhi [1999]. Further explanations are given in the text.

ics of melting, and parameters such as melting rate and the difference between solid and liquid upwelling rate have to be taken into account. The calculated ^{230}Th excesses created by melting of gt-peridotite and pyroxenite at similar porosity and upwelling rate are shown in Figure 2. It is obvious from Figure 2 that ^{230}Th excesses of Py melts are small, even for the low melting and upwelling rate (solid upwelling rate is 1 cm/yr, melt upwelling rate is 400 cm/yr) and low porosity (0.1%) assumed here (Figure 2). In summary, Py melts have higher $\delta_{(\text{Lu}/\text{Hf})}$ values, less variable $\delta_{(\text{Sm}/\text{Nd})}$ values, smaller ^{230}Th excesses,

and more enriched Nd and Hf isotopic compositions than Per melts for a given degree of melting.

2.3. Melting of Pyroxenite-Bearing Peridotite

[9] The key features of the melting, mixing, and melt extraction mechanisms of pyroxenite-bearing peridotites are summarized below. Since an understanding of these melting processes is key to understanding the characteristics of the aggregated melts from pyroxenite-bearing sources, the reader is also referred to [Prinzhofer *et al.* [1989], and references therein and Hirschmann and Stolper [1996], and references therein) for extended discussions on pyroxenite melting.

[10] As long as pyroxenites are not monomineralic, pyroxenites have a lower solidus temperature than peridotites, and the difference between solidus and liquidus temperature is smaller for pyroxenite than for peridotite. Therefore, during adiabatic decompression, melting of pyroxenite starts at a higher pressure than melting of peridotite, and pyroxenite melts to a larger extent than peridotite for a given pressure interval. After melting has been initiated, at any given pressure, Py melts are higher-degree melts than Per melts. Melting of a mixed lithology of pyroxenite and peridotite should therefore be investigated in terms of mixing low-degree Per melts and high-degree Py melts (solid mixing lines in Figures 1 and 2). These mixing lines between pure Py and pure Per melts can also be taken to represent varying amounts of pyroxenite in a peridotite source or mixing of melts derived from sources with different pyroxenite/peridotite ratios.

[11] In the following, we will discuss the chemical variations as a result of varying degrees of melting of sources with similar pyroxenite/peridotite ratios. Figures 1 and 2 show that Py and Per melts are clearly distinguishable in terms of their combined U-Th, Lu-Hf, and Sm-Nd systematics. These distinct geochemical characteristics in combination with the systematic rela-

tionships expected from melting a mixed pyroxenite-peridotite source, as described below, lead to correlations between $\delta_{(Sm/Nd)}$, $\delta_{(Lu/Hf)}$ and $(^{230}Th/^{238}U)$, $^{176}Hf/^{177}Hf$, and $^{143}Nd/^{144}Nd$. These systematic relationships are due to the higher melt

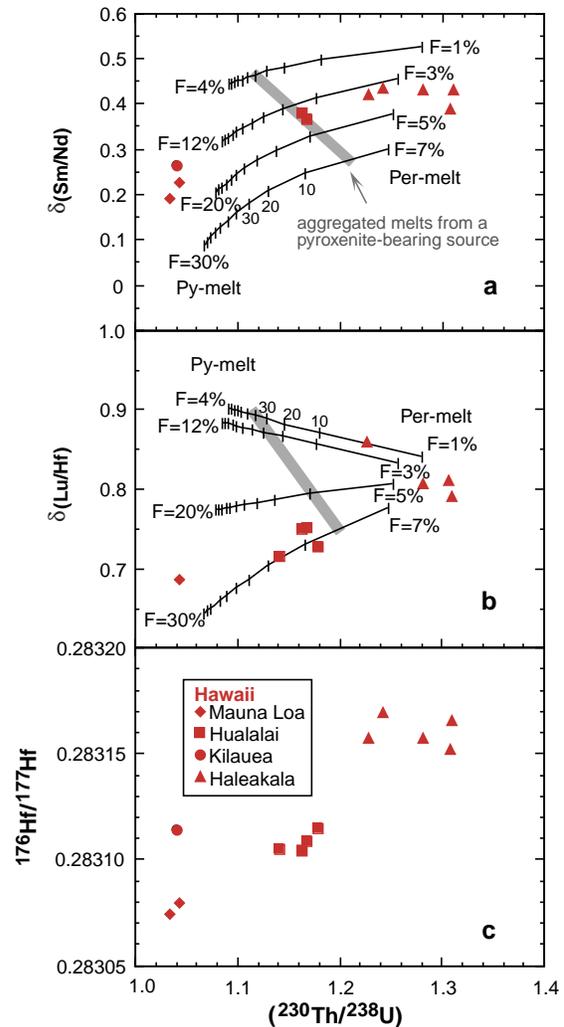


Figure 2. $(^{230}Th/^{238}U)$ versus (a) $\delta_{(Sm/Nd)}$, (b) $\delta_{(Lu/Hf)}$, and (c) $^{176}Hf/^{177}Hf$. Curves are mixing lines between Per melts and Py melts whereby Py melts represent a larger degree of melting. For the Hawaiian basalts the highest-degree melts (low $\delta_{(Sm/Nd)}$) with the least prominent garnet signature (low $\delta_{(Lu/Hf)}$) are the most enriched melts (low $^{176}Hf/^{177}Hf$) with the lowest ^{230}Th excesses. Mixing lines, symbols, and melting calculations as described in Figure 1. Melt transport times are equal for both melting of garnet-pyroxenite and garnet-peridotite (mantle matrix ascent rates are 1 cm/yr and melt ascent rates are 400 cm/yr for both components [Salters and Longhi, 1999]).

productivity of pyroxenite compared to peridotite resulting in varying proportions of Py melt in the aggregated melt as a function of the degree of melting. During progressive melting of a source in which the pyroxenite is a minor component compared to the peridotite, the amount of Per melt will eventually exceed the amount of Py melt owing to the larger abundance of the peridotite. As soon as melting of the peridotite is initiated, the Py melt is diluted by melt derived from the host peridotite, despite the fact that pyroxenite melts to a larger extent than peridotite for a given pressure interval. Additionally, the pyroxenite/peridotite ratio in the residual source decreases with increasing degree of melting due to the higher melt productivity and the lower abundance of the pyroxenite relative to the peridotite. The combination of the effects is to progressively dilute the amount of Py melt in the aggregated melt with increasing degree of melting. Aggregated melts representing low-degree melts will therefore contain a relatively larger proportion of Py melt than aggregated melts representing high-degree melts from a similar source. This relationship is shown qualitatively by the shaded bars in Figures 1 and 2. Therefore, in a suite of samples from sources with similar amounts of pyroxenite, there will be a systematic relationship between the average degree of melting and the amount of Py melt in the aggregated melt, simply as a consequence of melting. In aggregated melts generated from a similar pyroxenite bearing source but differing in degree of melting, Py melts are therefore most abundant in the lowest-degree melts, leading to high $\delta_{(Sm/Nd)}$, high $\delta_{(Lu/Hf)}$, low $^{176}\text{Hf}/^{177}\text{Hf}$, $^{143}\text{Nd}/^{144}\text{Nd}$, and $(^{230}\text{Th}/^{238}\text{U})$ values. In progressively higher-degree melts from this same source the abundance of Py melts in the aggregated melt decreases. Thus $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ decrease, and $^{176}\text{Hf}/^{177}\text{Hf}$, $^{143}\text{Nd}/^{144}\text{Nd}$, and $(^{230}\text{Th}/^{238}\text{U})$ increase in the aggregated melts with increasing degree of melting. This systematic relationship is shown qualitatively by the shaded bars in Figures 1 and 2.

3. The Hawaiian Case

[12] On the basis of major element and Re-Os systematics, it has recently been suggested that the isotopic variations in Hawaiian basalts be generated by melting of recycled oceanic crust components, preserved as gt-pyroxenite or eclogite in the source of Hawaiian basalts [Hauri, 1996; Hauri *et al.*, 1996; Lassiter and Hauri, 1998]. However, the recycled material in the source for Hawaiian basalts could also be well mixed within a peridotitic lithology and not add a distinct second lithology to the source. In this section, we use our model to test whether this recycled component is indeed associated with the inferred presence of a pyroxenitic or eclogitic lithology in the peridotitic mantle source of the Hawaiian volcanics.

3.1. U-Th, Lu-Hf, and Sm-Nd Systematics in Zero-Age Hawaiian Basalts

[13] The data presented in Table 1 are the first where Hf, Nd, and Th isotope systematics have been determined on the same samples. Our new isotope data for zero-age Hawaiian basalts from the Mauna Loa, Kilauea, Hualalai, and Haleakala volcanic series (Table 1) are in good agreement with results from previous studies compiled from the literature and cover most of the compositional range reported for the Hawaiian mantle plume (Figure 3). Hf, Nd, and Sr isotopes correlate well with trace element indices of source enrichment and melting in the presence of gt such as Th/U (Th/Nb, La/Sm) and (Sm/Yb)_n, respectively [Sims *et al.*, 1995, 1999]. All samples have $(^{230}\text{Th}/^{238}\text{U}) > 1$ with ^{230}Th excesses up to 30%, suggesting a significant amount of melting took place in the presence of residual gt. The $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ parameters correlate well with Hf, Nd, and Sr isotopes (Figures 1 and 3), confirming the relationship between source enrichment and recent trace element fractionation during melting observed in zero-age Hawaiian basalts by Sims *et al.* [1995]. The trends in $\delta_{(Sm/Nd)}$ versus $^{143}\text{Nd}/$

Table 1. Isotope and Trace Element Data for Zero-Age Hawaiian Basalts

Sample	ML-61	ML-07	HU-05	HU-13	HU-18	HU-24	KL-01	HK-02	HK-04	HK-06	HK-10	HK-11
Location	Mauna Loa	Mauna Loa	Hualalai	Hualalai	Hualalai	Hualalai	Kilauea	Haleakala	Haleakala	Haleakala	Haleakala	Haleakala
Eruption year	1935	1861	1801			1801	1990	1792				
Age (years)				1030±60	710±150				200-900	200-1000	200-1000	200-1000
Volcanic phase	shield	shield	postshield	postshield	postshield	postshield	shield	posterosional	posterosional	posterosional	posterosional	posterosional
Th-ID	0.552	0.568	1.611	1.53	1.56	1.654	0.867	3.472	2.369	3.423	3.35	3.761
U-ID	0.185	0.193	0.456	0.446	0.44	0.48	0.268	0.981	0.66	1.003	0.993	1.042
Lu-ICP	0.32	0.29	0.27	0.26	0.27	0.27	0.28	0.3	0.24	0.28	0.24	0.33
Hf-ICP		3.2	3.69	3.12	3.37	3.68		5.14	4.18		5.64	5.18
Sm-ID	4.79	4.67	5.04			5.12	5.33	8.08	6.67	8.72	9.15	10.57
Nd-ID	16.67	16.77	22.13			22.78	19.87	37.84	29.08	40.53	41.82	49.01
(²³⁰ Th/ ²³² Th)	1.051	1.076	1.002	1.009	1.009	1.024	1.041	1.099	1.105	1.106	1.102	1.102
(²³⁰ Th/ ²³⁸ U)	1.034	1.044	1.167	1.141	1.179	1.163	1.04	1.282	1.308	1.243	1.228	1.311
(²³⁴ U/ ²³⁸ U)	1.01	1.01			1.02	1.01			1.02	1.02		
¹⁷⁶ Lu/ ¹⁷⁷ Hf			0.0126	0.0102	0.0116	0.0112	0.0102	0.0081	0.008		0.0059	0.0089
¹⁷⁶ Hf/ ¹⁷⁷ Hf	0.283074	0.283079	0.283109	0.283105	0.283115	0.283104	0.283115	0.283157	0.283152	0.283169	0.283157	0.283165
ϵ_{Hf}	10.7	10.8	11.9	11.8	12.1	11.7	12.1	13.6	13.4	14	13.6	13.9
$\delta_{(Lu/Hf)}$		0.686	0.751	0.716	0.729	0.75		0.807	0.81		0.86	0.791
¹⁴⁷ Sm/ ¹⁴⁴ Nd	0.174	0.168	0.138			0.136	0.162	0.129	0.139	0.13	0.132	0.131
¹⁴³ Nd/ ¹⁴⁴ Nd	0.512864	0.512899	0.512905			0.51293	0.512951	0.513033	0.513022	0.513063	0.513043	0.513058
ϵ_{Nd}	4.4	5.1	5.2			5.7	6.1	7.7	7.5	8.3	7.9	8.2
$\delta_{(Sm/Nd)}$	0.188	0.223	0.364			0.379	0.264	0.431	0.386	0.432	0.418	0.429
⁸⁷ Sr/ ⁸⁶ Sr	0.7038	0.7036	0.7036			0.70357	0.70359	0.70325	0.70327	0.7031	0.70311	0.7031

Hf isotope ratios were obtained by multicollector plasma-source mass spectrometry on the Plasma 54 in Lyon. The ¹⁷⁶Hf/¹⁷⁷Hf ratios are reported relative to ¹⁷⁶Hf/¹⁷⁷Hf=0.282160 of the JMC-475 Hf standard. Other analytical details are reported by Sims *et al.* [1995], Blichert-Toft *et al.* [1997], and Sims *et al.* [1999]. Present bulk earth values for calculation of $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ are ¹⁴³Nd/¹⁴⁴Nd = 0.512638, ¹⁴⁷Sm/¹⁴⁴Nd = 0.1967, ¹⁷⁶Hf/¹⁷⁷Hf = 0.282772, and ¹⁷⁶Lu/¹⁷⁷Hf = 0.0332.

¹⁴⁴Nd and in $\delta_{(Lu/Hf)}$ versus ¹⁷⁶Hf/¹⁷⁷Hf in the zero age basalts originating from both the shield and postshield stages of Hawaiian volcanism are reproduced by the variations of basalts from Hawaiian shield volcanoes (Figure 4). This shows that the chemical variations observed in the zero-age Hawaiian basalts of this study (encompassing shield and postshield lavas) are representative of the Hawaiian plume. The isotopically most enriched melts (low ¹⁷⁶Hf/¹⁷⁷Hf) represent the highest degrees of melting (low $\delta_{(Sm/Nd)}$) and have the smallest gt signature (low $\delta_{(Lu/Hf)}$). (²³⁰Th/²³⁸U) are highest in low-degree melts with the largest gt signature (high $\delta_{(Sm/Nd)}$ and high $\delta_{(Lu/Hf)}$; Figure 2). It is evident from Figure 2c, however, that (²³⁰Th/²³⁸U) for Hawaiian basalts also vary systematically with source composition. The isotopically most enriched melts (lowest ¹⁷⁶Hf/¹⁷⁷Hf) have the smallest (²³⁰Th/²³⁸U). Sims *et al.* [1995, 1999] explained the systematic variations in the U-Th disequilibria as being a result of variations in porosity, melting rate, and mantle upwelling rate of a peridotitic mantle. These estimates of melting conditions, however, can only be made in the absence of pyroxenite. If pyroxenite is present in the source of Hawaiian basalts [Hauri, 1996; Hauri *et al.*, 1996; Lassiter and Hauri, 1998], the variations in the

U-Th disequilibria are additionally influenced by the amount of Py melt in the aggregated melt. The trends for the Hawaiian basalts in Figures 1 and 2 can only be explained if the highest-degree melts with the smallest gt signature (lowest ²³⁰Th excess and lowest $\delta_{(Sm/Nd)}$, $\delta_{(Lu/Hf)}$) have the isotopically most enriched signatures (lowest ¹⁷⁶Hf/¹⁷⁷Hf, ¹⁴³Nd/¹⁴⁴Nd) and contain the highest proportion of Py melt. The qualitative trends for increasing degrees of melting of sources with similar pyroxenite/gt-peridotite abundance ratio (shaded bars in Figures 1 and 2) are opposite to what is observed for the Hawaiian basalts. Before adding complexity to the model by resorting to varying amounts pyroxenite or before excluding pyroxenite from the Hawaiian source, we need to investigate the dependence of our calculations on some critical melting parameters.

3.2. Dependence of the Model on Input Parameters

[14] The relationship between the difference in melt productivity between gt-peridotite and pyroxenite determines the slopes of the mixing lines (solid lines) in Figures 1 and 2. Melt productivity is a function of both the location of the solidus and the difference between the solidus

and liquidus temperature; the smaller the difference between solidus and liquidus temperature, the larger the melt productivity. While it is widely accepted that the melt productivity of pyroxenite is higher than the melt productivity of gt-peridotite, the exact relationship is not well determined. A high melt productivity ratio (amount of Py melt over amount of Per melt at a given pressure) of at least 4 is required to produce mixing lines which approximate the trends of the Hawaiian basalts (solid mixing lines in Figures 1 and 2). Hirschmann and Stolper [1996] esti-

mate that the melt productivity of pyroxenite is only ~20–50% higher than the melt productivity of gt-peridotite. Decreasing the melt productivity ratio would decrease the difference between the degree of melting of the Py and Per melts at any given melting interval. At any given stage during melting, lower-degree Py melts would then be mixed with the Per melts, and these lower-degree Py melts have higher $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ and slightly larger ^{230}Th excesses. Thus lower melt productivity of the pyroxenite results in mixing trends that are inconsistent with the Hawaiian trend. Increasing the melt productivity ratio has the opposite effect. As a result of changing melt productivity, the slope of the solid mixing lines changes, and in response to that, the slope of the trend for the aggregated melts (shaded bar) also changes slightly but retains its negative slope even for very large pyroxenite/peridotite melt productivity ratios. Variations in the isotopic compositions or ages of the end-members will result in small changes in the absolute $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ values, but the quality and sense of the correlations between these and other parameters remain intact as long as the age difference between the enriched and depleted component is not large, e.g., as in the case of a 200 Ma enriched component and a 3 Ga depleted component. For example, if one would assume that depleted isotopic signatures of the posterosional basalts are derived from the lithosphere beneath Hawaii, then this should be easily be observed in the $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ parameters, as the lithosphere was depleted only 100 Myr ago. Basalts derived from this source would have small positive or negative δ values. Further details on the appropriateness of δ or α parameters to estimate trace element fractionation during melting independent of source composition and the dependence of δ or α parameters on source age are given by DePaolo [1988] and Salters [1996].

[15] On the basis of the assumption of a pure gt-peridotite source, Sims *et al.* [1999] explained the variation in the ($^{230}\text{Th}/^{238}\text{U}$) ratios of the Hawaiian basalts as being the result of melting with

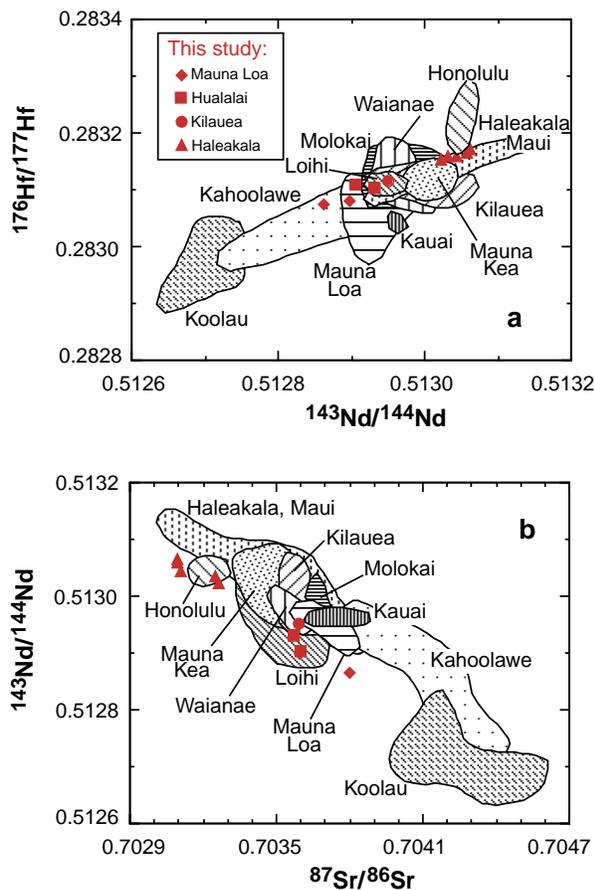


Figure 3. (b) Diagram of $^{87}\text{Sr}/^{86}\text{Sr}$ versus $^{143}\text{Nd}/^{144}\text{Nd}$ and (a) $^{143}\text{Nd}/^{144}\text{Nd}$ versus $^{176}\text{Hf}/^{177}\text{Hf}$. Our data (solid symbols) show a good correlation between $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ and between $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{176}\text{Hf}/^{177}\text{Hf}$. They cover most of the compositional range reported for the Hawaiian mantle plume. Data are compiled from the literature (data sources are available upon request from the authors).

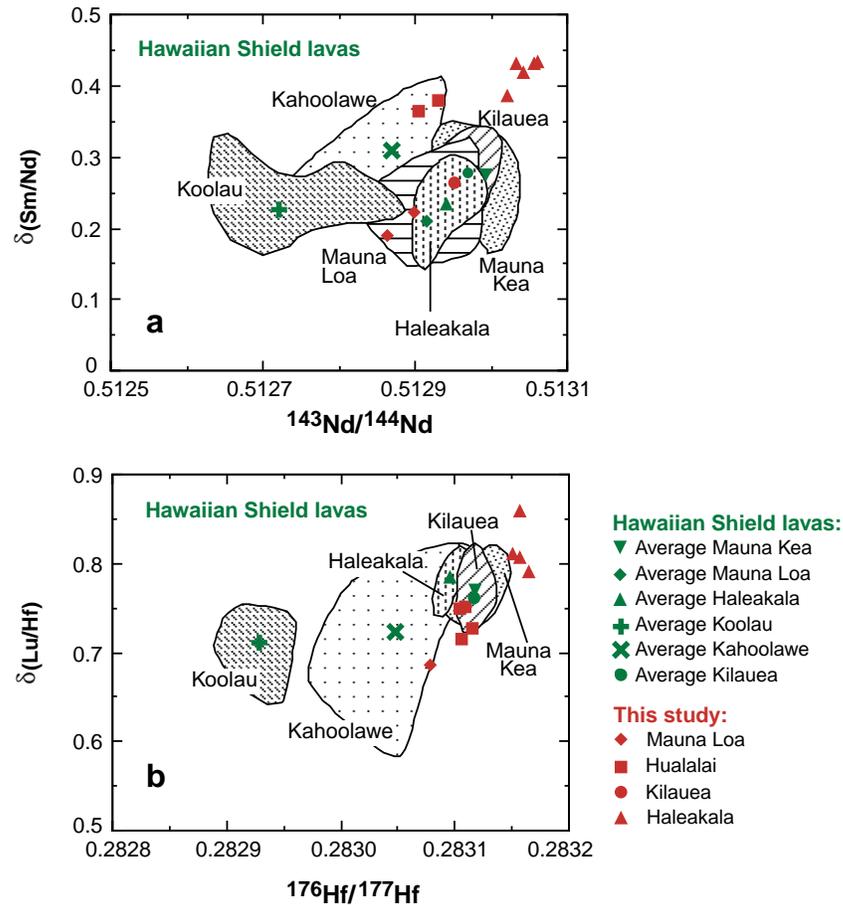


Figure 4. (a) Diagram of $^{143}Nd/^{144}Nd$ versus $\delta_{(Sm/Nd)}$ and (b) $^{176}Hf/^{177}Hf$ versus $\delta_{(Lu/Hf)}$ of Hawaiian shield lavas. For each volcano, there are no correlations between $^{143}Nd/^{144}Nd$ and $\delta_{(Sm/Nd)}$ or between $^{176}Hf/^{177}Hf$ and $\delta_{(Lu/Hf)}$ for Hawaiian shield lavas. Average compositions of the Hawaiian shield lavas from Mauna Kea, Mauna Loa, Kilauea, and Haleakala form a trend almost identical to that formed by lavas analyzed in this study (Figure 4a). Data are compiled from the literature (data sources are available upon request from the authors). Further explanations are given in the text.

varying residual porosity, melting, and upwelling rate. In *Sims et al.*'s model the Mauna Loa basalts represent the melts generated with the higher melting and upwelling rates and higher porosity compared to the Haleakala basalts. Although ($^{230}Th/^{238}U$) ratios for melts derived from a pyroxenite-bearing peridotite source depend to a large extent on the proportion of Py melt in the aggregated melt, upwelling and melting rate and porosity additionally influence the ($^{230}Th/^{238}U$) ratios of the melt. Upwelling and melting rate and porosity are identical for the two sources in Figure 2 and are chosen to create large ^{230}Th

excesses (low melting and upwelling rate and small porosity; see Figures 1 and 2) in order to assess the maximum tolerable amount of Py melt in the aggregated melts. Increasing melting or upwelling rates or increasing porosity results in a decrease in ^{230}Th excesses and a shift of the mixing trends in Figure 2 toward small or no ^{230}Th excesses. It is obvious from Figure 2 that the Haleakala basalts (basalts with large ^{230}Th excesses) require a low porosity, melting, and upwelling rate (as concluded by *Sims et al.* [1999]). Moreover, the Haleakala basalts cannot tolerate a significant pyroxenite component

in order to maintain their large ^{230}Th excesses. The small ^{230}Th excesses of the Mauna Loa basalts can be explained either by a larger porosity, upwelling, and melting rate or by melting of a source with a significant amount of a pyroxenite component. The small ^{230}Th excesses of the Mauna Loa basalts therefore do not allow us to strictly exclude a pyroxenite component in the Mauna Loa source. This would mean that the amount of pyroxenite in the source of Hawaiian basalts is variable, i.e., nonexistent at Haleakala and present at Mauna Loa.

3.3. Variable Pyroxenite/Peridotite Ratios in the Sources of Hawaiian Basalts?

[16] On the basis of major element and Re-Os systematics it has been suggested by Hauri [1996], Hauri *et al.* [1996], and Lassiter and Hauri [1998] that the chemical variations in Hawaiian basalts can be explained by varying amounts of pyroxenite in the source. According to these authors the amount of pyroxenite in the source of Hawaiian basalts is highest for the basalts that show the largest degree of melting: the Haleakala basalts are generated by small degrees of melting from a pyroxenite-free source, and the degree of melting and the amount of pyroxenite in the source increase systematically toward the Mauna Loa end of the trend. The Hawaiian data are bracketed by the solid mixing lines in Figures 1 and 2 and, to a first approximation, appear to be consistent with this model. However, this explanation requires a pyroxenite/gt-peridotite melt productivity ratio higher than experimentally found, at least as high as 4, which is perhaps not unrealistic. This possibility can be further tested by considering volcanoes or stages of volcanoes individually. At a single stage of a volcano the isotopic variation is smaller, and the source is more constant in composition; that is, there are no variations in the pyroxenite/peridotite ratio. Variations in degree of melting in each stage of a volcano should thus produce trends similar to the shaded bars in Figures 1 and 2. The exact location of each trend

would be dependent on the amount of pyroxenite in the source. In case of our zero-age Hawaiian data the data for each volcano do not form such a negative trend but rather appear to follow the general positive trend of all the zero-age basalts in Figures 1 and 2. However, a larger amount of samples would be needed for each volcano in order to reach a more decisive conclusion. There is, however, a large amount of data available for Mauna Loa and other Hawaiian shield lavas that allows assessment of the presence or absence of pyroxenite in the sources of individual Hawaiian shield volcanoes (Figure 4). However, because most of the Hawaiian shield lavas are older than 300,000 years, the constraints provided by the ($^{230}\text{Th}/^{238}\text{U}$) are lost. If the lavas from a single shield stage are derived from a pyroxenite-bearing source, then again, systematic variations similar to the shaded bars are expected. The shield lavas of each volcano show significant variations in the degree of melting, as well as melting in the presence of residual gt, as shown by their variations in $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ (Figure 4). These variations in degree of melting, however, are not correlated with the isotopic variations as would be required in case of a pyroxenite-bearing source (Figure 4). For shield volcanoes at the high $^{143}\text{Nd}/^{144}\text{Nd}$, high $^{176}\text{Hf}/^{177}\text{Hf}$ end of the array, the lack of correlation between $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ parameters and the isotopic composition might be explained as being due to negligible amounts of pyroxenite in their source. On the basis of their enriched Nd and Hf isotopic composition, especially the Kahoolawe and Koolau shield lavas are expected to have significant amounts of pyroxenite in their source. Although these volcanoes show significant variations in degree of melting ($\delta_{(Sm/Nd)}$), there is no apparent correlation between the isotopic composition and $\delta_{(Sm/Nd)}$ and $\delta_{(Lu/Hf)}$ as would be expected in case of a pyroxenite-bearing source (Figure 4). Therefore the observed variations in Hawaiian shield lavas also argue against the presence of pyroxenite in the source of Hawaiian basalts during the shield-building phase of these volcanoes.

4. Conclusions

[17] Owing to the compositional dependence of the partition coefficients, the combined U-Th, Lu-Hf, and Sm-Nd systematics in basalts can distinguish between peridotitic and pyroxenitic sources. U-Th, Lu-Hf, and Sm-Nd systematics in zero-age Hawaiian basalts have been shown to be inconsistent with a model where the extracted melts acquire their trace element and isotopic characteristics by different proportions of Py melt due to variations in degree of melting from a similar source. Furthermore, the uncorrelated variations in $\delta_{(Sm/Nd)}$ versus $^{143}\text{Nd}/^{144}\text{Nd}$ and $\delta_{(Lu/Hf)}$ versus $^{176}\text{Hf}/^{177}\text{Hf}$ for Hawaiian shield lavas, especially Koolau and Kahoolawe, argue against pyroxenite as a possible component in their source. If pyroxenite were present in the source of each of these volcanoes and allowing significant variations in the amount of pyroxenite in their sources, an array of negative trends would be expected, but is not observed. We therefore conclude that at the scale of melting as sampled by the U-Th, Sm-Nd, and Lu-Hf systematics, pyroxenite is not observed as a component in the source of Hawaiian basalts.

[18] The Os isotope systematics of Hawaiian basalts described by Hauri [1996], Hauri *et al.* [1996], and Lassiter and Hauri [1998], as well as the Hf isotope evidence as described by Blichert-Toft *et al.* [1999b], are strong arguments in favor of a recycled component in the source of Hawaiian basalts. We therefore conclude that the addition of a recycled component does not appear to be associated with a significant change in the lithologic composition of the source of the Hawaiian basalts at the scale of melting but is well mixed within the peridotitic lithology. The isotopic variability in Hawaiian basalts is the result of sampling of compositionally distinct peridotite components in the source of Hawaiian basalts. This is in agreement with variations in Na/Ti and rare earth element ratios in Hawaiian basalts [Putirka, 1999], which also suggest the absence of a pyroxenite component. In the absence of pyroxenite the Th-U disequilibria can

be used to estimate the residual porosity, melting, and upwelling rate in the manner described by Sims *et al.* [1999].

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