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Isotope and trace element evidence for depleted lithosphere in the source of enriched Ko'olau basalts

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Abstract We have measured the Hf and Nd isotopic compositions of 38 basalts from the Ko'olau drill hole, Hawai'i. The basalts show limited variations in both 176 Hf/¹⁷⁷Hf and 143 Nd/¹⁴⁴Nd (ε _{Nd} varies from +4.2 to +7.3 and ε_{Hf} from +8.0 to +12.3). Their correlated variation has an R^2 of 0.86. The data form an array with a slope of 1.2 on an $\varepsilon_{\text{Hf}}-\varepsilon_{\text{Nd}}$ isotope correlation diagram, while the slope of all Hawai'ian basalt data is 0.98. Both slopes are significantly shallower than that of the mantle array of 1.4 defined by ocean island basalts. Previous studies have shown that a shallow slope in Hf–Nd isotope space can be related to ancient pelagic sediments in the mantle source (Blichert-Toft et al. [1999](#page-13-0); Salters and White [1998\)](#page-14-0). However, the combined variations in Ko'olau basalts of Hf–Nd–Pb–Os isotopic compositions and trace element ratios, such as La/Nb, Th/La and Sr/ Nd, are not consistent with the simple addition of a sediment component to the mantle. We instead propose that the shallow slope on the Hf–Nd isotope correlation diagram for Ko'olau shield stage basalts can be better explained if the enriched endmember contains either an ancient oceanic lithosphere component or the high- 176 Hf/ 177 Hf component observed in the Salt Lake Crater (SLC) peridotite xenoliths (which also have a depleted lithosphere origin). Since Ko'olau basalts have high $187Os/188Os$ (0.135–0.160) and the SLC xenoliths have $187\text{Os}/188\text{Os}$ up to 0.13 (Lassiter et al. [2000\)](#page-14-0)

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Os-isotopes are consistent with the latter being a component in the enriched Ko'olau source.

Introduction

Of all the lavas of the Hawai'ian volcanoes those of the Ko'olau volcano show the widest range in chemical composition (Chen and Frey [1983](#page-13-0); Hauri [1996](#page-14-0); Roden et al. [1984](#page-14-0), [1994\)](#page-14-0). Lavas from Ko'olau volcano form the most ''enriched'' end of the chemical compositional spectrum of Hawai'ian volcanics and are characterized by low 143 Nd/¹⁴⁴Nd, low 176 Hf/¹⁷⁷Hf, low $^{206}Pb/^{204}Pb$, high ²⁰⁷Pb/²⁰⁴Pb for a given ²⁰⁶Pb/²⁰⁴Pb and are thought to have relatively high- $SiO₂$ contents compared to other Hawai'an volcanics. At the same time, the posterosional Honolulu Volcanic Series (HVS) of the Ko'olau volcano forms the most depleted end of the isotope spectrum of Hawai'ian volcanics and has been variously interpreted to be the result of assimilation of the approximately 90 Ma old oceanic lithosphere that overlies the plume (Chen and Frey [1983](#page-13-0)), incorporation of ancient depleted lithosphere (Frey et al. [2005\)](#page-13-0) or melting of garnet pyroxenites brought to the surface by these lavas (Lassiter et al. [2000](#page-14-0)). Previously, the enriched isotope characteristics of the Ko'olau basalts have been interpreted to be due to the influence of pelagic sediments (Blichert-Toft et al. [1999\)](#page-13-0), while their purported high-SiO₂ contents have been proposed to be related to a source with a relatively dominant pyroxenite component (Hauri [1996\)](#page-14-0). Eclogite or pyroxenite lithologies also have been invoked for the sources of other Hawai'ian volcanoes (Ren et al. [2004](#page-14-0); Takahashi and Nakajima [2002\)](#page-15-0). It has further been argued on the basis of high-Sr contents that a gabbroic component is present in the source of Hawai'ian lavas (Sobolev et al. [2000\)](#page-14-0). However, based on the correlation of Th-excesses with Nd and Hf isotopic compositions, Stracke et al. [\(1999\)](#page-15-0) argued against the presence of either a pyroxenite or an eclogite component in the source of Hawai'ian magmas.

Ko'olau volcano is also important because its posterosional HVS erupted through its apron carrying abundant xenoliths to the surface. Among these, both garnet pyroxenites and spinel peridotites have been observed (Jackson and Wright [1970](#page-14-0); Sen [1988](#page-14-0)). The isotopic composition of the spinel peridotites can only be accounted for by the involvement of depleted oceanic lithosphere (either ancient or young; >100 Ma; Bizimis et al. [2004a,](#page-13-0) [b](#page-13-0); Salters and Zindler [1995\)](#page-14-0). The origin of the garnet pyroxenites is not clear. Their Hf and Nd isotopic compositions place them at the depleted end of the ocean island basalt (OIB) array. Some pyroxenites (isotopic composition unknown) contain pseudomorphs after majorite indicating that they were once part of the Hawai'ian plume (Keshav and Sen [2001](#page-14-0)). However, the isotopic compositions of pyroxenites measured so far are all depleted and therefore cannot be the isotopically enriched component in Hawai'i (Bizimis et al. [2005\)](#page-13-0). Instead, the involvement of an ancient (100 Ma) depleted component intrinsic to the Hawai'ian plume has been proposed for these pyroxenites (Bizimis et al. [2005](#page-13-0)).

To further constrain the nature and composition of the source of the Ko'olau lavas as well as determine which period of the Ko'olau eruptive history shows the distinctive chemical signatures which make this volcano unique among Hawai'ian volcanoes, a previously rotary-drilled water well was cored and analyzed. Based on the chemistry of these lavas Haskins and Garcia ([2004](#page-14-0)) recognized three stages in the Ko'olau volcanic series. The main stage of the Ko'olau volcano, called the Kalihi stage (Haskins and Garcia [2004](#page-14-0)), is recovered in the bottom 400 m of the drill hole. These lavas are very similar in major element composition to the Mauna Loa lavas. By contrast, the Makapu'u stage, which follows the Kalihi stage, is characterized by lavas showing enriched trace element and isotopic signatures as well as high $SiO₂$ and FeO contents and high Al_2O_3/CaO ratios. In the H3 tunnels stage, following the Makapu'u stage, lavas with compositions intermediate to the two previous stages were erupted. The early lavas of the H3 stage are more Kalihi-like, whereas the basalts from the later stages have more Makapu'u-like compositions (Haskins and Garcia [2004\)](#page-14-0).

Table 1 Hf and Nd isotopic compositions of the Ko'olau drill core samples

| Unit | Depth (mbsl) | 176 Hf $/177$ Hf | 2σ | ϵ_{Hf} | 143Nd/144Nd | 2σ | ϵ_{Nd} |
|------------------|--------------|--------------------|-------------|--------------------------|-------------|----------------|-----------------|
| 1 | 350.7 | 0.283016 | $\,$ 8 $\,$ | 8.63 | 0.512860 | $\,$ 8 $\,$ | 4.41 |
| 6 | 363.5 | 0.282999 | 5 | 8.03 | 0.512847 | 9 | 4.16 |
| $\boldsymbol{7}$ | 371.9 | 0.283036 | 5 | 9.34 | 0.512903 | 8 | 5.25 |
| 8 | 375.9 | 0.283036 | 5 | 9.34 | 0.512909 | 10 | 5.36 |
| 9 | 379.3 | 0.283002 | 5 | 8.13 | 0.512856 | 9 | 4.33 |
| 13 | 384.9 | 0.283058 | 5 | 10.11 | 0.512936 | 9 | 5.89 |
| 15 | 390.7 | 0.283037 | 5 | 9.37 | 0.512917 | $\,$ 8 $\,$ | 5.52 |
| 17 | 397.3 | 0.283023 | 4 | 8.88 | 0.512900 | 8 | 5.19 |
| 19 | 404.7 | 0.283049 | 5 | 9.80 | 0.512944 | 11 | 6.05 |
| 20 | 406.1 | 0.283041 | 5 | 9.51 | 0.512933 | 10 | 5.83 |
| 22 | 410.8 | 0.283024 | 3 | 8.91 | 0.512913 | τ | 5.44 |
| 24 | 417.5 | 0.283038 | 5 | 9.41 | 0.512913 | 9 | 5.44 |
| 25 | 419.4 | 0.283044 | 5 | 9.62 | 0.512918 | 8 | 5.54 |
| 27 | 426.4 | 0.283044 | 5 | 9.62 | 0.512909 | 13 | 5.36 |
| 29 | 428.2 | 0.283055 | 5 | 10.01 | 0.512910 | 6 | 5.38 |
| 34 | 438.3 | 0.283068 | 6 | 10.47 | 0.512950 | 12 | 6.16 |
| 37 | 442.7 | 0.283078 | 4 | 10.82 | 0.512956 | τ | 6.28 |
| 38 | 445.5 | 0.283079 | 5 | 10.86 | 0.512960 | 6 | 6.36 |
| 41 | 455.7 | 0.283083 | 4 | 11.00 | 0.512967 | $10\,$ | 6.50 |
| 44 | 458.1 | 0.283086 | 5 | 11.10 | 0.512966 | 8 | 6.48 |
| 45 | 459.2 | 0.283062 | 5 | 10.26 | 0.512916 | 14 | 5.50 |
| 46 | 460.5 | 0.283059 | 5 | 10.15 | 0.512924 | 15 | 5.66 |
| 48 | 469.6 | 0.283062 | 5 | 10.26 | 0.512909 | 9 | 5.36 |
| 50 | 476.6 | 0.283020 | 6 | 8.77 | 0.512860 | 9 | 4.41 |
| 53 | 487.3 | 0.283044 | 5 | 9.62 | 0.512905 | 10 | 5.29 |
| 54 | 490.6 | 0.283066 | 4 | 10.40 | 0.512932 | 10 | 5.81 |
| 57 | 500.2 | 0.283063 | 5 | 10.29 | 0.512938 | 15 | 5.93 |
| 59 | 503.0 | 0.283065 | 4 | 10.36 | 0.512937 | $10\,$ | 5.91 |
| 61 | 516.2 | 0.283059 | 6 | 10.15 | 0.512914 | $\,$ 8 $\,$ | 5.46 |
| 62 | 523.3 | 0.283053 | 5 | 9.94 | 0.512934 | 9 | 5.85 |
| 63 | 529.1 | 0.283057 | 4 | 10.08 | 0.512934 | $\overline{7}$ | 5.85 |
| 65 | 542.8 | 0.283035 | 5 | 9.30 | 0.512909 | 9 | 5.36 |
| 67 | 558.0 | 0.283035 | 5 | 9.30 | 0.512902 | 8 | 5.23 |
| 68 | 561.1 | 0.283029 | 5 | 9.09 | 0.512884 | $\,$ $\,$ | 4.88 |
| 70 | 573.0 | 0.283039 | 4 | 9.44 | 0.512909 | 8 | 5.36 |
| 71 | 573.4 | 0.283055 | 5 | 10.01 | 0.512920 | 9 | 5.58 |
| 75 | 591.9 | 0.283070 | 5 | 10.54 | 0.512950 | $\,$ $\,$ | 6.16 |
| 76 | 597.3 | 0.283119 | 5 | 12.27 | 0.513009 | 9 | 7.32 |

The drill core samples, for which we here report Hf and Nd isotopic compositions, are, with the exception of two samples, all from the Kalihi stage (Haskins and Garcia [2004\)](#page-14-0). Major and trace element data for the Ko'olau Scientific Drilling Project (KSDP) samples have been reported by Haskins and Garcia ([2004](#page-14-0)) and Huang and Frey ([2005](#page-14-0)).

Analytical techniques

Splits of powdered samples, of which the preparation is described in Haskins and Garcia [\(2004](#page-14-0)), were obtained from Garcia (SOEST, Hawai'i). The sample identification used here corresponds to that reported by Haskins and Garcia [\(2004\)](#page-14-0). Hf and Nd isotopic compositions were analyzed on the same sample aliquot for all 38 basalts. The Hf separation chemistry was carried out in Lyon and followed the procedures described by Blichert-Toft et al. [\(1997\)](#page-13-0) and Blichert-Toft [\(2001\)](#page-13-0). The CaMg– fluoride precipitates left over from the Hf leaching step were recovered for the Nd separation chemistry, which was subsequently carried out at the NHMFL following the procedures originally described by Richard et al. ([1976\)](#page-14-0) and adapted by Zindler ([1980](#page-15-0)). The Hf-isotope analyses were done by MC-ICP-MS in Lyon (the Plasma 54) using 179 Hf $/177$ Hf of 0.7325 for instrumental mass bias correction. Sample ¹⁷⁶Hf/¹⁷⁷Hf ratios are reported relative to 176 Hf/¹⁷⁷Hf of 0.28216 for the Hf standard JMC-475. $\frac{143}{\text{Nd}}\frac{144}{144}\text{Nd}$ ratios were corrected for instrumental mass bias using 0.7219 for $146Nd/144Nd$ and are reported relative to 0.511850 for the La Jolla Nd standard. Hf and Nd total procedural blanks were both less than 20 pg, respectively.

Results

Nd and Hf isotopic compositions for the KSDP samples are given in Table [1](#page-1-0) and Fig. [1.](#page-3-0) Samples are identified by the unit designations used in Table 4 of Haskins and Garcia ([2004](#page-14-0)). The KSDP samples range in ε_{Hf} from $+8.0$ to $+12.3$, compared to the total range for Ko'olau volcano (including HVS) of $+4.4$ to $+17.6$ (Blichert-Toft et al. [1999](#page-13-0); Stille et al. [1983](#page-15-0)) and in ε_{Nd} from +4.2 to +7.3, compared to the total Ko'olau range of -1.9 to $+8.2$ (Frey et al. [1994](#page-13-0); Roden et al. [1984,](#page-14-0) [1994;](#page-14-0) Stille et al. [1983\)](#page-15-0). The KSDP samples thus display approximately 30% of the range of published data, but do not include the extreme endmember compositions. However, two samples at the high- $\varepsilon_{\rm HF}$ end (belonging to the HVS; Stille et al. [1983](#page-15-0)) are clearly outliers to the Ko'olau array and, if excluded, the upper limit on the range of Ko'olau volcanics reduces to $+14.2$. At the unradiogenic end, there also are several samples that clearly lie outside the Ko'olau array.¹ The KSDP samples show a well-correlated array in ε_{Hf}

 ε_{Nd} space (Fig. [2](#page-3-0)) with a slope of 1.2 and a high correlation coefficient (\overline{R}^2 =0.86). The correlation between Hf and Nd isotopes remains good (R^2 = 0.87) when the published data (excluding the five outliers) is included but the slope becomes shallower, 1.0. If the outliers are included, the correlation coefficient decreases to 0.74, while the slope stays unchanged at 0.99. In comparison, the global Hf– Nd array has a slope of 1.4 (epsilon notation; Vervoort et al. [1999](#page-15-0)). In addition, the KSDP data follow the trend of the published data for Ko'olau shield stage lavas and extend the trend toward more radiogenic Hf and Nd isotopic compositions. The KSDP data ''fill the gap'' between the previously published Ko'olau shield and post-erosional data.

The top 100 m of the drill core displays a trend toward lower Hf and Nd isotopic compositions (Fig. [1\)](#page-3-0), which is expected to continue in the Makapu'u stage. Units 6 and 9 are parts of the Makapu'u stage (Huang and Frey [2005](#page-14-0)) and have the lowest ^{176}Hf ^{[177}Hf and lowest $^{143}Nd/^{144}Nd$ of all the KSDP samples. The variations in isotopic composition are mimicked by variations in trace element ratios such as La/Nb and Sr/Nb. Makapu'u-stage lavas have higher La/Nb and Sr/Nb and the trend observed in the top 100 m shows a gradual change from the Kalihi to the Makapu'u stage (Huang and Frey [2005](#page-14-0)). The KSDP samples between 500 and 550 m below sea level display a trend toward more depleted isotopic compositions (Fig. [1\)](#page-3-0), i.e., higher 176 Hf/ 177 Hf and 143 Nd/ 144 Nd with increasing depth. If this part of the history of Ko'olau volcano was also controlled by variations in Kalihi and Makapu'u stage magmas, then this should lead to lower La/Nb and lower Sr/Nb. However, whereas Sr/Nb behaves as expected, La/Nb increases with increasing $176Hf/177Hf$ and $143Nd/144Nd$, which is opposite to what is observed for the top KSDP samples.

Discussion

Our new KSDP data in combination with the KSDP data obtained by other groups now allow for a more complete assessment of the origins and processes that resulted in the trace element and isotope variations observed for the Ko'olau basalts. Salters and White [\(1998\)](#page-14-0) first pointed out that the slope in the Hf–Nd isotope correlation diagram can be an indicator of the type of material present in the mantle source. A shallow slope in this diagram indicates decoupling of the Lu–Hf from the Sm–Nd system such that the source has $f_{(Lu/Hf)} > f_{(Sm)}$ N_d , where $f_{(Lu/Hf)}$ and $f_{(Sm/Nd)}$ are the fractionations of, respectively, Lu/Hf and Sm/Nd by a given process. If one assumes that the global Hf–Nd isotope array represents a smooth trace element pattern on a Coryell– Masuda diagram, then a shallower slope indicates either enhanced Lu (compared to Sm and Hf) or depletion of Hf (compared to Sm and Lu). If the Hf-depletion (or rare earth element, REE, enrichment) exists for some time, it will result in enhanced in-growth of 176 Hf and

¹The Hf isotope analyses in question are relatively low-precision thermal ionization measurements.

Fig. 1 Hf and Nd isotopic compositions versus depth (age) in meters below sea level for the Ko'olau drill core samples

thus higher 176 Hf/ 177 Hf than exhibited by the global array. Conversely, material with a relatively low Lu/Hf combined with relatively low Sm/Hf will retard its ingrowth of radiogenic Hf (compared to the in-growth of radiogenic Nd) and will fall below the global array. Midocean ridge basalts (MORBs) are an example of material that with time will develop a Hf–Nd isotopic signature that will lie below the mantle array (Salters and White [1998](#page-14-0)). As both the Ko'olau and the Hawai'ian data

arrays define slopes that are shallower than the global OIB regression line in a Hf–Nd isotope correlation diagram, this indicates that the isotopically least radiogenic end of the Ko'olau array is characterized by relatively high Lu/Hf for its Sm/Nd coupled with either high Sm/Hf or Lu/Sm. Red clays or pelagic sediments have these characteristics (Patchett et al. [1984](#page-14-0)). Patchett et al. ([1984](#page-14-0)) determined that in crustal rocks Hf resides mainly in the mineral zircon and suggested that since

Fig. 2 Hf–Nd isotope correlation diagram showing the Ko'olau Scientific Drilling Project (KSDP) data (circle) and all other published data for Hawai'i. The two regression lines are for the regression of the KSDP data (the steeper of the slopes) and for all Ko'olau data. Published data from Blichert-Toft and Albarede [\(1999](#page-13-0)), Blichert-Toft et al. [\(1999](#page-13-0), [2003\),](#page-13-0) Chen et al. [\(1990](#page-13-0)), Garcia et al. ([1993,](#page-13-0) [1996\),](#page-13-0) Hauri et al. ([1996\)](#page-14-0), Lassiter et al. [\(1996](#page-14-0)), Lassiter and Hauri [\(1998](#page-14-0)), Leeman et al. [\(1994](#page-14-0)), Roden et al. [\(1984,](#page-14-0) [1994\),](#page-14-0) Sims et al. [\(1995](#page-14-0), [1999\)](#page-14-0), Stille et al. [\(1983](#page-15-0), [1986\)](#page-15-0) and Stracke et al. [\(1999](#page-15-0))

zircon is refractory it is transported intact to the continental margins, where it is deposited in turbidites and remains there due to its high density. Sediments on the abyssal plains, such as red clays and pelagic sediments, therefore, are depleted in Hf relative to REEs. Based on this scenario, it has been suggested that the shallow slope on the Hf–Nd isotope correlation diagram of the Hawai'ian volcanics is the result of pelagic sediment involvement in their source (Blichert-Toft et al. [1999\)](#page-13-0). Furthermore, the radiogenic Os of Ko'olau basalts, as well as the coupled O and Os isotopic variations of Hawai'ian volcanics, can be explained if the Ko'olau enriched endmember were to consist of a peridotite source to which 30% of recycled oceanic crust and pelagic sediment had been added (Lassiter and Hauri [1998](#page-14-0)). With the larger data set now available, we can further test this hypothesis.

Assessment of previous models for the enriched Ko'olau endmember

Sediments on the abyssal plain uniquely distinguish themselves in several other chemical parameters than just those related to the Sm–Nd and Lu–Hf systems. Among the additional characteristics that allow us to identify this particular lithology, REE enrichment, a metalliferous nature (high Pb, low U/Pb), high U/Th and high La/Nb are some of the most useful (Plank and Langmuir [1998\)](#page-14-0). High-Pb contents combined with low U/Pb cause this type of sediment to develop unique Pb-isotopic characteristics upon aging. However, quantitative evaluation of the presence of pelagic sediments in the Ko'olau source requires several assumptions of which we make the

following. First, based on the linear Hf–Nd data array, we start with the assumption of binary mixing. Second, for the ambient mantle beneath Ko'olau, we have chosen a source that can yield the picritic magmas of Mauna Loa (see Table 2 of Norman and Garcia [\(1999\)](#page-14-0)) and has an isotopic composition similar to average Mauna Loa basalts. We consider this as a reasonable starting point because the trace element and isotope composition of the radiogenic-Nd-end of the Ko'olau shield basalt array is similar to that of Mauna Loa (Haskins and Garcia [2004](#page-14-0); Huang and Frey [2005\)](#page-14-0). Although the HVS falls at the extension of the Ko'olau shield basalt trend on most trace element and isotope plots, the time gap between these post-erosional and shield stage lavas renders their relationship less clear. However, the two trends seem to have the Mauna Loa picrites as a common end point. Third, for pelagic sediments, we used the red clay composition reported by Johnson and Plank [\(1999](#page-14-0)), which is a very good representative of this lithology (Plank and Langmuir [1998](#page-14-0)). This estimate is high in absolute trace element concentrations, but its trace element ratios are typical of pelagic sediments.

Among the different sediment-sensitive variables, the Hf–Nd isotope correlation is the one most sensitive to the amount of pelagic sediment involved. We will therefore first tune the components to obtain the correct mixing array for Hf–Nd. We subsequently test whether these calculated mixing arrays also provide the correct arrays in other parts of the investigated trace element– isotope space. Attempts are made to model trace element and isotope variations as simply as possible and "complications" are added only if needed. A pure pelagic sediment endmember derived from bulk silicate Earth (BSE) 2 Ga ago has $\varepsilon_{\text{Hf}} = +54$ and $\varepsilon_{\text{Nd}} = -43$.

Table 2 Trace element and isotopic compositions of the components used in the mixing calculations prior to alteration by subduction zone processes

Picrite from Norman and Garcia [\(1999](#page-14-0)), basaltic oceanic crust from Salters and Stracke [\(2004](#page-14-0)), gabbroic oceanic crust based on proposed gabbro composition by Sobolev et al. [\(2000\)](#page-14-0), pelagic sediment composition from Johnson and Plank ([1999\)](#page-14-0) and Guatemala sediment composition from Plank and Langmuir [\(1998](#page-14-0))

Clearly, such an endmember has high $\varepsilon_{\rm Hf}$ for a given $\varepsilon_{\rm Nd}$, but in its pure form its Hf-isotopic composition is too high to be a suitable endmember. Two obvious changes to be introduced are to modify the sediment by subduction and to combine it with other subducted components such as basaltic and gabbroic oceanic crust. Johnson and Plank ([1999](#page-14-0)) determined the changes occurring to pelagic sediment during melting and dehydration. We have used their experimental data to calculate the ''altered'' pelagic sediment composition using our previously published model (Stracke et al. [2003](#page-15-0)). If the sediments have a melt component removed during subduction, then ε_{Hf} increases to $+118$ with little accompanying change in ε_{Nd} and thus does not help to subdue the too-strong ε_{Hf} signature of our starting point composition. The removal of a fluid has little effect on the Sm/Nd and Lu/Hf ratios of the residue due to the refractory character of these elements and thus will not over time result in a significantly different isotopic composition either. Another variable is the age of the sediment. We have chosen a 2-Ga age and increasing the age will only serve to make the isotopic compositions even more extreme. Decreasing the age to 0.5 Ga will not affect the calculated mixing lines enough to allow for a ''pure'' sediment endmember and clearly another component in addition to pure sediment is required. None of these models are shown in the figures as the endmembers are too extreme and plot off scale.

"Sediment" is subducted as a package and especially near subduction zones this package contains several types of sediment. It is therefore not surprising that more than one component seems to be called for. Because it is the entire oceanic crust which is being subducted, then, in addition to other sediments, basaltic as well as gabbroic oceanic crust are potential ''contaminants'' of pelagic sediment. Ancient basaltic crust will develop a Hf–Nd isotopic signature that falls below the global array (Salters and White [1998\)](#page-14-0) and as such is a potential candidate for moderating the pelagic sediment signature toward less extreme compositions. However, both the igneous and the sedimentary part of the oceanic crust undergo changes during subduction. We have assumed that subduction causes dehydration of the igneous crust in a fashion similar to the experiments of Kogiso et al. ([1997\)](#page-14-0). For the sedimentary section, there is now abundant evidence that some portion of the sediment often melts during subduction (Elliott et al. [1997](#page-13-0); Turner et al. [2000,](#page-15-0) [1996](#page-15-0); Turner and Hawkesworth [1997](#page-15-0)). We have assumed that the melting of pelagic sediments during subduction is approximated by the experiments of Johnson and Plank [\(1999](#page-14-0)). Figure 3 shows that 1% of pelagic sediment added to basaltic oceanic crust and a recycling age of 2 Ga yield adequate Hf and Nd isotopic compositions and parent–daughter ratios. A source consisting of 40% of this recycled material mixed with the source of average Mauna Loa picrites could explain the most enriched end of the Ko'olau Hf–Nd array. However, this is not a unique solution. It has been proposed that gabbro is an

important component in the source of Hawai'ian volcanism (Sobolev et al. [2000](#page-14-0)) and the observed Hf–Nd isotope variations can be explained equally well by the proposed gabbroic composition of Sobolev et al. [\(2000\)](#page-14-0) with the addition of 0.1% pelagic sediments (Fig. 3). Furthermore, Huang and Frey [\(2005\)](#page-14-0) recently proposed that the enriched signature was caused by recycled oceanic crust with sediments similar in composition to those found in the Guatemala trench. If this sediment is used instead of the red clay used above, then a mixture of 20% sediment and basaltic oceanic crust or 2% sediment and gabbroic oceanic crust can explain the slope of the Ko'olau samples in Hf–Nd isotope space (Fig. 3). Depending on the exact composition of the recycled component, somewhere between 40 and 60% is required.

The Pb-isotopic composition and parent–daughter ratios of sediments and basaltic crust at the outset are such that they will very quickly develop a distinct isotopic composition (Hart and Staudigel [1989](#page-13-0)). The Hf– Pb isotope correlation diagram should therefore be instructive in determining whether one or several of the four combinations of sediments and oceanic crust that account for the Hf–Nd isotopic variations are indeed part of the Ko'olau source. As could be feared, none of the solutions that worked in Hf–Nd isotope space work straightforwardly when transposed into Hf–Pb isotope space (Fig. 3). There are, however, a number of parameters and assumptions involved in this model and we need to ascertain whether, without violating our geochemical intuitions, some of these can be changed in such a way that the model does fit. Since the Hf and Nd isotope variations have already been adequately mat-

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Fig. 3 Variations in selected isotopic compositions and trace element ratios for Ko'olau basalts as well as model mixing calculations. Solid circles are published data for Ko'olau (see Fig. 2 for references), squares are the Ko'olau Scientific Drilling Project (KSDP) data and open circles are the Honolulu Volcanic Series. Pb-isotope data for KSDP are unpublished data from Fekiacova, while trace element data for KSDP are from Huang and Frey [\(2005](#page-14-0)). For published data, the sources are Bennett et al. [\(1996](#page-13-0)), Clague and Frey [\(1982](#page-13-0)), Frey et al. ([1994\)](#page-13-0), Hart ([1988\)](#page-13-0), Jackson et al. ([1999](#page-14-0)), Jochum and Hofmann ([1997\)](#page-14-0), Jochum et al. [\(1993](#page-14-0)), Lassiter and Hauri [\(1998](#page-14-0)), Lassiter et al. [\(2000\)](#page-14-0), Roden et al. (1994) (1994) , Tatsumoto (1966) (1966) and Yang et al. (2003) (2003) . *Lines* are mixing curves for mixing of ambient mantle with different recycled components. Basalt composition from Salters and Stracke ([2004\)](#page-14-0); gabbro composition is the proposed recycled gabbro composition of Sobolev et al. ([2000\)](#page-14-0). Both recycled basalt and gabbro are altered by dehydration. Mobility of elements during dehydration from Kogiso et al. [\(1997](#page-14-0)) except where the text states the changes made to better fit the Ko'olau trend. PS stands for pelagic sediment; composition from Johnson and Plank [\(1999](#page-14-0)), GS stands for sediment package from the Guatamala trench (adapted from Plank and Langmuir [1998\)](#page-14-0). Isotopic and trace element compositions are calculated using the model of Stracke et al. ([2003\)](#page-15-0). Ambient Hawai'ian mantle is calculated using the Mauna Loa picrite composition (adapted from Norman and Garcia [1999](#page-14-0)) assuming the ambient mantle can yield these picrites by 20% melting. Symbols on mixing curves indicate amount of recycled material and are at 0, 1, 2, 4, 6, 10, 20, 30, 40, 50,60, 70, 80, 90 and 100%. Numbers at mixing curves correspond to the numbered compositions in the figure legend

ched, we will not make any further changes to the model to affect these compositions. In the following, we consider each of the four possible scenarios separately. The trace element compositions of the individual components are listed in Table [2](#page-4-0). Also listed in Table [2](#page-4-0) are the

present-day isotopic compositions of the endmembers used in the mixing calculations. The isotopic compositions of the oceanic crust components at the time of recycling (subduction) lie on growth curves that result in these present-day isotopic compositions. The isotopic

compositions of the gabbro and the basalt is that of MORB, assuming the history of the MORB reservoir can be represented by a two-stage evolution to its present-day isotopic composition with a 3-Ga age for the second stage. Sediments follow the two-stage Stacey and Kramers model for Pb, which would result in the present-day isotopic compositions as detailed by Stracke et al. ([2003](#page-15-0)). For Sr, Nd and Hf, we have used singlestage growth curves to their present-day compositions. More complex growth histories do not provide a better fit to the data.

Basaltic crust and pelagic sediment The ε_{Hf} versus $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ mixing trends (Fig. [3](#page-5-0)) can be made to mimic the Ko'olau trend by decreasing Umobility during subduction. Kogiso et al. ([1997](#page-14-0)) measured a 71% loss of U from basalt during dehydration, while Pb-loss was measured to be 81%. If the U-loss is lowered to 30%, then the mixing trend fits the Ko'olau data and the U–Pb behavior is similar to what is proposed by Kelley et al. ([2005](#page-14-0)). The decrease in U-loss would also require a decrease in Th-loss from 38% as slab dehydration is associated with U-excesses. This would render Th similar to Nb in immobility, which does not conform to observations (Elliott et al. [1997\)](#page-13-0). Therefore, the adjustments in Pb–Th–U mobilities during slab dehydration required to allow for basaltic crust $+$ pelagic sediments in the Ko'olau source contradict observations from arc volcanism. The variation in Sr-isotopic composition can only be matched by reducing Rb mobility to \sim 40% (down from 80%) during the dehydration of basaltic crust. Lowering the Sr-isotopic composition of the sedimentary material does not have a significant effect on the mixing curves. This implies that Sr and Rb have similar mobilities, which again contrasts with the robust observation of LILE enrichments in arcs. In addition, the high-Sr/Nd ratio at lower ε_{Hf} cannot be reproduced. Therefore, a mixture of basalt and pelagic sediment is not the appropriate enriched endmember composition for Ko'olau.

Gabbroic crust and pelagic sediment This scenario also requires a decrease in the loss of U, but contrary to the previous case, the U-loss must be reduced for both the gabbroic portion and the sediment and again in such a way that U mobility is similar to that of Ce. In addition, the mobility of Pb during sediment melting needs to increase; sediment U-loss should be 30% instead of 71% and sediment Pb-loss should be 30% instead of 13%. During dehydration of the gabbroic crust, U-loss needs to decrease from 76 to 30% and Th should be reduced from 30 to 5%. With these "tweakings", ²⁰⁶Pb/²⁰⁴Pb versus ε _{Hf} variations</sub> are successfully reproduced, but the $207Pb/204Pb$ versus ϵ_{Hf} variations are not. However, the all-overshadowing problem with this scenario is posed by the Rb–Sr system. In order to mimic the variations of Sr/Nd as a function of ε_{Hf} , and ε_{Nd} as a function of ${}^{87}\text{Sr}/{}^{86}\text{Sr}$, Rb will have to be quantitatively retained in the slab,

whereas Sr-loss during dehydration of the gabbroic crust needs to be increased from 40 to 90%. Changing the Sr-isotopic composition of the sediments or gabbros does not change the curves significantly. Again these are conditions that are unlikely to be met during subduction. In addition, the ε_{Hf} versus La/Nb variations cannot be explained by this model. The mixing curve in Fig. [3](#page-5-0) is for reduced mobilities of U and Th, immobile Rb and increased Sr and Pb mobilities.

Basaltic crust and sediment from the Guatemala trench Contrary to the previous scenarios, this example requires a large amount of sediment (20%), implying there will be some very distinctive trace element and isotopic compositions. The observed variations in Pb isotopes again require a decrease in U-loss during basalt dehydration such as to become similar in behavior to Ce. In addition, the loss of Pb during sediment melting needs to increase from 14 to 80%, while the basalt component loses only 30% of its Pb (compared to 80% in the quoted experiments). These ''adjustments'', however, again do not result in appropriate mixing curves for the $^{207}Pb/^{204}Pb$ versus ε_{Hf} variations. Additionally, the sediment composition used here has a very high-Sr content (1,540 ppm), while the amount of sediment required for this scenario is also high (20%). Consequently, the Rb– Sr system again is very instructive. The $87\text{Sr}/86\text{Sr}$ versus ε_{Nd} variations of Ko'olau basalts can be reproduced if 99% of the Sr is removed from the sediment, while all the Rb needs to be retained. Furthermore, during dehydration of the basaltic crust only 10% of Sr can be lost versus the experimentally observed 41%. However, this model does not provide the right sense of variations of Sr/Nd versus ε_{Hf} and is also unable to fit the ε_{Hf} versus La/Nb variations. We therefore argue that this scenario is unlikely.

Gabbroic crust and sediment from the Guatemala trench Although significantly less sediment is involved compared with the previous scenario, the U–Pb part of both models is very similar. Again, U behaves as Ce for both gabbro dehydration and sediment melting, and again, the behavior of the Rb–Sr system is very diagnostic. For the gabbroic part of the crust, it is required that $>90\%$ of the Sr inventory is lost, while for the sediments a 60% loss is necessary. This then results in the correct variations on an Sr/Nd versus ε_{Hf} diagram, but the variations of Sr and Hf isotopes and Sr and Nd isotopes cannot be reproduced. Lastly, this model also is unable to fit the ε_{Hf} versus La/Nb variations. Therefore, this scenario is considered as unlikely as the previous three.

Changing the age of the recycled material does not result in the appropriate variations either as younger ages lead to Hf-isotopic compositions that are too radiogenic, while older ages suffer the same misfits for the U–Pb and Rb–Sr systems. As the misfits are similar for gabbroic and basaltic crust scenarios, a combination of the two, which constitutes a more realistic oceanic crust composition, also does not provide adequate solutions. It is more probable that altogether different material is required to explain the observed variations. Global subducting sediment (GLOSS) (Plank and Langmuir [1998\)](#page-14-0) has a composition that, if unaltered by subduction processes, will fall at the extension of the global array in Hf–Nd isotope space, i.e., define a somewhat steeper trend than the Ko'olau array. There have not been any experiments carried out on GLOSS to determine the changes associated with either dehydration or partial melting and the experiments of Johnson and Plank [\(1999\)](#page-14-0) are not readily applicable to this composition. For example, in their experiments Hf is more mobile than Rb, which is not appropriate for GLOSS, which contains zircon. Best estimates for dehydration or melting of GLOSS always result in compositions that fall on or below the OIB array in Hf–Nd isotope space and none lie above it. Thus, GLOSS by itself or combined with igneous oceanic crust is not a suitable component for the Ko'olau array. Contrary to earlier studies that have examined only a subset of the geochemical parameters investigated here (Blichert-Toft et al. [1999;](#page-13-0) Huang and Frey [2005\)](#page-14-0), or altogether different geochemical parameters (Lassiter and Hauri [1998](#page-14-0)), which in both cases were applied to far fewer Ko'olau samples than now allowed for by the KSDP samples, we find that the observed Hf–Nd–Pb–Sr isotope variations combined with trace element variations are incompatible with a source consisting of any combination of oceanic crust and sediment previously subjected to alteration in a subduction zone. We emphasize, however, that we are not suggesting that there is no component in the source that had sediment as a precursor, but only that changes in addition to processing in a subduction zone must have happened to this material. The next important step therefore becomes to re-examine what is the nature of these changes that may account for the source variations of Ko'olau basalts?

The nature of the Ko'olau enriched component

Although the previously proposed scenarios for the enriched Ko'olau endmember do not seem to reproduce all the observed trace element and isotope characteristics, involvement of a pelagic sediment component nevertheless cannot be excluded. However, the previous section has shown that if pelagic sediment is involved in the source of the Ko'olau basalts, then its composition has been altered by processes other than dehydration and melting during subduction. Because this scenario is severely underconstrained, we here propose an alternative scenario involving material, which has been observed at Ko'olau, to explain the shallow slope on the Hf–Nd isotope correlation diagram.

Depleted lithosphere at Ko'olau?

The peridotite xenoliths of Salt Lake Crater (SLC) clearly fall above the global mantle array (Bizimis et al. [2004b](#page-13-0); Salters and Zindler [1995](#page-14-0)) and it should therefore be investigated whether these could be a suitable alternative to pelagic sediment. In addition, the Hf and Os isotope systematics of these xenoliths indicate that they contain an ancient depleted lithosphere component (Bi-zimis et al. [2004a\)](#page-13-0). As the SLC xenoliths have been metasomatized recently, we also must consider whether instead of the SLC xenoliths, depleted lithosphere can provide the appropriate signal. Both SLC peridotite and ancient depleted lithosphere have high $143\text{Nd}/144\text{Nd}$ and low $87\text{Sr}/86\text{Sr}$. Therefore, this material would have to be added to an existing enriched (e.g., unradiogenic Nd isotopic composition) component. Norman and Garcia ([1999](#page-14-0)) also argued that the Hawai'ian plume contained a depleted lithosphere component. Their arguments were mainly based on the constant Sr/Pb , Sr/Y and Ba/Th in picrites with varying isotopic compositions. We propose that the Ko'olau enriched endmember lies on a mixing trajectory between a depleted lithosphere and an enriched component within the OIB array. We will assume the enriched component to be somewhat lower in both 143 Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf than the Ko'olau array, but to fall within the OIB array. Figure 4 shows the mixing trajectories for both SLC peridotites and ancient (2 Ga) depleted lithosphere. We used BSE Hf and Nd concentrations (McDonough and Sun [1995](#page-14-0)) for the enriched

Fig. 4 Hf–Nd isotope variations for Ko'olau basalts (symbols as in Fig. 3) with slopes of the Hf–Nd isotope variations shown for ocean island basalt and the Hawai'ian volcanics. Also shown are mixing curves for mixing of a component on the mantle array with Salt Lake Crater (SLC) peridotite or ancient depleted lithosphere. The ancient depleted lithosphere component is the average abyssal peridotite composition (adapted from Johnson and Dick [1992;](#page-14-0) Johnson et al. [1990](#page-14-0)) aged for 2 Ga. The SLC component is sample SLC326-4 (adapted from Bizimis et al. [2004b](#page-13-0)). The low- $143\text{Nd}/144\text{Nd}$ and low-¹⁷⁶Hf/¹⁷⁷Hf component has bulk silicate Earth trace element contents. Mixing with depleted lithosphere results in steep mixing lines

endmember. The depleted lithosphere composition was calculated using abyssal peridotite clinopyroxene trace element data (Johnson and Dick [1992;](#page-14-0) Johnson et al. [1990](#page-14-0)) assuming chondritic Zr/Hf and Yb/Lu . Bulk trace element concentrations were calculated using mineral modes and assuming a Hf concentration ratio of 25 between clinopyroxene and orthopyroxene, with pyroxenes being the only phases to contain Hf. For Sm, Nd and Lu, clinopyroxene was assumed to be the only phase containing a significant amount of these elements. As orthopyroxene can in fact contain some Lu, the clinopyroxene Lu/Hf only underestimates the bulk Lu/Hf and thus 176 Hf/ 177 Hf. The calculated composition is an average of the data reported by Johnson and Dick [\(1992\)](#page-14-0) and Johnson et al. ([1990\)](#page-14-0). Thus, although potentially underestimated, the isotopic composition of this component is extreme: ε_{Hf} = +1,387 and ε_{Nd} = +187. We also calculated mixing trajectories using SLC326-4 for

which data is reported by Bizimis et al. [\(2004b](#page-13-0)) for the SLC endmember (ε_{Hf} = +65.7 and ε_{Nd} = +8.35; note that even more radiogenic values of ε_{Hf} = +115 have been measured for SLC peridotites; M. Bizimis, unpublished data). Both scenarios produce satisfactory mixing trends in Hf–Nd isotope space (Fig. [4](#page-8-0)). However, the amount of material required is different in the two cases. Addition of approximately 30% of SLC peridotite is required to produce the Ko'olau endmember, whereas for average depleted lithosphere only 10% would be needed. Alternatively, smaller amounts of low degree melts from the depleted lithosphere could also create the increase in Hf-isotopic compositions. Figure 4 shows the location of the mixing curves relative to the Ko'olau data and demonstrates the ability of either scenario to produce the Hf–Nd isotope characteristics of the enriched Ko'olau endmember. As the SLC component and depleted lithosphere have high Sm/Hf, 4 and 8, respectively,

it is important next to check whether the Sm/Hf ratio of the calculated endmember fits with the observations. The calculations indicate that depleted lithosphere could indeed be part of the enriched Ko'olau endmember. The part of the Ko'olau array that contains the largest amount of depleted lithosphere is the low ε_{Hf} -low ε_{Nd} end as it plots furthest above the mantle array.

Pb and Sr isotope systematics do not provide any additional constraints on the depleted peridotite scenario. Measured Sr concentrations in clinopyroxenes from abyssal peridotites are low and calculated wholerock Sr concentrations average 117 ppb. Furthermore, the Rb content of the depleted lithosphere will be low, resulting in limited in-growth of radiogenic Sr. Thus, by virtue of the incompatible behavior of both Rb and Sr during melting, the depleted residue has very little leverage on the Sr-isotopic composition. The U–Pb system behaves similarly, as these elements are too incompatible and therefore too depleted in the residue to significantly affect the Pb-isotopic composition of the mixture. For the mixing scenario with the SLC peridotite (with a higher Sr content), the Rb–Sr isotope systematics are matched by the proposed mixing endmembers. This scenario, however, now requires a mixing chronology in that first depleted lithosphere has to mix with enriched mantle to form the Ko'olau endmember and then this mixture combines with a more depleted Hawai'ian component to produce the Ko'olau array. Bennett et al. ([1996\)](#page-13-0), based on Os–Pb isotope variations, also argued for the presence of depleted mantle in the source of Hawai'ian volcanism, although their depleted mantle component was more akin to a MORB source than to a MORB residue, which is what we call for here.

The remainder of the Ko'olau source

Although we have modeled the Ko'olau endmember as having an isotopic composition close to BSE, it has to be distinctive in trace element ratios, i.e., low Nb/Th and Th/La, and high Sr/Nd and La/Nb. Since the concentrations of all these elements are low in depleted lithosphere, the observed deviations from the chondritic reference must have been created by another process.

Os isotopes potentially provide an additional constraint on the makeup of the source (Lassiter and Hauri [1998](#page-14-0)). Hawai'ian basalts in general have high $187Os/188Os$ and Ko'olau basalts in particular are among those with the highest $187Os/188Os$. The high $187Os/188Os$ for all Ko'olau basalts indicates that the source has experienced an ancient melt enrichment event and that this could correspond to a pyroxenite component. The radiogenic Os-isotope composition requires a high timeintegrated Re/Os, which would argue against a peridotitic depleted lithosphere component. Lassiter and Hauri ([1998\)](#page-14-0) interpreted the high Os isotopic compositions as a recycled oceanic crust component, and noted that in order for this to ''work'' in Pb-isotope space, most of the Pb has to be removed during subduction. Their conclusions on the Pb-isotope systematics is similar to our modeling here, which removes 80% of the Pb from the subducted slab, while also removing most of the U. These large changes during subduction are required to prevent this endmember from developing HIMU type characteristics.

Only a limited number of samples have been analyzed for both Hf and/or Nd and Os isotopes. However, Os and Hf isotopes do not correlate with each other, although they each correlate with the same trace element ratios (Fig. [5\)](#page-9-0). Figure [5](#page-9-0) shows trace element ratios distinctive for Ko'olau basalts plotted against Os and Hf isotopic compositions. These combined data show that both the high- $187\text{Os}/188\text{Os}$ and high- $176\text{Hf}/177\text{Hf}$ (and 143 Nd/¹⁴⁴Nd) endmembers have low La/Nb (though still higher than chondritic) and high Th/La (but lower than chondritic). This would then indicate that the enriched low - 176 Hf/ 177 Hf end of the Ko'olau array should have lower $187Os/188Os$. However, the number of data points defining this correlation is still limited and until more combined Hf–Os isotope data become available it should be viewed with caution.

Is pyroxenite present in the Ko'olau source?

Several studies have used the variations of selected trace elements and/or Os-isotopic composition with $SiO₂$ as evidence for the presence of pyroxenite (Huang and Frey [2005;](#page-14-0) Lassiter and Hauri [1998](#page-14-0); Lassiter et al. [2000\)](#page-14-0). $High-SiO₂$ magmas are expected to have the largest component of pyroxenite melt. Stracke et al. [\(1999\)](#page-15-0) provided the first comprehensive investigation of the effects on trace element and isotope variations as a function of varying degree of melting in a mixed peridotite–pyroxenite lithology. Stracke et al.'s [\(1999\)](#page-15-0) arguments were based mainly on the Lu–Hf, U–Th and Sm–Nd systematics as these systems show different trace element fractionations for pyroxenite versus peridotite melting. If radiogenic Os isotopes are interpreted as reflecting ancient melt enrichment, then this melt enrichment is associated with a lowering of Lu/Hf and Sm/Nd, which in turn results in lower time-integrated 176 Hf/¹⁷⁷Hf and 143 Nd/ 144 Nd. Os isotopes exhibit systematically radiogenic signatures for Hawai'i as a whole, but for individual volcanoes there are no good correlations between Os isotopes and the other isotope systems.

The low- 143 Nd/ 144 Nd Ko'olau endmember contains depleted lithosphere and at least one enriched component. The question thus becomes: does this enriched component contain pyroxenite, especially in the form of recycled oceanic crust (Lassiter and Hauri [1998](#page-14-0); Sobolev et al. [2000,](#page-14-0) [2005\)?](#page-14-0) It has been suggested that the high- $SiO₂$ contents of Ko'olau lavas are an indicator of pyroxenite melt involvement (Hauri et al. [1996](#page-14-0); Lassiter et al. [2000](#page-14-0), [1996](#page-14-0); Lassiter and Hauri [1998;](#page-14-0) Pertermann and Hirschmann [2003\)](#page-14-0). However, experimental evidence suggests that high- $SiO₂$ magmas can result from high

pressure melting of quartz–eclogite or a hydrous amphibole-bearing eclogite (Green [1982;](#page-13-0) Klemme et al. [2002](#page-14-0); Rapp et al. [1991;](#page-14-0) Wyllie [1982;](#page-15-0) Pertermann and Hirschmann [2003](#page-14-0)). Recent experimental work have shown that melting of anhydrous pyroxenite results in silica-undersaturated magmas (Hirschman et al. [2003](#page-14-0); Keshav et al. [2004](#page-14-0)). Moreover, the larger the amount of pyroxenite component the further this component will fall below the mantle array on a Hf–Nd isotope correlation diagram and, consequently, progressively more depleted lithosphere is required to compensate. It therefore seems improbable that the low- $^{143}Nd/^{144}Nd$ Ko'olau component contains any pyroxenite.

There is a weak correlation of Os isotopes with $SiO₂$ content for lavas that have been corrected to be in equilibrium with Fo87 (Hauri et al. [1996](#page-14-0)), whereas the other isotope systems do not correlate with $SiO₂$. However, Huang and Frey [\(2005\)](#page-14-0) found that if the Ko'olau basalt compositions are adjusted to be in equilibrium with Fo90 olivine, then the Pb, Hf and Nd isotopic

compositions for Ko'olau basalts do correlate with $SiO₂$. They use this correlation as evidence for the presence of pyroxenite. Either correction, however, requires large additions of olivine (up to 25%) because the basalts are in equilibrium with olivine down to Fo80. Olivine in Hawai'ian peridotites range from Fo88 to Fo91, while pyroxenites have olivine ranging from Fo71 to Fo85 (Sen [1988\)](#page-14-0). Therefore, pyroxenite involvement would increase the Fe content, which is not observed. If magmas are aggregate melts derived from both pyroxenite and peridotite lithologies or if a pyroxenite-derived melt has fertilized a peridotite, then the assumption that all basalts are in equilibrium with Fo90 is not necessarily correct and is prone to introducing artificial variations. For example, Huang and Frey ([2005](#page-14-0)) interpreted the Fo90 corrected Ko'olau compositions to contain a pyroxenitederived melt component with $>64\%$ SiO₂. Such a melt, however, would be too FeO rich (Mg $\# \sim 35$; see Table 3 of Perterman and Hirschmann [2003](#page-14-0)) and the addition of up to 20% of such a melt to the Ko'olau lavas (Huang

Fig. 6 MgO (wt.%) versus $SiO₂$ $(wt.^{0}\%)$ and Ni (ppm) for Ko'olau shield stage basalts, tholeiites and picrites. Open symbols are literature data and closed symbols are Ko'olau Scientific Drilling Project (KSDP) data (adapted from Haskins and Garcia [2004\)](#page-14-0). Data sources as in Fig. 3. Squares labeled KS and MS are proposed sources for KSDP and Makapu'u stage volcanics as proposed by Sobolev et al. [\(2005](#page-14-0)). The two most mafic and isotopically enriched Ko'olau samples KOO-CF and KOO-17A are also indicated. Norman and Garcia [\(1999](#page-14-0)) proposed KOO-CF as the most primitive of the Ko'olau magmas. The primitive Ko'olau magma composition proposed in this study is indicated by a large open circle

and Frey [2005\)](#page-14-0) clearly is at odds with a Fo90-correction. We therefore do not find the high- $SiO₂$ content and the correlations between $SiO₂$ content and isotope composition to be particularly strong evidence in favor of pyroxenite involvement.

The KSDP data allows for a better assessment of the parental lavas of the Ko'olau shield stage. Figure 6 shows MgO versus Ni and $SiO₂$ for Ko'olau and the proposed parental compositions from Norman and Garcia [\(1999\)](#page-14-0) and Sobolev et al. ([2005](#page-14-0)). It is clear from Fig. [6](#page-11-0) that the new KSDP data requires a readjustment of the Ko'olau parental compositions. Based on the new data, we suggest a parental magma with 15.0 wt.% MgO, 550 ppm Ni and 49.1 wt.% $SiO₂$ (Fig. [6\)](#page-11-0). These compositions are similar to melts derived from peridotites (Sobolev et al. [2005\)](#page-14-0), and thus based on these major elements there is no need for a contribution from a high- $SiO₂$, olivine-free source such as pyroxenite or eclogite. The $MgO-SiO₂$ variation can be modeled by simple crystal fractionation. However, the linear relationship between MgO and Ni indicates that crystal fractionation alone cannot be responsible for the range in MgO content. The parental composition for Ko'olau is very similar to the most primitive magmas of Kilauea (Wagner and Grove [1998](#page-15-0)). Wagner and Grove [\(1998\)](#page-15-0) concluded that these types of magmas could not be direct melts from a garnet peridotite as such melts would be more undersaturated and have lower silica contents. They instead proposed mantle–melt interaction during melt transport whereby the melt replaces pyroxene with olivine in the matrix. Evidence for such a process is also found in the xenoliths from Oahu (Sen and Presnall [1986](#page-14-0)). Hauri and Kurz ([1997\)](#page-14-0) argued against plume– lithosphere interaction based on the combined Os, Sr and He isotope systematics. However, the occurrence of melt–mantle interaction in the enriched Ko'olau samples would indicate that this process takes places within the plume, in t80urn implying that the plume has eroded and rejuvenated the lithosphere. Indeed, a recent study suggests that the plume has thinned the lithosphere down to 60 km (Li et al. [2004\)](#page-14-0), which would allow for the existence of ''pure'' plume melts down to that depth, which is well below the garnet stability field.

 $SiO₂$ content is also expected to vary as a function of both degree and depth of melting, as well as of water content (Baker and Stolper [1994;](#page-13-0) Baker et al. [1994](#page-13-0); Hirose and Kushiro [1993;](#page-14-0) Parman et al. [2001](#page-14-0)). Lowdegree melts with $SiO₂$ contents in excess of 56 wt. $\%$ have been observed experimentally (Baker et al. [1994\)](#page-13-0). A low-Hf, low-Nd, high-Os isotope endmember is also characterized by high $\delta_{(Sm/Nd)}$ and high $\delta_{(Lu/Hf)}$ indicating a low degree of melting. It is therefore not unlikely that the $187\overline{\mathrm{Os}}/188\overline{\mathrm{Os}}$ -SiO₂ correlation is reflective of the variations in degree of melting of a peridotite source.

The pyroxenite xenoliths from HVS have radiogenic Nd and Os isotopic compositions. Most of them are interpreted to be high-pressure cumulates from the HVS basalts (Bizimis et al. [2005;](#page-13-0) Frey [1980\)](#page-13-0). However, pseudomorphs after majorite have been observed in

some pyroxenites (Keshav and Sen [2001](#page-14-0)) making it a possibility that they were once part of the plume material. If these pyroxenites contributed to the melts, this would imply that the amount of pyroxenite is larger in the source of the radiogenic Hf and Nd basalts than in that of the unradiogenic Hf and Nd basalts. Lassiter et al. [\(2000](#page-14-0)), based on the Os–Nd isotopic compositions of SLC pyroxenites, also argued for the presence of a pyroxenite component in the high- $187Os/188Os$ endmember of the post-erosional basalts. Stracke et al. ([1999](#page-15-0)) argued against the presence of pyroxenite based on the assumption that the pyroxenites would have relatively unradiogenic Hf and Nd isotopes. However, the SLC pyroxenites with their radiogenic isotopic composition have different characteristics than assumed by Stracke et al. [\(1999\)](#page-15-0) and thus can potentially be part of the Ko'olau basalts. As explained by Lassiter et al. ([2000](#page-14-0)), the high 143 Nd/¹⁴⁴Nd and high 187 Os/¹⁸⁸Os indicate a very young origin for this component as the low-Sm/Nd ratio cannot support such a high $143Nd/144Nd$ (see also Bizimis et al. [2005](#page-13-0)). Therefore, we argue that SLC pyroxenites could contribute to the high- 176 Hf/ 177 Hf, high- 143 Nd/ 144 Nd post-erosional basalts, which are silica-undersaturated in accordance with experimental evidence of pyroxenite melts (Hirschmann [1995;](#page-14-0) Keshav et al. [2004\)](#page-14-0).

Although Ko'olau is the enriched endmember of the Hawai'ian volcanics, its isotope characteristics are still relatively close to BSE. The trace element pattern of Ko'olau basalts, however, indicates a significant enrichment over BSE. Although depleted lithosphere may be an important source component for Hf isotopes, its significance for trace element systematics is minimal. The enriched character and non-chondritic trace element ratios, such as Th/La, Sr/Nd and La/Nb are most likely caused by an internal mantle differentiation process. It should be noted that the distinctive trace element ratios of this endmember have similarities to those of carbonatites (Bizimis et al. [2003\)](#page-13-0). However, the REE pattern of carbonatites is too steep to permit as a realistic scenario simple addition of carbonatite to either BSE or depleted mantle. Norman and Garcia ([1999](#page-14-0)) argued in favor of the addition of a melt component to a depleted mantle. They used the correlated 186 Os/ 188 Os and 187 Os/ 188 Os (Brandon et al. [1999\)](#page-13-0) to explain the high $187Os/188Os$ not as a melt addition, but as a contribution from the core. The possibility of a core component has been corroborated by high Fe/Mn ratios in Hawai'ian basalts (Humayun et al. [2004](#page-14-0)). If the Os-isotope signature is indeed due to core addition this then also explains its poor correlation with the other radiogenic isotopes.

The evidence derived here from modeling of the combined trace element and isotope variations is inconclusive in that none of the components present in the Ko'olau source are directly identifiable. Instead, the Ko'olau lavas exhibit a hybrid character, with some characteristics of pelagic sediments (notably high La/Nb), some of depleted lithosphere (relatively radiogenic Hf isotopes at low $143Nd/144Nd$) and some of ancient basaltic components (high Os isotopes). However, within the range of the potential variabilities in these endmembers neither one by itself nor a combination of the three can explain all the observed variations. This is not to say that neither of these components are present in the Ko'olau source, but that the sediment and the ancient basaltic component cannot be straightforwardly recognized as such, either in their unaltered form or after alteration due to subduction processes. The depleted lithosphere component may be present in the source, but it does not explain all the features of the Ko'olau basalts either. Thus, intra-mantle differentiation processes must have played a dominant role in creating the enriched Ko'olau source from a combination of lithologies. Unraveling the exact nature of these processes requires further work.

Conclusions

Our new data confirm that the Hf and Nd isotope variations in Ko'olau basalts define a slope which is less steep than that of the OIB array. The Hf–Nd–Pb– Sr isotope variations combined with trace element variations for Ko'olau basalts cannot be readily reconciled with only a recycled pelagic sediment component. Rather, the relatively high 176 Hf/ 177 Hf (at low $143\text{Nd}/144\text{Nd}$ of the Ko'olau enriched endmember indicates that ancient depleted lithosphere, similar to residual lithosphere after MORB melting, is a component in the Ko'olau mantle source. The new KSDP data further allows for a re-assessment of the high-MgO Ko'olau endmember, which is found to be similar in composition to Kilauea parental magmas. These most likely represent melts from the garnet stability field that have interacted with plume-type mantle. The Ko'olau isotopic signatures, including the radiogenic Os, could be an integral part of a peridotite mantle that has undergone some episode of melt infiltration which is not mineralogically distinct.

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