

# Lead isotopes reveal bilateral asymmetry and vertical continuity in the Hawaiian mantle plume

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The two parallel chains of Hawaiian volcanoes ('Loa' and 'Kea') are known to have statistically different but overlapping radiogenic isotope characteristics. This has been explained by a model of a concentrically zoned mantle plume, where the Kea chain preferentially samples a more peripheral portion of the plume. Using high-precision lead isotope data for both centrally and peripherally located volcanoes, we show here that the two trends have very little compositional overlap and instead reveal bilateral, non-concentric plume zones, probably derived from the plume source in the mantle. On a smaller scale, along the Kea chain, there are isotopic differences between the youngest lavas from the Mauna Kea and Kilauea volcanoes, but the 550-thousand-year-old Mauna Kea lavas are isotopically identical to Kilauea lavas, consistent with Mauna Kea's position relative to the plume, which was then similar to that of present-day Kilauea. We therefore conclude that narrow (less than 50 kilometres wide) compositional streaks, as well as the larger-scale bilateral zonation, are vertically continuous over tens to hundreds of kilometres within the plume.

Much of the worldwide isotopic heterogeneity in basalts from ocean island volcanoes, thought to be derived from mantle plumes, can be found in basalts derived from a single plume. Such is the case for the basalts from the Hawaiian islands. Recent Hawaiian volcanoes form a double chain widely referred to as the Loa and Kea trends, named after their largest volcanoes — Mauna Loa and Mauna Kea<sup>1–3</sup>. It has long been known that systematic differences in Pb, Sr and Nd isotopic compositions exist between these two chains, but the published isotope data show large areas of overlap (see GEOROC (Geochemistry of Rocks of the Oceans and Continents) database <http://georoc.mpch-mainz.gwdg.de> and references therein), and the significance of these two trends remains unclear<sup>4,5</sup>. One of the objectives of the Hawaii Scientific Drilling Project (HSDP) was to test the idea that the plume's isotopic and chemical composition is concentrically zoned as a result of the entrainment of external mantle material during plume ascent<sup>6–9</sup>. Such entrainment might help to explain the compositional differences between volcanoes from the Kea and Loa chains. Phase 2 of HSDP drilling reached a depth of ~3 km and samples approximately 0.3 million years (Myr) of plume history<sup>10</sup>, corresponding to about 40 km of the horizontal traverse of Mauna Kea across the plume.

Recent HSDP-2 results have led to contrasting views of the geometry of the compositional heterogeneities across the plume conduit. Eisele *et al.*<sup>11</sup> interpreted the three distinct Pb isotope arrays (labelled Kea-lo8, Kea-mid8 and Kea-hi8) as reflecting vertical streaks at least several tens of kilometres in length within the Hawaiian plume stem. This interpretation implies that the volcano samples successive vertical streaks within the plume as it is carried across the plume by the Pacific plate. The alternative model<sup>12</sup> suggests that these heterogeneities are distributed vertically as a stack of 'pancakes', which are sampled consecutively by the volcano stratigraphy. Both vertical and horizontal variability are likely, but dynamic modelling suggests a dominance of vertical stretching during plume ascent<sup>13</sup>.

To address both questions—that of the larger-scale, possibly concentric plume zonation expressed by the Loa and Kea chains, and the significance of the smaller-scale variability observed within

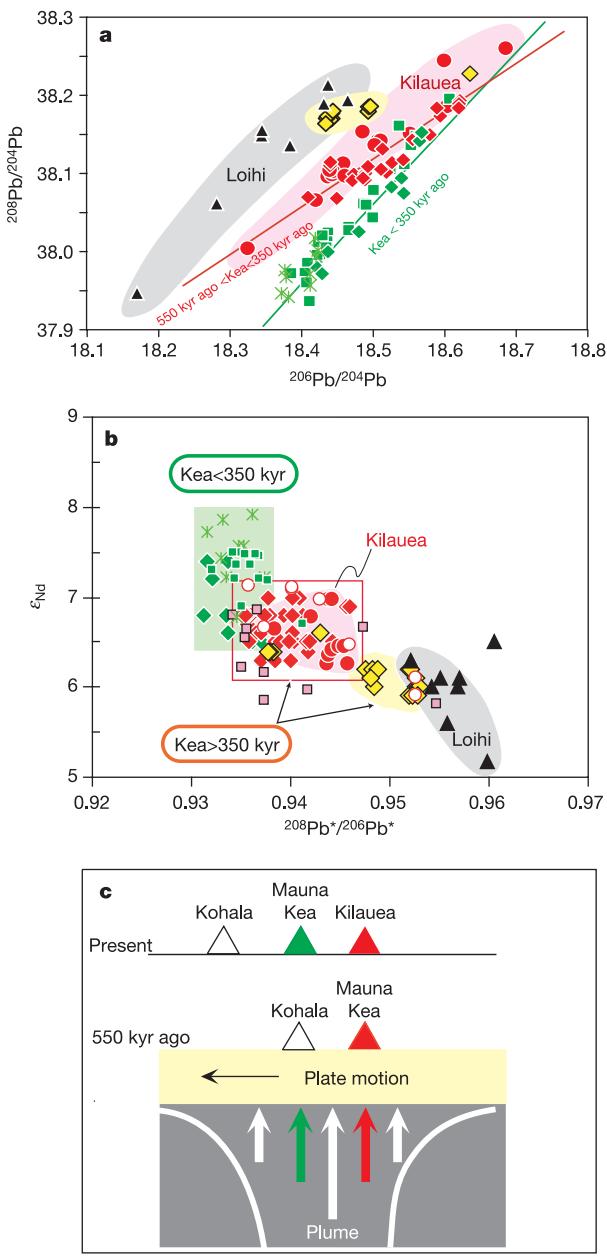
a single Kea volcano (HSDP)—we report here high-precision Pb isotope ratios from nine recent Hawaiian volcanoes belonging to the Loa (Loihi, Mauna Loa, Lanai, Kahoolawe, Koolau) and Kea (Kilauea, Mauna Kea, Kohala) chains using a Pb triple-spike technique<sup>14</sup>. The sampling of these volcanoes includes early, pre-shield stage (Loihi), main-phase shield building (Kilauea, Kohala, Mauna Kea, Mauna Loa, Lanai, Kahoolawe, Koolau) and late-stage, tholeiitic to alkaline lavas (Mauna Kea) (Supplementary Table 1).

On the basis of these data, we demonstrate geographic and temporal isotopic variations in several Hawaiian volcanoes which indicate that the heterogeneities within the plume resemble vertically drawn-out 'streaks' rather than flat pancakes. We further show that the Hawaiian plume is compositionally not purely concentric. Rather, there exists a lateral chemical asymmetry, as expressed in the Loa–Kea chains. We discuss two scales of plume heterogeneity, beginning with local variations within the Kea chain.

## Small-scale heterogeneity in the plume

Our data have a bearing on the origin of short-term heterogeneities observed within the stratigraphic section of Mauna Kea<sup>11,15</sup>, which reveals small-scale features of the Hawaiian plume anatomy. In particular, the Pb isotopic composition of the submarine section of Mauna Kea is nearly identical to that of present-day Kilauea. Figure 1a shows a Pb isotopic comparison of different portions of the HSDP-1<sup>15</sup> and HSDP-2<sup>11</sup> drill cores in Mauna Kea with those of recent lavas from Kilauea (red circles)—including Mauna Ulu and Hilina lavas—as well as Loihi (black triangles).

Among Mauna Kea lavas, there is a temporal decrease in  $^{208}\text{Pb}/^{204}\text{Pb}$  at a given  $^{206}\text{Pb}/^{204}\text{Pb}$  ratio (Fig. 1a). That is, the youngest (<350 kyr) Mauna Kea lavas, the uppermost 800 m of the HSDP drill core (green diamonds and squares) and subaerially exposed postshield lavas (green crosses) have the lowest  $^{208}\text{Pb}/^{204}\text{Pb}$ , whereas older (350–550 kyr) Mauna Kea lavas, 800–3,000 m within the HSDP core, have higher  $^{208}\text{Pb}/^{204}\text{Pb}$  (red and yellow diamonds)<sup>11</sup>. The upper part of the HSDP core (green symbols) contains Pb isotope signatures that are quite distinct from those of recent Kilauea lavas (red circles). By contrast, the lower HSDP



**Figure 1** Small-scale heterogeneity in the Hawaiian plume revealed by the HSDP core, Mauna Kea. **a**,  $^{208}\text{Pb}/^{204}\text{Pb}$  versus  $^{206}\text{Pb}/^{204}\text{Pb}$  in HSDP-2 (ref. 11) (diamonds) and HSDP-1 lavas<sup>15</sup> (green squares). Also plotted are Kilauea (red circles), Loihi (black triangles) and Mauna Kea post-shield lavas (green crosses). The HSDP-2 data are subdivided following Eisele *et al.*<sup>11</sup> into ‘Kea-hi8’ (yellow diamonds), ‘Kea-mid8’ (red diamonds) corresponding to samples with ages 350–550 kyr, and ‘Kea-lo8’ (green diamonds) with ages younger than 350 kyr. Most of the 350–550-kyr-old lavas fall in the field of Kilauea, with the exception of the Kea-hi8 samples, which have Pb isotopic compositions trending towards those of Loihi. Error bars on Pb isotope ratios are similar to the symbol sizes (2 s.d. = 100 p.p.m.). **b**,  $\varepsilon_{\text{Nd}}$  versus  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  ratios in HSDP-2 lavas. Same symbols as in **a**. Pb isotope data are from refs 11 and 15, and Nd isotope data from ref. 18, except for the red open circles, which correspond to HSDP-2 Nd data from this work. Comparison between HSDP-2 and Kilauea data obtained in our laboratory confirm the similarities between 350–550-kyr-old Kea samples and present-day Kilauea lavas. Like Loihi lavas, the Kea-hi8 samples (yellow diamonds) display high  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios and low  $\varepsilon_{\text{Nd}}$ . Pb and Nd isotope data on submarine Mauna Kea glasses (pink squares) are from ref. 21. **c**, Schematic illustration of the position of Mauna Kea relative to the plume 550 kyr ago, that is, the same location above the plume occupied by Kilauea today. The red arrow illustrates how similar plume sources are sampled by the two volcanoes at different times.

core (red diamonds) mostly comprises Kea-mid8 compositions<sup>11</sup> that are indistinguishable in Pb isotope space from young Kilauea lavas. The only exceptions are the apparently anomalous, Loihi-like lavas from very restricted stratigraphic levels in the vicinity of 1,800–2,100 m and 2,200–2,500 m depths in the HSDP core (yellow diamonds, Fig. 1a). These lavas share the low SiO<sub>2</sub> content that is typical of Loihi lavas<sup>16</sup>, as well as other Loihi characteristics, such as lower  $\varepsilon_{\text{Nd}}$  values and high  $^3\text{He}/^4\text{He}$  ratios<sup>17</sup>. These exceptional compositions are enigmatic at present, and we can only speculate that they represent a local heterogeneity within the plume resembling Loihi sources in several respects. The recurrence of these lavas as intercalated flows in the deeper sections of the core suggests that this heterogeneity must have been a short-term feature of magma production during the main shield-building stage of Mauna Kea. We emphasize that, except for these anomalous lavas, the remainder of the 800–3,000 m section of Mauna Kea, with estimated eruption ages of 350–550 kyr, are indistinguishable from recent Kilauea lavas in  $^{207}\text{Pb}/^{204}\text{Pb}$ ,  $^{208}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$  space.

Strontium and Nd isotope data do not provide the same level of volcano-specific resolution. Strontium isotope ratios in Mauna Kea, Kilauea and Loihi lavas are almost indistinguishable, ranging from 0.7034 to 0.7036 (see the GEOROC database). In contrast, published Nd isotope data and those reported here (Supplementary Table 1) indicate that Kilauea lavas have, on average, lower  $\varepsilon_{\text{Nd}}$  values (+6 to +7) than those measured in HSDP-1 (ref. 8) (mean =  $7.19 \pm 0.57$ ,  $n=26$ ). HSDP-2 Mauna Kea lavas show a range of  $\varepsilon_{\text{Nd}}$  between 7.4 and 5.9 with an average of  $6.6 \pm 0.69$  ( $n=96$ )<sup>18</sup>, lower than that reported for HSDP-1 (ref. 8). We also measured Nd isotopic compositions in seven HSDP-2 samples to compare these directly with Nd data on Kilauea lavas obtained in our laboratory. In  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ – $\varepsilon_{\text{Nd}}$  space (Fig. 1b)—where  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  represents the time-integrated  $^{232}\text{Th}/^{238}\text{U}$  ratio since the formation of the Earth and is defined as:  $^{208}\text{Pb}^*/^{206}\text{Pb}^* = (^{208}\text{Pb}/^{204}\text{Pb} - 29.475)/(^{206}\text{Pb}/^{204}\text{Pb} - 9.306)$  (ref. 19)—the 350–550-kyr-old samples also fall in the field defined by Kilauea lavas. Thus, Nd isotope data, including our own, provide additional evidence for the similarities in sources sampled by 350–550-kyr-old Mauna Kea and present-day Kilauea. Submarine Mauna Kea glasses have also been shown to have radiogenic isotopic characteristics and trace-element ratios similar to those of Kilauea and Loihi lavas<sup>20,21</sup> (Fig. 1b), and they are consistent with these observations.

The possibility that the 350–550-kyr-old samples are actually intercalated Kilauea lava flows can be ruled out, because the lowermost HSDP-2 samples are much older than the oldest-known Kilauea lavas (Hilina section dated 200–300 kyr ago (ref. 22); East Rift Zone, 350 kyr ago) (ref. 23). Moreover, recent data indicate that volcanism at Kilauea was in the early alkalic phase 220–275 kyr ago (ref. 24). The most straightforward explanation for the isotopic similarities is that the Kilauea-like lavas found in the HSDP-2 core are derived from Kilauea-type sources within the Hawaiian plume. The overall resemblance between young Kilauea and old Mauna Kea lavas can therefore be reasonably interpreted to result from the position of Mauna Kea relative to the upwelling plume centre. Reconstruction of the palaeo-distance of Mauna Kea from a hypothetical plume centre<sup>11,25</sup> indicates a movement of the volcano by about 40 km during the past 550 kyr (from ~20 km 550 kyr ago to ~60 km at present), implying that Mauna Kea occupied almost the same relative spot then as Kilauea does now. This reconstruction provides a natural explanation for the isotopic similarities shared by the two volcanoes (Fig. 1a, c), and implies that the surface expression of the Hawaiian plume reflects precisely the position of the volcano relative to the plume axis at a given evolutionary stage of the volcano.

If old Mauna Kea sources are identical to recent Kilauea sources, we can use this information to estimate the vertical extent of this particular isotopic fingerprint within the plume, assuming we know something about ascent rates in the plume conduit. These rates have been estimated by a variety of methods<sup>11,26</sup> that suggest an extreme

minimum given by the velocity of the Pacific plate of about  $10 \text{ cm yr}^{-1}$  and a conservative maximum of about  $1 \text{ myr}^{-1}$  near the centre of the plume<sup>27–29</sup>. Using these upwelling velocities, the vertical distance travelled by plume material over  $\sim 0.5 \text{ Myr}$  would be  $50\text{--}500 \text{ km}$ , implying that the minimum vertical extent of the specific ‘Kilauea-like’ signature common to Kilauea and Mauna Kea lies somewhere between 50 and 500 km. These considerations suggest that the Hawaiian plume contains relatively narrow compositional streaks (‘spaghetti’) that are drawn out by laminar flow in the conduit over a significant part of the vertical extent of the plume<sup>13</sup>. The sampling of different plume streaks by a volcano as it moves across the plume is illustrated in Fig. 1c. Because the characteristic isotopic signature of each streak is a linear array in Pb isotope space, each streak can be envisioned as consisting of two isotopically distinct endmember components, mixing in variable proportions during melting to produce a linear array<sup>11,15</sup>. The fluctuations along these linear arrays reflect an additional, still smaller scale of isotopic variations, corresponding to a dominant frequency of about 10,000 yr (ref. 11) and a spatial scale of 5–50 km, depending on the plume ascent rate, but this feature is beyond the scope of this paper. The occurrence of several Pb isotope arrays during the lifetime of Mauna Kea reflects sampling of isotopically distinct streaks as the volcano moves laterally over the plume owing to plate motion (Supplementary Fig. S2).

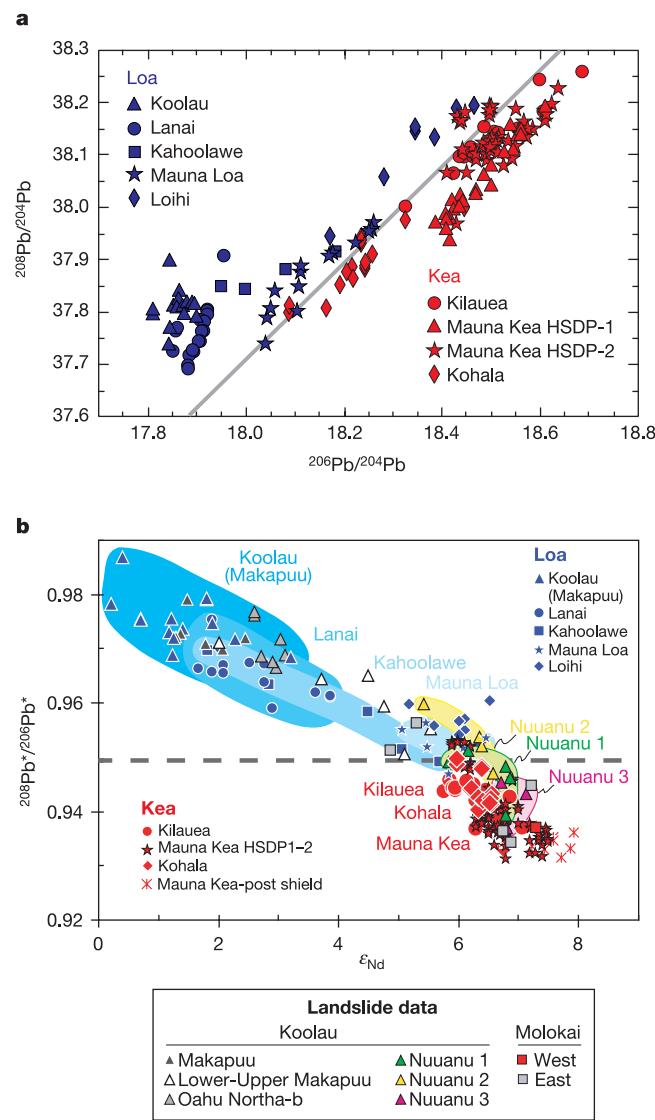
### Large-scale heterogeneity of the plume

A much larger-scale heterogeneity of the Hawaiian plume is revealed by the isotope data shown in Fig. 2a. The striking systematic differences between Loa (blue symbols) and Kea (red symbols) volcanoes demonstrate a clear left-right compositional division within the Hawaiian plume. In  $^{208}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$  space (Fig. 2a), most of the data from individual volcanoes define distinct linear Pb isotopic arrays, resolved outside the analytical error ( $\sim 100 \text{ p.p.m.}$ ), indicating that there exists a significant heterogeneity in the components present within each of the two trends. The high  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios of Loa volcanoes in general, and Koolau and Lanai in particular, indicate the presence of an EM-1 component<sup>30</sup>, the nature of which is discussed in the literature<sup>31</sup>. Here we note that these data rule out possible interpretations whereby Hawaiian melts represent simple two-component mixtures<sup>32</sup>. This is supported by both the HSDP-2 results, which, even when taken alone, indicate the presence of four distinct Pb components<sup>11</sup>, and the distinct slopes of the individual Hawaiian Pb isotope arrays (Supplementary Table S2 and Fig. S1).

We performed a statistical analysis of the Pb isotope data (Supplementary Table S3) demonstrating that the populations of the Kea and Loa trends are significantly different from the pooled population at a significance level higher than 99.9% in  $^{208}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$  space. The regression analyses of the Pb isotope data for individual volcanoes are reported in Supplementary Table S2 and Fig. S1.

The Pb isotopic compositions of the two trends are clearly distinct, with overlap restricted to the anomalous samples from the submarine section of the HSDP-2 drill core (Kea-hi8, yellow diamonds in Fig. 1). However, in the absence of drill holes other than HSDP, sampling is necessarily limited. Nevertheless, the Loa–Kea differences exist regardless of the location of the volcanoes with respect to the plume centre. Our data for Loa trend volcanoes include early (Loihi), main- and late-shield (Mauna Loa) volcanism. Data for Kea volcanoes include main-shield (central location; old Mauna Kea and recent Kilauea) and late-shield (peripheral location; youngest Mauna Kea and Kohala) volcanism. We therefore argue that these data rule out previous interpretations in which the distinction between the Loa and Kea chains has been ascribed to the more central position of the Loa chain relative to the plume centre, and a more peripheral track of the Kea chain volcanoes.

In Fig. 2b, the data are plotted in  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ – $\epsilon_{\text{Nd}}$  space, together with recently published Pb isotope data on Koolau landslide blocks<sup>33</sup>, which are of similar precision to those using the Pb triple-spike technique, the  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  ratio being used as a distinction criterion between the Loa trend and Kea trend. Despite the fact that the data form a broad negative correlation, the boundary between Loa (blue) and Kea (red) trends can be pinned down at a  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  value of about 0.95, which is similar to that inferred from the slope of the Loa–Kea boundary in  $^{208}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$  space (Fig. 2a). Some overlap does occur across the  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  boundary—in particular, the Kea-hi8



**Figure 2** The Loa–Kea compositional division of the Hawaiian plume. **a**, Triple-spike Pb isotope data of Hawaiian shield stage lavas (except Loihi, which are early-shield stage) plotted in  $^{208}\text{Pb}/^{204}\text{Pb}$  versus  $^{206}\text{Pb}/^{204}\text{Pb}$  space. The grey line marks the boundary between Kea and Loa volcanoes, with higher  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios for Loa relative to Kea volcanoes at a given  $^{206}\text{Pb}/^{204}\text{Pb}$  ratio. The only exceptions are a few lava flows from deeper levels in the Mauna Kea HSDP-2 core (red stars) which have Loihi-like features. Error bars ( $2\text{s.d.} = 100 \text{ p.p.m.}$ ) on Pb isotope ratios are similar to symbol size. **b**,  $\epsilon_{\text{Nd}}$  versus  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  ratios in Hawaiian volcanoes. Also plotted are the data for submarine landslide blocks from Koolau and Molokai<sup>33</sup>. The grey dashed horizontal line marks the  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  boundary value between Loa- (blue) and Kea- (red) trend volcanoes. Notable exceptions include Kea-hi8 lavas (red stars on Loa side) and Nuuanu 3 (Koolau) landslide blocks, which partly fall on the Kea side.

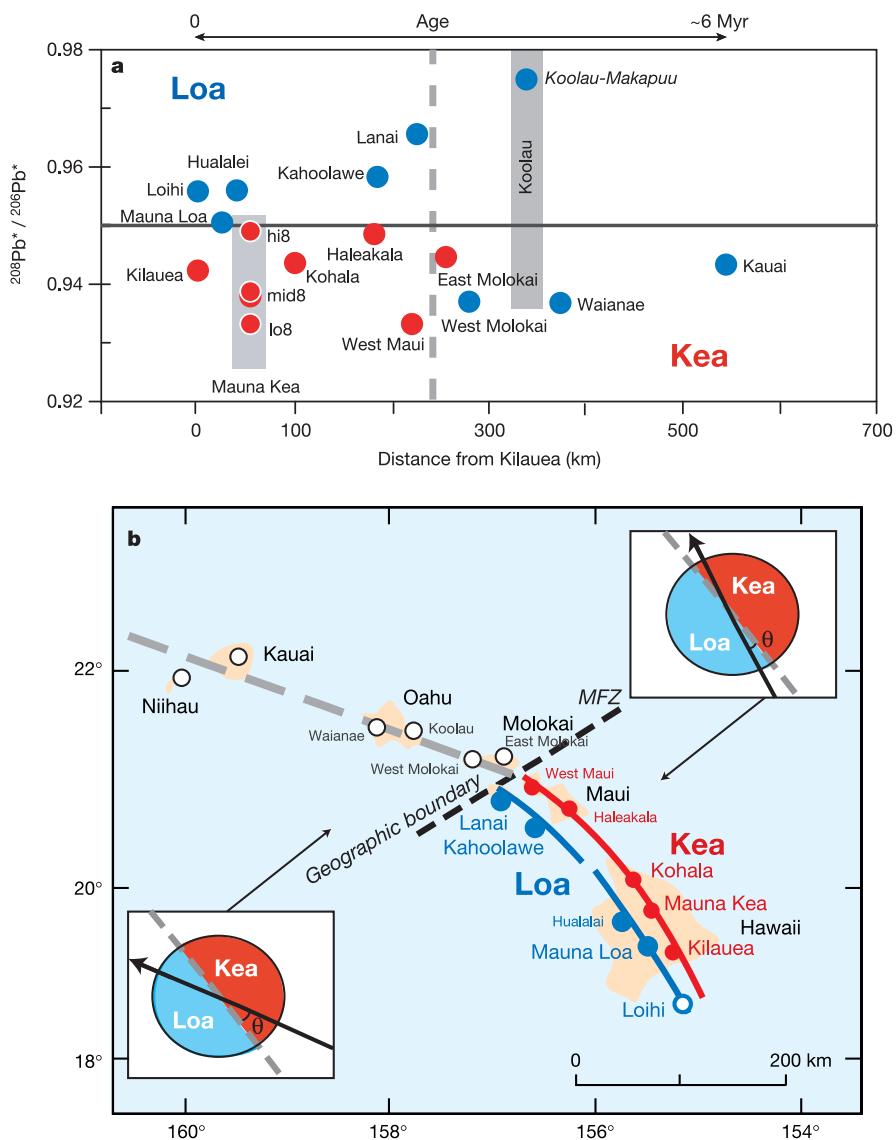
samples from Mauna Kea plot on the Loa side and the Koolau landslide blocks partly fall on the Kea side of the boundary.

The greater Pb isotope compositional variability within the Loa trend (a factor of  $\sim 3$ ) compared with that observed along the Kea trend (Fig. 2b) suggests larger source heterogeneities within the left (Loa) side compared with the right (Kea) side of the Hawaiian plume. The progressive increase in  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  ratios and decrease in  $\varepsilon_{\text{Nd}}$  along the Loa chain with increasing volcano age further imply that sampling of the Loa heterogeneities is time-dependent. This is also reflected in the isotopic variations found in Koolau lavas (subaerial and submarine landslide blocks), which span the entire range found in Hawaiian volcanoes<sup>33</sup>. There also appears to be a temporal evolution in Koolau Volcano from ‘old’, Kea-like to ‘young’, Loa-like isotope characteristics, although the

significance of this feature relies upon the validity of the reconstruction of the landslides, their origin and especially the relative ages between and within the Nuuanu landslide blocks (see ref. 33 and references therein). Recent Pb isotope data (our unpublished work) on Koolau Scientific Drilling Project (KSDP) lavas still show a consistent ‘Loa’ signature in the deeper stratigraphic levels of Koolau Volcano<sup>34</sup>, suggesting that caution should be exercised in interpreting the landslide blocks data.

### Timing and location of the Loa–Kea boundary

Using the  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  ratio as a distinction criterion between the Loa and Kea trends and considering the geographic distribution of this parameter (Fig. 3a), we show that the extension of the Loa and Kea trends terminates at the Molokai fracture zone. In particular,



**Figure 3** Geographic extension of the Loa–Kea compositional boundary. **a**,  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  mean values versus distance from Kilauea for Hawaiian volcanoes. The upper x axis gives a corresponding timescale. Note that the Koolau data, shown as a grey shaded box<sup>33</sup>, span the entire range of  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  displayed by Hawaiian volcanoes. The blue circle labelled ‘Koolau’ corresponds to the mean  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  of subaerial Makapuu stage lavas. The Loa–Kea distinction is disturbed beyond Lanai, as shown by the vertical dashed line. West Molokai, Waianae, Koolau (main-shield and landslide blocks) and Kauai, generally referred to as Loa chain volcanoes, have Kea-type  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  ratios. Data are from this study and the GEOROC database. **b**, Map of the Hawaiian islands showing

that the distinct Loa–Kea chains, based on the spatial distribution of the  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  ratio, terminate at the Molokai fracture zone, shown by the dashed black line. Beyond this geographic boundary, a single line passes through the volcanoes (black open circles) indicating that the Loa–Kea subdivision does not hold. The insets are schematic drawings illustrating the change in angle between the plate motion direction (solid black arrow) and the compositional Loa–Kea boundary (dashed grey line). The lower left inset corresponds to the case of Koolau volcano (2–3 Myr ago) and the upper right inset to the modern situation.

some of the main shield-stage lavas from Oahu (Koolau and Waianae) and Kauai volcanoes do not have the high  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  ratios typical of Loa trend volcanoes (Fig. 3a), but rather have Kea-type features. We thus propose revising the geographic location of the Loa and Kea trend lines on the basis of the Loa–Kea compositional Pb isotopic boundary, which we infer does not extend beyond Lanai and Haleakala (Fig. 3b). Although this may appear to be at odds with the age-progression map<sup>35</sup>, the coincidence of the compositional Pb isotopic transition with the bend of the Hawaiian islands chain near Molokai may be significant.

Geometric relocation of hotspots in the Pacific indicates that the Loa–Kea chains may be a recent feature that developed only about 2–3 Myr ago<sup>36</sup>. This feature may have resulted from a change in the plate motion, as indicated by a bend of the Hawaiian chain near Molokai, and the effects of lithospheric flexure due to a volcanic load placed off-axis from the hotspot<sup>37</sup>. The geographic location and the timing of the compositional Pb isotopic change agree well with these models, once the age uncertainties have been taken into account. Thus, the sampling of two compositionally different sides of the Hawaiian plume, with the left (Loa) side characterized by high  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$  and the right (Kea) side by low  $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ , reflects the asymmetry of the Hawaiian plume. Such an asymmetry can be perpetuated over long distances and timescales by a dual line of volcanoes—with volcano spacing dependent upon lithospheric thickness<sup>38</sup>—developing in response to changes in the direction of plate motion<sup>37</sup>.

An explanation is required, however, for the isotopic transition of Koolau from a Kea-type signature (in the early shield-building stage) to a Loa-type signature (in the late shield-building stage)<sup>33</sup>. Provided that the reconstruction and relative ages of the landslide blocks are valid<sup>33</sup>, this transition can be explained if the azimuth of the compositional boundary between Loa and Kea-type plume chemistry is at a slight angle to the azimuth of the plate motion. Figure 3b illustrates how this may determine which side of the compositional boundary is sampled during a given evolutionary stage of a volcano. In the case of Koolau (left inset), the switch from Kea- to Loa-like chemistry implies that the angle between the vectors defined by the Loa–Kea compositional boundary and the plate motion was greater than the present-day angle (right inset). Such an explanation is reasonable given the uncertainties over the velocity of the Pacific plate relative to the plume and the direction of plate motion over the past 2 Myr (ref. 25). It is also quite possible that the change in the direction of plate motion 2–3 Myr ago may also have caused a change in the position of the Koolau volcano in such a way that its magma supply switched from one side of the plume to the other.

## Bilateral asymmetry of the plume

The overall consistency of the Pb isotopic signature shown by the Loa and Kea chains, together with small-scale heterogeneities evident in the stratigraphic section of both Mauna Kea and Koolau, rules out models ascribing the compositional variations to purely concentric zoning of the plume<sup>6–9,39</sup>. Instead, we infer that the plume possesses a gross lateral zonation (Supplementary Fig. S2). This model is consistent with the numerical simulations of plume evolution of Farnetani *et al.*<sup>13</sup>, showing that plumes draw compositional heterogeneities from a low-viscosity source layer into the plume stem by laminar flow, which should preserve any incidental, lateral, initial heterogeneities of the source layer. This source layer (perhaps located above the core–mantle boundary) is inferred to contain large-scale heterogeneities extending to perhaps hundreds of kilometres<sup>40</sup>, which are drawn into, compressed and vertically stretched in the plume stem, and ultimately sampled by the two parallel trends on the Hawaiian islands. It is important, however, to emphasize that the lateral variations in the Hawaiian plume Pb isotope chemistry represent first-order observations and are thus independent of plume structure models. Rather, our data

demonstrate the existence of different scales of heterogeneities which, assuming a plate velocity of  $10 \text{ cm yr}^{-1}$  and upwelling velocities of  $1 \text{ m yr}^{-1}$ , can be translated into length scales varying from tens of kilometres for the short-term (10 kyr) oscillations in HSDP-2 core to  $\sim 50 \text{ km}$  (Loa–Kea difference). Consequently, these results are inconsistent with a purely concentric zonation of the Hawaiian plume, as inferred from the He (and Nd) isotope distribution in the context of the dynamical plume structure model of refs 25 and 41. Recent three-dimensional numerical results show that radial variations in vertical velocity and induced shear stress across plume tails can generate long and distinct filaments leading to laterally heterogeneous rather than concentrically zoned plume tails<sup>42</sup>. In addition, fluid-dynamical tank experiments demonstrate that any left–right geochemical asymmetry in the plume source's region will be preserved and reflected in the geochemistry of the erupted lavas<sup>43</sup>. Altogether, these results provide strong support for our conclusion, which is essentially based on the Pb isotope anatomy of the Hawaiian plume.

Entrainment of mantle material overlying the plume-source layer<sup>26</sup> should lead to a concentric zonation, but this pattern is not reflected in our Pb isotope data. However, our sampling does not as yet include very-late-stage, post-erosional lavas that might be derived from the outermost parts of the plume and might therefore represent entrained ‘ambient’ mantle material. Therefore, we cannot firmly rule out the presence of entrained material in the outermost parts of the plume, but we can exclude concentric entrainment as the prime mechanism for explaining the occurrence of the Loa and Kea chains.

High-precision Pb isotope data on the scale of the Hawaiian islands chain demonstrate a large-scale left–right asymmetry in the Hawaiian plume. This asymmetry is reflected in the spatial distribution of the volcanoes along the Loa–Kea chain. Smaller-scale heterogeneities, revealed in the temporal records of Mauna Kea and Koolau lavas, also exist in the Hawaiian plume. These heterogeneities appear to be stretched out vertically within the plume and are sufficiently long-lived to be sampled by successive volcanoes. To progress beyond these general conclusions and develop a more detailed model of the plume anatomy, new high-resolution dynamic modelling will have to be carried out, incorporating the various scales of isotopic plume heterogeneity. □

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