Cretaceous-to-recent record of elevated ^{3}He/^{4}He along the Hawaiian-Emperor volcanic chain

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[1] Helium isotopes are a robust geochemical tracer of a primordial mantle component in hot spot volcanism. The high ^{3}He/^{4}He (up to 35 R_A, where R_A is the atmospheric ^{3}He/^{4}He ratio of 1.39×10^{-6}) of some Hawaiian Island volcanism is perhaps the classic example. New results for picrites and basalts from the Hawaiian-Emperor seamount chain indicate that the hot spot has produced high ^{3}He/^{4}He lavas for at least the last 76 million years. Picrites erupted at 76 Ma have ^{3}He/^{4}He (10–14 R_A), which is at the lower end of the range for the Hawaiian Islands but still above the range of modern mid-ocean ridge basalt (MORB; 6–10 R_A). This was at a time when hot spot volcanism was occurring on thin lithosphere close to a spreading ridge and producing lava compositions otherwise nearly indistinguishable from MORB. After the hot spot and spreading center diverged during the Late Cretaceous, the hot spot produced lavas with significantly higher ^{3}He/^{4}He (up to 24 R_A). Although ^{3}He/^{4}He ratios stabilized at relatively high values by 65 Ma, other chemical characteristics such as La/Yb and ^{87}Sr/^{86}Sr did not reach and stabilize at Hawaiian-Island-like values until ~45 Ma. Our limited ^{3}He/^{4}He record for the Hawaiian hot spot shows a poor correlation with plume flux estimates (calculated from bathymetry and residual gravity anomalies [*Van Ark and Lin*, 2004]). If ^{3}He is a proxy for the quantity of primordial mantle material within the plume, then the lack of correlation between ^{3}He/^{4}He and calculated plume flux suggests that variation in primordial mantle flux is not the primary factor controlling total plume flux.

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Theme: Movement, Dynamics, and Geochemical Evolution of the Hawaiian Hot Spot
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1. Introduction

[2] High ^{3}He/^{4}He ratios (up to nearly 50 R_A [*Stuart et al.*, 2003]) found at ocean islands such as Hawaii and Iceland are fundamental evidence for the existence of mantle plumes rising from regions deep within the Earth [e.g., *Kurz et al.*, 1982; *Lupton*, 1983; *Allègre et al.*, 1983; *O'Nions*, 1987], although a few have challenged this viewpoint [e.g., *Anderson*, 1998; *Meibom et al.*, 2003]. A limitation to the utility of helium isotopes in improving our understanding of mantle plumes has been the lack of studies of long-term ^{3}He/^{4}He variability at ocean island hot spots. Almost all reported analyses from hot spots are of young rocks (<10 Ma), so unlike other radiogenic isotope systems (e.g., Sr-Nd-Pb [*White et al.*, 1990; *Weis et al.*, 1992; *Class et al.*, 1993; *Keller et al.*, 2000; *Regelous et al.*, 2003]), little information exists on the long-term temporal evolution of hot spot ^{3}He/^{4}He ratios. Here we present new helium isotope data for lavas from the Hawaiian-Emperor chain of seamounts and islands which show that the Hawaiian hot spot has been capable of producing lavas with ^{3}He/^{4}He higher than upper mantle (MORB source) values for at least the last 76 million years.

[3] The trace of the Hawaiian hot spot is marked by more than 100 islands and seamounts that make up the Hawaiian-Emperor volcanic chain in the northwest Pacific Ocean (Figure 1). Geochemical studies revealed significant temporal variability of the Hawaiian hot spot over >80 million years of its history [*Lanphere et al.*, 1980; *Basu and Faggart*, 1996; *Keller et al.*, 2000; *Regelous et al.*, 2003]. The oldest dated Emperor Seamount (Detroit at 76–81 Ma) has ^{87}Sr/^{86}Sr values essentially indistinguishable from MORB [*Keller et al.*, 2000]. Other geochemical features of Detroit Seamount are also MORB-like, although triple-spike Pb isotope data [*Regelous et al.*, 2003] and Ba/Th data from glasses [*Huang et al.*, 2004] are outside of the ranges of modern MORB. With time, the hot spot produced lavas that were less MORB-like. For example, La/Yb and ^{87}Sr/^{86}Sr ratios in Late Cretaceous and Early Tertiary tholeiitic basalts (81–43 Ma) increase steadily southward along the Emperor chain, from MORB-like values (as low as 1.1 and 0.70262, respectively, at ODP Site 884 on Detroit Seamount), to Hawaiian Island-like values (mean of 6.4 and 0.70346) in the southern Emperor Seamounts. These higher values continue southeast of the Hawaiian-Emperor bend all the way to the Hawaiian Islands, although the range of values increases in the well-sampled islands. The temporal trend away from MORB-like compositions in the older Emperor seamounts was attributed to increasing distance between the hot spot and a seafloor spreading center, and the resultant increase in lithospheric thickness [*Lanphere et al.*, 1980; *Keller et al.*, 2000; *Regelous et al.*, 2003]. Younger (thinner) lithosphere permits a taller melting column in the ascending mantle plume, leading to larger extents of partial melting [*Ellam*, 1992; *Keller et al.*, 2000]. This increases the likelihood that depleted mantle, whether entrained from the surrounding upper mantle [*Keller et al.*, 2000] or intrinsic to the plume [*Regelous et al.*, 2003], would make a greater contribution to hot spot volcanism.

[4] Individual ocean islands often show spatial and temporal variability in ^{3}He/^{4}He that is usually explained by intrinsic isotopic heterogeneity within the plume, or by mixing between high-^{3}He/^{4}He plume-derived material and lower-^{3}He/^{4}He material ($\sim 6-10$ R_A) derived from the MORB source region in the upper mantle [*Graham*, 2002]. However, little is known about the long-term ^{3}He/^{4}He evolution of mantle plumes because of difficulties in unambiguously measuring the initial ^{3}He/^{4}He in old volcanic rocks, due to factors such as post-eruptive radiogenic production of ^{4}He, low ^{3}He contents resulting from diffusive loss, and a lack of unaltered helium-retentive minerals such as olivine or pyroxene. Tertiary lavas from the Iceland hot spot have been successfully studied [*Graham et al.*, 1998; *Marty et al.*, 1998; *Hilton et al.*, 1999; *Breddam and Kurz*, 2001; *Stuart et al.*, 2003], but we are not aware of published data from pre-Tertiary hot spot lavas.

[5] Suitably fresh samples from three Late Cretaceous-Early Tertiary seamounts along the Emperor Seamount chain (Figure 1) now make it possible to study the ^{3}He/^{4}He evolution of the Hawaiian hot spot. Samples used in this study include tholeiitic picrites recovered during Leg 197 of the Ocean Drilling Program from the shield-building phase of Detroit Seamount at Site 1203 (76 Ma), and from Koko Seamount at Site 1206 (49 Ma) (see *Tarduno et al.* [2003] for a summary). We also obtained picrite recovered from the shield-building phase of Suiko Seamount at Site 433 (65 Ma [*Dalrymple et al.*, 1980]) by Leg 55 of the Deep Sea Drilling Project. Combined with alkalic and tholeiitic basalt samples from the northwest Hawaiian Islands (including Niihau, Kaula, Nihoa, and Necker islands, and La Perouse and Gardner pinnacles, as well as
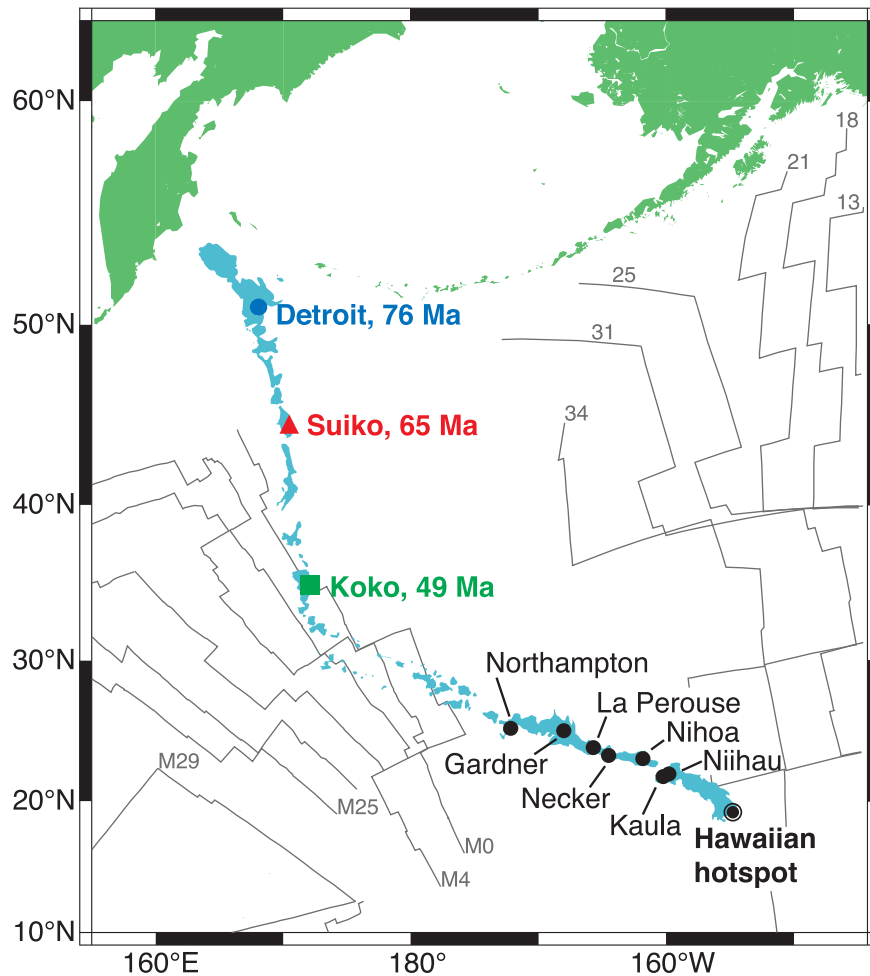
Figure 1. The Hawaiian hot spot track in the North Pacific as shown by the Hawaiian-Emperor chain of islands and seamounts. Names are shown for the islands and seamounts sampled for this study. Ages are shown for the three Emperor seamounts from which picrites have been recovered. Modified from *Shipboard Scientific Party* [2002]. Suiko Seamount age is from *Dalrymple et al.* [1980]; Detroit and Koko ages are from *Duncan and Keller* [2004].

Northampton Bank), and previously published data from the main Hawaiian Islands (compiled by *Farley and Neroda* [1998]), a sufficient number of olivine-phyric lavas is now available to allow a first-order determination of the ^{3}He/^{4}He history of the Hawaiian hot spot since the Cretaceous. We recognize that a few samples from a few seamounts does not fully characterize the nature of volcanism at the Hawaiian hot spot, but until a full suite of samples is available from several of the Emperor Seamounts, the best comparison we can make is using similar rock types from similar stages in the volcanic development of the seamounts. We therefore concentrated on tholeiitic picrites from late in the shield-building stage of the Emperor Seamounts.

[6] The Emperor samples are picrites containing 20–30% olivine, and whole rock analyses gave 24–25 wt.% MgO for the Detroit samples [*Shipboard Scientific Party*, 2002] and 22–26 wt.% MgO for the Suiko samples [*Shipboard Scientific Party*, 1980]. The Koko core intervals analyzed for He have not been analyzed for major elements, but nearby core intervals contain 16.5–18 wt.% MgO (P. Thompson, personal communication, 2003), which suggests that these lavas may not be true picrites (i.e., >18 wt.% MgO), although we refer to them as picrites here for convenience. During hand-picking of olivine for the He isotope analyses, we found that the Detroit samples contained two distinct subpopulations of olivine distinguished by their light and dark color. We therefore separated each Detroit picrite into two aliquots for He isotopic analysis (light olivine versus dark olivine in Table 1). Representative olivine crystals from all three of the Emperor seamounts were analyzed for major element compositions by elec-

Table 1. Helium Analyses of Olivines From Emperor Seamount Picrites[a]

Location	Age, Ma	Depth, mbsf	Sample	Phase	Fo of Olivine	Crushed			Melted Powder		
						^{3}He/^{4}He	2σ	[He], ccSTP/g	^{3}He/^{4}He	2σ	[He], ccSTP/g
Detroit Seamount (50°57.00'N, 167°44.40'E)	76	591.9	197-1203A-32R-3,100-105cm	lt olivine	89–90	10.35	0.42	3.03×10^{-9}	2.10	0.08	1.15×10^{-8}
				dk olivine	80–82	10.68	0.55	3.00×10^{-9}	0.49	0.07	1.27×10^{-8}
		593.6	197-1203A-32R-4,50-65cm	lt olivine	86–90	11.08	0.24	1.91×10^{-8}	1.38	0.07	1.44×10^{-8}
				dk olivine	78–87	(7.30)	0.61	2.18×10^{-9}	0.29	0.04	1.90×10^{-8}
		637.5	197-1203A-38R-1,43-48cm	lt olivine	86–90	10.91	0.27	8.68×10^{-9}	1.64	0.05	2.29×10^{-8}
				dk olivine	84–86	13.78	0.25	4.07×10^{-8}	7.91	0.16	3.37×10^{-8}
Suiko Seamount (44°46.63'N, 170°1.23'E)	65	315.8	55-433C-24-7,138-145cm	olivine	85–87	23.52	0.41	5.78×10^{-8}	5.54	0.15	1.05×10^{-8}
		338.9	55-433C-27-2,138-144cm	olivine	85–89	16.70	0.45	8.88×10^{-9}	2.28	0.12	1.45×10^{-8}
Koko Seamount (34°55.55'N, 172°8.75'E)	49	104.5	197-1206A-10R-1,13-16cm	olivine	82–87	24.13	0.54	1.21×10^{-8}	0.38	0.05	1.31×10^{-8}
		306.6	197-1206A-40R-1,10-17cm	olivine	87–89	19.85	0.39	4.22×10^{-8}	3.10	0.12	2.10×10^{-8}

[a] The ^{3}He/^{4}He values are reported relative to the atmospheric ratio of 1.39×10^{-6}. Analytical uncertainties are the 2-sigma quadrature sum of uncertainties in analyses of the sample, standards, and blanks. Detroit and Koko ages are from Duncan and Keller [2004]; Suiko age is from Dalrymple et al. [1980]; and other ages are from numerous sources compiled by Clague and Dalrymple [1987].

tron microprobe at Oregon State University, and their forsterite contents are included in Table 1. Olivine from Koko Seamount ranged between Fo_{82-89}, and that from Suiko Seamount between Fo_{85-89}. Olivine from Detroit Seamount formed two groups with virtually no overlap, the light olivine being Fo_{86-90} and the dark olivine being Fo_{78-87}.

2. Analytical Procedures

[7] Olivine was separated from basaltic groundmass by crushing and sieving to 0.85–1.7 mm size fraction, and hand-picking under a binocular microscope for fresh appearance and lack of attached groundmass. Olivine separates were then ultrasonically cleaned in deionized water, rinsed in acetone and air-dried before hand-picking again for the freshest grains. The ODP and DSDP samples were analyzed using procedures outlined by Graham et al. [1998]. To ensure that we were looking at the magmatic component of the helium, gas extractions were first performed by crushing the olivine in vacuum and analyzing the released helium. The crushed powders were then retrieved and melted in a high-temperature vacuum furnace and the helium released during melting was also analyzed. During the period in which these analyses were performed, line blanks were run before and after all samples. The crushing blanks were $<1 \times 10^{-10}$ ccSTP for ^{4}He, and the furnace blanks at 1800°C were $<5 \times 10^{-10}$ ccSTP ^{4}He. Samples from the $\leq$27 Ma section of the Hawaiian-Emperor chain were analyzed by in vacuo crushing at Scripps Institution of Oceanography in 1987 (A. R. Basu and H. Craig, manuscript in preparation, 2004). Analytical procedures used for those samples are as described by Farley et al. [1992, 1993].

[8] The posteruptive addition of ^{4}He by radioactive decay of U and Th, and its potential for biasing the measured ^{3}He/^{4}He to lower values, is a concern for such old samples. The initial ^{3}He/^{4}He ratio (i.e., the magmatic value at the time of eruption) can, in principle, be distinguished by comparing the crushing and melting results for the ODP and DSDP samples. Most of the melting experiments released an amount of ^{4}He that was similar to or greater than the amount released by crushing of the same sample (Table 1). More importantly, melting always released gas with a lower ^{3}He/^{4}He ratio, from about 1%–50% of the crush ^{3}He/^{4}He ratio (Figure 2a). If a significant amount of posteruptive radiogenic ^{4}He had been transferred into the crushable fraction, then samples with lower [He] during crushing would

display lower ^{3}He/^{4}He. Our results demonstrate that significant amounts of radiogenic He, while present in the crystal lattice, were not released until the crushed powder was melted. The fact that the three Fo_{86-90} olivine samples from Detroit gave very similar ^{3}He/^{4}He values despite nearly an order of magnitude difference in [He] also bears this out. The crushed ODP and DSDP analyses in Table 1 therefore accurately represent original magmatic values at the time of eruption, and have not been significantly modified by ^{4}He ingrowth. The only exception appears to be the lowest ^{3}He/^{4}He value (7.3 R_A) obtained during crushing of the dark olivine from Detroit sample 1203A-38R-1. In this case, almost 90% of the ^{4}He sample budget (crushed + melted) was present as radiogenic helium in the melted fraction (Figure 2b). This very high proportion of radiogenic ^{4}He, coupled with the relatively low ^{3}He/^{4}He during crushing, suggests that a significant fraction of radiogenic ^{4}He within the crystal lattice or dissolved in melt inclusions may have also been released during the crushing. While we note that the measured value of 7.3 R_A for this crushing analysis is within the range of modern MORB, we do not assign petrogenetic significance to this value given

the melting results, and we do not include this sample further in our discussion.

3. Results

[9] Olivines from 76 Ma Detroit Seamount picrites have ^{3}He/^{4}He ratios of 10.4–13.8 R_A, which is at the low end of the observed range for young Hawaiian lavas (7–35 R_A), but higher than the modern MORB range (6–10 R_A; Figure 3). Exactly how these Detroit ^{3}He/^{4}He values compare to those of the Cretaceous MORB upper mantle is unknown, but on the basis of detailed modeling of coupled He-N-Ne-Ar isotopes [*Tolstikhin and Marty*, 1998], the ^{3}He/^{4}He of the upper mantle does not appear to have decreased significantly over the past 500 m.y., suggesting that values >10 R_A are not appropriate for Cretaceous MORB mantle. ^{3}He/^{4}He ratios of 16.7–23.5 R_A for picritic lavas from 65 Ma Suiko Seamount, and 19.8–24.1 R_A for 49 Ma
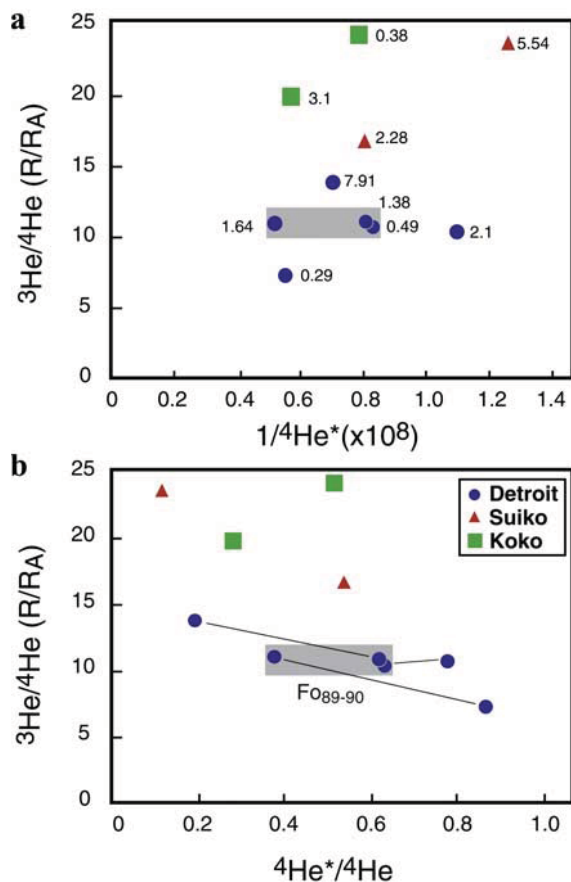

Figure 2. (a) Reciprocal of radiogenic ^{4}He concentration versus ^{3}He/^{4}He (relative to the atmospheric ratio R_A) for Emperor seamount samples. This diagram shows that the ^{3}He/^{4}He upon melting the crushed powder was always lower then the crushed ^{3}He/^{4}He ratio. This indicates that significant radiogenic ^{4}He was released in the melted fraction but not in the crushed fraction. Data points are plotted at the crushed ^{3}He/^{4}He ratio and are labeled with the melting ^{3}He/^{4}He ratio. The crushing ^{3}He/^{4}He values thus appear to closely represent the original ^{3}He/^{4}He of the magmas (with one exception; see below). (b) The proportion of radiogenic helium, ^{4}He*/^{4}He (where ^{4}He* is the amount of radiogenic ^{4}He in a sample and ^{4}He is the total crushed+melted ^{4}He), versus ^{3}He/^{4}He (relative to the atmospheric ratio R_A) for Emperor seamount samples. This diagram shows that Detroit samples consistently have the lowest ^{3}He/^{4}He despite a wide range of ^{4}He*/^{4}He. ^{4}He*, the amount of radiogenic helium, is given by ^{4}He* = ^{4}He$_{melt}$ (1 − R_{crush}/R_{melt}), where R is ^{3}He/^{4}He [*Graham et al.*, 1987]. Tie lines connect olivine separates that came from the same rock sample but were split into light (Fo_{86-90}) and dark (Fo_{78-87}) olivine fractions prior to isotopic analyses. All three of the Detroit samples contained these two populations of olivine, but there were no systematic isotopic differences between the light and the dark olivine. The sample with the lowest ^{3}He/^{4}He (7.3 R_A) contains the highest proportion of radiogenic He. We suspect that that sample has been disturbed by posteruptive alteration and/or ingrowth of radiogenic ^{4}He and do not use it in our plots or our interpretations. The four Detroit samples in the 10.3–11.1 R_A range have a range of He concentrations, which is especially compelling evidence that these are reliable magmatic ratios.
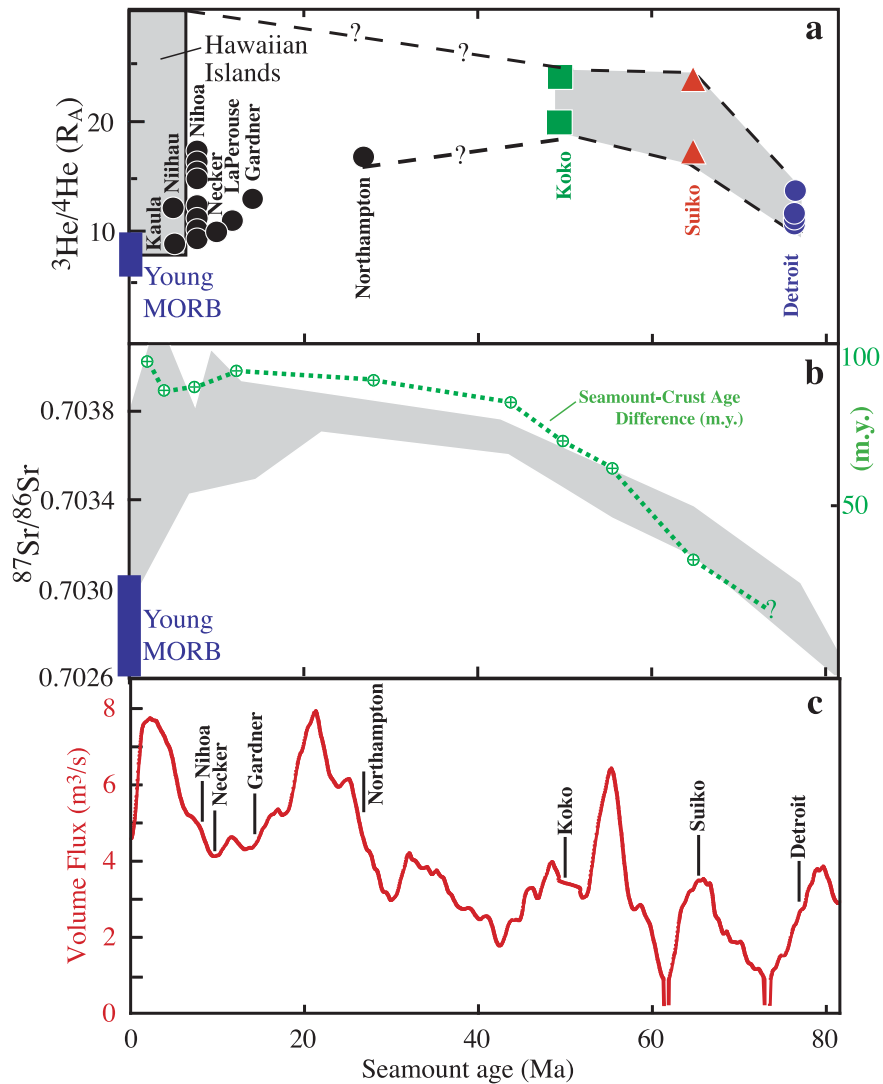
Figure 3. Temporal trends in $^{87}Sr/^{86}Sr$ and $^{3}He/^{4}He$ (relative to atmospheric ratio R_A) for the Hawaiian-Emperor chain, compared to plume volume flux. Age differences between the seamounts and their underlying crust are shown by the dotted green line in Figure 3b. The $^{87}Sr/^{86}Sr$ of tholeiitic basalts (shaded field in Figure 3b) from Detroit Seamount are indistinguishable from MORB but by about 45 Ma $^{87}Sr/^{86}Sr$ transitioned to values equivalent to modern Hawaii. In contrast, all $^{3}He/^{4}He$ values are distinct from MORB, even at 76 Ma, when the $^{87}Sr/^{86}Sr$ was MORB-like. Modeling suggests that Cretaceous MORB mantle also had $^{3}He/^{4}He$ values <10 R_A [*Tolstikhin and Marty*, 1998]. Hawaiian-Emperor $^{87}Sr/^{86}Sr$ data are from *Lanphere et al.* [1980], *Basu and Faggart* [1996], *Keller et al.* [2000], *Regelous et al.* [2003], and *Huang et al.* [2004], plus the Hawaiian Islands data compilation by *Farley and Neroda* [1998]. Helium isotope data points from the <27 Ma part of the Hawaiian Ridge are unpublished data of A. R. Basu and H. Craig (A. R. Basu, personal communication, 2003). Calculated plume crustal volume flux based on bathymetry and residual gravity anomalies is from *Van Ark and Lin* [2004].

Koko Seamount (Table 1) are much higher than MORB $^{3}He/^{4}He$ values (Figure 3), but in the middle part of the range of Hawaiian Island values. Lavas from the 27 to 4 Ma part of the Hawaiian-Emperor chain (including the northwest Hawaiian Islands) have $^{3}He/^{4}He$ from 8.6–17.5 R_A, which overlaps with the lower half of the main Hawaiian Islands range. These northwest Hawaiian Islands values are included in Figure 3 for

comparison, but will be published elsewhere (A. R. Basu and H. Craig, manuscript in preparation, 2004). The lower limit of $^{3}He/^{4}He$ for the northwest Hawaiian Island lavas appears to decrease with decreasing age, with only Kaula and Nihoa samples extending below 10 R_A and approaching the low $^{3}He/^{4}He$ values that characterize the end stage of shield building volcanism on Hawaii [*Kurz et al.*, 1983, 1996; *Kurz and*
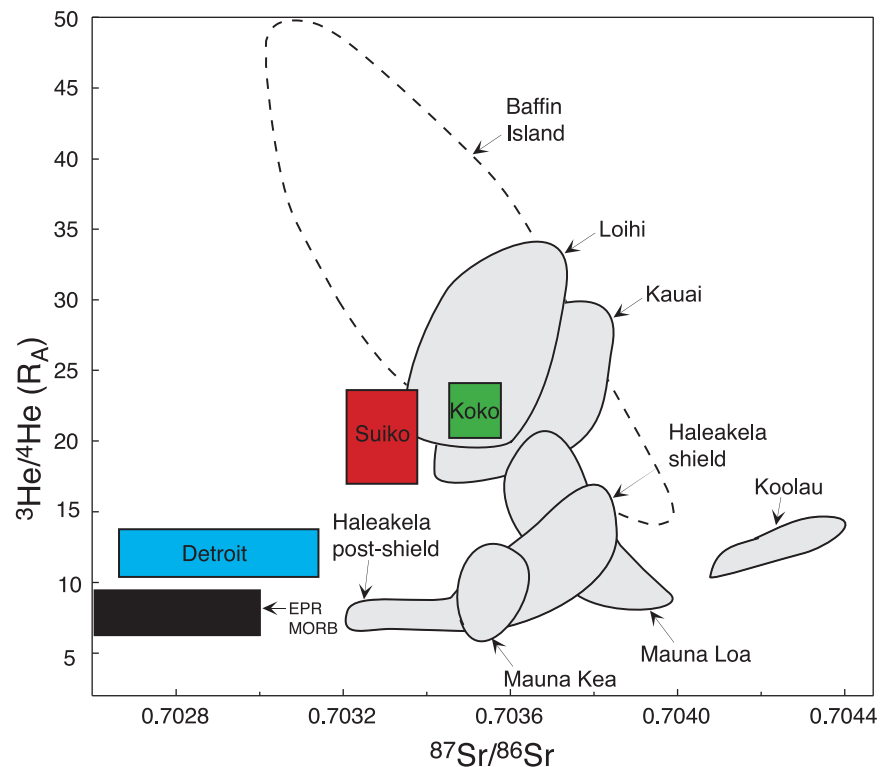
Figure 4. The $^{87}Sr/^{86}Sr$ versus the $^{3}He/^{4}He$ of samples from the Hawaiian Islands compared to data from Detroit, Suiko, and Koko seamounts. The field for Baffin Island picritic basalts (the proto-Iceland plume) is shown for comparison [from *Stuart et al.*, 2003]. Hawaiian Islands data fields are modified from *Mukhopadhyay et al.* [2003]. Emperor Seamounts data are plotted as boxes using Sr data from *Keller et al.* [2000] and *Regelous et al.* [2003] because the same samples have not been analyzed for Sr and He, although the high-$^{87}Sr/^{86}Sr$ end of the Detroit field includes data from *Huang et al.* [2004] for ODP Site 1203 samples very close to the picrites we analyzed for He.

Kammer, 1991]. The lack of very high $^{3}He/^{4}He$ (>17.5 R_A) in the northwest Hawaiian Islands samples is not surprising given that even on the well-studied main Hawaiian Islands, $^{3}He/^{4}He$ >20 R_A are rare.

4. Discussion

[10] In contrast to the MORB-like La/Yb and $^{87}Sr/^{86}Sr$ values previously found at Detroit Seamount [*Keller et al.*, 2000, *Regelous et al.*, 2003], there is a clear plume signal in the elevated $^{3}He/^{4}He$ ratios at Detroit (although Detroit $^{3}He/^{4}He$ values are subdued compared to the highest Hawaiian Island values; Figure 3). As with $^{87}Sr/^{86}Sr$ [*Lanphere et al.*, 1980; *Keller et al.*, 2000; *Regelous et al.*, 2003], the $^{3}He/^{4}He$ signal increased with time along the Emperor Seamounts, although unlike for $^{87}Sr/^{86}Sr$, almost all of the increase in $^{3}He/^{4}He$ occurred between 76 Ma and 65 Ma as the hot spot and a seafloor spreading center diverged. While it is possible that the intrinsic geochemical and isotopic char-

acteristics of the plume coincidentally changed as a spreading ridge passed over it, such a model is untestable and therefore unsatisfactory. Besides, several other hot spots also show less-radiogenic $^{87}Sr/^{86}Sr$ values early in their history (Kerguelen: *Class et al.* [1993]; Reunion: *White et al.* [1990]; Iceland: *Stuart et al.* [2003]). Iceland is the only other hot spot where the amount of He isotope data is sufficient to reveal trends in its long-term history. In contrast to the Emperors, the highest $^{3}He/^{4}He$ values for the Iceland hot spot occur very early and are accompanied by low $^{87}Sr/^{86}Sr$ [*Stuart et al.*, 2003] (Figure 4). However, the high-$^{3}He/^{4}He$ samples from the Iceland hot spot are associated with the initiation of the Iceland plume (West Greenland: *Graham et al.* [1998]; Baffin Island: *Stuart et al.* [2003]). The time of initiation of the Hawaiian plume is unknown because our volcanic record from the Hawaiian hot spot only goes as far back as the oldest remaining Emperor Seamounts. If there were Emperor Seamounts older than the ones we see today, or any evidence for the initiation of the Hawaiian plume,

those features have apparently been subducted into the Kamchatka-Aleutian Trench.

[11] The obvious explanation for the MORB-like La/Yb, ^{87}Sr/^{86}Sr, and ^{3}He/^{4}He of Detroit Seamount is the proximity of the hot spot to a seafloor spreading center at the time Detroit formed in the Late Cretaceous central Pacific basin [*Keller et al.*, 2000; *Regelous et al.*, 2003]. The exact mechanism may relate to the increased decompressional melting within the plume as it was able to ascend to shallower depths beneath very young and thin lithosphere (<10 m.y. old at the time Detroit Seamount formed [*Caplan-Auerbach et al.*, 2000]). The greater extent of melting would have exhausted the fertile, more readily fusible parts of the plume first, and then began melting the depleted (lower La/Yb, ^{87}Sr/^{86}Sr, and ^{3}He/^{4}He), normally refractory portions of the ascending plume. These normally refractory portions may have been intrinsic to the plume [*Regelous et al.*, 2003] or they may have been derived from depleted material entrained from the upper mantle [*Keller et al.*, 2000]. The 17–24 R_A and 20–24 R_A values for ^{3}He/^{4}He at Suiko and Koko seamounts, respectively, also fit with this thin-lithospheric lid model, as the slightly lower ^{3}He/^{4}He for Suiko compared to Koko is consistent with the fact that Suiko was erupted onto 35 m.y. old crust, whereas Koko erupted onto 72 m.y. old crust (Figure 3b). Alternatively, these minor differences in ^{3}He/^{4}He could be an artifact of our very limited sampling for the Emperor Seamounts.

[12] While the helium isotope systematics of the Hawaiian hot spot were clearly affected by some process related to the proximity of the hot spot to a spreading center, this tectonic setting had an even stronger influence on other geochemical characteristics of the hot spot. For example, the REE and ^{87}Sr/^{86}Sr of Detroit Seamount are nearly indistinguishable from MORB, and recovered more gradually than the ^{3}He/^{4}He as the spreading center and the hot spot diverged (Figure 3b). The cause of this difference in behavior is unclear, but on the basis of a comparison of Figures 3a and 3b, ^{3}He/^{4}He appears nearly immune to the affects of thin lithosphere once that lithosphere is older than about 10 million years. In contrast, the ^{87}Sr/^{86}Sr field in Figure 3b is steepest at the old end of the Emperor Seamounts, but then slowly levels off at recent Hawaiian Islands values. The suggestion that He can be a more sensitive plume tracer than Sr is not surprising given results from the Mid-Atlantic Ridge where it interacts with the

Iceland plume. South of Iceland, the high ^{3}He/^{4}He anomaly on the Mid-Atlantic Ridge extends much farther south than the Sr isotope anomaly [*Schilling*, 1973; *Poreda et al.*, 1986; *Taylor et al.*, 1997; *Schilling et al.*, 1998; *Hilton et al.*, 2000]. Thus, in places on the northern MAR there is evidence for a plume-derived He isotope signal in the absence of a plume-derived Sr isotope signal (similar to what is observed at Detroit Seamount). Alternatively, the depleted upper mantle component entrained by the plume beneath Detroit Seamount may have been recently degassed during melting beneath the nearby ridge. In this case, the entrained upper mantle component, given its low [He], would have little affect on the overall He isotopes of the plume. Such ridge-related melting would deplete Sr as well, but to a lesser degree because of the greater compatibility of Sr during melting (higher solid-melt partition coefficient) as compared to He.

[13] Differences between the Emperor samples and the main Hawaiian Islands are most dramatic on a ^{87}Sr/^{86}Sr versus ^{3}He/^{4}He plot (Figure 4, modified from *Mukhopadhyay et al.* [2003]). Taken together, the Hawaiian Islands shield-stage lavas (Kauai, Mauna Loa, and Mauna Kea shield) form a negatively sloping array. In contrast, our picritic lavas from the Emperor Seamounts form a positively sloping array directed toward the MORB field. Thus both the Hawaiian and Emperor samples appear to share a high-^{3}He/^{4}He-moderate-^{87}Sr/^{86}Sr component similar to Loihi. The other dominant component in the Hawaiian samples is low-^{3}He/^{4}He-high-^{87}Sr/^{86}Sr, whereas the other dominant component in our Emperor samples is low-^{3}He/^{4}He-low-^{87}Sr/^{86}Sr. Although the postshield Hawaiian samples show evidence for a low-^{3}He/^{4}He-low-^{87}Sr/^{86}Sr component similar to that in our Emperor samples, the high-^{3}He/^{4}He component is largely subdued in the postshield samples. The data field for Baffin Island picritic basalts from the proto-Iceland plume [*Stuart et al.*, 2003] has a negative slope, and the inferred mixing components are more akin to the Hawaiian Islands data array than to the Emperor Seamounts array, despite the fact that the Emperor samples, like the Baffin Island samples, are from early in the history of their plume (although for the Emperor samples we do not know how early). We therefore conclude that the life-stage of the plume does not appear to be the controlling factor for the Emperor data, but rather the melting regime permitted by thin, young lithosphere.

[14] Plume buoyancy flux is an additional variable that could have influenced the ^{3}He/^{4}He signal of the Hawaiian hot spot over time. A positive correlation between plume flux and ^{3}He/^{4}He might be expected because the plume carries helium with high ^{3}He/^{4}He from relatively undegassed deep mantle. The long-term crustal volume flux of the Hawaiian plume, on the basis of bathymetry and residual mantle Bouguer anomalies (Figure 3c) [*Van Ark and Lin*, 2004], shows a general increase over time. These plume flux calculations are similar to calculations based on bathymetry alone [*Vidal and Bonneville*, 2004], but also incorporate crustal thicknesses constrained by gravity data. There does not appear to be a simple correlation between ^{3}He/^{4}He and plume flux, except there may be a relationship between times of lower ^{3}He/^{4}He and decreasing plume flux. Although our rock sampling is obviously limited, the only two time periods when we have no ^{3}He/^{4}He values higher than 14 R_A are times of decreasing plume flux (76 Ma at Detroit and 12–10 Ma at Gardner-to-Necker; Figure 3). Shortly after Necker formed at 10 Ma, the plume flux began to increase, and Nihoa and younger locations all have ^{3}He/^{4}He > 17 R_A. Northampton Bank, and Koko and Suiko Seamounts all formed at times when the plume flux was relatively stable or increasing, and these times all include samples with ^{3}He/^{4}He $\geq$ 17 R_A. An obvious caveat of these observations is that all of our samples are from times of moderate to high plume flux, at least in part because times of low flux are much less likely to have produced features conducive to sampling.

[15] Our study shows that it is possible to determine magmatic ^{3}He/^{4}He values from submarine volcanic rocks as old as 76 Ma. This demonstrates the potential to study the long-term helium isotopic behavior of hot spots, and perhaps mid-ocean ridges. In the case of the Hawaiian hot spot, elevated ^{3}He/^{4}He values can be found at least as far back as 76 Ma, and support the hypothesis that the Hawaiian hot spot originates as a deeply rooted mantle plume that taps an undegassed source of primordial helium. Of the many geochemical indicators associated with the Hawaiian mantle plume, helium appears to be one of the most sensitive, as it was the least affected by depletion processes associated with proximity of the plume to a seafloor spreading center during the Late Cretaceous.

Acknowledgments

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