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Origin and temporal evolution of Koʻolau Volcano, Hawaiʻi: Inferences from isotope data on the Koʻolau Scientific Drilling Project (KSDP), the Honolulu Volcanics and ODP Site 843

Z. Fekiacova^{a,*}, W. Abouchami^a, S.J.G. Galer^a, M.O. Garcia^b, A.W. Hofmann^a

^a Max-Planck-Institut für Chemie, Abteilung Geochemie, Postfach 3060, 55020 Mainz, Germany ^b Department of Geology and Geophysics, University of Hawai'i, Manoa, Honolulu, Hawai'i 96822, USA

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Abstract

The "Ko'olau" component of the Hawaiian mantle plume represents an extreme (EM1-type) end member of Hawaiian shield lavas in radiogenic isotope space, and was defined on the basis of the composition of subaerial lavas exposed in the Makapu'u section of Ko'olau Volcano. The 679 m-deep Ko'olau Scientific Drilling Project (KSDP) allows the long-term evolution of Ko'olau Volcano to be reconstructed and the longevity of the "Ko'olau" component in the Hawaiian plume to be tested. Here, we report triple spike Pb isotope and Sr and Nd isotope data on KSDP core samples, and rejuvenation stage Honolulu Volcanics (HV) (together spanning ~ 2.8 m.y.), and from ~ 110 Ma basalts from ODP Site 843, thought to be representative of the Pacific lithosphere under Hawai'i.

Despite overlapping ranges in Pb isotope ratios, KSDP and HV lavas form two distinct linear arrays in ²⁰⁸Pb/²⁰⁴Pb–²⁰⁶Pb/²⁰⁴Pb isotope space. These arrays intersect at the radiogenic end indicating they share a common component. This "Kalihi" component has more radiogenic Pb, Nd, Hf, but less radiogenic Sr isotope ratios than the "Makapu'u" component. The mixing proportions of these two components in the lavas oscillated through time with a net increase in the "Makapu'u" component upsection. Thus, the "Makapu'u" enriched component is a long-lived feature of the Hawaiian plume, since it is present in the main shield-building stage KSDP lavas. We interpret the changes in mixing proportions of the Makapu'u and Kalihi components as related to changes in both the extent of melting as well as the lithology (eclogite vs. peridotite) of the material melting as the volcano moves away from the plume center. The long-term Nd isotope trend and short-term Pb isotope fluctuations seen in the KSDP record cannot be ascribed to a radial zonation of the Hawaiian plume: rather, they reflect the short length-scale heterogeneities in the Hawaiian mantle plume.

Linear Pb isotope regressions through the HV, recent East Pacific Rise MORB and ODP Site 843 datasets are clearly distinct, implying that no simple genetic relationship exists between the HV and the Pacific lithosphere. This observation provides strong evidence against generation of HV as melts derived from the Pacific lithosphere, whether this be recent or old (100 Ma). The depleted component present in the HV is unlike any MORB-type mantle and most likely represents material thermally entrained by the upwelling Hawaiian plume and sampled only during the rejuvenated stage. The "Kalihi" component is predominant in the main shield building stage lavas but is also present in the rejuvenated HV. Thus this material is sampled throughout the evolution of the

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^{*} Corresponding author. Present address: Ecole Normale Supérieure de Lyon, 46, allée d'Italie, 693 64 Lyon Cedex 07, France. Tel.: +33 4 72 72 85 68; fax: +33 4 72 72 86 77.

E-mail address: zfekiaco@ens-lyon.fr (Z. Fekiacova).

volcano as it moves from the center (main shield-building stage) to the periphery (rejuvenated stage) of the plume. The presence of a plume-derived material in the rejuvenated stage has significant implications for Hawaiian mantle plume melting models. © 2007 Elsevier B.V. All rights reserved.

Keywords: Hawai'i; Ko'olau; Honolulu volcanics; isotopes; plume

1. Introduction

Hawaiian Island volcanoes are the most thoroughly studied in the world. They sample the most active mantle plume (Sleep, 1992), which is centered between Kilauea and Lo'ihi volcanoes. Geochemical data on Hawaiian volcanoes have been interpreted in terms of binary or ternary mixing between end member components referred to as Lo'ihi, Kea and Ko'olau (Chen and Frey, 1983; Staudigel et al., 1984; Hauri, 1996; Bennett et al., 1996; Eiler et al., 1996; Lassiter and Hauri, 1998; Blichert-Toft et al., 1999). It is generally accepted that a depleted upper mantle and a recycled slab of oceanic crust material are present in the source of the Hawaiian mantle plume (e.g. Tatsumoto, 1978; Hofmann and White, 1982; Stille et al., 1986; Eiler et al., 1996; Hauri, 1996; Lassiter et al., 1996; Lassiter and Hauri, 1998). High precision Pb isotope data have proven important in delimiting the number of

isotopically distinct components present in the source and length-scales of heterogeneity (Abouchami et al., 2000; Eisele et al., 2003). Recent drill cores and underwater samples from submarine landslides have allowed access to the shield's interior, thus permitting the temporal evolution of a single volcano to be unravelled. Such a record for Mauna Kea shield building has been made available by the Hawaiian Scientific Drilling Project (HSDP) and has shown the existence of short-term changes in source composition (Stolper et al., 1996; Abouchami et al., 2000; Tanaka et al., 2002; Shinozaki et al., 2002; Eisele et al., 2003; Blichert-Toft et al., 2003; Stolper et al., 2004; Abouchami et al., 2005). Similarly, lead isotopes at the scale of the whole Hawaiian chain have provided important information on the chemical structure of the Hawaiian plume (Abouchami et al., 2005).

Ko'olau is an extinct volcano forming the eastern part of O'ahu Island in the Hawaiian archipelago (Fig. 1). Its



Fig. 1. Map of the island of O'ahu showing the location of Ko'olau volcano after Haskins and Garcia, 2004. The Ko'olau Scientific Drilling Project (KSDP) drill site is located in the Western part of the lower Kalihi valley. Location of the ODP 843 site is also shown (coordinates from Shipboard Scientific Party, 1992).

shield stage lasted from 2.9 to 2.1 Ma (Haskins and Garcia, 2004; Ozawa et al., 2005) and produced only tholeiitic lavas; no post-shield stage volcanic activity has been found (Macdonald et al., 1983). After ~1.3 m.y. of erosion and at least 1 km of subsidence of Ko'olau shield (Ozawa et al., 2005), a rejuvenated stage of volcanism produced alkaline lavas referred to as the Honolulu Volcanics (HV) (Winchell, 1947). The geochemical and isotopic compositions of subaerial lavas exposed in the Makapu'u section define the so-called "Ko'olau" component which bears the classical EM1 signature. It is characterized by the highest SiO2 content, the highest ⁸⁷Sr/⁸⁶Sr and ¹⁸⁷Os/¹⁸⁸Os isotope ratios, and the lowest $\varepsilon_{\rm Nd}$, $\varepsilon_{\rm Hf}$ and ${}^{206}{\rm Pb}/{}^{204}{\rm Pb}$ isotope ratios (Stille et al., 1983; Roden et al., 1984; Stille et al., 1986; Lassiter and Hauri, 1998; Blichert-Toft et al., 1999). These characteristics have been interpreted as reflecting contributions from old recycled oceanic crust (Lassiter and Hauri, 1998) and/or the presence of ancient (3 Ga) pelagic sediments in the source (Blichert-Toft et al., 1999). The Makapu'u lavas, however, appear to form only a veneer. Compositions similar to those of Mauna Loa are found in lavas from the submarine flanks of Ko'olau volcano (Jackson et al., 1999; Tanaka et al., 2002; Shinozaki et al., 2002) and in the Ko'olau Scientific Drilling Project core (Haskins and Garcia, 2004).

The Ko'olau Scientific Drilling Project (KSDP) was aimed at understanding the geochemical and temporal evolution of Ko'olau volcano (Haskins and Garcia, 2004). The KSDP drill core is located on the western margin of the lower Kalihi Valley (21°20'39.36" N, 157° 52'22.36" W). The KSDP samples studied come from the cored section (from 300 to 632 mbsl), which is subdivided into 104 petrologic units, described in detail by Haskins and Garcia (2004). The lowermost part of the KSDP core, defined as the "Kalihi" stage of the Ko'olau shield, has major and trace element compositions resembling those of Mauna Loa lavas (Haskins and Garcia, 2004; Huang and Frey, 2005).

Here, we report radiogenic isotope data (triple spike Pb, Sr and Nd) on lavas representative of the shield building and rejuvenated stages of Ko'olau Volcano in order to evaluate the long-term isotopic evolution of Ko'olau Volcano. This new dataset complements existing geochemical and isotope data (Haskins and Garcia, 2004; Huang and Frey, 2005; Salters et al., 2006). The sampling includes the Ko'olau drill core (KSDP), the Honolulu Volcanics (HV), and also basalts from ODP Site 843 (Leg 136, Hole B, 19°20.53'N, 159°5.68'W). The ODP Site 843 is located on the Hawaiian arch, about 250 km south of the island of O'ahu (see insert Fig. 1) and provide samples represen-

tative of the 110 Ma-old Pacific lithosphere (King et al., 1993; Waggoner, 1993). With the exception of Nd isotopic compositions of KSDP lavas (Salters et al., 2006), all isotope data are from this study.

The rejuvenated Honolulu lavas analysed here include twelve samples dated by unspiked K–Ar with ages ranging from 0.06 to 0.8 Ma (Ozawa et al., 2005) and five samples (labelled OA) previously analyzed by Stille et al. (1983). In addition, major and trace element and isotope data were obtained on four samples cored from ODP Site 843. These data shed light on the sources of Honolulu Volcanics whose origin has been attributed to melts derived from the lithosphere that overlies the Hawaiian plume (Clague and Frey, 1982; Stille et al., 1983; Lassiter et al., 2000).

2. Results

Lead, Sr and Nd isotopic compositions of KSDP lavas, Honolulu Volcanics and ODP Site 843 basalts are reported in Table 1 together with Pb isotopic compositions of replicate measurements and/or dissolutions as well as leaching results. The leaching results are discussed in Supplementary Material 1. Major and trace element data on Honolulu Volcanics, along with the data on four ODP Site 843 samples, are reported in Supplementary Material 2.

2.1. Isotope stratigraphy

The isotopic variations of KSDP lavas with depth are represented in Fig. 2 along with the isotopic compositions of subaerial Makapu'u stage lavas and Honolulu Volcanics. KSDP lavas generally have higher ²⁰⁶Pb/²⁰⁴Pb (\geq 18.0) (Fig. 2a) and ε_{Nd} (+4.2 to+7.3) (Fig. 2d), and lower ⁸⁷Sr/⁸⁶Sr (0.70361–0.70386) (Fig. 2c) than Makapu'u stage lavas $(^{206}Pb/^{204}Pb \le$ 17.9; ε_{Nd} = +0.2 to +3.2; ⁸⁷Sr/⁸⁶Sr = 0.7040 to 0.7044), indicating a change in source composition during the transition from the main to late shield building of Koʻolau Volcano. Despite the small isotopic variability in 206 Pb/ 204 Pb ratios (~0.15) of KSDP lavas, a general increase from the core top to \sim 450 mbsl, followed by a decrease down core, can be resolved. Superimposed on this "bell" trend, oscillations in ²⁰⁶Pb/²⁰⁴Pb ratios occur recurrently at ~ 25 m depth intervals (Fig. 2a). The Pb isotopic spike observed at 450 mbsl is associated with spikes in Sr, Nd and Hf isotopic compositions (Fig. 2c,d), indicating changes in magma composition at this depth. Indeed, samples above 450 mbsl tend to have more Makapu'u-like isotopic compositions than those below this depth, which corresponds (within our

Tabl	e 1						
Pb, S	Sr and Nd isotopic compositions	of KSDP lava	s, Honolulu	Volcanics and	ODP	843	basalts

Sample	Rock type	Material	Depth (mbsl)	²⁰⁶ Pb/ ²⁰⁴ Pb	$\pm 2\sigma$	²⁰⁷ Pb/ ²⁰⁴ Pb	$\pm 2\sigma$	²⁰⁸ Pb/ ²⁰⁴ Pb	$\pm 2\sigma$	²⁰⁸ Pb*/ ²⁰⁶ Pb*	⁸⁷ Sr/ ⁸⁶ Sr	$\pm 2\sigma$	€ _{Nd}	$\pm 2\sigma$
Koʻolau Scientific Drilling Project units														
KSDP-U1	Picrite	Chips	350.7	18.0239	9	15.4498	9	37.8254	26	0.9582	0.703841	7	4.41	
KSDP-U6	Picrite	Chips	363.5	18.0147	10	15.4489	12	37.8193	37	0.9582			4.16	
KSDP U8-d1	Ol basalt	Chips	375.9	18.0112	10	15.4519	9	37.7771	27	0.9538			5.36	
KSDP U8-d2	Ol basalt	Chips	375.9	18.0118*	11	15.4542*	13	37.7761*	42	0.9536				
KSDP U8 mean				18.0115		15.4530		37.7766		0.9537				
KSDP-U9	Ol basalt	Chips	379.3	17.9997	10	15.4467	10	37.8264	28	0.9607	0.703860	11	4.33	
KSDP-U15	Ol basalt	Chips	390.7	18.0726*	11	15.4550*	14	37.8400*	44	0.9543			5.52	
KSDP-U17	Basalt	Chips	397.3	18.0661	8	15.4547	8	37.8325	23	0.9541			5.19	
KSDP-U20	Picrite	Chips	406.1	18.0655	9	15.4565	9	37.8238	25	0.9532			5.83	
KSDP-U24	Basalt	Chips	417.5	18.0936	8	15.4572	8	37.8527	25	0.9534	0.703780	6	5.44	
KSDP-U27	Basalt	Chips	426.4	18.0423	11	15.4482	11	37.8316	30	0.9566	0.703806	9	5.36	
KSDP-U34	Ol basalt	Chips	438.3	18.0926	7	15.4539	7	37.8305	23	0.9510			6.16	
KSDP-U38	Basalt	Chips	445.5	18,1237	8	15.4581	8	37.8516	24	0.9500			6.36	
KSDP-U41	Basalt	Chips	455.7	18,1485	8	15.4614	8	37.8661	25	0.9490	0.703614	8	6.5	
KSDP-U46	Basalt	Chips	460.5	18.0223	7	15.4505	8	37.8165	24	0.9571	0.703693	12	5.66	
KSDP-U49	Basalt	Chips	473	18.1142	9	15.4540	8	37.8477	25	0.9506	0.703666	12		
KSDP-U51	Basalt	Chips	480.3	18.0460	8	15.4545	8	37.8200	24	0.9549	0.703734	9		
KSDP-U54	Basalt	Chips	490.6	18.0711	7	15 4537	8	37 8377	24	0.9542	01/05/01	-	5.81	
KSDP-U57	Basalt	Chips	500.2	18 0961	, 7	15 4556	8	37 8584	25	0.9538	0 703653	12	5.93	
KSDP-U61	Basalt	Chips	516.2	18 0788	8	15 4534	8	37 8486	24	0.9546	017000000	12	5 46	
KSDP-U62	Basalt	Chips	523.3	18.0631	6	15 4508	7	37 8380	22	0.9551			5.85	
KSDP-U63	Basalt	Chips	529.1	18.0577	8	15 4475	8	37 8287	22	0.9546	0 703669	11	5.85	
KSDP-U66	Basalt	Chips	553.9	18 0352	6	15 4502	7	37 8234	21	0.9564	0.102000		0.00	
KSDP-U68	Ol basalt	Chips	561.1	18 0324	9	15 4483	10	37 8372	28	0.9583	0 703859	11	4 88	
KSDP-U71	Basalt	Chips	573.4	18 0705	6	15 4531	8	37 8475	24	0.9553	0 703771	6	5 58	
KSDP-U74	Basalt	Chips	584.8	18 0278	8	15 4533	8	37 8402	25	0.9592	01/05//1	0	5.50	
KSDP-U76 (3H)	Basalt	Chips	597.3	18 1293	10	15 4505	11	37 8084	35	0.9445			7 32	
KSDP-U82 (3H)	Basalt	Chips	605.6	18 0604	12	15 4480	11	37.8216	31	0.9535			1.02	
KSDP U88-d1	Ol basalt	Chips	616.4	18 0271	13	15 4482	13	37 9215	41	0.9686				
KSDP U88 (3H)-d2	ol basalt	Chips	616.4	18 0218	13	15 4440	12	37 9012	32	0.9668				
KSDP U88 mean	or ousure	emps	01011	18 0245	10	15 4461		37 9113	52	0.9677				
KSDP-U92 (3H)-d1	nicrite	Chins	628 7	18 1098	16	15 4428	14	37.8152	36	0 9474				
L-U76 (3H)	pierite	emps	02017	18 1740*	16	15 4653*	19	37.8612*	60	0.9457				
L-U82 (3H)				18 1163	9	15 4588	12	37.8637	34	0.9522				
L-U88 (3H)-d2				18 0846*	59	15 4651*	51	37 9645*	128	0.9671				
L-U92 (3H)-d1				18 1629	9	15 4603	9	37 8816	27	0.9492				
L-U92 (3H)-d2				18 1588*	11	15 4644*	13	37 8900*	41	0.9506				
L-U92 (3H) mean				18 1609		15 4624	10	37 8858		0.9499				
2 0)2 (011) 110411				1011009		1011021		5710020		0.0 100				
Honolulu Volcanics														
OA3	Nephelinite	Powder	0.47	18.1670	10	15.4634	11	37.8332	32	0.9433	0.703335	9	7.67 [£]	
OA4-d1	Nephelinite	Powder	0.41	18.2061	10	15.4698	12	37.8807	35	0.9445	0.703349	13	8.23 [£]	
OA4-d2	Nephelinite	Powder	0.41	18.2024*	12	15.4667*	14	37.8610*	44	0.9427				
OA4 (3H)-d3	Nephelinite	Powder	0.41	18.2022*	30	15.4704*	38	37.8696*	123	0.9437				
OA4-mean				18.2036		15.4690		37.8704		0.9436				
OA5	Nephelinite	Powder	0.58	18.1545	11	15.4601	11	37.8149	32	0.9426	0.703308	10	8.04^{\pounds}	
OA6	Alkol. basalt	Powder	0.1	18.0925	12	15.4547	12	37.7682	34	0.9439	0.703337	8	7.84 [£]	

OA10	Alkol. basalt	Powder	0.06	18.1216	7	15.4568	8	37.7941	25	0.9437	0.703358	10	8.02 [£]	
MKMN-d1	Basalt	Chips	0.7	18.1554	7	15.4566	8	37.8202	24	0.9431	0.703393*	6	7.21 ^{\$}	
MKMN (3H)-d2	Basalt	Chips	0.7	18.1556*	20	15.4565*	24	37.8170*	76	0.9427				
MKMN-mean		-		18.1555		15.4565		37.8186		0.9429				
MKL	Basalt	Chips	0.58	18.1374	8	15.4563	9	37.7863	28	0.9412	0.703304*	6	8.09 ^{\$}	
HV02-03-d1	Basalt	Chips	0.8	18.1680	6	15.4591	8	37.8205	23	0.9418	0.703450*	6	8.20 ^{\$}	
HV02-03 (3H)-d2	Basalt	Chips	0.8	18.1698*	16	15.4625*	19	37.8254*	57	0.9421				
HV02-03-mean				18.1689		15.4608		37.8230		0.9420				
HV02-04b	Basalt	Chips	0.6	18.1825	10	15.4609	10	37.8284	29	0.9411	0.703318*	6	7.82 ^{\$}	
HV02-05-d1	Basalt	Chips	0.79	18.2317	7	15.4597	7	37.8733	23	0.9410	0.703358*	7	7.71 ^{\$}	
HV02-05-d2	Basalt	Chips	0.79	18.2346*	13	15.4634*	16	37.8797*	53	0.9414				
HV02-05-mean				18.2332		15.4616		37.8765		0.9412				
HV02-07	Basalt	Chips	0.10	18.1465	7	15.4615	7	37.8120	23	0.9431	0.703350*	5	7.81*	5
HV02-08	Basalt	Chips	0.07	18.1335	11	15.4580	12	37.7982	38	0.9429	0.703345*	6	7.79*	4
HV02-14-d1	Basalt	Chips	0.45	18.1687	6	15.4600	6	37.8205	20	0.9417	0.703496*	6	7.73*	5
HV02-14 (3H)-d2	Basalt	Chips	0.45	18.1726*	23	15.4652*	29	37.8326*	96	0.9427	0.703514*	3		
HV02-14 (3H)-d3	Basalt	Chips	0.45	18.1781*	34	15.4674*	43	37.8452*	141	0.9435				
HV02-14-mean				18.1731		15.4642		37.8328		0.9426				
HV02-19-d1	Basalt	Chips	0.11	18.0492	38	15.4561	34	37.7437	84	0.9458			7.97*	4
HV02-19 (3H)-d2	Basalt	Chips	0.11	18.0509*	28	15.4592*	35	37.7497*	113	0.9463				
HV02-19-mean				18.0501		15.4577		37.7467		0.9460				
RH	Basalt	Chips	0.06	18.0490	11	15.4512	10	37.7360	28	0.9449			7.96*	3
HV02-16	Basalt	Chips	0.48	18.1842*	29	15.4623*	28	37.8432*	76	0.9426	0.703314*	6	7.79*	3
X208	Basalt	Chips	0.43	18.1489*	32	15.4586*	36	37.8060*	110	0.9422	0.703316*	6	7.85*	4
ODP 843 basalts														
First dissolution, 3 h leaching														
136 843 B 1R1/13-17	Basalt	Powder	265.63	18.6490*	19	15.5202*	23	38.2090*	73	0.9304	0.702685*	4	10.64*	9
136 843 B 1R1/59-62 basalt		Powder	266.09	19.4763*	11	15.5117*	14	38.0542*	44	0.9356	0.703662*	3		
136 843 B 2R2/75-79	Basalt	Powder	296	19.2459*	15	15.5481*	16	38.2647*	51	0.8843	0.702834*	4		
136 843 B 2R1/106-109	Basalt	Powder	295.06	18.6355*	19	15.5160*	20	38.0969*	61	0.9242	0.702704*	51	10.70	13
L-136 843 B 1R1/13-17				18.5146*	57	15.5230*	50	38.1335*	125	0.9403				
Second dissolution, step-wise leaching														
136 843 B 1R1/13-17	Basalt	Chips	265.63	18.7487*	24	15.5188*	30	38.2434*	97					
136 843 B 1R1/59-62 basalt		Chips	266.09	18.6330*	26	15.5139*	32	38.1621*	106					
136 843 B 2R2/75-79	Basalt	Chips	296	19.1083*	18	15.5414*	22	38.1906*	71					
136 843 B 2R1/106-109	Basalt	Chips	295.06	18.5570*	20	15.5160*	25	38.0704*	82					

Samples were leached 1 h, unless stated otherwise.

Samples were analyzed using the TIMS Finnigan MAT 261, unless stated otherwise.

"L" stands for leachate; "d1" and "d2" stands for dissolution 1 and 2, respectively; "3H" stands for three hour-leaching (see Supplementary Material 1 for explanation). Star (*) indicates samples which were analyzed with TIMS ThermoElectron, Triton.

 ϵ_{Nd} data on HV marked with "\$" were analyzed with multicolector ICP MS (Nu-plasma).

 ε_{Nd} data on HV marked with "£" are from Stille et al. (1983).

 ϵ_{Nd} data on KSDP in italics are from Salters et al. (2006). Ages for Honolulu Volcanics are from Ozawa et al. (2005).

Sr isotopic ratios analyzed with MAT 261 were normalized to SRM987=0.71025; all Nd isotopic ratios were normalized to La Jolla=0.511858.

Average standard values measured, as follows: *Finnigan MAT 261*: SRM987=0.710184±42 (N=8), SRM981: $^{206}Pb/^{204}Pb=16.9399\pm35$, $^{207}Pb/^{204}Pb=15.4962\pm45$ and $^{208}Pb/^{204}Pb=36.7228\pm116$ (N=51). The external reproducibility for MAT 261 calculated on the basis of repeated measurements of the SRM981 standard (N=51) yielded 207 ppm, 290 ppm and 316 ppm for $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios, respectively. *Nu-plasma*: La Jolla=0.511839 (N=3), Aldrich=0.511405 (N=6). *ThermoElectron, Triton*: SRM987=0.710250±13 (N=6), La Jolla=0.511842±12 (N=6), SRM981: $^{206}Pb/^{204}Pb=16.9426\pm22$, $^{207}Pb/^{204}Pb=15.5000\pm26$ and $^{208}Pb/^{204}Pb=36.7273\pm83$ (N=13). The external reproducibility for ThermoElectron, Triton calculated on the basis of repeated measurements of the SRM981 (N=13) yielded 131 ppm, 171 ppm and 225 ppm for $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ ratios, respectively.



Fig. 2. Depth variations of ${}^{206}\text{Pb}{}^{204}\text{Pb}$ (a), ${}^{208}\text{Pb}{}^{*}{}^{206}\text{Pb}{}^{*}$ (b), ${}^{87}\text{Sr}{}^{86}\text{Sr}$ (c) and ε_{Nd} (d) in Ko'olau Volcano. The isotopic variations of subaerial Makapu'u lavas and rejuvenated Honolulu Volcanics are also shown. Note that a zero-depth assigned to the Honolulu Volcanics (HV) samples is fictive and illustrate the evolution stage rather than actual stratigraphic position. The dashed line at ~575 mbsl marks the limit between unaltered and altered samples (Haskins and Garcia, 2004). Data for Makapu'u subaerial lavas are from Abouchami et al. (2005) (Pb), Stille et al. (1983) (Nd), Roden et al. (1994) (Sr, Nd); KSDP Nd isotope data are from Salters et al. (2006) and HV Nd data are from Stille et al. (1983). Age span for HV is from Ozawa et al. (2005) and age for one KSDP sample is from Haskins and Garcia (2004).

sampling resolution) to the transition from Kalihi-type composition to Makapu'u-type composition (Huang and Frey, 2005). Precise age constraints are not available for the KSDP core; but using the growth rate of 2 mm/year inferred indirectly by Huang and Frey (2005), the depth-related Pb isotopic changes can be translated into a time-scale. In this case, the Pb isotope changes over ~ 25 m depth intervals correspond to ~ 12 kyr cyclicity. This

period is comparable to the 10 kyr cycle reported for the HSDP Mauna Kea record (Eisele et al., 2003). However, this result should be treated with caution given the assumptions made in estimating the growth rate (see Huang and Frey, 2005).

It has been shown that the radiogenic ²⁰⁸Pb*/²⁰⁶Pb* ratio (Galer and O'Nions, 1985) is an important discriminator of Loa versus Kea trend volcanoes (Abouchami



Fig. 3. Pb isotope covariations in KSDP lavas, Honolulu Volcanics (HV) and ODP Site 843 basalts. Present-day Pb isotope ratios are plotted for the 110 Ma-old ODP Site 843 samples. Pb isotope data on Makapu'u subaerial lavas (Abouchami et al., 2005) are shown for comparison. Three distinct Pb isotope arrays, labelled "KSDP", "HV" and "ODP 843" are resolved in 208 Pb $^{-206}$ Pb space (a). In 207 Pb $^{-206}$ Pb space (b), the KSDP and HV arrays overlap but remain distinct from the ODP 843 array. 2σ error bar similar to the symbol size.

et al., 2005) and a useful tracer of compositional changes during the lifetime of a single volcano (e.g. Mauna Kea (Eisele et al., 2003)). Significant differences in $^{208}\text{Pb*}/^{206}\text{Pb*}$ ratios are also observed during the evolution of Ko'olau Volcano, with an increase from the main shield building KSDP lavas (0.949 to 0.961) to the late subaerial Makapu'u stage (0.968 to 0.987) and a reversal to low $^{208}\text{Pb*}/^{206}\text{Pb*}$ values during the rejuvenated stage represented by the Honolulu Volcanics ($^{208}\text{Pb*}/^{206}\text{Pb*}=0.941$ to 0.946) (Fig. 2b). Honolulu Volcanics also exhibit the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ (Fig. 2c) and the highest ε_{Nd} (Fig. 2d), features typical of rejuvenated Hawaiian lavas (Lassiter et al., 2000; Yang et al., 2003; Frey et al., 2005).

2.2. Isotopic covariations

The total range of Pb isotope variations in KSDP and Honolulu lavas is relatively small (see Table 1 and

Fig. 3a,b). However, KSDP and HV lavas form two distinct, well-defined linear arrays in 208 Pb/ 204 Pb vs. 206 Pb/ 204 Pb which intersect at the radiogenic end (Fig. 3a). In 207 Pb/ 204 Pb $-^{206}$ Pb/ 204 Pb space, however, the KSDP and HV arrays cannot be distinguished and the 207 Pb $-^{206}$ Pb regression slopes are similar within analytical error (Fig. 3b) (see Supplementary Material 2, Table 3 for linear regression parameters). The relationships in Pb isotope spaces are consistent with binary mixing and involvement of at least three distinct Pb end members: two unradiogenic components and a radiogenic component common to both KSDP and HV samples (Fig. 3).

The present-day Pb isotopic compositions of ODP Site 843 basalts, which are representative of the old Pacific lithosphere beneath Hawai'i are more radiogenic than those of both KSDP and HV lavas, and define a distinct Pb isotope array in both Pb isotope spaces (Fig. 3a,b). If the HV were derived from old oceanic



Fig. 4. Comparison of Ko'olau isotope systematics with that of recent EPR MORB glasses and 110 Ma-old Pacific lithosphere (ODP Site 843 basalts). (a) 208 Pb/ 204 Pb vs. 206 Pb/ 204 Pb; (b) 207 Pb/ 204 Pb vs. 206 Pb/ 204 Pb; (c): ε_{Nd} vs. 87 Sr/ 86 Sr. For comparison we also show isotope data on Emperor Seamount lavas (Regelous et al., 2003; Huang et al., 2005b), gabbroic crustal xenoliths recovered from Hualãla'i lavas (Lassiter and Hauri, 1998), Salt Lake Crater xenoliths (O'ahu) (Okano and Tatsumoto, 1996; Lassiter et al., 2000) and glasses from Hawaiian North Arch (Frey et al., 2000). Pb isotopes on EPR MORB (Galer et al., 1999) and Emperor Seamount lavas (Regelous et al., 2003) are triple spike data.

lithosphere as commonly hypothesized (e.g. Class and Goldstein, 1997; Lassiter et al., 2000), the present-day Pb isotope ratios of 110 Ma oceanic lithosphere would

be relevant for generating the <1 My-old HV. Note that the age-corrected Pb isotopic compositions of ODP Site 843 basalts (assuming a μ value of 8, a κ value of 3.5 and an age of 110 Ma), in particular the 207 Pb/ 204 Pb ratios, are not significantly different from the measured values. We also note that the present-day Pb isotopic compositions of ODP Site 843 basalts fall within the range of EPR MORB (Fig. 4a,b), demonstrating that the Pb isotope characteristics of the Pacific lithosphere did not undergo major changes over at least the last ~100 Ma.

The ODP Site 843 data form a well-defined array in ²⁰⁷Pb/²⁰⁴Pb vs. ²⁰⁶Pb/²⁰⁴Pb space, whereas they scatter in ²⁰⁸Pb/²⁰⁴Pb vs. ²⁰⁶Pb/²⁰⁴Pb space, suggesting U addition due to post-eruption alteration effect rather than Pb removal, as attested by their MORB-like Ce/Pb ratios (18 to 25; Supplementary Material 2). This is consistent with their high U concentrations relative to those of average MORB, as seen in the primitive-mantle normalized trace element patterns of two out of four samples (Supplementary Material 2, Fig. 2). In general, the trace element patterns of ODP Site 843 basalts are similar to those of average N-MORB and exhibit variable depletion in the most incompatible elements (e.g. Nb, Th, La). The large variability in Cs and Rb contents of ODP Site 843 samples indicate they have been affected by post-eruption alteration processes.

Strontium, Nd and Pb isotopes are generally correlated at the scale of Hawaiian volcanoes. By comparison, KSDP samples have Sr and Nd isotopic compositions intermediate between those of Makapu'u stage lavas, which lie at the high Sr - low Nd end of the global Hawaiian correlation and HV which have the lowest Sr and the highest Nd isotope ratios (Fig. 4c). In ²⁰⁸Pb/²⁰⁴Pb vs. ²⁰⁶Pb/²⁰⁴Pb space (Fig. 5a), the KSDP isotopic compositions overlap the fields defined by Mauna Loa and Nu'uan landslide block samples (see insert Fig. 5a). This indicates some similarities between Mauna Loa and main shield stage KSDP lavas, as suggested by their chemical compositions (Haskins and Garcia, 2004; Huang and Frey, 2005; Salters et al., 2006). However, their distinct slopes in ²⁰⁸Pb/²⁰⁴Pb vs. ²⁰⁶Pb/²⁰⁴Pb space (Fig. 5a) are inconsistent with this conclusion.

3. Discussion

3.1. Sources and mixing relationships in Ko'olau

The isotope variations in Hawaiian shield volcanoes have been explained in terms of mixing two (Bennett et al., 1996; Lassiter and Hauri, 1998; Blichert-Toft et al., 1999) or three components (Chen and Frey, 1983; Hauri, 1996; Eiler et al., 1996). Case studies of Hawaiian shield volcanoes have also revealed the existence of short-term (Pietruszka and Garcia, 1999) and long-term isotopic heterogeneities (Abouchami et al., 2000; Eisele et al., 2003). The long-term variations observed within the 1-3 km-long Hawaiian Scientific Drilling Project cores requires up to four components in the source of Mauna Kea over the past 320 ka (Abouchami et al., 2000; Eisele et al., 2003).

The linear Pb isotope array for the 328 m-thick KSDP section indicates the involvement of only two components in the Ko'olau source during the shield building — an unradiogenic and a radiogenic Pb component (Fig. 3a). The end member components can be described in terms of (1) an enriched EM1-type or "Ko'olau" component, represented by Makapu'u subaerial lavas, which lie at the unradiogenic extreme end of the KSDP array, and (2) a second more radiogenic Pb component, but still less radiogenic than Kea-type components. We will refer in the following to these inferred source end members as the "Makapu'u" and "Kalihi" components, respectively. Fluctuations in the mixing proportion of these two components occurred through time as shown by the oscillations of Pb isotope ratios with depth (Fig. 2a,b), while still lying along the Pb–Pb mixing line shown in Fig. 3.

The new results indicate that the Makapu'u component is not restricted to the Makapu'u section, but is clearly a long-lived feature of the Hawaiian mantle plume, waxing and waning throughout the volcano's evolution (Fig. 2). The waxing-waning effect is supported by the observation that KSDP samples with the least radiogenic Pb isotopic compositions (e.g. units 6 and 9) also have the lowest ε_{Nd} , ε_{Hf} and highest Sr isotopic compositions — features that are indeed typical of Makapu'u lavas. In addition, these samples belong to the group identified by Huang and Frey (2005) as having the most obvious Makapu'u trace element features. Thus, the short-scale Pb isotope fluctuations in the KSDP stratigraphic record (Fig. 2a,b) support relatively rapid changes in end member mixing proportions occurring with a cyclicity of ~12 kyrs.

The Kalihi component is found in the main and late shield-stage lavas, and rejuvenated Honolulu Volcanics. In addition to its elevated Pb isotope ratios (Fig. 5a), the Kalihi component has lower Sr and higher Nd isotope ratios than the Makapu'u component (Fig. 5b,c). On the basis of isotope data obtained on Nu'uanu land-slide blocks, Tanaka et al. (2002) suggested that a Mauna Kea-like source contributed to the mixing that produced Ko'olau shield lavas. This so-called "Kea" component is characterized by the highest ²⁰⁶Pb/²⁰⁴Pb and ¹⁴³Nd/¹⁴⁴Nd, and the lowest ⁸⁷Sr/⁸⁶Sr and ¹⁸⁷Os/¹⁸⁸Os ratios amongst Hawaiian shields (Tatsumoto, 1978; Stille et al., 1986; Hauri, 1996; Lassiter and

Hauri, 1998) — isotope features that have been interpreted as reflecting the involvement of old recycled oceanic lithosphere (Stille et al., 1986) or entrained upper mantle during plume ascent (Hauri, 1996; Lassiter et al., 1996). The HSDP Pb isotope data, however, have demonstrated that there is no unique "Kea" component. Rather, the long-term Pb isotopic evolution of Mauna

Kea requires a diversity of variably depleted components mixing with a young "high- μ " source (Eisele et al., 2003). In a ²⁰⁸Pb/²⁰⁴Pb vs. ²⁰⁶Pb/²⁰⁴Pb plot, the KSDP and HV Pb arrays intersect at a point slightly on the Keatrend (i.e. low ²⁰⁸Pb/²⁰⁴Pb) side of the dividing line between Loa and Kea trends defined by Abouchami et al. (2005) (Fig. 5a). However, none of three Kea trends seen



in Fig. 5a converges toward the Kalihi end member of the Koʻolau lavas.

The systematic differences in isotopic composition between KSDP and Makapu'u stage lavas - more radiogenic Pb. Nd and Hf but less radiogenic Sr isotope ratios in KSDP - reflect changes in compositional sources and their mixing proportions: the main shieldbuilding has a predominantly high ²⁰⁶Pb/²⁰⁴Pb (radiogenic) component, whereas a low ²⁰⁶Pb/²⁰⁴Pb (unradiogenic) component is present in the late shield-building stage. Contributions of both components vary on a short-time scale (Fig. 2a) with a gradual increase of the proportion of the low ²⁰⁶Pb/²⁰⁴Pb (unradiogenic) endmember as Ko'olau tends towards the late-shield stage of its evolution. Because a major shift in these components is associated with a specific time of the volcano's evolution, it must reflect variations in the material/lithology being melted. Such an inference is consistent with the gradual changes in major and trace element ratios documented during the transition from the Kalihi to the Makapu'u stage (Haskins and Garcia, 2004; Huang and Frey, 2005).

The EM-1-type isotopic characteristics and the high SiO2 content of Makapu'u stage lavas have been related to a source in the plume consisting of eclogite (Hauri, 1996; Lassiter et al., 2000; Pertermann and Hirschmann, 2003; Sobolev et al., 2005), derived from old recycled oceanic crust (Eiler et al., 1996; Lassiter and Hauri, 1998; Blichert-Toft et al., 1999; Sobolev et al., 2000; Huang and Frey, 2005; Sobolev et al., 2005) and also perhaps containing small amounts of ancient pelagic sediments (Blichert-Toft et al., 1999; Huang and Frey, 2005). Huang and Frey (2005) called upon an increased contribution of a sedimentary component (up to 3%) in SiO₂-rich melts (dacitic) derived from a garnet pyroxenite source during the late shield-building Makapu'u stage. On the other hand, the Hf-Nd isotope relationships found in KSDP lavas (Salters et al., 2006) have been explained in terms of ancient depleted lithosphere present in the Hawaiian plume, an idea initially proposed

by Norman and Garcia (1999) and supported by recent studies on rejuvenated volcanics (Frey et al., 2005) and Salt Lake Crater xenoliths (Bizimis et al., 2003, 2005).

The coexistence of both fertile eclogite and depleted peridotite in the Hawaiian plume, and changes in the relative proportions of these components in the melts as the volcano moves away from the plume center has been demonstrated by both experimental and geochemical studies (Ren et al., 2004; Sobolev et al., 2005; Herzberg, 2006). In particular, the geochemical characteristics of Makapu'u stage lavas have been interpreted in terms of an increased contribution of eclogitic component to the peridotite-derived melts (e.g. Norman and Garcia, 1999; Haskins and Garcia, 2004; Huang and Frey, 2005; Herzberg, 2006). The Ko'olau Pb isotope data are consistent with such a view: the Makapu'u and Kalihi components identified in Pb isotope space correspond, broadly, to the lithological components "eclogite" and "peridotite", respectively. These components coexist in the plume source and contribute simultaneously but in variable proportion during shield building. We performed binary mixing calculations using the extreme measured Pb isotopic compositions of KSDP and Makapu'u lavas as representative of the pure end member Kalihi and Makapu'u components (see Supplementary Material 3). These calculations indicate that the Makapu'u eclogite component (unradiogenic Pb, Nd, Hf and high Sr isotope ratios) contributed 10% of melt to the main shield building of Ko'olau, increasing drastically to over 80% towards the end of shield building. The Sr and Nd isotope composition of the components, extrapolated from the relationships between Sr, Nd and Pb isotope ratios (Fig. 5), are very similar to those reported by Bianco et al. (2005) and provide similar mixing proportions to those obtained with Pb isotopes (see Supplementary Material 3). These figures are consistent with the results of Gaffney et al. (2004) and Sobolev et al. (2005) who inferred up to 80% of garnet pyroxenite-derived melts in Makapu'u stage lavas.

Fig. 5. Comparison of isotope systematics for Ko'olau Volcano with other Hawaiian shields and the Loa–Kea compositional boundary. (a): in 208 Pb/ 204 Pb vs. 206 Pb/ 204 Pb diagram, KSDP lavas plot on the Loa side of the Loa–Kea boundary (dashed black line) defined by Abouchami et al. (2005) and define a trend whose slope is distinct from that of Mauna Loa array. Also shown are the three Mauna Kea arrays of Eisele et al. (2003). The insert shows the Nu'uanu landslide blocks data from Tanaka et al. (2002) and illustrate the similarity of KSDP (filled circles) with the Nu'uanu 2 landslide blocks (grey field). Kalihi, Makapu'u and Rej. depleted ("Rej.": rejuvenated) correspond to the three mixing components inferred from the KSDP and HV Pb arrays (see text). (b): 87 Sr/ 86 Sr vs. 206 Pb/ 204 Pb. (c) ε_{Nd} vs. 206 Pb/ 204 Pb. The inferred average Sr and Nd isotopic compositions for the three components are plotted as large grey circles. Lead triple spike, Sr and Nd isotopes data on EPR MORB are from Galer et al. (1999). Lead triple spike, Sr and Nd data on Hawaiian shield volcanoes are from Abouchami et al. (2005) and Eisele et al. (2003) (Mauna Kea HSDP-1 core). Additional data (Pb, Sr and Nd) were compiled from the literature as follows: Mauna Kea from Blichert-Toft et al. (2003) and Bryce et al. (2005), Kilauea from Pietruszka and Garcia (1999) and Kimura et al. (2006), Lana'i from Gaffney et al. (2005), West Mau'i from Gaffney et al. (2004), East Moloka'i from Xu et al. (2005), Kaho'olawe from Huang et al. (2005a) and Haleakala from Ren et al. (2006). Nd data on KSDP lavas are from Salters et al. (2006).

The time-dependent changes in endmember mixing proportions in Ko'olau lavas (Fig. 5) can be accounted for by changes in the melting process, temperature and pressure (depth) as the volcano drifts away from the plume axis. In the main shield building, high-degree melting in the center of the plume led to preferential melting of the Kalihi (i.e., peridotite) component whose contribution waned towards the end of the shield building. In the late shield building stage, when the volcano is located at the peripheral, cooler part of the plume, lower degree melting resulted in enhanced proportions of the Makapu'u component (i.e., eclogite) in the melts generated to produce the Makapu'u lavas. The interpretation that the proportions of eclogite (Makapu'u) to peridotite (Kalihi) fluctuate during shield building is consistent with the model of Pertermann and Hirschmann (2003) which predicts that peridotite will dominate melting in the central, hottest part of the plume, whereas eclogite melting is more prevalent at the cooler plume periphery.

Most likely, the eclogite Makapu'u component of Ko'olau is irregularly distributed within the Hawaiian plume. Its EM-like Pb isotope fingerprint has been detected only in volcanoes from the Loa track older than 1 Ma (Kaho'olawe, Lana'i, Ko'olau) (Abouchami et al., 2005) and Wai'anae (Coombs et al., 2005, Abouchami, unpub.). Most important, it appears to be sampled predominantly during the late stage of shield growth. This observation is at odds with the decrease in the proportion of "eclogite" (recycled oceanic crust) from the plume center ($\sim 30\%$) to the edge (nearly 0%) inferred by Sobolev et al. (2005), who already pointed out that there exists an inconsistency for Ko'olau. They called upon the presence under Ko'olau of a large eclogitic body, as shown in the melting model of Takahashi and Nakajima (2002) where eclogitic blocks may survive in the plume and generate melts similar to those of Makapu'u late shield stage. The Ko'olau Pb isotope data, however, indicate clearly that the eclogitic component dominates the late shield building phase, suggesting that this body is located at the plume margin, rather than near the plume center, where the peridotitic source has been nearly "exhausted" due to the high extent of melting during earlier shield building. The fact that this component dominates the late stages of five consecutive volcanoes (Hualãlei, Kaho'olawe, Lana'i, Ko'olau, Wai'anae) indicates that it has a considerable vertical continuity within the Hawaiian plume and cannot simply be an isolated "block", which in any case would be rheologically untenable (see Farnetani and Samuel, 2005). This continuity resembles that previously observed by Abouchami et al. (2005) for Kilauea and Mauna Kea lavas.

3.2. Source(s) of Honolulu Volcanics

The source of the Honolulu Volcanics has been a long-standing subject of discussion. Honolulu lavas have low Sr and high Nd isotope ratios compared to Ko'olau shield stage lavas, and show a relative homogeneity of Sr, Nd, Pb and Hf isotopic ratios (Stille et al., 1983; Roden et al., 1984). These isotope features indicate a long-term history of incompatible element depletion of the source which contrasts with the enrichment in highly incompatible elements observed in the lavas (Abouchami et al., 2005). Roden et al. (1984) suggested that mixing of high proportion of a relatively enriched mantle with a few percent ($\sim 2.5\%$) of a very low degree ($\sim 0.26\%$) melt of a MORB source could generate the HV lavas. Other hypotheses for the origin of HV include melts derived from the ~100 Ma-old oceanic lithosphere beneath Hawai'i (Chen and Frey, 1983; Class and Goldstein, 1997; Lassiter et al., 2000) or ancient depleted lithosphere (Frey et al., 2005) similar to that sampled by Emperor Seamount lavas (Regelous et al., 2003). The isotopic similarities of HV with Salt Lake Crater (SLC) xenoliths (Oahu) have also been used to argue that they are genetically related (Frey et al., 2005; Bizimis et al., 2005), but whether the xenoliths are a product or source of the HV remains an open question.

So, are the HV magmas derived from the Pacific plate (or the underlying Pacific asthenosphere) or do the sources reside in the Hawaiian plume? If the HV isotopic signatures are derived from melting or interaction of melts with the Pacific lithosphere this is readily testable. In such a case, one (or both) of the Pb isotope end-members found in the HV (Fig. 3) would constitute a "Pacific Plate" component. Samples of the ~ 100 Ma-old basaltic basement near to Hawai'i are only available from ODP Site 843. Pb isotope data from a previous study by King et al. (1993) scatter widely and lack sufficient analytical precision to be useful in this regard. Our new data, presented in Table 1, are a proxy for the present-day Pb isotopic composition of the Pacific Plate, as it might contribute to the HV. Similarly, East Pacific Rise (EPR) MORB represents the composition of the present-day accreting Pacific lithosphere (Galer et al., 1999).

Comparison of HV Pb isotope data with those of two proxies for the composition of the Pacific lithosphere under Hawai'i demonstrates convincingly that the HV magmas are not derived from the lithospheric plate (see Fig. 4). In both Pb isotope spaces, the Honolulu Volcanics, EPR MORB and ODP Site 843 basalts arrays can be distinguished (Fig. 4a,b). At a given ²⁰⁶Pb/²⁰⁴Pb ratio, HV have higher ²⁰⁸Pb/²⁰⁴Pb but lower ²⁰⁷Pb/²⁰⁴Pb ratios than both EPR MORBs and ODP Site 843 basalts,

providing evidence against derivation of HV from the Pacific lithosphere (Figs. 3 and 4). Strontium and Nd isotopes data confirm the distinct characteristics of Honolulu Volcanics and Pacific MORB. At a given ²⁰⁶Pb/²⁰⁴Pb ratio, Honolulu Volcanics have higher ⁸⁷Sr/⁸⁶Sr and lower ¹⁴³Nd/¹⁴⁴Nd ratios than those measured in both EPR MORB and ODP Site 843 basalts (Fig. 5b,c).

We thus conclude that the rejuvenated stage Honolulu Volcanics are not derived from melting of any known (young or old) "normal" MORB-related lithospheric source. Instead, they sample components intrinsic to the Hawaiian plume: (1) a radiogenic plume component (Kalihi) shared by KSDP and Makapu'u stage lavas, and (2) a depleted component with an unradiogenic Pb isotopic composition that is distinct from that of the ~ 100 Ma old Pacific lithosphere under Hawai'i. The latter depleted component is found only in the HV lavas and is clearly unrelated to the Pacific upper mantle. Binary mixing calculations indicate that the depleted rejuvenated component contributed variably, 10-50% melt, to the HV with the remainder derived from the Kalihi plume component (Supplementary Material 3).

Our interpretation corroborates the findings of Frey et al. (2005) who suggested that a depleted component, similar to that found in the Emperor Seamounts lavas (Regelous et al., 2003), is sampled by rejuvenated volcanism in Hawai'i. The Pb isotope characteristics of the HV lavas analysed here overlap the field of lavas from the Emperor Seamounts (Fig. 4). Our results show that (1) both the rejuvenated "depleted" component and the radiogenic Kalihi component reside in the Hawaiian plume, and (2) the HV are plume-derived melt mixtures of these two components. This conclusion is at odds with claims that there is no significant Hawaiian plume influence on rejuvenated volcanism, which are based on the higher ¹⁸⁷Os/¹⁸⁸Os ratios of rejuvenated lavas and SLC pyroxenites xenoliths compared to Hawaiian shield lavas (Lassiter et al., 2000). On the other hand, the Hf-Nd relationships in the SLC xenoliths has been interpreted in terms of mixing between melts derived from an ancient depleted lithosphere (high Hf ratios) - similar to the rejuvenated depleted component identified by Frey et al. (2005) - and plume-derived melts (less radiogenic Hf) (Bizimis et al., 2005). No high-precision Pb isotope data are yet available on the SLC xenoliths, and the few existing data have similar ²⁰⁶Pb/²⁰⁴Pb but lower 208 Pb/ 204 Pb than those of HV samples (Fig. 4), which is may well be the result of instrumental mass fractionation. If this were the case, then the SLC xenoliths and their host lavas HV would share a common depleted

Pb component, in agreement with the inference from Hf–Nf isotope data (Bizimis et al., 2005).

The high incompatible element contents of HV lavas (e.g., Lassiter et al., 2000; Yang et al., 2003, this study) might be explained by an enrichment process, most likely via a metasomatic agent. Interaction between ancient metasomatized lithosphere and Hawaiian plume melts has been suggested to explain the origin of the HV (Roden et al., 1984; Class and Goldstein, 1997). We have, however, shown that the lithosphere can be definitely ruled out as a potential source for the HV. Instead, we propose that the HV lavas are generated by very low-degree melting of a depleted mantle source intimately associated with the Hawaiian plume. The most plausible origin is that it represents deep-seated ambient mantle thermally accreted onto the sides of the plume core during its ascent. Such an explanation has already been put forward for Hawaiian post-shield volcanism by Hauri et al. (1994) to explain the MORB-like (i.e., "depleted") isotopic signature of magmas erupted away from the plume axis. Ribe and Christensen (1999) considered that shield stage lavas stem from a primary melting zone located in the hottest central part of the plume, while rejuvenated lavas are derived from a secondary melting zone 300 and 500 km downstream. In this model, there is no melting of either the ambient mantle or the lithosphere. Our Pb isotope data show in fact that both "plume" core material (Kalihi component) and "entrained" mantle (rejuvenated depleted component) contribute to the Honolulu Volcanics.

3.3. Koʻolau evolution and plume structure

Two basic models for the geochemical structure of the Hawaiian mantle plume are: (1) radial zoning whereby the Kea trend volcanoes sample the peripheral parts of the plume while the Loa trend samples the central portions of the plume (Hauri et al., 1994; DePaolo and Stolper, 1996; Bryce et al., 2005), and (2) bilateral asymmetry and vertical continuity with bundles of vertically stretched-out "filaments" in the plume conduit, each being sampled successively as the volcano traverses the plume (Eisele et al., 2003; Abouchami et al., 2005; Farnetani and Samuel, 2005, see also Blichert-Toft et al., 2003). The temporal evolution of Ko'olau Volcano bears directly on these plume models.

The first question to address is whether the KSDP data support a temporal evolution of Ko'olau Volcano from Kea-like to Loa-like compositions through time, as suggested by Tanaka et al. (2002). In an attempt to explain this feature, Abouchami et al. (2005) suggested that the Loa–Kea boundary might stop at the Moloka'i Fracture



Fig. 6. KSDP and Hawaiian rejuvenated lavas in Pb isotope space. Dashed line represents the Loa/Kea boundary (Abouchami et al., 2005). Ko'olau belongs to the Loa-trend volcances (Abouchami et al., 2005) and KSDP lavas plot on the left (Loa-trend) side of the Loa–Kea boundary. However, the rejuvenated lavas from Ko'olau (Honolulu Volcanics) plot together with rejuvenated lavas from West Maui (Lahaina Volcanics) and East Moloka'i (Kalaupapa Volcanics) on the Kea side of the boundary. The rejuvenated Koloa Volcanics from Kaua'i straddle the Loa/Kea boundary but predominantly plot in the Kea field. Literature source data: Kalaupapa (Xu et al., 2005), Kaua'i (Sun, 1980; Lassiter et al., 2000) and West Maui (Gaffney et al., 2004).

Zone, and that the Ko'olau transition from Kea- to Loalike features might reflect a change in the azimuth of the Loa-Kea compositional boundary and plate motion. Our results indicate that KSDP lavas differ isotopically from Makapu'u late stage lavas: they have Sr and Nd isotope ratios ranging from Loa-like to intermediate between Loa and Kea trends (Fig. 5b,c). But Sr and Nd isotopes are less diagnostic of the two trends, since there is considerable overlap between Kea and Loa trends in Sr-Nd isotope space. In contrast, lead isotopic compositions of the KSDP lavas lie completely in the field of the Loa-trend volcanoes (Fig. 6). Thus, the KSDP Pb isotope record shows a persistent "Loa" signature in the main shield stage Ko'olau lavas, providing support for the extension of the Loa-Kea compositional boundary beyond the Molokai Fracture Zone. However, the possible existence of Kea-like features in lavas older than those sampled by the KSDP core cannot be ruled out.

The isotopic shifts seen in Ko'olau Volcano cannot be ascribed to plume radial zonation based on two lines of evidence. First, the Pb isotope variations seen in the stratigraphic KSDP record at 25 m depth intervals (Fig. 2), corresponding to ~ 12 kyr cycle, are comparable to the 10 kyr — cycle described in the HSDP2 record of Mauna Kea (Eisele et al., 2003). These short duration shifts are probably related to melt transport and magma chamber processes and thus have little bearing on the radial plume structure (Eisele et al., 2003; Bryce et al., 2005). Second, the Ko'olau Nd isotope data exhibit the reverse trend to that predicted by the radially zoned plume, namely, as was noted for the HSDP2 Nd isotope record, values increase with decreasing age, from the center (older lavas) to the periphery (younger lavas) of the plume (Bryce et al., 2005). The KSDP record shows the opposite trend — that is a decrease in ε_{Nd} from the early shield building (ε_{Nd} =+6) to the late shield building stage (ε_{Nd} =-2) (Fig. 2d) — with a noticeable hiatus in the isotope values.

On the basis of the KSDP data, we propose a model for the evolution of Ko'olau Volcano shown schematically in Fig. 7. Vertically distributed peridotite filaments (black) and embedded eclogitic bodies (light grey) are stretched out by shearing in the plume center (see Farnetani and Samuel, 2005) and these components melt and mingle to form binary mixtures seen in the Pb isotope systematics. The isotopic shift from the early to the late shield-building can be explained in terms of changes in the proportions of the peridotite and eclogite components in the plume. In the first stage, mixing between a peridotite matrix and eclogite melts produces the KSDP main shield stage lavas, with high degree melting leading to a larger proportion of peridotite-derived melts. In the second stage, as the volcano drifts away from the plume center, the proportion of eclogite-derived melts increases in the Makapu'u late shield lavas, possibly due to the presence of a large eclogitic body located at the plume margin (see



Fig. 7. Cartoon illustrating the evolution of Ko'olau Volcano from the main shield to late shield and rejuvenated stage from the point of view of potential sources in and around the Hawaiian plume. The diagram shows a schematic cross-section with depth, slicing in a roughly E–W direction; plate motion is from right to left (thin arrow in the upper part of the cartoon). The overall plume structure consists of fast-upwelling material from a deep plume source forming a plume core (upward black arrows), surrounded by sheaths of thermally-entrained mantle (thick side arrows on the left of the cartoon) — see Hauri et al. (1994). The plume core is postulated to consist of a vertically stretched-out mixture of peridotite and eclogite/ pyroxenite (light grey lines) which have contrasting solidi. These lithologies correspond, respectively, to the Kalihi and Makapu'u isotopic components (Fig. 5). The isotopic signatures of the Ko'olau main shield stage lavas (1) are mostly derived by melting of the peridotite, the late-stage lavas (2) from pyroxenite, while the rejuvenated stage HV (3) are from low-degree melts of entrained mantle. Importantly, the large degrees of melting involved in shield-building effectively strip out the plume core of highly incompatible elements, leaving a "barren zone" flowing right-to-left beneath the lithosphere. Although the "barren zone" may contribute melts to the HV, the isotopic signature of the HV is derived mostly from materials unmelted during shield building, lying outside the plume core.

Section 3.1). We infer that this eclogite body was "consumed" prior to the rejuvenation stage based on its absence in HV lavas (Fig. 3).

Comparison of rejuvenated lavas from Ko'olau and other Hawaiian volcanoes shows that Honolulu Volcanics, Lahaina Volcanics (West Maui) (Gaffney et al., 2004), Kalaupapa Volcanics (East Moloka'i) (Xu et al., 2005) and Koloa Volcanics (Kaua'i) (Sun, 1980; Lassiter et al., 2000) share some similarities and have a relatively restricted range in isotopic compositions. For example, these lavas have lower ²⁰⁸Pb/²⁰⁴Pb ratio, at a given ²⁰⁶Pb/²⁰⁴Pb, compared to their underlying shield lavas (Fig. 6). In addition, rejuvenated lavas have consistently less radiogenic Sr and more radiogenic Nd isotopic compositions than the associated shield lavas. Because of this, most previous authors (e.g. Roden et al., 1984; Lassiter et al., 2000) suggested the rejuvenated lavas are derived from the depleted Pacific lithosphere (or asthenosphere). However, as we have already discussed, the HV lavas cannot be derived from the Pacific

lithosphere. Rather, the HV sample a distinct, depleted component of the Hawaiian plume itself. This component does not conform to the established isotopic distinction between shield lavas derived from the Loa and Kea trends (Abouchami et al., 2005).

The question remains as to why this depleted component is sampled only during the rejuvenated stage of volcanism? Regelous et al. (2003) proposed that the depleted refractory Hawaiian plume component is preferentially sampled due to relatively high extent of melting, which would only occur when the plume rises under young, thin oceanic lithosphere. Recent seismic data indicate that the lithosphere thins markedly along the Hawaiian chain from 100 km to 50-60 km beneath Kaua'i (Li et al., 2004). As the Pacific plate overrides the plume, the plume material is dragged along by the lithosphere; since the plume heats the base of the lithosphere, the lithosphere-asthenosphere boundary becomes shallower (Li et al., 2004). Thus, the plume material that had ceased melting can rise to shallower levels beneath the thinned lithosphere - this by itself may be sufficient to trigger the "rejuvenated" melting of the plume. Alternatively, melting could be consistent with flexture-induced upwelling and decompression (Ten Brink, 1991; Bianco et al., 2005).

Bianco et al. (2005) consider the rejuvenated stage volcanism to derive from a region above the plume core near to the base of the lithosphere; however, we consider the region of entrained mantle lying below the core of the plume to be equally as plausible if not more so (Fig. 7). First, if the "upper parts" of the plume were to melt to form the HV, it is hard to envision why some deep Pacific lithosphere should not also contribute — a situation we can firmly exclude. Second, the upper parts of the plume have been cooled off by conduction to the lithosphere and are less likely to melt.

Our preferred explanation is shown schematically in Fig. 7. Because melting in the inner plume core has nearly "exhausted" its incompatible element budget during the early shield stage, a "barren zone" is created, which is simply the downstream extension of the extensively melted core material. The trace element signatures in the rejuvenated lavas are little derived from material within the barren zone, even if some melting takes place there, simply because it is largely devoid of incompatible elements. The plume-derived melt signatures present in the HV are instead derived mostly from the (originally) outermost parts of the plume which still retain a reasonable incompatible element content. This region is lying above and below the plume core -i.e., in the parts which formed initially the periphery of the plume. From a practical standpoint, in order to generate the HV, melts from the "depleted" thermally accreted parts of the plume (Fig. 7) must mix with some nonbarren melts derived from more central parts (Kalihi component). Lastly, it is conceivable that some melting of the plume core contributes major elements to the HV, but effectively none of its incompatible element character. Thus, the trace element signature of posterosional volcanism should be used with caution for delimiting where and by how much melting takes place, or forming the basis for dynamical models (e.g. Ribe and Christensen, 1999).

4. Conclusions

The temporal isotope evolution of Ko'olau Volcano shows similarities to, and differences from, that found for Mauna Kea in the HSDP record, but in both cases, isotopic heterogeneites on both short and large-scale are persistent features of the Hawaiian mantle plume.

On the basis of Pb, Sr and Nd isotope data on lavas from KSDP and Honolulu Volcanics, we show that source variations occurred throughout the evolution of Ko'olau Volcano. Three isotopically distinct components - Makapu'u, Kalihi and rejuvenated depleted component - can be identified from the Pb isotope systematics. The enriched (EM1-type) Makapu'u component - characterized by unradiogenic Pb and Nd, and radiogenic Sr isotope ratios - is a long-lived feature of the Hawaiian plume, despite its predominance in the lateshield building stage. The Kalihi component - with radiogenic Pb and Nd but unradiogenic Sr - persisted throughout Ko'olau's evolution, since it contributed in variable proportions to the main shield KSDP lavas, the late shield subaerial Makapu'u lavas and the rejuvenated HV.

We show, in contrast to previous studies, that the rejuvenated-stage HV do not share any Pb isotopic similarities with the Pacific lithosphere. Instead, the HV source samples two distinct components (1) the longlived Kalihi component, and (2) a "depleted" component, characterized by unradiogenic Pb isotopic compositions, low ⁸⁷Sr/⁸⁶Sr and high ¹⁴³Nd/¹⁴⁴Nd ratios. Due to its lack of similarity to East Pacific Rise MORB and basaltic basement close to Hawai'i, it is exceedingly unlikely that the "depleted" component in the HV has a source in the Pacific Plate or shallow asthenosphere. This depleted rejuvenated component appears to be instead part of the overall plume structure. We suggest it represents material from the deeper part of the mantle which becomes thermally accreted laterally to the plume stem on its ascent through the mantle. The rejuvenated stage of melting - producing the HV - mixed smalldegree melts of the depleted component with the more plume-like Kalihi component, and may be the result of lithospheric thinning or flexure.

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Appendix A. Supplementary Material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j. epsl.2007.06.005.

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