The link between Hawaiian mantle plume composition, magmatic flux, and deep mantle geodynamics

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Abstract

Oceanic island basalts sample mantle reservoirs that are isotopically and compositionally heterogeneous. The Hawaiian-Emperor chain represents ~85 Myr of volcanism supplied by a deep mantle plume. Two geographically and geochemically delineated trends, Kea and Loa, are well documented within the Hawaiian Islands. Enriched Loa compositions originate from subduction recycled or primordial material stored in deep mantle reservoirs such as the large low shear velocity province (LLSVP) below Hawai‘i. Loa compositions have not been observed along the Emperor Seamounts (>50 Ma), whereas lavas on the Hawaiian Islands (<6.5 Ma) sample both Kea and Loa sources. Lead isotopes in shield lavas along the Northwest Hawaiian Ridge (NWHR) spanning ~42 Myr between the bend in the chain and the Hawaiian Islands record the geochemical evolution of the Hawaiian mantle plume over a time period when many geophysical parameters (volcanic propagation rate, magmatic flux, mantle potential temperature) increased significantly. Along the NWHR, the Loa geochemical component appears ephemerally, which we link to the sampling of different lower mantle compositional domains by the Hawaiian mantle

plume. The plume initially sampled only the deep Pacific mantle (Kea component) from outside the LLSVP during the formation of the Emperor Seamounts. Southward migration and anchoring of the plume on the LLSVP led to entrainment of increasing amounts of LLSVP material (Loa component) along the NWHR as documented by an increase in $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ with decreasing age. The correlation between $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ and magmatic flux suggests source composition affects the magmatic flux, and explains why the Hawaiian mantle plume has dramatically strengthened through time.

1. Introduction

Long-lived intraplate oceanic islands and chains such as Iceland, Kerguelen, and Hawai‘i are typically explained as the products of mantle plumes (Morgan, 1972; Boschi et al., 2007). Hawai‘i is one of the most studied examples of such volcanism because it has a well-documented age progression along the chain (O’Connor et al., 2013; Garcia et al., 2015), is far from continent and mid-ocean ridge sources of contamination, has a deep mantle source (French and Romanowicz, 2015), and, on the main islands, exhibits two geographical trends, Kea and Loa, with distinct geochemical signatures (Abouchami et al., 2005; Tanaka et al., 2008; Weis et al., 2011). Hawai‘i, the archetypal mantle plume, is anomalous in many of its dynamic and geochemical features. For example, intraplate lavas worldwide are dominantly alkalic in composition, reflecting lower degrees of partial melting, in comparison to Hawai‘i where ~98% of eruptive products are of tholeiitic composition (Garcia et al., 2015). In addition, mantle plumes typically exhibit higher magmatic production at their initial arrival at the base of the lithosphere that dramatically diminishes with time, i.e. from melting the plume head to the plume tail (White, 1993; Jellinek and Manga, 2004; Kumagai et al., 2008). The volume flux of Hawaiian eruptive products, conversely, has increased ~650% along the NWHR and an additional ~375% during the

formation of the Hawaiian Islands to an all-time high at the current locus of volcanism centered on Kilauea and Lō‘ihi volcanoes (Wessel, 2016). At the same time, mantle potential temperature has increased (Tree, 2016), along with the volcano propagation rate (i.e. the frequency that a new volcano is created as the Pacific Plate moves over the Hawaiian mantle plume; O’Connor et al., 2013). Thus, on the basis of most major physical parameters, the Hawaiian mantle plume has strengthened, an unexplained observation that is a very unusual feature among mantle plumes worldwide (Tree, 2016). No modern geochemical study has addressed this issue or determined the isotopic variation along the NWHR, to evaluate whether source changes have played a role.

The source of the Loa component in Hawaiian lavas is more enriched (higher Th/U, $^{208}\text{Pb}^*/^{206}\text{Pb}^*$, and Sr isotopic ratios; lower Nd and Hf isotopic ratios) than that of the Kea component. This signature is predominately explained by the presence of subduction recycled material (Weis et al., 2011) or primordial material in the deep source (Li et al., 2015). Lead isotopes provide the most robust tool for delineating the Loa and Kea components in Hawaiian basalts: at a given $^{206}\text{Pb}/^{204}\text{Pb}$, Loa-trend volcanoes will have a higher $^{208}\text{Pb}/^{204}\text{Pb}$ than Kea trend volcanoes (Abouchami et al., 2005; Weis et al., 2011). Emperor Seamount lavas (85-51 Ma) are Kea-like in geochemical affinity, whereas both Kea and Loa compositions are present in lavas from the Hawaiian Islands, and furthermore are clearly distributed along two geographical trends of volcanoes (Fig. 1; Keller et al., 2000; Regelous et al., 2003; Abouchami et al., 2005; Tanaka et al., 2008; Weis et al., 2011). On the basis of the only three Pb isotope analyses previously available for the NWHR, Loa geochemical compositions have been observed at only one seamount, Daikakuji, near the bend in the Hawaiian-Emperor chain (Regelous et al., 2003). It was unknown whether this is an isolated case or represents the first arrival of the Loa geochemical component (Garcia et al., 2015). Furthermore, because the Loa composition typically is

associated with a more fusible source component, its presence may be linked to the dramatic increase in volcano volume (lowest at Daikakuji with 30 km³ to highest at Gardner with 540 km³; Bargar and Jackson, 1974), magmatic flux (0.4 m³/s at Daikakuji to a maximum of 4 m³/s at Gardner; Wessel, 2016), and volcanic propagation rate (Detroit to Midway 57 ± 2 km/Myr, from which increases to 80-100 km/Myr afterwards; O'Connor et al., 2013) observed along the ~2800 km long ridge (Pertermann and Hirschmann, 2003; Garcia et al., 2015).

Lead isotopic compositions were measured on 22 shield-stage samples from 13 NWHR volcanoes spanning from ~47 Ma at the bend to ~7 Ma at Nīhoa (Fig. 1; Dalrymple et al., 1974; O'Connor et al., 2013). There were no Pb isotopic measurements for NWHR volcanoes younger than Daikakuji Seamount; this study fills this critical 40 million year gap. The Pb isotopic evolution of the entire Hawaiian-Emperor Seamount chain was examined to identify when the enriched Loa component appears and to assess its involvement in magmatic flux variations. Finally, we discuss the implications of these results for the Hawaiian deep mantle source and the potential contribution of material from the Pacific LLSVP to account for flux variations.

2. Geological setting and sampling

Fifty-one volcanoes erupted over ~42 million years between the bend in the Hawaiian-Emperor chain and the Hawaiian Islands constitute the Northwest Hawaiian Ridge (NWHR; Garcia et al., 2015). We focus on shield stage tholeiitic lavas because they are most likely to record the plume source composition variation (i.e. Loa and Kea geochemical variation), whereas the later post-shield and rejuvenated Hawaiian lavas present a uniform depleted isotopic composition regardless of the geochemical affinity of shield stage lavas at the same volcano (Frey et al., 2005; Hanano et al., 2010). Study of the Hawaiian Islands show that single islands are composed of several volcanic centers which may erupt different geochemical signatures from as

close as ~50 km apart (e.g. Mauna Loa and Mauna Kea on Hawai‘i; Abouchami et al., 2005). One to four samples from selected volcanoes were available for this large-scale study, which consequently provides average isotopic trends along a previously unstudied section of the Hawaiian-Emperor chain.

Sampling of the NWHR is currently highly restricted by the Papahānaumokuākea Marine National Monument, a recently expanded ~1,508,870 km² U.S. Marine Conservation Area and UNESCO World Heritage Site that stretches from Middle Bank to Hancock Seamount (~80% of the NWHR). Because of this access impediment, samples in this study originate from pre-2006 dredge or island sampling expeditions stored at the University of Hawai‘i and Scripps Institution of Oceanography rock collections or from samples collected by the University of Hawai‘i’s human occupied vehicle (HOV) submersible *Pisces V* reconnaissance survey (2003 and 2007) of the new monument. Supplementary Table 1 summarizes sample locations.

Samples from near the bend of the Hawaiian-Emperor chain (Yuryaku and Daikakuji) were dredged by the research vessel *Thomas Washington* in August of 1971 during the Scripps Institution of Oceanography expedition Aries VII. This expedition dredged Koko, Kimmei, Yuryaku and Daikakuji Seamounts. Published data from this cruise includes ages, some major and trace element data, and isotopic compositions (Davies et al., 1971; Clague et al., 1973; Clague, 1974; Clague et al., 1975; Dalrymple and Clague, 1976; Clague and Dalrymple, 1989; Lanphere et al., 1980; Regelous et al., 2003; Sharp and Clague, 2006).

Multiple expeditions by the Hawai‘i Institute of Geophysics’ research vessel *Ka ‘imikai-O-Kanaloa* dredge sampled the NWHR from 1972-1984 (identified by the prefix KK- on the sample name). Some of these samples have published XRF major and trace elemental analyses (Garcia et al., 1987, 2015). Recently in 2003 and 2007 the *Ka ‘imikai-O-Kanaloa* has been used for several

NWHR sampling expeditions to Pioneer, Laysan, and Twin Banks by the Hawai‘i Undersea Research Laboratory (HURL). These samples are identified by the prefix P5- and were collected using the submersible *Pisces V*.

Two sample cores were drilled at Midway Island, one on Sand Island (Sand Island hole) and one in the lagoon of Midway Atoll (reef hole) in 1965 by the Hawai‘i Institute of Geophysics at the University of Hawai‘i in partnership with the U.S. Geological Survey and the U.S. Office of Naval Research. Samples analyzed in this study are from the reef hole, which drilled 1261 feet of limestone, dolomite, marl, and volcanoclastic sedimentary rock before penetrating approximately 387 feet of tholeiitic basalt (Ladd et al., 1967, 1970; Macdonald, 1969).

Finally, subaerial samples from Mokumananana (formerly known as Necker Island) and Nīhoa Island were collected by Harmon Craig during the 1985 ALCYONE expedition of Scripps Institution of Oceanography. This same cruise also dredged Nīhoa, recovering manganese crust, volcanoclastic pebblestone, and volcanic rock. Strontium and neodymium isotopes on unleached powders from some of these Nīhoa and Mokumananana samples were previously reported (Clague, 1973; Basu and Faggart, 1996). Some of these samples were reanalyzed in this study.

3. Analytical techniques and age correction

Lead isotopic compositions of 22 Northwest Hawaiian Ridge basalts were analyzed at the Pacific Centre for Isotopic and Geochemical Research (PCIGR) at the University of British Columbia. In addition to isotopic analyses, major elements were analyzed at the University of Massachusetts (Rhodes and Vollinger, 2004) and this data is reported in Garcia et al. (2015).

For isotopic analyses, whole rock powders were sequentially leached in HCl 6N in steps of 20 minutes of acid-leaching in an ultrasonic bath followed by immediate decantation before fine particles have time to settle. This leaching step was repeated until the supernatant was clear,

typically between 6 and 14 times. Step-wise acid leaching is a necessary procedure to remove possible post-eruption alteration and/or contamination (Weis et al., 2005, 2006; Nobre Silva et al., 2009, 2010). After leaching, samples were digested using HF and HNO₃ and purified on chromatographic ion exchange columns following the procedures of Fourny et al. (2016). Lead isotopic ratios were determined on a Nu Plasma (Nu Instruments) MC-ICP-MS NP II 214 or Nu 1700 that were cross-calibrated by multiple analysis of standards NIST SRM 981 and reference materials of comparable composition (Kil-93 and BHVO-2). A Tl spike was added to monitor internal mass fractionation (White et al., 2000) and all analyses were corrected on-line to a ²⁰⁵Tl/²⁰³Tl=2.3885 (Weis et al., 2005). Analyses were then normalized off-line to NIST SRM 981 triple spike values of Galer and Abouchami (1998) using the sample-standard bracketing method. For the duration of this study the measured ratios for the NIST SRM 981 were: ²⁰⁸Pb/²⁰⁴Pb=36.7188 ± 0.0107, ²⁰⁷Pb/²⁰⁴Pb=15.4983 ± 0.0041, and ²⁰⁶Pb/²⁰⁴Pb=16.9418 ± 0.0028 (2SD, n=102). On a daily basis, the standard errors on the analyses of the NIST SRM 981 are smaller, in the range of ~100 ppm. Mercury 204 was monitored for possible interference on mass ²⁰⁴Pb by analysis of the signal on the 202 mass. Total procedural blanks contained 3-88 picograms Pb, negligible compared to the amount of Pb present in samples (on average ~100-300 nanograms Pb).

Repeated analyses of U.S Geological Survey (USGS) reference material BHVO-2, a Hawaiian basalt, resulted in average Pb isotopic compositions of ²⁰⁸Pb/²⁰⁴Pb=38.2091 ± 0.0107, ²⁰⁷Pb/²⁰⁴Pb=15.4893 ± 0.0023, and ²⁰⁶Pb/²⁰⁴Pb=18.6482 ± 0.0001 (2SD, n=3), comparable to recent published values (Weis et al., 2005; Fourny et al., 2016). In-house PCIGR reference material Kil-93, a Hawaiian basalt from the 1993 Kīlauea eruption, resulted in average Pb isotopic compositions of ²⁰⁸Pb/²⁰⁴Pb=38.0679 ± 0.0038, ²⁰⁷Pb/²⁰⁴Pb=15.4741 ± 0.0017, and

$^{206}\text{Pb}/^{204}\text{Pb}=18.4090 \pm 0.0035$ (2SD, $n=4$), also comparable to recent published values (Nobre Silva et al., 2013a). All data compiled from the literature are normalized to NIST SRM 981 standard values used in this study to ensure comparability: 16.9405 for $^{206}\text{Pb}/^{204}\text{Pb}$, 15.4963 for $^{207}\text{Pb}/^{204}\text{Pb}$, and 36.7219 for $^{208}\text{Pb}/^{204}\text{Pb}$ (Weis et al., 2011).

Trace element concentrations (e.g. Pb, Th, U, La, and Yb) of samples were measured at the PCIGR using a Thermo Finnigan Element2 high resolution ICP-MS (Schudel et al., 2015; Fourny et al., 2016). Powdered samples were digested in a mixture of 10:1 HNO_3 :HF with no prior leaching. After digestion, samples were fluxed in 6N HCl to eliminate insoluble fluorides, dried down and diluted 5000 times in a 5% HNO_3 -1% HF-1 ppb In solution, and analyzed on the Element2 using the USGS basaltic standard BCR-2 for external calibration (Schudel et al., 2015; Fourny et al., 2016). BHVO-2 was analyzed with all samples, and resulting values were within two standard deviations of published values for all elements (Schudel et al., 2015; Fourny et al., 2016). Full procedural duplicates and replicate measurements showed excellent agreement, and most elements were analyzed with one relative standard deviation (RSD) of less than 5%.

Radiogenic Pb, a powerful discriminator of Loa and Kea trends, (in this paper, radiogenic Pb refers to $^{208}\text{Pb}^*/^{206}\text{Pb}^*$) is a measure of the time integrated Th/U ratio of the source throughout the entire history of the Earth (Galer and O’Nions, 1985; Tatsumoto et al., 1973; White, 2015). It is calculated for each sample based on the equation below where the initial Pb isotopic values are taken as the measured isotopic compositions of the Canyon Diablo troilite, assumed to be representative of the initial isotopic compositions of the Earth.

$$\frac{^{208}\text{Pb}^*}{^{206}\text{Pb}^*} = \frac{\left(\frac{^{208}\text{Pb}}{^{204}\text{Pb}}\right)_{\text{sample}} - \left(\frac{^{208}\text{Pb}}{^{204}\text{Pb}}\right)_{\text{initial}}}{\left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}}\right)_{\text{sample}} - \left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}}\right)_{\text{initial}}}$$

$$\text{where } \left(\frac{{}^{208}\text{Pb}}{{}^{204}\text{Pb}} \right)_{\text{initial}} = 29.476 \quad \left(\frac{{}^{206}\text{Pb}}{{}^{204}\text{Pb}} \right)_{\text{initial}} = 9.307$$

Identifying differences in Pb isotopic ratios for rocks that are older than a few million years is contingent upon the accurate and precise determination of initial isotopic ratios of Pb at the time of eruption. Samples in this study span an age range from 8.5 to ~47 Ma (O'Connor et al., 2013; Dalrymple et al., 1974, 1981; Garcia et al., 1987, 2010). Age correction of measured isotope ratios is necessary in the older NWHR samples to account for in situ radiogenic decay of parent to daughter isotopes since eruption. This correction is very small for young samples, but for samples older than ~15-20 Ma age corrections of measured isotopic ratios are outside the range of analytical error. The accuracy of this correction is also affected by the fact that both subaerial weathering while the volcano was above sea level and seawater alteration once the volcano subsided below sea level might have altered many of these samples, disturbing the magmatic elemental concentrations and ratios. All NWHR volcanoes older than Midway Island are currently below sea level. Loss-on-ignition of analyzed NWHR rocks ranges from less than 1 to 5 weight percent (Garcia et al., 2015), certainly not as altered as some samples analyzed from the Emperor Seamounts (Regelous et al., 2003), but greater than what is considered fresh for a Hawaiian basalt (LOI <3 weight percent; Garcia et al., 2015).

In the U-Th-Pb decay systems, U and Pb are the most sensitive to secondary alteration and as a result, the elemental concentrations measured in unleached powders are not always representative of the primary magmatic concentrations. The degree to which samples have been compromised by alteration can be assessed using element ratios that vary very little in oceanic island basalts (OIBs). Useful ratios for the U-Th-Pb systems include Ce/Pb (25 ± 5), Nb/U (47 ± 10), and Th/U (4 ± 0.5 ; Hofmann et al., 1986; O'Nions and McKenzie, 1993; Nobre Silva et al.,

2013a). Here we use the Th/U ratio of samples to estimate primary U concentrations in unleached sample powders following the method of Nobre Silva et al. (2013a) that considers Th immobile during basaltic weathering and used the Th/U ratio to assess U loss or gain. The average Th/U ratio of Hawaiian shield lavas is ~ 3 ($n=294$), not significantly different from NWHR lavas or OIBs globally (O’Nions and McKenzie, 1993; Nobre Silva et al., 2013a). Correcting measured U concentration values back to a Th/U of 3 (assuming only U has been gained or removed by alteration) provides a robust estimation of magmatic U concentrations and a better approximation of initial Pb isotopic ratios. In addition to using this method on our new isotopic data, we also re-correct Emperor Seamount literature data using this technique for overall comparability of the dataset. Previous researchers addressed secondary alteration elemental mobility by measuring concentrations of leached sample powders for age correction purposes (Regelous et al., 2003; Huang et al., 2005; Shafer et al., 2005). However, this method assumes that secondary alteration was a closed system and did not fractionate elements between the primary and secondary phases (Staudigel et al., 1996). More recently it has been shown that acid leaching differentially removes some elements (Nobre Silva et al., 2009, 2010; Hanano et al., 2009). The impact of elemental mobility during alteration is particularly well displayed by the oldest sampled Emperor Seamount, Meiji (~ 85 Ma), which is significantly altered and has proven notoriously problematic for accurate age correction (Regelous et al., 2003; Frey et al., 2005; Huang et al., 2005). Using the U/Th estimation method described here to age-correct previous data provides age-correct Pb isotopic compositions that are more coherent than previously reported for Emperor Seamount data. To illustrate, radiogenic Pb of Meiji ranges from 0.88-0.92 after applying our correction method versus unrealistically high 0.96-1.01 in published data (Regelous et al., 2003). For comparison, average $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ of the most enriched Hawaiian volcano Ko‘olau is 0.96 ± 0.02

(n=36) while East Pacific Rise MORB is 0.92 ± 0.03 (n=146). Because Meiji erupted under the thinnest lithosphere of any Hawaiian volcano with high degrees of melting near a mid-ocean ridge, it is highly unlikely that lavas have such highly radiogenic Pb isotopic compositions, overlapping with Ko‘olau Makapu‘u compositions that are only observed on O‘ahu (Keller et al., 2000; Regelous et al., 2003; Frey et al., 2005). Also, the use of measured elemental concentrations on leached sample powders of such altered rocks tends to over-correct measured isotopic ratios and is not the adequate solution to correct for in situ decay of radioactive parents (Frey et al., 2005). Finally, the robustness of our age correction procedure is illustrated in Figure S1, which shows $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ of the NWHR, Emperor Seamounts, and major Hawaiian Island volcanoes (see supp. info. references for data sources in Figures 2, 3, S1, and S2). The limited range of $^{207}\text{Pb}/^{204}\text{Pb}$ of NWHR lavas that overlaps with non-age corrected young Hawaiian Island data supports the robustness of our leaching and age-correction procedure to recover accurate initial isotopic ratios of plume sourced basalts.

4. Results

Extreme Ko‘olau Makapu‘u-type Pb isotopic signatures were found only once along the NWHR; Daikakuji Seamount has the most unradiogenic $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, and $^{208}\text{Pb}/^{204}\text{Pb}$ among NWHR lavas (17.869, 15.418, and 37.809, respectively; Fig. 2a; Table 1). The most radiogenic lead isotopic compositions are observed for lavas from Midway in $^{206}\text{Pb}/^{204}\text{Pb}$ (18.669) and Pioneer Seamount in $^{207}\text{Pb}/^{204}\text{Pb}$ (15.493) and $^{208}\text{Pb}/^{204}\text{Pb}$ (38.206) (Fig. 2a). Samples from Daikakuji, Mokumanamana, and West Nīhoa plot on the Loa side of the Loa-Kea divide line in $^{208}\text{Pb}/^{204}\text{Pb}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ isotopic space (Fig. 2a). After Daikakuji, there are no Loa-like compositions until the youngest section at Mokumanamana and Nīhoa (less than ~11 Ma; Dalrymple et al., 1974). The Pb isotopic composition of Nīhoa lavas span about two-thirds of the

entire range of $^{208}\text{Pb}/^{204}\text{Pb}$ present in young Hawaiian lavas (37.913 – 38.144) and define a trend that crosses the Loa-Kea divide (Fig. 2a), an intriguing hint that Nīhoa may be a composite island with two volcanic centers of distinct compositions. Further work is needed to explore this possibility.

The majority of NWHR lavas are distinctly Kea-like in their geochemical affinity (Fig. 2a; Table 1). These lavas span nearly the entire range in $^{208}\text{Pb}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ observed in Kea-trend shield lavas on the Hawaiian Islands (Fig. 2b). The variation in Pb isotopic composition can be explained by mixing 40-91% of the Kea end-member component with an isotopically depleted plume source such as the Depleted Makapu‘u (DMK) component, one of the four end-member components identified in Hawaiian lavas (i.e. Kea, EMK, DMK, Lō‘ihi; Tanaka et al., 2002, 2008; see Fig. 2b). All of the Hawaiian geochemical components are thought to be intrinsic to the plume and sourced from the deep mantle (Tanaka et al., 2002, 2008; Regelous et al., 2003; Weis et al., 2011).

Some NHWR lavas (Daikakuji and Nīhoa) have Loa-like Pb isotopic compositions whose range of variations may be explained by mixing 35-40% of the Enriched Makapu‘u (EMK) with the Kea component (Fig. 2b; Tanaka et al., 2002). The Enriched Makapu‘u end member, proposed to contain mafic recycled crustal material (Tanaka et al., 2008), is more fusible and thus likely to be observed under lower melting conditions (Fig. 2a inset; Pertermann and Hirschmann, 2003). High primitive-mantle normalized $\text{La}/\text{Yb}_{\text{PM}}$ values (>5) correspond to lower degrees of partial melting because La is more incompatible and highly sensitive to degree of partial melting whereas Yb is nearly constant in Hawaiian lavas due to melting in the garnet stability field (Hofmann and Farnetani, 2013). This is not an effect of regional changes in lithospheric thickness because the NWHR erupted on oceanic lithosphere older than ~70 Ma (White, 1993; Caplan-

Auerbach et al., 2002), which is the age when oceanic lithospheric thickness reaches a stable maximum of ~90 km based on the plate tectonic model (Parsons and Sclater, 1977). The isolated occurrence of enriched Loa-type Pb isotopic composition at Daikakuji suggests it is not related to a first arrival of a sustained Loa component in the Hawaiian mantle plume as previously proposed (Garcia et al., 2015). Daikakuji is characterized by anomalously high La/Yb_{PM} ratios compared to Hawaiian shield basalts (La/Yb_{PM} of 6.9-8.5 for Daikakuji lavas versus 2.0-5.1 for Mauna Loa and Mauna Kea lavas; primitive mantle values from McDonough and Sun, 1995). Many NWHR Loa and Kea-type lavas are high in La/Yb_{PM} compared to Hawaiian island and Emperor Seamount shield lavas and may reflect lower degrees of melting, at least locally. Previous authors argued that Emperor Seamount lavas are characterized by higher ²⁰⁶Pb/²⁰⁴Pb at a given ²⁰⁸Pb/²⁰⁴Pb because they were initially erupted onto thin, young crust that grew progressively thicker with time and increasing distance from the Pacific-Kula spreading ridge (Keller et al., 2000; Regelous et al., 2003). The geochemistry of Emperor Seamount lavas reflects this thickening lid effect, with a possible contribution from a depleted MORB mantle source for the oldest seamounts, Detroit and Meiji (Fig. 2; Keller et al., 2000; Regelous et al., 2003; Whittaker et al., 2015). Conversely, all NWHR volcanoes erupted onto oceanic crust older than 70 Ma without interaction with a nearby spreading ridge (supplementary fig. S2; White, 1993; Whittaker et al., 2015). Thus the depleted component (DMK) involved in mixing with the Kea component to produce NWHR Kea-type lavas is different from the one involved in the formation of the Emperor Seamounts.

Lead isotopes generally vary systematically in time and space along the NWHR (Fig. 3). They become more radiogenic (i.e. increase in ²⁰⁸Pb*/²⁰⁶Pb*) from the oldest to the youngest volcanoes. This represents a fundamental difference from the Emperor Seamounts, which on

average are uniformly unradiogenic. Radiogenic Pb correlates with the overall increase in magmatic flux along the NWHR, except for Gardner and Daikakuji Seamounts, which are respectively local maxima and minima of magmatic flux along the ridge (Fig. 3 inset). Furthermore, volcano compositions separate into three compositional trends (Kea, Loa, and Enriched Loa, Fig. 3 inset) only for the main Hawaiian Islands at a critical threshold of magmatic flux ($\sim 4 \text{ m}^3/\text{s}$ based on the most recent magmatic flux model; Wessel, 2016). NWHR volcanoes do not present a clear geographical distribution as exhibited by the parallel chains of Loa and Kea paired volcanoes on the Hawaiian Islands younger than O‘ahu ($\sim 3 \text{ Ma}$). For much of the NWHR, there are only single, widely spaced volcanoes, while in other places there are as many as four volcanoes abreast; thus geochemistry is the only predictor of Loa or Kea-type affinity rather than the spatial positioning of NWHR volcanoes.

5. Discussion

5.1 Role of the Lithosphere in Magmatic Flux and Pb Composition of NWHR Basalts

The thickness of the lithosphere is postulated to have an effect on magmatic flux because thinner, warmer lithosphere allows higher degrees of melting than thick, cold lithosphere (White, 1993; Keller et al., 2000; Regelous et al., 2003; Niu et al., 2011). Higher degrees of melting results in lavas that are less enriched in incompatible elements with lower radiogenic Pb due to dilution of the signature of small, enriched heterogeneities by productive melting of large volumes of peridotite (Pertermann and Hirschmann, 2003; Garcia et al., 2010). Thus if lithospheric thickness did exert a primary control over magmatic flux, we would also expect it to impact the erupted Pb isotopic signature of basalts. Along the NWHR, there is no correlation of radiogenic Pb with lithospheric thickness (see Supp. Fig. S2), regardless of the type of lithospheric thickness model employed (i.e. a half space cooling model or a plate tectonic model;

Parker and Oldenberg, 1973; Parsons and Sclater, 1977). This indicates that lithospheric thickness variations are not as significant as source composition changes in explaining the exponential increase in magmatic flux along the NWHR. Emperor Seamount geochemistry, conversely, correlates with changes in lithospheric thickness, although it is important to note that increases in lithospheric thickness from the oldest to youngest Emperor Seamounts did not result in any significant decrease in magmatic flux (Supp. Fig. S2; Regelous et al., 2003; Garcia et al., 2015). Furthermore, both the Emperor Seamounts and all of the Hawaiian chain younger than the bend exhibit the same magnitude of variation in radiogenic Pb: Emperor Seamounts $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ range from 0.89 at Meiji to 0.94 at Detroit, 59.4% of total range of Hawaiian-Emperor chain; younger than the bend range from $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ 0.92 at Midway to 0.97 at Ko‘olau, 59.6% of total range. This corresponds to vastly different ranges of variations of lithospheric thickness (Emperor Seamounts 22.4-92.2 km, 92% of total range of Hawaiian-Emperor chain; younger than the bend 89.4-98.2 km, 10% of the total range) and magmatic flux (Emperor Seamounts ~1% of variation; younger than the bend ~99% of variation). These results are inconsistent with the hypothesis that changes in geochemical signature of erupted basalts and in magmatic flux are principally controlled by lithospheric thickness.

Physical variations of the lithosphere such as changes in temperature and thickness between the major Pacific fracture zones (e.g. Murray, Mendicino, and Molokai fracture zones; Fig. 1 inset) could have an effect on magmatic flux (Van Ark and Lin, 2004; Wessel, 2016). However, NWHR geochemistry does not alter across these structures and instead gradually increases in radiogenic Pb across all fracture zones of the NWHR and Hawaiian Islands (Fig. 3). It has also been hypothesized that variations between extensional and compressional stress in the Pacific Plate by changes in absolute plate motion could have an effect on how much magma is

able to pass through the lithosphere and erupt (Wessel and Kronke, 2008; O'Connor et al., 2013; Wessel, 2016). Potentially, this could have impact the degree of melting along the NWHR, but both the steady increase in radiogenic Pb and in magmatic flux along the NWHR after Midway Island would require a steady transition into an increasingly tensional stress regime for the Pacific Plate from ~20 Ma to the present. This is unlikely as geologic events that alter absolute plate motion around the Pacific are more episodic (Wessel and Kronke, 2008). It has also been hypothesized that small-scale sub-lithospheric rolls caused by sub-lithospheric convection is the cause of the Loa and Kea bilateral trends on the Hawaiian Islands (Ballmer et al., 2011). However, this theory breaks down along the NWHR because the entire NWHR is erupted onto oceanic crust mature enough to have small-scale sub-lithospheric convection, yet there is only an episodic presence of the Loa component and no evidence for a bilateral plume along the NWHR.

It is possible that smaller timescale fluctuations in magmatic flux (e.g. 1-2 and 10-15 Myr variations; Wessel, 2016; see Fig. 3) could be caused by variations in plume dynamics (e.g. solitary waves; Whitehead and Helfrich, 1988; Ito, 2001) possibly generated by pulses of core-mantle heat transfer (Mjelde et al., 2010). However, our sample resolution is not fine enough to assess the potential correlation of geochemistry with such a short periodicity. The current resolution of sampling does not show any correlation of radiogenic Pb with these small fluctuations in flux, while there is a clear general trend of increasing magmatic flux. The same limitation in sampling resolution applies to assess the possible effect of local fluctuations in the thickness or stress field of the lithosphere. Below, we propose a deep mantle geodynamic conceptual model that accounts for the variation of Pb isotopes with time along the Hawaiian-Emperor chain and also explains geophysical changes in magmatic flux.

5.2 Role of the LLSVP in Magmatic Flux and Pb Isotopic Composition of NWHR Basalts

The arrangement of compositionally heterogeneous lower mantle structures in the source region of deep-sourced plumes is thought to control the geochemistry of erupted oceanic island basalts (Farnetani and Hofmann, 2009; Weis et al., 2011). The Hawaiian mantle plume is located above an ultra-low velocity zone (ULVZ) at the edge of the large low shear velocity province (LLSVP) located in the Pacific lower mantle near the core-mantle boundary (McNamara et al., 2010; Cottaar and Romanowicz, 2012; French and Romanowicz, 2015; Garnero et al., 2016). LLSVPs are thousands of kilometers wide, sharp-sided (~400-600 km thick), chemically heterogeneous, long-lived piles of anomalously seismically slow material in the lower mantle below the Pacific Ocean and African continent (Garnero et al., 2016). ULVZs are much smaller, denser features (~10 km thick and <100 km across, except underneath Hawai'i where it is ~900 km across; Cottaar and Romanowicz, 2012) that may contain partial melts (Jellinek and Manga, 2004; McNamara et al., 2010; Garnero et al., 2016). Mantle plumes may drift across or around these lower mantle structures on long timescales triggered by dynamic re-equilibration from the arrival of subducting slabs (McNamara et al., 2010; Bower et al., 2013; Garnero et al., 2016; Hassan et al., 2016). It has also been suggested that the enriched geochemical compositions of some oceanic island basalts originate from these lower mantle reservoirs, which are either the remnants of a primordial magma ocean or accumulations of subduction recycled crustal material (Weis et al., 2011; Li et al., 2015; Garnero et al., 2016). ULVZ material is denser than surrounding peridotitic mantle and only small amounts are likely entrained by plumes in addition to LLSVP material (Jellinek and Manga, 2004; McNamara et al., 2010; Weis et al., 2011; Cottaar and Romanowicz, 2012; Li et al., 2015). Increasing radiogenic Pb along the NWHF suggests an increasing contribution from these enriched sources with time. Dynamic changes in the lower mantle may control the amounts of LLSVP, ULVZ, and ambient mantle entrained into the

Hawaiian mantle plume. We propose that the Hawaiian plume initiated close to the margin of the Pacific LLSVP, sampling only the Kea component (the deep Pacific mantle) during Emperor Seamount formation, then drifted up and anchored at the edge of the Pacific LLSVP to entrain enriched material in increasing quantities as the NWHR formed (Fig. 4).

Motion of the Hawaiian mantle plume has the potential to sample different geochemical sources over its 85 million year lifetime. Plumes worldwide migrate slowly through the mantle at velocities of ~ 1.1 cm/year (Davies and Davies, 2009), an observation supported by the better fit of dynamic models with moving-source plumes than fixed-source plumes to global seismic tomographic models (Boschi et al., 2007). The lower mantle has been moving south at a rate of 1-4 cm/year (seismic anisotropy studies; Walker et al., 2011) or 1-2 mm/year (plume experiments; Davaille et al., 2003). Dynamic global mantle convection and hotspot track reconstructions of the last 230 Myr identified south-southeast flow with varied strength for the base of Hawaiian mantle plume towards the central Pacific (Steinberger, 2000, 2004; Davaille et al., 2003; Burke and Torsvik, 2004; Hassan et al., 2016). Assuming the base of the Hawaiian mantle plume was advected in that flow at the lowest rate (~ 1 mm/year; Steinberger, 2000, 2004; Garnero et al., 2016; Hassan et al., 2016), it could have drifted ~ 80 kilometers in the lower mantle. Plume movement tends to be episodic (Davies and Davies, 2009; Hassan et al., 2015) while still allowing the sampling of different lower mantle domains. Dynamic models and plume experiments have established that plumes anchor on topographic features such as ULVZs or LLSVP ridges (Jellinek and Manga, 2004; Steinberger et al., 2004; Boschi et al., 2007; McNamara et al., 2010; Bower et al., 2013; French and Romanowicz, 2015). Finally, the mass conservation law requires compensation of upper mantle wind by equal and opposite movement in the lower mantle (Steinberger, 2000; Farnetani and Hofmann, 2009), likely resulting in plume drift.

It has been proposed that the highest temperatures at the core mantle boundary are located along the rim of LLSVP; this is where P-waves begin to slow, many mantle plumes are generated, and ULVZs are detected (Jellinek and Manga, 2004; Kumagai et al., 2008; Davies and Davies, 2009; McNamara et al., 2010; Cottaar and Romanowicz, 2012; Hassan et al., 2015). The northern edge of the Pacific LLSVP underneath Hawai'i is not steep sided, but has a 25-35° north-northwest-facing gentle slope to a topographic high ~450 km above the core mantle boundary (He and Wen, 2012; Frost and Rost, 2014; Zhao et al., 2015). The Hawaiian mantle plume initiated slightly to the north of the present plume location where the highest temperatures occur (Fig. 4b). After formation, the flow of ambient mantle south up the edge of the LLSVP would push the plume to the most dynamically stable place, anchored at an LLSVP ridge above a ULVZ (Fig. 4c, 4d; Davaille et al., 2003; Jellinek and Manga, 2004; Zhao et al., 2015; Garnero et al., 2016). During this time, the Hawaiian mantle plume sampled peridotitic mantle from outside the LLSVP (Sobolev et al., 2005; Tanaka et al., 2008), which corresponds to the Kea component, also present in nearly all Pacific oceanic plateaus (e.g. Ontong Java, Shatsky Rise, and Wrangellia) and oceanic islands (Nobre Silva et al., 2013b). It is also the same composition as prevalent mantle (PREMA), an ubiquitous component in the mantle where oceanic island basalt isotope arrays converge (White, 2015 and references therein). In addition, the dominance of this material during the initiation of the Hawaii mantle plume (e.g. Emperor Seamounts) would account for the low magmatic flux during this time because peridotite is less productive than enriched, mafic lithologies (Pertermann and Hirschmann, 2003; Van Ark and Lin, 2004; Vidal and Bonneville, 2004). This is in agreement with the theoretical flux for a fully peridotitic Hawaiian plume ($\sim 3.5 \text{ m}^3 \text{ s}^{-1}$ for peridotite only, rather than the observed $8 \text{ m}^3 \text{ s}^{-1}$) and within the range of other Pacific plume fluxes (Courtillet et al., 2003; Sobolev et al., 2005). Conversely, if the Hawaiian plume

was both generated and fixed on the Pacific LLSVP, we would expect to observe Loa compositions in lavas from the entirety of the Hawaiian-Emperor chain. Although it is possible that the participation of the Loa component was concealed in Emperor Seamount lavas by dilution from voluminous melting beneath young, thin lithosphere (supplementary figure S2; Regelous et al., 2003; Hofmann and Farnetani, 2013), we would then expect the Emperor Seamount data to exhibit mixing relationships that include the Loa enriched end member (EMK), which is not observed (Figure 2b).

After the plume was anchored on a LLSVP ridge above the ULVZ, it entrains initially small amounts of enriched material from the LLSVP and/or ULVZ (Fig. 4d). Incorporation of increasing amounts of this material in NWHR volcanoes gradually elevates radiogenic Pb (Fig. 3), changes the slope of magmatic flux growth (Wessel, 2016; Van Ark and Lin, 2004; Vidal and Bonneville, 2004), and accelerates the volcanic propagation rate (O'Connor et al., 2013). These changes all commenced ~20 Myr after the bend around Midway Island. Assuming a 15-25 Myr mantle plume transit time, this enriched material was entrained around the time of the bend (Hassan et al., 2015; 2016). Plate reconstruction studies have found that absolute plate motion change was likely 4-31° in the Pacific around the time of the bend (Steinberger et al., 2004; Tarduno et al., 2009; Wright et al., 2015), not enough to account for the ~120° change in bearing of the Hawaiian-Emperor chain at the bend (i.e. a ~60° change in absolute Pacific plate motion; Hassan et al., 2016; Tarduno et al., 2009). Previous studies hypothesized that interaction of the Hawaiian mantle plume with the Kula-Pacific mid-ocean ridge around ~85 Ma tilted the plume drastically. With both increasing distance from and waning in spreading rate of the ridge, the plume moved back to a more vertical position, resulting in hotspot motion (Tarduno et al., 2009). This documented plume movement in the upper mantle coupled with the hypothesized southward

plume motion in the lower mantle stabilized around the time of the bend and likely contributed to the sharpness of the bend and to the transition in Hawaiian plume geochemical signature (Steinberger, 2000; 2004; Tarduno et al., 2009; Hassan et al., 2016).

We speculate that the longer the plume is fixed on a ridge of the LLSVP, the more sustained is the sampling of LLSVP and/or ULVZ enriched material, resulting in both higher radiogenic Pb and magmatic flux as Loa-type source lithologies are more fusible pyroxenites and eclogites (Pertermann and Hirschmann, 2003; Sobolev et al., 2005). The bilateral Kea-Loa trends of the Hawaiian Islands appear once the plume has been anchored on the Pacific LLSVP for enough time to provide a sustained supply of both enriched material on the southwest edge of the plume (Loa) and ambient Pacific mantle on the northeast side of the plume (Kea; Fig. 4e; Farnetani and Hofmann, 2009; Weis et al., 2011; Nobre Silva et al., 2013b). The ephemeral presence of the Loa composition along the NWHZ suggests that a simple concentric or bilateral zonation of the Hawaiian mantle plume is overly simplistic, as concluded for the Hawaiian Islands (Xu et al., 2014). The isolated and highly radiogenic lavas erupted at Daikakuji volcano right after the Hawaiian-Emperor bend may be explained by a combination of unique melting conditions of this volcano coupled with the presence of a small mantle heterogeneity. Daikakuji exhibits the coldest mantle potential temperature of any studied Hawaiian volcano (i.e. 1335 ± 26 °C, versus the current range of estimated mantle potential temperatures on the Island of Hawai'i 1632-1690 °C; Tree, 2016), resulting in low degrees of melting. This is also supported by the fact that Daikakuji has the highest $(La/Yb)_{PM}$ of the entire Hawaiian-Emperor chain (Fig. 2a inset). These extremely low degrees of melting allowed the signature of a finite and isolated heterogeneity to be expressed for a short period of time. The mantle has been shown to be heterogeneous on scales varying between melt inclusion scale (Ren et al., 2005) to the hundreds

of kilometers scale (Garnero et al., 2016), and periodic entrainment of plum-pudding type heterogeneities is not unexpected.

6. Conclusion

New high-precision Pb isotopic data closes the 2700 km (~42 Myr) gap in the Hawaiian-Emperor chain geochemical record and shows a transition in lava composition commencing ~20 million years after the bend. During this period, the flux of the Hawaiian plume, mantle potential temperature, and volcanic propagation rate all increased. The correlation of radiogenic Pb with magmatic flux indicates that all of these changes are related, and we suggest may be triggered by the movement of the deep source of the Hawaiian plume through different lower mantle compositional domains, from sampling dominantly ambient Pacific mantle outside of the LLSVP to entraining enriched material from the ULVZ and Pacific LLSVP. Mounting geochemical and geodynamical data on oceanic islands and chains suggest the lower mantle is both dynamic and compositionally heterogeneous in space and time. Long-lived mantle plumes such as Hawai'i provide a window into this fundamental Earth reservoir and future integrated geochemical and geophysical studies of long island chains will provide the key insight needed to decipher the relationship between geochemical components and geophysical anomalies in the deep mantle.

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746 **Figure Captions**

747 **Figure 1. Bathymetric map of the ~51 seamounts and islands of the Northwest Hawaiian**

748 **Ridge and sample locations.** Bathymetry is 2-minute Gridded Global Relief Data ETOPO2v2

749 satellite altimetry dataset (U.S. Department of Commerce, National Oceanic and Atmospheric

750 Administration, National Geophysical Data Center, 2006) and new multibeam bathymetry (Smith

et al., 2014). White circles show sample locations. References for ages are in yellow superscript and are as follows: 1 – O’Connor et al., 2013; 2 – Dalrymple et al., 1974; 3 – Dalrymple et al., 1981; 4 – Garcia et al., 1987; 5 – Garcia et al., 2010. Inset figure is a traverse Mercator projection of the Hawaiian-Emperor chain modified from Garcia et al., 2015. Dashed lines in this figure indicate major Pacific fracture zones (FZ).

Figure 2. Pb-Pb isotopic diagrams for Northwest Hawaiian Ridge shield-stage basalts (error bars are smaller than data points). **a** Plot of $^{206}\text{Pb}/^{204}\text{Pb}$ versus $^{208}\text{Pb}/^{204}\text{Pb}$ and **b** $^{207}\text{Pb}/^{206}\text{Pb}$ versus $^{208}\text{Pb}/^{206}\text{Pb}$ (data sources in supplementary material). For reference the Loa-Kea divide line (Abouchami et al., 2005) is shown in black, and the four Hawaiian end member components are shown by empty white circles (KEA, DMK, EMK, and Lō‘ihi; Tanaka et al., 2002). Mixing lines are calculated between the KEA end member and enriched Makapu‘u (EMK; blue line) and depleted Makapu‘u (DMK; red line). Tick marks are at intervals of 10%. Inset to figure a shows a histogram of primitive mantle normalized (McDonough and Sun, 1995) La/Yb of the Emperor Seamounts (orange), Mauna Loa (blue), Mauna Kea (red), the Northwest Hawaiian Ridge (yellow), and Daikakuji (purple). The black line is the average of Mauna Loa and Mauna Kea, representative of average Hawaiian shield basalt ($\text{La}/\text{Yb}_{\text{PM}} \approx 3.4$) and the dashed line is average EPR MORB ($\text{La}/\text{Yb}_{\text{PM}} \approx 0.8$). Inset to figure b gives the composition of the end members used in the calculation of mixing lines (Tanaka et al., 2002). All literature data in this and following figures is normalized to the same NIST SRM 981 values to ensure comparability.

Figure 3. Northwest Hawaiian Ridge $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ variations with Hawaiian Plume estimated volume flux (in m^3/s assuming flexural compensation; Van Ark and Lin, 2004; Vidal and Bonneville, 2004; Wessel, 2016). The black curve is the average flux variation with uncertainty shown in red (Wessel, 2016). The flux of the Emperor Seamounts is from Van Ark

and Lin (2004; green line) and Vidal and Bonneville (2004; purple line) or not calculated (Wessel, 2016). Radiogenic Pb increases gradually with age along the NWHR (dashed regression lines). Emperor seamount triangles are averages of all data from each seamount with 2SD error bars. Hawaiian Island data is the average of each volcano. Inset shows in detail $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ versus estimated volume flux (m^3/s ; Wessel, 2016). Age correction of $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ is smaller than datapoints.

Figure 4. Model of the evolution of the Hawaiian plume source at the CMB. Figure shows the lowermost 800-1000 kilometers of mantle (not to scale; blue lens is an ULVZ). Black and grey arrows represent generalized flow in the mantle (Steinberger, 2000; Davaille et al., 2003; Walker et al., 2011; Hassan et al., 2016) and LLSVP (McNamara et al., 2010; Zhao et al., 2015; Garnero et al., 2016). **A:** Pacific LLSVP **B:** The Hawaiian plume is created at the LLSVP edge where temperature anomalies are greatest and large viscosity gradients stabilize an initially thermal plume (Jellinek and Manga, 2004; Davies and Davies, 2009; Garnero et al., 2016). **C:** The plume will migrate in mantle flow up the sloped northern edge of the LLSVP to a ridge where plumes are dynamically stabilized over geologic time (Steinberger, 2000, 2004; Davaille et al., 2003; Burke and Torsvik, 2004; Jellinek and Manga, 2004; Davies and Davies, 2009; Bower et al., 2013; Hassan et al., 2015; Zhao et al., 2015). Flow into thermals perturbs ULVZ material below (Li et al., 2015). **D:** LLSVP material is entrained in the plume and $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ values increase in erupted lavas (Weis et al., 2011; Li et al., 2015). **E:** A significant amount of LLSVP (+/- ULVZ) is consistently continuously entrained into the southeast side of the Hawaiian plume, resulting in presence of the bilateral Loa and Kea trends observed on the Hawaiian Islands.

Figure1
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Figure3
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Figure4

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Table 1: Pb Isotopic Composition and Pb, U and Th Concentrations of Northwest Hawaiian Ridge Basalts

Sample	Seamount	Distance from Kilauea (km)	Est. Vol. Flux (m ³ /s) ⁸	Age (Ma)	Age Ref.	La/Yb _{PM} ^a	Measured Ratios ^c								Pb ^{ICP} (ppm) ^a	Th ^{ICP} (ppm) ^a	U ^{ICP} (ppm) ^a	Th/U ^d	U and Age-Corrected Isotopic Ratios ^d			
							²⁰⁸ Pb/ ²⁰⁴ Pb	2σ	²⁰⁷ Pb/ ²⁰⁴ Pb	2σ	²⁰⁶ Pb/ ²⁰⁴ Pb	2σ	²⁰⁸ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb					²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb*/ ²⁰⁶ Pb*		
A-53-1	Yuryaku	3541	0.4	46.9	1	3.17	38.0527	21	15.4718	8	18.5140	8	0.74	0.71	0.314	2.27	37.904	15.465	18.363	0.9306		
A-55-1	Daikakuji	3520	0.4	46.8	1	6.86	37.9374	22	15.4244	8	17.9992	7	1.53	1.31	0.436	3.02	37.809	15.418	17.869	0.9732		
A-55-2	Daikakuji	3520	0.4	46.8	1	8.53	37.9977	26	15.4372	11	18.0915	12	2.22	2.18	0.672	3.24	37.850	15.430	17.942	0.9698		
A-55-4	Daikakuji	3520	0.4	46.8	1	8.42	38.0094	23	15.4402	14	18.0972	11	2.16	2.22	0.689	3.22	37.855	15.433	17.941	0.9705		
84-28E	Unnamed	2801	0.6	31.7	6	5.38	38.2106	27	15.4958	8	18.7045	10	1.70	1.46	0.474	3.08	38.119	15.492	18.612	0.9289		
84-30G	Academician Berg	2608	1.3	31.0	6	5.41	38.2009	27	15.4923	9	18.7037	11	1.24	1.42	0.438	3.25	38.079	15.487	18.580	0.9277		
R-1316	Midway	2447	1.8	27.6	1	5.09	38.2085	29	15.4890	8	18.7484	16	1.06	1.08	0.368	2.93	38.117	15.485	18.656	0.9243		
R-1295.6	Midway	2447	1.8	27.6	1	4.82	38.2173	22	15.4890	8	18.7601	11	1.03	1.04	0.375	2.77	38.127	15.485	18.669	0.9241		
P5-524-42	Pioneer	1998	3.4	28.0	7	4.81	38.2475	20	15.4949	5	18.7008	6	2.63	1.20	0.817	1.47	38.206	15.493	18.659	0.9335		
76-5-4 DD	Northampton	1846	3.6	26.6	3	3.81	38.2028	19	15.4818	7	18.6307	8	0.83	0.83	0.275	3.01	38.126	15.478	18.553	0.9355		
76-5-4A	Northampton	1846	3.6	26.6	3	3.50	38.2039	23	15.4813	8	18.6348	8	0.84	0.77	0.277	2.79	38.133	15.478	18.564	0.9353		
76-6-7-F	Gardner	1449	5.8	12.3	4	3.12	38.1315	68	15.4785	28	18.5903	30	0.61	0.53	0.196	2.69	38.097	15.477	18.555	0.9322		
84-40E	Gardner	1449	5.8	12.3	4	3.04	38.0246	29	15.4690	12	18.4651	13	0.75	0.64	0.264	2.44	37.991	15.467	18.431	0.9332		
84-39B	Gardner	1449	5.8	12.3	4	3.30	38.1808	33	15.4858	14	18.5857	13	1.91	0.93	0.287	3.24	38.161	15.485	18.566	0.9380		
NEC-3A	Mokumanamana	1080	3.1	10.0	2	2.79	37.9611	28	15.4714	10	18.3017	11	1.31	0.91	0.284	3.22	37.934	15.470	18.274	0.9432		
72-49A	Keoea	963	2.0	11.0	7	3.49	38.0024	23	15.4754	8	18.4213	9	0.98	0.85	1.041	0.81	37.972	15.474	18.390	0.9353		
P5-688-1	Twin Banks	920	2.1	9.6	4	3.59	38.0459	28	15.4792	10	18.4813	9	0.71	0.55	0.854	0.64	38.024	15.478	18.459	0.9340		
76-9-11	West Nihoa	825	2.6	8.8	1	6.23	37.9402	25	15.4724	7	18.1919	7	66.2 ^b	1.38	0.694	1.98	37.940	15.472	18.191	0.9527		
NIH-D4	Nihoa	794	2.6	7.0	2	3.88	38.0444	19	15.4789	7	18.4540	10	0.78	0.70	0.211	3.32	38.020	15.478	18.429	0.9366		
NIH-W11-1	Nihoa	794	2.6	7.0	2	5.72	38.1702	27	15.4827	9	18.6103	9	0.78	0.74	0.246	3.02	38.144	15.481	18.584	0.9344		
NIH-F9	Nihoa	794	2.6	7.0	2	4.69	38.1109	24	15.4744	8	18.5338	9	0.74	0.73	0.153	4.76	38.084	15.473	18.507	0.9357		
NIH-D-1-2	Nihoa	794	2.6	7.0	2	8.17	37.9256	27	15.4752	9	18.2092	10	2.53	1.16	0.429	2.70	37.913	15.475	18.200	0.9488		

¹⁻⁸References to ages and melt flux: 1- O'Connor et al., 2013; 2- Dalrymple et al., 1974; 3- Dalrymple et al., 1981; 4- Garcia et al., 1987; 5- Garcia et al., 2010; 6- Sharp and Clague, 2006; 7- Age inferred by interpolating between existing ages and assuming a linear age progression of NWHR volcanoes; 8- Wessel, 2016.

^aTrace element concentrations (in ppm) were measured on unleached whole rock powders using a Thermo Finnigan Element2 high resolution ICP-MS at the PCIGR at UBC. USGS Reference Material BCR-2 was used for external calibration and a 1 ppb In spike was used as an internal standard. RSDs are less than 5% or better for all elements and measured concentrations of USGS Standard BHVO-2 are within 2SD of published values.

^bThis sample was run in duplicate. The high Pb appears to come from the rock itself, likely contained in secondary minerals that are removed by acid leaching before isotopic analysis. This is supported by Pb isotopic values from leached whole rock powder for this sample that plot within the field for Hawaiian shield basalts.

^cPb isotopes were measured on acid leached whole rock powders (after being purified twice on ion exchange columns) using a Nu Plasma II multicollector ICP-MS at the PCIGR at UBC. Pb data were corrected for fractionation by a Tl spike and 2σ error is the absolute error values of the individual sample analysis (internal digit) reported to the significant digit. Pb standard NIST SRM-981 values during the course of this study were ²⁰⁸Pb/²⁰⁴Pb=36.7188 ± 0.0107, ²⁰⁷Pb/²⁰⁴Pb=15.4983 ± 0.0041, and ²⁰⁶Pb/²⁰⁴Pb=16.9418 ± 0.0028 (n=102).

^dTh/U ratio is corrected to a Th/U value of 3 (average Th/U of the mantle, all OIB, and average of Hawaiian Island shield tholeiites) based on the assumption of U mobility only. These adjusted U concentrations are then used to age correct measured Pb isotopic ratios avoiding major complications due to secondary alteration.