

# The chemical structure of the Hawaiian mantle plume

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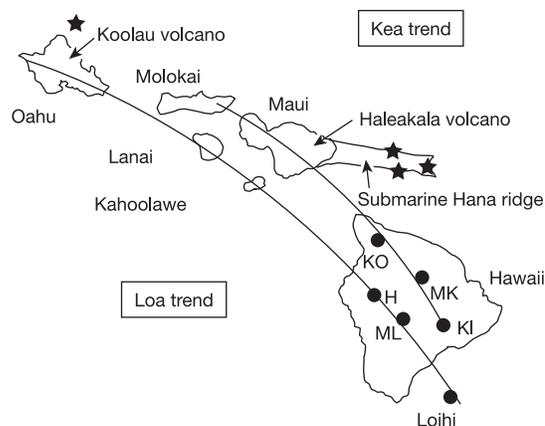
The Hawaiian–Emperor volcanic island and seamount chain is usually attributed to a hot mantle plume, located beneath the Pacific lithosphere, that delivers material sourced from deep in the mantle to the surface<sup>1–5</sup>. The shield volcanoes of the Hawaiian islands are distributed in two curvilinear, parallel trends (termed ‘Kea’ and ‘Loa’), whose rocks are characterized by general geochemical differences<sup>2–5</sup>. This has led to the proposition that Hawaiian volcanoes sample compositionally distinct, concentrically zoned, regions of the underlying mantle plume<sup>4,5</sup>. Melt inclusions, or samples of local magma ‘frozen’ in olivine phenocrysts during crystallization, may record complexities of mantle sources<sup>6</sup>, thereby providing better insight into the chemical structure of plumes. Here we report the discovery of both Kea- and Loa-like major and trace element compositions in olivine-hosted melt inclusions in individual, shield-stage Hawaiian volcanoes—even within single rock samples. We infer from these data that one mantle source component may dominate a single lava flow, but that the two mantle source components are consistently represented to some extent in all lavas, regardless of the specific geographic location of the volcano. We therefore suggest that the Hawaiian mantle plume is unlikely to be compositionally concentrically zoned. Instead, the observed chemical variation is probably controlled by the thermal structure of the plume.

Most of our current understanding of the geochemistry of mantle plumes originates from data on whole rocks. Yet, whole rocks record only an ‘average’ composition produced by complex petrogenesis involving partial melting in the mantle, subsequent aggregation of melts on their way to the surface, and mixing of melts in shallow magma chambers<sup>6</sup>. In contrast, melt inclusions can provide important information on compositions of magmas before extensive magma mixing<sup>6–9</sup>. To test the homogeneity of the mantle plume source sampled in the Kea and Loa trends by Hawaiian shield volcanoes, we examined major and trace element compositions of olivine-hosted melt inclusions from Hawaiian shield lavas, using electron microprobe and laser ablation inductively coupled plasma source mass spectrometry (see Methods). The data are presented in Supplementary Table 1. We selected lava samples from the submarine Hana ridge, Haleakala volcano (Maui), and the submarine exposures of the Makapuu stage, Koolau volcano (Oahu), because these two volcanoes are believed to represent the sampling of the two distinct compositions, having erupted above the Kea trend and Loa trend, respectively<sup>4</sup> (Fig. 1). Detailed sample information, including specific locality and whole rock geochemistry, has been presented elsewhere<sup>10–12</sup>.

Primary information about the mantle source composition beneath Hawaii can, in theory, be obtained from data on melt inclusions, provided the host olivines are derived exclusively from

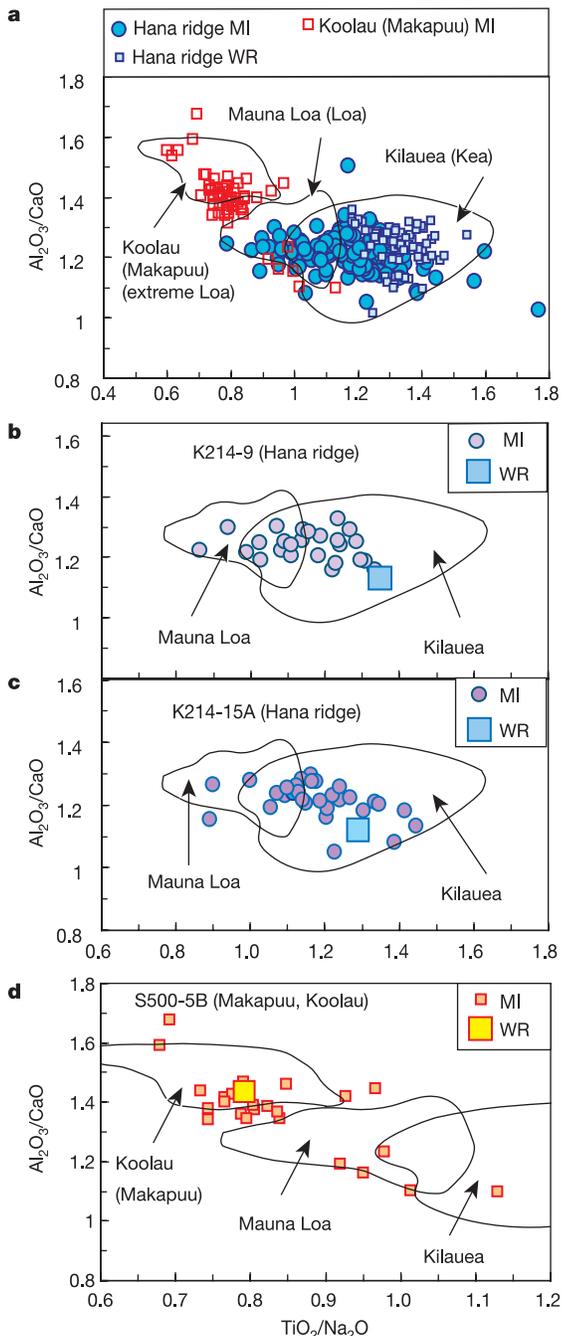
Hawaiian plume melts. All host olivines have CaO contents (0.16–0.29 wt%) that are typical of magmatic olivine and significantly higher than those of mantle (that is, xenocrystic) olivine<sup>10,13</sup>. Furthermore, we exclusively use major element ratios (such as Al<sub>2</sub>O<sub>3</sub>/CaO and TiO<sub>2</sub>/Na<sub>2</sub>O) and incompatible trace element ratios (such as Zr/Nb and Sr/Nb) in order to circumvent problems associated with olivine fractionation in the magmas, possible modification of inclusions from exchange with olivine during eruption, and re-equilibration between the host olivine and inclusion before analysis<sup>8,14</sup>.

Major element ratios (such as Al<sub>2</sub>O<sub>3</sub>/CaO and TiO<sub>2</sub>/Na<sub>2</sub>O), together with incompatible trace element ratios (such as Zr/Nb and Sr/Nb), of melt inclusion from submarine Hana ridge lavas overlap the fields for both Kilauea and Mauna Loa lavas (Figs 2a, 3a). Most major and trace element compositions of melt inclusions from the Makapuu stage of Koolau olivines are similar to data for whole rocks from Koolau (Makapuu stage, extreme Loa) lavas<sup>3,12</sup>, however, other melt inclusions extend well into the fields for Mauna Loa and Kilauea lavas (Figs 2d, 3d). Most importantly, melt inclusions from individual rock samples at both Hana ridge and Makapuu display not only significant heterogeneity, but also span the compositional fields for both Kilauea and Mauna Loa lavas (for example, K214-9, K214-15A, S500-5B, S500-1; Figs 2b–d, 3b–d). Multiple melt inclusions from individual olivine phenocrysts show much more limited variability than observed in the population from whole rocks.



**Figure 1 | Map of the Hawaiian islands.** Prominent volcanoes, and the Kea and Loa volcano trends, are shown. KI, Kilauea; ML, Mauna Loa; MK, Mauna Kea; H, Hualalai; KO, Kohala. Sample locations from the submarine Hana ridge, Haleakala volcano and the submarine Makapuu stage of Koolau volcano (on the Kea and Loa trends, respectively) are labelled with stars.

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**Figure 2** | Variation of major element ratio in bulk rocks compared to melt inclusions. **a**,  $\text{TiO}_2/\text{Na}_2\text{O}$  versus  $\text{Al}_2\text{O}_3/\text{CaO}$  for the melt inclusions from the submarine Hana ridge (Haleakala shield) and the submarine Makapuu stage of Koolau volcano span both Kea- and Loa-like compositions in single, shield-stage volcanoes (Haleakala and Koolau). Even melt inclusions from some individual samples (for example, K214-9 (**b**), K214-15A (**c**) and S500-5B (**d**)) overlap both Kilauea and Mauna Loa fields. Bulk rock data for Hana ridge and Koolau Makapuu are from refs 10 and 12, respectively. Fields for Kilauea and Mauna Loa whole rocks are from literature sources available at the GEOROC database (<http://georoc.mpch-mainz.gwdg.de>). The field for Koolau (Makapuu stage) lavas is from refs 3 and 12. Only whole rock data from fresh lavas ( $\text{K}_2\text{O}/\text{P}_2\text{O}_5 > 1$ ) are used for the fields. Data for submarine Hana ridge bulk lavas are from ref. 10. All whole rock and melt inclusion major element data have been normalized to 100% without volatiles and with all iron as FeO. MI, melt inclusion; WR, whole rock.

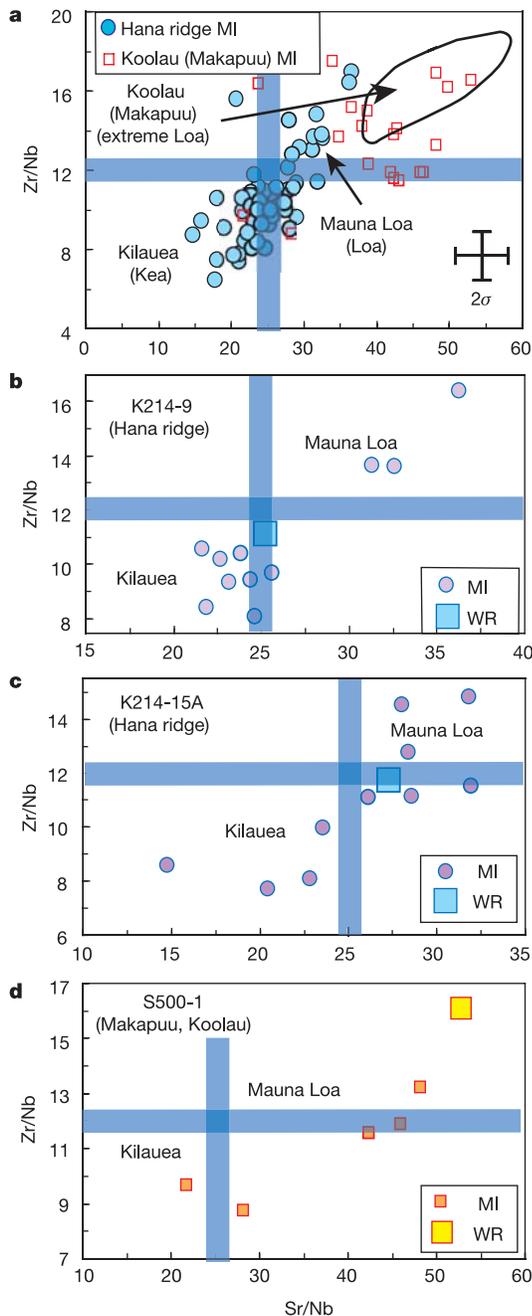
Potential processes that might cause the observed major and trace element ratio variations among the melt inclusions include the assimilation of ocean crust basalt by Hawaiian plume-derived magmas, or mixing of evolved magmas with primitive magmas. Ocean crust basalt typically has low Sm/Yb values as melting occurs extensively and at shallow depths, whereas Hawaiian shield lavas usually have higher Sm/Yb values because of deep melting of a garnet-bearing source. The melt inclusions have relatively constant Sm/Yb values that are high, much higher than those associated with ocean crust basalt (Fig. 4), making assimilation an unlikely scenario<sup>11</sup>. Mixing of primitive and evolved (for example, plagioclase-fractionated) magmas in the magma plumbing system could also cause variations in  $\text{Al}_2\text{O}_3$ , CaO and Sr, because of the affinity of these elements for plagioclase. However, Zr and Nb should not be affected as they are highly incompatible in both plagioclase and clinopyroxene. Melt inclusions reported here have  $\text{Al}_2\text{O}_3/\text{CaO}$  and Sr/Nb that are well correlated with Zr/Nb (not shown), indicating that mixing of primitive and evolved magmas is unlikely to have occurred.

Previous work has consistently demonstrated that trace element ratios, such as Zr/Nb and Sr/Nb, are correlated with radiogenic isotope ratios in Hawaiian shield lavas, and it is widely accepted that these trace element ratios reflect mantle source compositions<sup>2,3,15,16</sup>. Also some major element ratios, such as Na/Ti, vary systematically with isotope compositions<sup>17</sup>. Major element ratios ( $\text{Al}_2\text{O}_3/\text{CaO}$ ,  $\text{TiO}_2/\text{Na}_2\text{O}$ ) and incompatible trace element ratios (Zr/Nb, Sr/Nb) are correlated in our melt inclusion data, implying that these ratios reflect source geochemical characteristics and the variations, therefore, must reflect source heterogeneity. Therefore, in the same way that isotopic compositions (for example, Pb isotopic ratios) are used, these major and trace element ratios may be sufficient in distinguishing the Kea and Loa signatures of the Hawaiian shield lavas. The Kea and Loa geochemical trends among the Hawaiian shield volcanoes are commonly believed to reflect melting above a compositionally concentrically zoned<sup>4,5</sup> or compositionally left-right asymmetrically zoned<sup>18</sup> mantle plume. Here, we demonstrate that both Kea and Loa components co-exist in a single shield at both Haleakala and Koolau; therefore, a compositionally zoned mantle plume is inconsistent with our data. Overall, general geochemical differences exist in whole rocks between Kea- and Loa-trend volcanoes, but in detail these differences can also exist within a single shield<sup>19</sup>, and can vary relative to eruption age<sup>11,12,16,20</sup>, suggesting a more complicated structure for the Hawaiian mantle plume<sup>20,21</sup>. The presence of both Kea and Loa compositions in melt inclusions from individual shield volcanoes, and even in single lava samples, indicates that whole rocks may sample one mantle source component dominantly, but that the two (or more) source components are consistently represented to some extent, regardless of the specific geographic locations of the volcano (for example, above the 'Kea' zone).

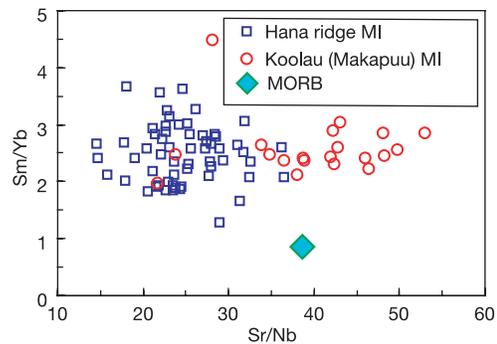
To best explain both the overall geochemical trends and the newer, more detailed whole rock and melt inclusion data, we propose a Hawaiian mantle plume characterized by more random heterogeneity than would be present in a compositionally zoned mantle plume. The compositional variations in melt inclusions in individual rock samples from Haleakala (Kea trend) and Koolau (Loa trend) suggest that the length-scale of chemical heterogeneity is remarkably smaller than that estimated on the basis of bulk rock geochemistry. The plume may be envisioned as having a matrix characterized by one composition (for example, Kea) with streaks or ribbons of another composition (for example, Loa) distributed throughout the entire plume. Experimental petrology shows that, for a given degree of partial melting, depleted peridotite generates melts having higher Ca/Al, lower  $\text{SiO}_2$  and higher FeO contents than does fertile peridotite<sup>22</sup>. Therefore, changes in major element compositions of melt inclusions from Kilauea- to Mauna Loa- to Koolau (Makapuu)-like probably reflect changes in the source available for melting (that is, from relatively depleted to relatively fertile peridotite). A final

implication is that melt compositions are 'fingerprinted' by the source and transported independently before final mixing and homogenization in the magma chambers of individual volcanoes<sup>23</sup>.

The geochemistry of Hawaiian lavas, particularly isotopically 'enriched' lavas from Koolau, has been explained by the presence of a recycled eclogite (or pyroxenite) component in the Hawaiian plume<sup>4,24</sup>. Partial melting at 2–3 GPa of eclogite with a composition like that of mid-ocean-ridge basalt produces silica-rich basaltic andesites low in FeO and with low CaO/Al<sub>2</sub>O<sub>3</sub> (refs 25, 26). Major



**Figure 3 | Variation of trace element ratios in bulk rocks compared to melt inclusions.** **a**, Melt inclusions from Hana ridge and Koolau Makapuu have Sr/Nb and Zr/Nb values that overlap those of Kilauea and Mauna Loa lavas. Melt inclusions from some individual samples (for example, K214-9 (**b**), K214-15A (**c**) and S500-1 (**d**)) overlap both the Kilauea field and Mauna Loa field. Delimitations for Kilauea and Mauna Loa are from ref. 2, and for Koolau (Makapuu stage) lavas are from refs 3 and 12. The  $2\sigma$  error bars are indicated in **a**.



**Figure 4 | Variation of trace element ratios in melt inclusions compared to oceanic crust basalt.** The wide range of Sr/Nb, and relatively constant and higher Sm/Yb, in the melt inclusions implies that their compositions are unlikely to have been derived from assimilation of ocean crust basalt by plume magmas. Data for ocean crust basalt are from ref. 33. MORB, mid-ocean-ridge basalt.

element compositions of melts derived from recycled eclogite would be expected to be associated with enriched isotopic signatures<sup>4</sup>. Lavas suspected of derivation from a source containing recycled oceanic gabbroic crust have Th/La and Th/Ba values less than the primitive mantle<sup>27</sup>, and excesses of Eu and Sr<sup>28</sup>. These characteristics are also present in melt inclusions examined in this study. Likewise, the presence of Sr-enriched, light rare earth element-depleted melt inclusions from Mauna Loa also suggests a recycled lower oceanic crustal component in the Hawaiian plume<sup>29</sup>. This recycled ancient oceanic crust may remain distinct geochemically<sup>4</sup>, forming streaks or ribbons that are deformed and stirred by stretching and folding as the plume rises through the mantle<sup>30</sup>.

The dominant component sampled at a given shield volcano may reflect melting processes related to the position of the volcano relative to the hot centre of the plume. As the volcano grows, it migrates away from the hot plume axis with plate motion. At higher temperatures, basaltic eclogite melts will react with peridotite, producing Kilauea-like melts with a composition higher in MgO and lower in SiO<sub>2</sub> than melts formed at lower temperatures<sup>31</sup>. In other words, in the plume core, higher temperatures are able to generate melts from the more refractory component (that is, depleted peridotite), but at plume margins, lower temperatures permit melting of only the more fertile material. Bulk lavas from some younger stages of the Hawaiian shields have enriched isotopic characteristics (for example, Mauna Loa, the Haleakala shield (Honomanu), and the Koolau shield Makapuu stage), suggesting that the proportion of fertile pyroxenite is higher than the proportion of depleted peridotite sampled by the melts. This is most easily explained if melting zones for lavas from the younger stages of Hawaiian shields are located farther from the hot mantle plume axis. Far from the plume's centre, temperatures would be lower, and the more fertile, isotopically enriched material (for example, eclogite) may be sampled to a greater extent.

## METHODS

**Homogenization of melt inclusions.** Olivine-hosted melt inclusions were subjected to experiments designed to homogenize them by melting and quenching, using a 1-atm gas mixing furnace following accepted procedures and conditions<sup>7</sup>. The oxygen fugacity was kept at the quartz–fayalite–magnetite (QFM) buffer. Olivines from single samples were loaded into Pt capsules that were gradually lowered, over 10 min, from the top of the furnace (100 °C) to the hottest place in the furnace (1,250 °C) and kept at that temperature for 10 min before quenching.

**EPMA and LA-ICP-MS analyses.** We measured major element compositions for 147 melt inclusions from 13 rock samples of submarine Hana ridge, Haleakala volcano and 63 melt inclusions from 3 rock samples of submarine Koolau volcanoes. Inclusions and host olivine were analysed by electron probe microanalysis (EPMA) with a JEOL-8800 instrument at the Tokyo Institute of

Technology, following procedures described elsewhere<sup>10</sup>. Each melt inclusion was measured using 3–20 points, the compositions of which were then averaged. Analytical uncertainty is 1–2% for major elements, and 5–10% for minor elements. We measured trace element compositions for 58 melt inclusions from 13 Hana ridge lavas and 21 melt inclusions from 3 Koolau lavas. Trace element abundances were measured using laser ablation inductively coupled plasma source mass spectrometry (LA-ICP-MS) coupled with a ThermoElemental PQ2 $\Omega$  instrument at the Tokyo Institute of Technology, following procedures described elsewhere<sup>32</sup>. The resulting precision, determined by analysing a standard as an unknown, is better than about 10% for most elements.

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**Supplementary Information** is linked to the online version of the paper at [www.nature.com/nature](http://www.nature.com/nature).

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