

An isotopically depleted lower mantle component is intrinsic to the Hawaiian mantle plume

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Most ocean island basalts sample an isotopically depleted mantle component, but the origin of this component is unclear. It may come from either the entrained upper mantle or from a reservoir intrinsic to the plume, sourced from the lower mantle. For Hawaii, the isotopically depleted component is primarily sampled during the secondary rejuvenated-stage volcanism, 0.5-2 million years after the initial shield-stage volcanism. However, it is also inferred in shield and post-shield lavas. We analyse the radiogenic isotopic and trace element compositions of a suite of Mauna Kea shield-stage tholeiites, and found that they have the same isotopic compositions as rejuvenated-stage lavas. We use trace element models to show that these shield-stage basalts can be explained as higher degree partial melts of a rejuvenated-stage source. Our data, therefore, show that the depleted rejuvenated-stage component was directly sampled during shield-stage volcanism. The common source for both shield-stage and secondary rejuvenated volcanism implies that the depleted rejuvenated component is intrinsic to the Hawaiian mantle plume. It is further inferred that the mantle region from which the Hawaiian plume originates, probably in the lower mantle, is also isotopically depleted, similar but not identical to the upper mantle.

he compositions of ocean island basalts (OIBs) and mid-ocean ridge basalts (MORBs) reflect the compositional evolution of the Earth's interior. MORBs tend to have lower 87Sr/86Sr and higher ¹⁴³Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf in comparison to OIBs. MORBs sample the upper mantle, which is 'depleted' in highly incompatible trace elements. OIBs, such as those from Hawaii, Iceland, Samoa, Galapagos and Kerguelen, have higher 87Sr/86Sr and lower 143Nd/144Nd and ¹⁷⁶Hf/¹⁷⁷Hf, which are described as isotopically 'enriched'. Pb isotope ratios (206,207,208Pb/204Pb) are more complicated; although many OIB values overlap with MORB values, OIBs range to more radiogenic Pb isotopes. Some OIBs are produced by mantle plumes from the lower mantle²⁻⁵ and provide a rare opportunity to investigate the composition of the lower mantle and the long-term geochemical differentiation of the Earth. Although the majority of OIBs have enriched isotopic and elemental characteristics relative to MORBs, many OIBs contain a distinct isotopically depleted component with low 87Sr/86Sr and high 143Nd/144Nd and 176Hf/177Hf, for example, Hawaii⁶⁻¹³, Iceland¹⁴⁻¹⁶, Kerguelen^{17,18}, Samoa^{19,20} and Galapagos²¹⁻²³.

Isotopically depleted components in OIBs

The origin of the isotopically depleted components in hotspots globally is contentious, but critical to understand the long-term evolution of the Earth's mantle. There are three main hypotheses: (1) the depleted source component is intrinsic to the plume⁹⁻¹³, (2) entrained upper mantle⁶⁻⁸ or (3) metasomatized lithospheric mantle^{24,25}. These hypotheses can be distinguished by their Nd and Hf isotope ratios, because depleted plume components are expected to have distinct Nd-Hf isotopic signatures from the upper mantle sampled by MORBs²⁶. All three models were suggested for Galapagos and Iceland 14-16,21-23,27,28, where the depleted components have been attributed to either the local lithospheric and upper mantle contributions or an intrinsic plume component. At Ninetyeast Ridge (Kerguelen) the depleted component is isotopically and chemically distinct from the Indian MORB, and probably intrinsic to the plume¹⁸, whereas at Samoa the depleted component is thought to be entrained depleted mantle (DM)^{20,29}. Understanding the isotopically depleted end members of hotspots is critical to understanding the long-term evolution of the Earth's mantle.

The origin of the isotopically depleted component at Hawaii is still under debate. At Hawaii, isotopically depleted lavas commonly erupt during rejuvenated-stage volcanism, 0.5–2 Myr after the end of the isotopically enriched shield-stage volcanism^{30–33}. Rejuvenated-stage volcanism occurs on the islands of Kaula, Kauai, Niihau, Oahu, Molokai and Maui (Fig. 1). Lavas erupted off-axis of the plume, called the North and South Arch Volcanic Field, are also grouped with rejuvenated-stage lavas because they have similar radiogenic isotope ratios and trace element abundances. The cause of these eruptions is probably flexural arching of the lithosphere caused by the building of Hawaiian volcanoes^{34,35}. The mantle source of Hawaiian rejuvenated-stage lavas may be the Pacific MORB source⁶, local lithospheric mantle⁷ and also an isotopically depleted intrinsic plume component^{8–13,36–39}.

The isotopically depleted rejuvenated-stage components were also inferred in lavas from the pre-shield stage of Loihi¹³, the shield stage of Kauai^{13,36} and the post-shield stages of Hualaiai³³ and Mahukona^{33,37}. However, in these cases, it is debatable whether the depleted component sampled by these shield lavas is DM or a depleted plume component. For example, some Kauai shield lavas form a positive 206Pb/204Pb versus 87Sr/86Sr trend, which points to the rejuvenated-stage lava field or MORB field (see Fig. 8 of Mukhopadhyay et al.¹³). However, Kauai shield lavas still have isotope ratios within the shield-stage range. Lavas from the Hana Volcanics of Haleakala have isotopic signatures similar to those of rejuvenated-stage lavas. However, it is debatable whether the Hana Volcanics belongs to post-shield-stage volcanism or rejuvenatedstage volcanism40. Nevertheless, post-shield and pre-shield volcanism sample the edge of the plume; consequently, it is difficult to unambiguously assess whether the depleted components sampled by the above-mentioned lavas are intrinsic to the Hawaiian plume.

The Emperor Seamounts, which were also created by the Hawaiian plume, contain an isotopically depleted component, similar to that sampled by Hawaiian rejuvenated-stage lavas^{8,10}. Owing to

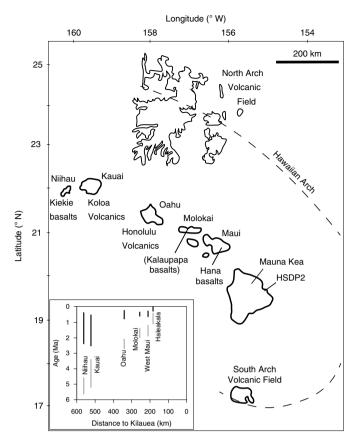


Fig. 1 | Map of the Hawaiian Islands and the Hawaiian Arch. Inset, Age and duration of rejuvenated-stage volcanism (black bars) and shield-stage volcanism (grey bars) at each respective island. Data are from Ozawa et al. ²². The dashed line is the approximate trace of the Hawaiian Arch, a flexing of the lithosphere caused by the building of the Hawaiian volcanoes^{24,35}. The label in brackets indicates a location not discussed in this study.

the proximity of the Hawaiian plume to a spreading centre when they formed, the Emperor Seamounts may sample the depleted upper mantle⁶, although others argue that the depleted source component is intrinsic to the plume^{8,10}. However, plume–ridge interaction cannot explain isotopically depleted Hawaiian rejuvenated-stage lavas in the middle of the Pacific plate, far removed from any spreading centre⁴¹. Hence, the location and nature of the isotopically depleted reservoir sampled by Hawaiian volcanism remains debated and critical to understanding the nature of the Hawaiian plume.

Depleted isotopic signature in Mauna Kea high-CaO lavas

In this study, we focus on lavas from 1,760-1,810 m.b.s.l. (meters below sea level) from the Hawaii Scientific Drilling Project (HSDP) that have elevated CaO (wt%) at a given MgO (wt%), which are referred to as high-CaO basalts42-45 and are tholeitic shield-stage lavas from Mauna Kea. HSDP cored 3.5km of lavas from Mauna Kea volcano in Hilo, Hawaii, which represent ~500 kyr of volcanic growth⁴⁶. A reference sample suite from the drill core⁴⁷ was investigated, which shows considerable variation in source composition and petrogenetic processes (Fig. 2). However, for the high-CaO basalts, major and trace element data exist only for seven glasses^{42,45} and 24 whole rocks⁴⁴, and they are an important end member to understand the composition of the Hawaiian plume. Herzberg⁴³, for example, argued that the high-CaO basalts are the only true melts of peridotite and that most other Hawaiian basalts are the result of pyroxenite melting. Compared to other Mauna Kea shield tholeiites, the HSDP high-CaO basalts are enriched in the most and least incompatible elements, and their mantle source may be a mixed lithology of pyroxenite and peridotite⁴⁵.

Here we report Sr, Nd, Hf and Pb isotope ratios (Supplementary Table 1) and trace element concentrations (Supplementary Table 2) for 24 whole-rock high-CaO basalts that were previously analysed for their major and some trace element concentrations by X-ray fluorescence⁴⁴ (Supplementary Fig. 1). The high-CaO basalts have the most unradiogenic Pb and Sr isotopes and the most radiogenic Hf and Nd isotope ratios among the Hawaiian shield-stage lavas (Fig. 3). The high-CaO lavas have a higher $\varepsilon_{\rm Hf}$ at a given $\varepsilon_{\rm Nd}$ than modern Pacific MORBs and the estimated 100 million years old Pacific lithosphere (field outlined in Fig. 3c and details given in the Supplementary Information), suggesting that neither the local lithospheric mantle nor the Pacific mantle contribute to the high-CaO lavas (Fig. 3c).

These high-CaO lavas have the same Sr, Nd, Hf and Pb isotope compositions as the Hawaiian rejuvenated-stage alkalic lavas (Fig. 3), which shows that the depleted rejuvenated-stage component is also present during the Hawaiian shield-stage volcanism, in agreement with previous studies that have detected it in the pre-shield stage of Loihi¹³, the shield stage of Kauai^{13,36} and the post-shield stage of Hualaiai³³, Mahukona^{33,37} and Haleakala³⁸. Our high-precision Pb isotope data for high-CaO lavas fall on the same ²⁰⁶Pb/²⁰⁴Pb versus ²⁰⁸Pb/²⁰⁴Pb trends as those defined by lavas and high-pressure pyroxenite cumulates associated with the rejuvenated-stage Honolulu Volcanics on Oahu^{9,11} (Fig. 4). This conclusion agrees with previous observations of heterogeneity within the depleted component^{11,12,48} and reinforces the isotopic connection between the rejuvenated-stage component and Mauna Kea shield-stage high-CaO basalts.

Trace element evidence

With the isotopic similarity between high-CaO lavas and Hawaiian rejuvenated-stage lavas but their difference in major and trace element compositions, we hypothesized that HSDP tholeiitic high-CaO basalts represent high-degree partial melts of a depleted rejuvenated mantle source. That is, the rejuvenated-stage source component(s) was sampled by smaller degrees of partial melting during the Hawaiian rejuvenated-stage volcanism (to produce alkalic basalts) than the HSDP high-CaO lavas (tholeiitic basalts). To test this hypothesis, we used both non-modal fractional and batch melting models of the depleted rejuvenated-stage sources to reproduce their trace element patterns. Three groups of well-studied rejuvenated-stage lavas were used: Honolulu Volcanics9 from Oahu, Kiekie basalts12 from Niihau and lavas from the North Arch Volcanic Field⁹. A filter that excluded samples with MgO < 6.5 wt% was applied to avoid samples that crystallized more than just olivine. Data for samples with MgO > 6.5 wt% were then corrected to be in equilibrium with Fo₉₀ to remove the olivine fractionation/accumulation effect. The source compositions were calculated assuming that rejuvenated-stage lavas are the products of low degrees of partial melting^{9,30} (0.5-4.0%). The calculated source compositions were then melted to a higher degree⁸ (6.0-9.5%) to see if they could reproduce the trace element patterns of the high-CaO basalts. The results of this model are shown in Fig. 5, and additional modelling details are given in the Supplementary Information. Our results show that the trace element patterns of the high-CaO basalts are consistent with those of high-degree partial melts of a rejuvenatedstage source. Our calculated depleted rejuvenated-stage source compositions are similar to those of the carbonatite-metasomatized source of Dixon et al. 12, who reconstructed the depleted rejuvenatedstage source compositions by metasomatizing a depleted peridotite with small amounts of incipient silicate or carbonatite melts⁴⁹ from the Hawaiian plume (Fig. 5d), a model first proposed by Chen and Frey³⁰ and also adopted here (Supplementary Fig. 4). Here we find that the same depleted component melted during the shield stage of volcanism.

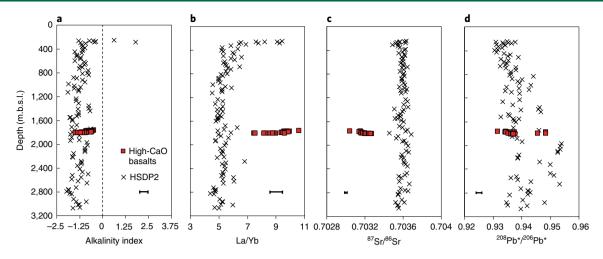


Fig. 2 | HSDP depth profiles. a-d, Depth profiles of the HSDP samples 44,61 versus the alkalinity index (**a**), La/Yb (**b**), 87 Sr/ 86 Sr (**c**) and 208 Pb* * 206Pb* * 206Pb* * 204Pb_{sample} $^{-206}$ Pb/ 204 Pb_{sample} $^{-206}$ Pb/ 204 Pb_{sample}

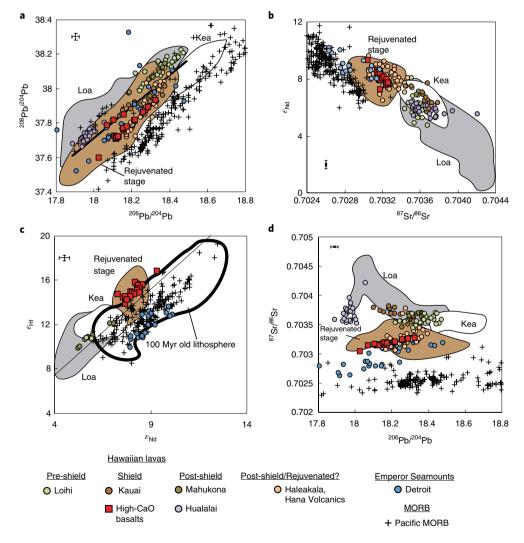


Fig. 3 | Radiogenic isotope data of Mauna Kea high-CaO basalts relative to shield-stage basalts of both Hawaiian Kea and Loa-trend volcanoes, rejuvenated-stage basalts and Pacific MORBs. a-d, 208 Pb/ 204 Pb versus 206 Pb/ 204 Pb (a), ε_{Nd} versus 87 Sr/ 86 Sr (b), ε_{Hf} versus ε_{Nd} (c) and 87 Sr/ 86 Sr versus 206 Pb/ 204 Pb (d). The thick line in a is the dividing line between Kea- and Loa-trend shield basalts 58 . The diagonal black line in c is the terrestrial array 62 and the field outlined with a thick black line in c is the calculated 100 Myr old lithosphere after MORB generation (Supplementary Information gives details). Error bars represent the two-standard error of measurements. Literature data sources are given in the Supplementary Information.

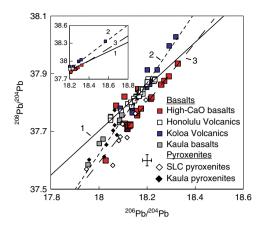


Fig. 4 | High-precision Pb isotope data for Hawaiian rejuvenated-stage lavas. The data are from the rejuvenated-stage basalts from Oahu (Honolulu Volcanics 39), Kaula 11 and Koloa Volcanics on Kauai 36,48 . High-pressure pyroxenite cumulates are from Salt Lake Crater (SLC), which is associated with Honolulu Volcanics at Oahu 9,11 and Kaula 11 . Honolulu Volcanics define a slope of $y = 0.7625 \times +23.885$ that three high-CaO basalts fall on (line 1). High-CaO basalts also fall on line 3 defined by the SLC pyroxenites ($y = 1.0338 \times +18.983$). Koloa Volcanics define a slope ($y = 1.2427 \times +15.251$) that some Kaula pyroxenites and Kaula basalts fall on (line 2). Inset, Koloa Volcanics with more radiogenic Pb isotope ratios. Error bars are the two standard error of measurements.

DM⁵⁰ was also considered as a possible source, but we could not reproduce the trace element patterns of the high-CaO basalts by melting the DM. Specifically, partial melts of DM are too depleted in incompatible elements compared to high-CaO lavas (Supplementary Fig. 5). Compared to modern Pacific MORBs, the high-CaO lavas have a higher $\varepsilon_{\rm Hf}$ for a given $\varepsilon_{\rm Nd}$ (Fig. 3c), consistent with the inference that DM is not the source of the high-CaO lavas.

The depleted intrinsic plume component

We therefore conclude that the high-CaO basalts are high-degree partial melts of a rejuvenated source that is intrinsic to the Hawaiian plume, consistent with plume models of previous studies^{8–13,34,35,51}. The Hawaiian mantle plume thus consists of enriched shield-stage components embedded as 'plums' inside a refractory matrix that is the source of depleted rejuvenated-stage lavas. During shield-stage volcanism, the enriched low-solidi shield-stage components start to melt first, and the refractory depleted component provides heat to enhance the melting of the enriched component is limited and overwhelmed by partial melting of the enriched components during shield-stage volcanism.

Hawaiian rejuvenated-stage volcanism and arch volcanism are thought to be produced by flexural arc decompression melting^{34,35}. Because of the volcanic loading, the Pacific plate is bent so that a flexural uplift of up to 100 m occurs at a distance of 200–400 km from the volcanic loading centre. This flexural uplift can produce a small degree of partial melting of the depleted component spread out beneath the lithosphere during shield-stage volcanism, which produces onshore rejuvenated-stage volcanism and offshore arch volcanism^{34,35}. The enriched plume components are consumed during shield-stage volcanism, which allows the refractory depleted component to melt and produce isotopically depleted, highly alkalic lavas^{18,34,35,51} during the rejuvenated-stage and arch volcanism.

In the case of the high-CaO basalts at Mauna Kea, it is possible that the portion of the plume that contributed to high-CaO lavas did not contain a significant amount of enriched shield components. That is, the mantle source of high-CaO basalts is dominated by the

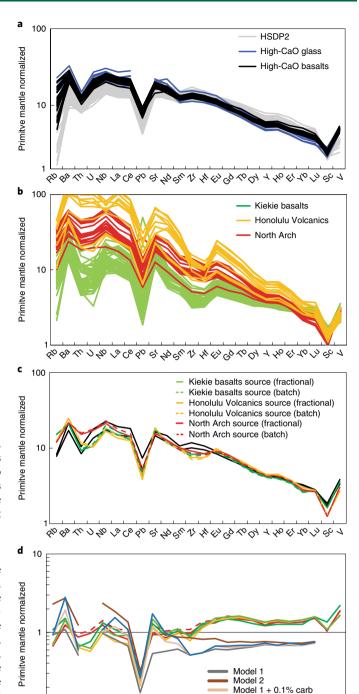


Fig. 5 | Trace element patterns and models. **a**, Primitive mantle-normalized trace element patterns for the HSDP reference suite samples (data references are in the Supplementary Information), high-CaO basalts (this study) and high-CaO glasses⁴² corrected to Fo₉₀ by adding or subtracting olivine. **b**, Rejuvenated-stage lavas from Honolulu Volcanics on Oahu, North Arch Volcanic Field and Kiekie basalts from Niihau, corrected to Fo₉₀ by adding or subtracting olivine, used for the partial melting model. **c**, Results of our partial melting model relative to the range of values for high-CaO basalts enveloped by the two black lines. **d**, Calculated source compositions in this study (same key in **c**) compared to those of Dixon et al.¹² (key in **d**). 'Carb' in **d** is carbonatite¹². The Supplementary Information give details and data sources.

depleted rejuvenated-stage component. Because of the higher temperature during the shield-stage volcanism, this refractory depleted component melted to a large degree and generated tholeiitic lavas with rejuvenated-stage isotopic signatures. In this case, we obtain a rare glimpse of the true isotopic composition and nature of the depleted component intrinsic to the Hawaiian plume.

The duration of high-CaO lavas is ~2,400 years according to the age-depth model of Bryce et al.⁵⁴. Using a plume upwelling rate of ~30 cm yr⁻¹ (ref. ^{54,55}), the estimated size of the geochemical heterogeneity is ~720 m. That is, the Hawaiian plume is heterogeneous at the subkilometre scale. For comparison, the radius of the magma capture zone at Hawaii is estimated to be 25–30 km (refs. ^{56,57}). Apparently, the compositions of erupted lavas reflect the sum of different plume components sampled by the melting events, and the short-scale source heterogeneity is homogenized to some degree during melting process^{54,56}. The observed Loa-Kea geochemical difference⁵⁸⁻⁶⁰ between the two Hawaiian volcanic trends must thus reflect the fact that their plume sources are more different than those shown in the lavas.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41561-019-0348-0.

Received: 27 September 2018; Accepted: 6 March 2019; Published online: 22 April 2019

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Acknowledgements

This project was supported by NSF grants EAR-1524387 and NSF Ocean Sciences grant no. 1355932. C.D. acknowledges the UNLV Department of Geoscience for support through the Jack and Fay Ross fellowship.

Author contributions

C.D. and S.M. were responsible for the geochemical and isotopic measurements. All of the authors contributed to the data interpretation and model calculations and wrote the paper.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information is available for this paper at https://doi.org/10.1038/s41561-019-0348-0.

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Methods

Data compilation. All the geochemical data for the Hawaiian shield-sta ge^{5,10,13,03,63,63,64,68,65-112} and rejuvenated-stage lavas^{7,9,11,12,28,93,113-115} were collected from GEOROC (http://georoc.mpch-mainz.gwdg.de/georoc/) and compiled. Pacific MORB radiogenic isotope data were collected from the Geochemical Earth Reference Model (GERM) database (https://earthref.org/GERM/) on 24 January 2018 and compiled¹¹⁶⁻¹³⁹. The search parameters set using the GERM database were the tectonic setting to spreading centres between 31.11 °N, 68.11 °S, 211.82 °W and 77.52° E. Sample types included were glass or whole-rock analyses of volcanic origin, classified as a basalt with major oxides, rare earth elements, trace elements or isotopic ratio data. The trace element data for rejuvenated-stage lavas used in the model were collected from GEOROC. North Arch Volcanic Field data are from Yang et al.⁹ and Clague et al.¹⁴⁰ and those for Honolulu Volcanics are from Fekiacova et al.³⁹ and Clague and Frey¹⁴¹, and for Kiekie basalts from Dixon et al.¹².

Sample preparation. Millipore ultrapure water (resistivity = $18\,\mathrm{M}\Omega$), double distilled acids using a subboiling PFA distiller and Baseline/Optima grade acids were used for the sample preparation.

Trace element analytical procedure. Trace element data for high-CaO basalts are reported in Supplementary Table 2. The sample of rock powder (~50 mg) was dissolved in acid-cleaned Savillex beakers using 3 ml of a 1:1 mix of HF:HNO and placed on a hot plate for 2 wk at 100 °C. Samples were then dried down and rehydrated with 1 ml of 50% HNO₃ (by mass) and dried back down three separate times. Then, 6 ml of 50% HNO3 was added to each sample and no undissolved residue was observed. An aliquot (2 ml) of each sample solution was pipetted into acid-cleaned 125 ml plastic bottles, in which the sample was diluted with 2% HNO₃ to a dilution factor of ~5,000. The samples were analysed for major and trace elements using a Thermo Scientific iCAP-Q inductively coupled plasma mass spectrometer (ICP-MS) at the University of Nevada, Las Vegas (UNLV) in kinetic energy discrimination (KED) mode to reduce polyatomic interferences. A typical analytical procedure of drift monitor-standard-unknown-unknown-unknownunknown-drift monitor was used to monitor the sensitivity drift during analysis⁶¹. Three USGS rock standards were used, BHVO-1, BCR-1 and AGV-1, to produce linear calibration curves and BHVO-2 was measured as an unknown sample for quality control. For the elements that were measured by both UNLV ICP-MS and UMass X-ray fluorescence, data were compared, and our data agree with published data from the UMass XRF Lab44 (Supplementary Fig. 2).

Radiogenic isotope analytical procedure. Sr, Nd, Hf and Pb were purified using ion exchange chromatography techniques. Sample dissolution and purification were performed in the PicoTrace clean lab at Brown University. About 500 mg of whole-rock powders were acid leached following the technique of Huang et al.41 using 6N double distilled HCl in an ultrasonic bath for 30 min. The acid was changed until the leachate was clear or pale yellow. Then, the samples were rinsed twice by adding Millipore water and sonicating for 30 min. Samples were then dried. About 100 mg of acid-leached sample was dissolved in a 3:1 HF:HNO₃ mixture following the procedure described above. Interested elements were extracted from the same sample aliquot in the order Pb, Hf, Sr/Nd (Supplementary Fig. 3). Pb was extracted first using AG1-X8 resin (100-200 mesh (BIO-RAD)). The matrix that remained from the Pb column was used to purify the remaining elements. First, Hf was removed from the matrix using Ln resin (Eichrom). Rb-Sr-REEs (rare earth elements) cut from the Hf column went through TRU-spec resin (Eichrom) to separate Rb-Sr from the REEs. The Rb-Sr cut from the TRUspec resin then went through Sr-spec resin to separate Sr from Rb. The remaining matrix from the TRU-spec column that contained REEs was passed through Lnspec resin to purify Nd. A schematic diagram (Supplementary Fig. 3) illustrates the order in which the columns were processed.

Sr. Nd. Hf and Pb isotope measurements were done at the Mass Spectrometer Analytical Facility at Brown University using a Thermo Scientific NEPTUNE PLUS multicollector ICP-MS. Sr, Nd and Hf were introduced to the plasma using a PFA nebulizer at a rate of ~70 µl min⁻¹ coupled with a glass spray chamber. Pb was introduced to the plasma using an APEX-IR introduction system to enhance the sensitivity. The instrument was equipped with an H-skimmer cone and H-sampler cone. The baseline measurement was taken off peaks at -0.5 AMU. 87Sr/86Sr, $^{143}\mathrm{Nd}/^{144}\mathrm{Nd}$ and $^{176}\mathrm{Hf}/^{177}\mathrm{Hf}$ were corrected for instrumental mass fractionation using 88 Sr/ 86 Sr = 0.1194, 146 Nd/ 144 Nd = 0.7219 and 179 Hf/ 177 Hf = 0.7325 with the exponential law. The Pb solution was spiked with the NBS SRM-997 Tl standard prior to analysis with a Pb/Tl=4 to correct for the instrumental mass fractionation using the exponential law with ²⁰³Tl/²⁰⁵Tl = 0.418922. Sr, Nd and Hf were measured at 200 ppb concentration, whereas Pb was measured at a 75 ppb concentration level. 87Sr/86Sr, 143Nd/144Nd and 176Hf/177Hf of the samples are reported relative to NBS SRM 987 87 Sr/ 86 Sr = 0.71024, the JNd-i Nd standard 143 Nd/ 144 Nd = 0.512115 and the JMC-475 Hf standard $^{176} Hf/^{177} Hf = 0.282160. \,^{206} Pb/^{204} Pb, \,^{207} Pb/^{204} Pb$ and 208 Pb/ 204 Pb are reported relative to the NBS 981 standards 206 Pb/ 204 Pb = 16.9409, $^{207}\text{Pb}/^{204}\text{Pb} = 15.4985$ and $^{208}\text{Pb}/^{204}\text{Pb} = 36.7222$ (ref. 142). The external precision on the ratios over the course of two years is 30 ppm for Sr (2σ , n = 75) and Nd $(2\sigma, n=104)$, 40 ppm for Hf $(2\sigma, n=89)$ and 45–80 ppm for Pb $(2\sigma, n=75)$. The Sr and Pb blanks were <50 pg, and the Nd and Hf blanks were <30 pg. To ensure

accuracy, three high-CaO basalt samples and two other HSDP samples that were studied previously^{63,65-67} were dissolved again and the same procedure as described above was followed in a blind test in which the origins of the samples being processed were unknown until after they were measured. These values are reported in Supplementary Table 3 and compared with the original measurements. Duplicated samples agree with original values within the 2-standard error of their measurements (Supplementary Table 3).

Calculation for a 100 Myr old lithosphere. To calculate the isotopic composition of a 100 Ma lithosphere after MORB generation, we first calculated the Nd-Hf isotopic compositions of the DM at 100 Ma using present-day Nd-Hf isotopic compositions of MORB (references are in the data compilation above), and Sm/Nd and Lu/Hf of DM50. Then, the parent/daughter ratios (Sm/Nd and Lu/Hf) of the melting residue after MORB generation were calculated using partition coefficients given in Dixon et al.¹² and assuming a degree of partial melting of 10%. It was assumed that the melting residue retains 4% melt due to porosity (outer border of the field in Fig. 3c). A second field was calculated with 8% partial melting and 4% melt retained, but their values are close to those of the first calculation and almost overlap (inner border of the field in Fig. 3c). The retained melt is consistent with previous interpretations of a recently metasomatized depleted source for the rejuvenated-stage9-12. Then, we used the calculated Sm/Nd and Lu/Hf in the melting residue and the calculated DM Nd-Hf isotopic compositions at 100 Ma to calculate their present-day Nd-Hf isotopic compositions, which are plotted as fields in Fig. 3c.

Partial melting models. To test whether or not high-CaO trace element patterns can be produced by higher degrees of partial melting of a rejuvenated source, we calculated rejuvenated-stage source compositions and melted them to a higher degree. In detail, rejuvenated-stage lavas filtered for MgO > 6.5 wt% from the North Arch Volcanic Field, Honolulu Volcanics on Oahu and Kiekie basalts from Niihau were selected for this model because they have complete major and trace element data $^{0.11,12,140,141}$. Their major and trace elements were corrected to be in equilibrium with Fo $_{90}$ by adding or subtracting equilibrium olivine to the samples.

We back calculated their source compositions by using non-modal batch melting (equation (1)):

$$\frac{C_1}{C_2} = \frac{1}{D + F(1 - P)} \tag{1}$$

where C_o is the initial source composition, C_1 is the basalt composition normalized to Fo₉₀. D is the bulk partition coefficient and P is the bulk partition coefficient for the phases entering the melt (see Supplementary Table 4). The values for D and P are from refs. ^{12,45,61,143}. Equation (1) was rearranged to solve for the initial source composition (equation (2)):

$$C_0 = C_1[D + F(1-P)]$$
 (2)

The same method was applied using non-modal fractional melting and the source compositions back calculated by solving equation (3):

$$\frac{C_1}{C_o} = \frac{1}{F} \left[1 - \left(1 - \frac{FP}{D} \right)^{\left(\frac{1}{P} \right)} \right] \tag{3}$$

Equation (3) was rearranged to solve for the initial source composition (equation (4)):

$$C_{o} = \frac{(C_{1}F)}{1 - \left(1 - \frac{(DF)}{P}\right)^{\left(\frac{1}{P}\right)}} \tag{4}$$

A 'mismatch value' was calculated by summing the absolute values of the log difference of each potential basalt composition to the average high-CaO basalt composition and taking the absolute value to assess the potential match. Lower 'mismatch' values indicate a better fit to the average primitive mantle-normalized value of the high-CaO basalts. The best fits from both batch and fractional melting were produced using the same starting compositions and the same F values for the source calculations and the melts of the calculated source (details about best fits are given below). Here we model the aggregated liquids of fractional melting, which produces results similar to batch melting. However, the calculated source compositions using batch melting and fractional melting for a single sample are slightly different (Fig. 5).

Best-fit sources for Honolulu Volcanics, North Arch and Kiekie basalts were chosen (Fig. 5c). Specifically, Honolulu Volcanics sample 69KAL-2 was used to calculate the rejuvenated mantle source with a partial melting degree of 3.5%. Then this calculated mantle source was melted by 6.0% to reproduce a trace element pattern that is similar to that observed in HSDP high-CaO lavas. Similarly, the HSDP high-CaO trace element pattern can also be reproduced by 6.0% partial melting of a mantle source calculated using North Arch sample 9-1 with a partial

melting degree of 3.0, and 6.5% partial melting of a mantle source calculated using Kiekie basalts sample T318-R19 with a partial melting degree of 3.5%. Comparisons of the calculated sources are shown in Fig. 5d. Fractional melting produces a similar result because the aggregated liquid produced from fractional melting closely resembles equilibrium batch melting with a different degree of partial melting. Major element compositions of high-CaO basalts agree with this interpretation; they are tholeitic and have lower concentrations of Na₂O and K₂O than rejuvenated-stage lavas (Supplementary Fig. 2).

High-CaO basalts have Ba/Th and La/Yb values that are intermediate among all the rejuvenated-stage lavas and within the shield-stage field. Ba is mobile during alteration. High-CaO basalts have a loss on ignition up to 10% (ref. ⁴⁴), which makes it difficult to ascertain whether they are the result of incipient silicate metasomatism or carbonatite metasomatism, as shown in Fig. 12 of Dixon et al. ¹² (Supplementary Fig. 4). Melting of a pure DM source ⁵⁰ was also considered and is a poor fit (Supplementary Fig. 5). DM melts at higher degrees of partial melting do not reproduce the pattern of the high-CaO basalts.

Data availability

The authors declare that all the data supporting the findings of this study are available within the article and its Supplementary Information.

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