REPORT

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Tungsten-182 heterogeneity in modern ocean island basalts

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New tungsten isotope data for modern ocean island basalts (OIB) from Hawaii, Samoa, and Iceland reveal variable $^{182}W/^{184}W$, ranging from that of the ambient upper mantle to ratios as much as 18 parts per million lower. The tungsten isotopic data negatively correlate with $^{3}He/^{4}He$. These data indicate that each OIB system accesses domains within Earth that formed within the first 60 million years of solar system history. Combined isotopic and chemical characteristics projected for these ancient domains indicate that they contain metal and are repositories of noble gases. We suggest that the most likely source candidates are mega–ultralow-velocity zones, which lie beneath Hawaii, Samoa, and Iceland but not beneath hot spots whose OIB yield normal ^{182}W and homogeneously low $^{3}He/^{4}He$.

The short-lived ^{182}Hf-^{182}W isotopic system [$^{182}Hf \rightarrow {}^{182}W + 2\beta^-$, half-life ($t_{½}$) = 8.9 million years] (*1*) is a powerful tool for studying planetary differentiation processes that occurred within the first ~60 million years of solar system history, while ^{182}Hf was extant (*2*). The system is particularly sensitive to the timing of metal-silicate segregation, such as occurs during planetary core formation, given that tungsten (W) is moderately siderophile and hafnium (Hf) is strongly lithophile (*3*). This results in most W residing in metallic cores and all Hf being sited within the silicate portions of differentiated bodies. In the case of Earth, the main stage of core formation occurred while ^{182}Hf was alive, resulting in the core (very low Hf/W and low $^{182}W/^{184}W$) and bulk silicate Earth (BSE) (comparatively high Hf/W and high $^{182}W/^{184}W$) having $^{182}W/^{184}W$ ratios that differ by ~220 parts per million (ppm) (*4*).

For Earth, excesses in ^{182}W relative to the modern upper mantle have been found in ancient Archean (≥2.5 billion years old) magmatic and supracrustal rocks (*5*–*7*). Such rocks have $\mu^{182}W$ values (deviation of $^{182}W/^{184}W$ from terrestrial standards in parts per million) as high as +20. These excesses have been interpreted to reflect large-scale metal-silicate or silicate-silicate fractionation processes that occurred within the first 60 million years of solar system history (*7*, *8*). Alternately, they may reflect derivation from mantle that had not yet received its full inventory of late accreted materials, which could be characterized by high W concentrations and low $\mu^{182}W$ (*5*, *6*).

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Tungsten isotopic heterogeneity is not limited to ancient rocks. $\mu^{182}W$ values as high as +48 have been reported in flood basalts as young as 60 million years (*9*), indicating long-term preservation of W isotopic signatures formed early in Earth history. Although several studies have examined the W isotopic compositions of modern mid-ocean ridge basalts (MORB) and ocean island basalts (OIB) (*5*, *7*, *9*, *10*), no anomalous isotopic compositions have yet been reported for these types of rocks.

Here, we report W isotopic data for MORB and OIB that range in age from ~20 million years to recently erupted lavas, to further examine the W isotopic composition in the modern mantle. In our examination of young, broadly basaltic rocks, we include samples from the major mantle components defined by variations in long-lived radiogenic isotope systems (*11*): enriched mantle 1 (EM1), EM2, high-μ (HIMU), and depleted MORB mantle (DMM). Attention is focused on rocks from the Hawaiian and Samoan OIB hot spots because of the wide range of chemical and isotopic compositions sampled by lavas at these hot spots, including the presence of ancient recycled crustal materials as well as high $^{3}He/^{4}He$ components. These results are compared with data for other radiogenic isotopic systems and helium (He) isotopic compositions.

Tungsten concentrations for the OIB we examined range from 9.4 to 967 parts per billion (ppb) (Table 1). In general, W concentrations are consistent with the combined effects of partial melting and crystal-liquid fractionation of melt derived from mantle with normal W abundances (*12*). When comparing the Hawaiian and Samoan suites, concentrations for Samoan basalts, on average, are slightly higher than that of the Hawaiian suite (fig. S1). However, the highest concentrations for individual rocks are for Kilauea Iki samples KI67-2-85.7 and KI81-2-88.6, which are late-stage fractionates of the lava lake that formed in 1959 (*13*).

We determined $\mu^{182}W$ values for OIB ranging from +3 to −18 (Table 1 and Fig. 1). Analytical uncertainties for individual measurements are typically ±4 ppm (2 SD), so some of these compositions are well resolved from what is presumed to be the ambient, modern upper mantle with a $\mu^{182}W$ value of zero (2 SD for Alfa Aesar standard = ±4 ppm; 2 SE = ±0.5 ppm; number of analyses (n) = 57). Samples from Mangaia (HIMU), Pitcairn (EM1), and the Canary Islands ("HIMU-like") have values that are indistinguishable from those of the ambient modern mantle. Our new datum for KNOX-11-RR, a sample of MORB from the Central Indian Ridge (Marie Celeste fracture zone), is also normal, which is consistent with a previously reported datum for MORB from the East Pacific Rise (*9*). In contrast, the suites from Samoa (EMII) and Hawaii, as well as two Icelandic basalts, are isotopically heterogeneous, with $\mu^{182}W$ values ranging from normal (~0) to strongly negative.

Reservoirs characterized by deficits in ^{182}W can be produced through several processes. As discussed in (*8*), crystal-liquid fractionation in an early silicate magma ocean can result in the creation of a mantle domain with a low Hf/W ratio relative to that of the modern upper mantle, eventually leading to a deficit in ^{182}W. There is some evidence for this type of process in ancient mantle-derived rocks. Puchtel *et al.* (*14*) reported an average $\mu^{182}W$ value of −8.4 ± 4.5 (2 SD) for the 3.55-billion-year-old Schapenburg komatiites from southern Africa, which they interpreted to be the result of silicate crystal-liquid fractionation in a magma ocean during the first 30 million years of solar system history. Alternatively, early formation (while ^{182}Hf was alive) and subduction of an incompatible trace element enriched protocrust, followed by long-term isolation in the deep mantle, is another means by which a mantle reservoir with $\mu^{182}W < 0$ could be formed (*15*). If either of these two processes is responsible for the negative anomalies in the Hawaiian, Samoan, and Icelandic systems, the preservation of the isotopic evidence in modern rocks would require isolation of the chemically distinct mantle domain from the ambient mantle for >4.5 billion years. In addition to processes that may have occurred within the silicate shell of Earth, minor present-day chemical or isotopic interactions between the lower mantle and the W-rich, strongly ^{182}W-depleted outer core (W = ~500 ppb; $\mu^{182}W$ = ~−220) could also impart a negative ^{182}W anomaly to the affected portion of the mantle (*10*). For this scenario, negative ^{182}W anomalies can be created in the mantle at any time during Earth history because of minor mixing between the core and the silicate Earth.

Consideration of complementary data for the long-lived Rb-Sr, Sm-Nd, U-Pb, and Re-Os isotopic systems available for Hawaiian and Samoan samples analyzed in this study (*16*–*27*) reveals no obvious correlations with $\mu^{182}W$ (fig. S2). In contrast, $\mu^{182}W$ values are negatively correlated with $^{3}He/^{4}He$, and all samples with $^{3}He/^{4}He$ >20 R/R_A (where R/R_A is the measured ratio divided by the atmospheric ratio of 1.384×10^{-6}) are characterized by negative $\mu^{182}W$ values that are well resolved from the ambient upper mantle (Table 1 and Fig. 2). This

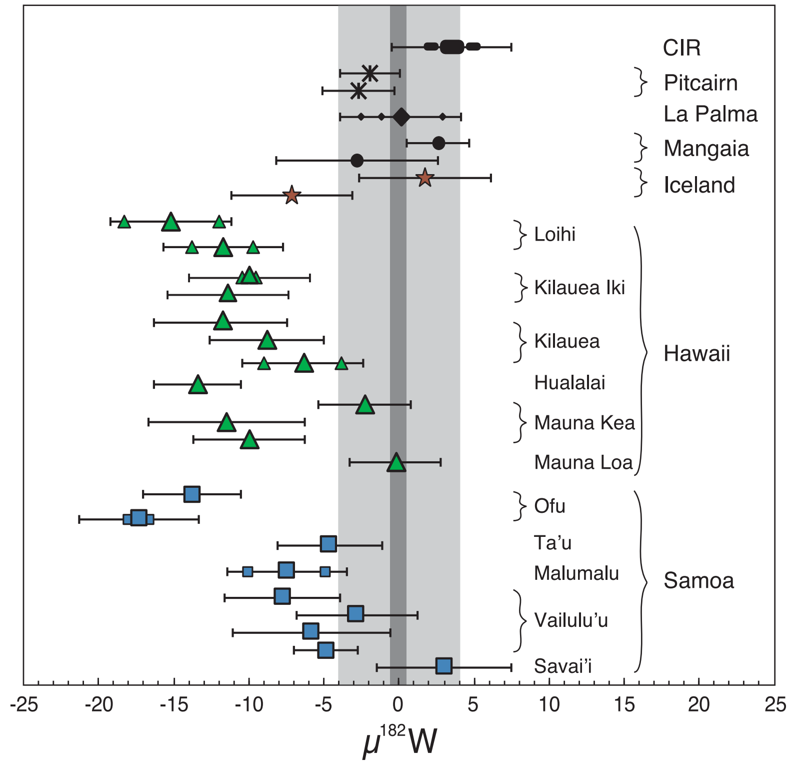

Fig. 1. μ^{182}W for MORB from the Central Indian Ridge (CIR) and a variety of OIB. Light gray area represents the 2 SD long-term external reproducibility of Alfa Aesar tungsten standard (4 ppm; number of analyses (n) = 57). Where applicable, small symbols indicate individual samples, and large symbols indicate the average of multiple analyses of one sample. The dark gray area shows the respective 2 SE of the standard (0.5 ppm; n = 57). Sample error bars represent 2 SE of individual analyses and 2 SD of multiple analyses (n = 2, except for LP15, where n = 5), respectively.

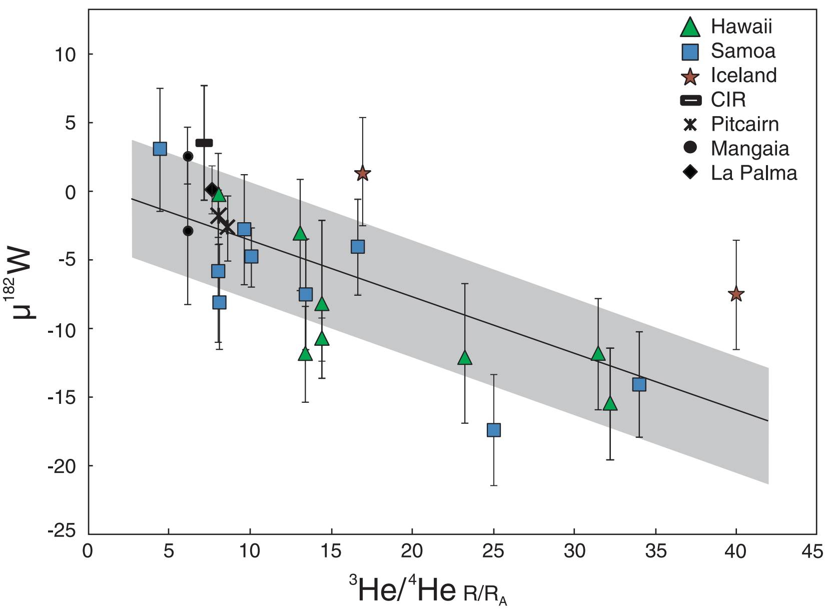

Fig. 2. μ^{182}W versus ^{3}He/^{4}He (R/R_A) for OIB and a MORB from the CIR. Error bars represent 2 SE of individual analyses or long-term external precision (2 SD) for average analyses of multiple samples, respectively. The average ^{182}W data are plotted for Kilauea Iki samples (KI67-2-85.7 and KI81-2-88.6) and Kilauea samples (BHVO-2 and KIL3-1). Error bars for ^{3}He/^{4}He are smaller than symbols. The black line represents a trendline for all samples. The gray area shows the error envelope for the trend line by using the long-term external precision for μ^{182}W isotopic measurements of ±4 ppm.

is also true for basalt sample 408614 from Iceland, with ^{3}He/^{4}He of 40 R/R_A. The negative correlations between ^{3}He/^{4}He and μ^{182}W observed for lavas from the Hawaiian, Samoan, and Icelandic hot spots must reflect their derivation from mantle that underwent variable extents of mixing between mantle with ambient He-W isotopic characteristics and a primordial reservoir characterized by high ^{3}He/^{4}He and negative μ^{182}W.

Although other causes have been proposed (*28–30*), OIB lavas with high ^{3}He/^{4}He ratios have most commonly been interpreted to sample primitive, undegassed regions of the mantle. By this reasoning, large differences in ^{3}He/^{4}He of MORB and OIB sources are generally interpreted to reflect different degassing histories (*31*), with some deep-seated OIB sources being less degassed and having higher ^{3}He/(U+Th) as compared with upper mantle MORB sources. The Central Indian Ridge MORB as well as OIB from Pitcairn, Mangaia, and the Canary Islands all have normal ^{182}W compositions, corresponding with comparatively low ^{3}He/^{4}He between 6.1 and 8.6 R/R_A (Fig. 2).

Mantle reservoirs that exhibit ^{182}W deficits created by elemental fractionation during early melting or crystal-liquid fractionation processes in the silicate Earth should also be enriched in highly incompatible elements, such as uranium (U) and thorium (Th). Whereas ^{3}He is considered primordial, terrestrial ^{4}He is mainly produced from the decay of ^{238}U, ^{235}U, and ^{232}Th. Assuming that Th and U are more incompatible than He (*28*), a reservoir with low Hf/W that formed during the crystallization of a magma ocean would be characterized by low ^{3}He/(Th+U) and, over time, by low ^{3}He/^{4}He. Thus, the negative correlation between μ^{182}W and ^{3}He/^{4}He argues against an origin via silicate crystal-liquid fractionation, either in the mantle or as a result of early formation of protocrust.

The strongly negative estimated μ^{182}W value for the outer core (–220) makes core-mantle interaction a potential candidate to account for the negative anomalies in the Hawaiian, Samoan, and Icelandic suites. Some experimental studies have suggested that substantial amounts of primordial He, characterized by ^{3}He/^{4}He ≈ 120 R/R_A, could have been incorporated into Earth's core during its formation (*32*). The presumed low concentrations of U and Th in the core would mean that the core has retained a primordial He isotopic composition to the present. Thus, transfer of the high ^{3}He/^{4}He and low μ^{182}W signature of the outer core to a lower mantle domain, which eventually rises in the form of a plume from which OIB are generated, could result in the production of lavas with the observed isotopic characteristics. However, the direct transfer of core metal to a rising plume would result in some collateral chemical effects, most notably an increase in the abundances of highly siderophile elements (HSEs; Re, Os, Ir, Ru, Pt, Rh, Pd, and Au) (*10*). For example, ~0.2 weight % (wt %) of outer core metal would have to be added to ambient mantle to yield a μ^{182}W value of –18. This amount of metal addition would more than double the HSE concentration of the hybrid mantle, relative to the ambient mantle. The HSE data for the ^{182}W-depleted lavas, however, provide no

Table 1. $^3He/^4He$ (R/R_A), $\mu^{182}W$ values, and W abundances for OIB and one MORB (in parts per billion). The reference sources for the helium data are included in the table. Tungsten concentration data were measured in this study or sourced from (*21*) or (*26*), as noted in the table. dup., duplicate analysis. All duplicates represent separate sample dissolutions.

Location	Sample	$^3He/^4He$ (R/R_A)	2 SD	$\mu^{182}W$	2 SE	W (ppb)
OIB						
Samoa						
Savai'i	ALIA 115-18	4.5 (*50*)	0.1	3.0	4.5	844
Vailulu'u	AVON3-63-2	10.1 (*18*)	0.1	–4.9	2.1	306
	AVON3-70-9	8.1 (*19*)	0.2	–5.8	5.2	523
	AVON3-71-22	9.6 (*19*)	0.2	–2.8	4.0	342
	AVON3-73-1	8.1 (*19*)	0.2	–7.7	3.8	693
Malumalu	AVON3-77-1	13.4 (*19*)	0.2	–4.9	3.1	720
	AVON3-77-1 dup.			–10.0	6.6	
Ta'u	T33	16.6 (*19*)	0.2	–4.6	3.5	304
Ofu	OFU-04-14	25.0 (*20*)	0.4	–18.0	3.5	213
	OFU-04-14 dup.			–16.6	5.9	
	OFU-05-18	33.8 (*20*)	0.4	–13.8	3.3	414
Hawaii						
Mauna Loa	ML 1868-9	8.0 (*22*)	1.6	–0.3	3.1	103 (*21*)
Mauna Kea	MK 1-6	13.4	0.4	–2.3	3.1	209
	SR0891-15.10	14.0 (*51*)	0.2	–10.0	3.7	90
	SR0750-12.45	23.2 (*51*)	0.4	–11.5	5.2	114
Hualalai	H-2			–13.4	2.9	97
Kilauea	BHVO-2	14.4 (*52*)*	0.8	–9.0	3.0	251 (*26*)
	BHVO-2 dup.			–3.9	3.1	
	KIL 3-1	14.4 (*52*)*	0.8	–8.8	3.8	155 (*21*)
	KIL1840-2	13.3	0.8	–11.9	4.4	182 (*21*)
Kilauea Iki	KI81-2-88.6	14.4 (*52*)*	0.8	–11.4	3.6	935
	KI81-2-88.6 dup.			–11.4	3.3	
	KI67-2-85.7	14.4 (*52*)*	0.8	–9.5	2.9	967
	KI67-2-85.7 dup.			–10.4	3.5	
	Kilauea Iki average	14.4 (*52*)*	0.8	–10.7	1.8	
Loihi	LO-02-02	31.6 (*53*)	1.2	–9.7	3.5	173
	LO-02-02 dup.			–13.8	3.6	
	J2-374-R5	32.2	0.8	–18.4	6.2	205
	J2-374-R5 dup.			–11.9	3.8	
Iceland	408614	40.2	1.2	–7.1	4.0	59.3
	ICE-14-32A	17.8	0.1	1.7	4.4	9.4
Mangaia	MG 1001-1	6.3 (*54*)†	1.4	–2.8	5.4	346
	MG 1001-8	6.3 (*54*)†	1.4	2.6	2.1	305
La Palma	LP-15			–1.2	3.5	
	LP-15 dup.			–2.6	3.0	
	LP-15 dup.			0.4	4.7	
	LP-15 dup.			2.8	3.7	
	LP-15 dup.			0.2	5.4	
	La Palma average	7.6 (*55*)‡	1.6	0.1	1.8	855
Pitcairn	PIT-1	8.6 (*56*)	1.0	–2.7	2.4	385
	PIT-8	8.0 (*56*)	1.0	–1.9	2.0	178
MORB						
Central Indian Ridge	KNOX-II-RR-D9	7.3 (*57*)	0.2	4.9	2.5	178
	KNOX-II-RR-D9 dup.		2.1	2.1		

*Average data for Kilauea volcano. †Average data for Mangaia. ‡Average data for La Palma.

evidence for HSE enrichment in the sources of the ^{182}W-depleted Hawaiian lavas (*21*) or for other OIB (table S1 and fig. S3). Osmium-187 of the outer core is probably not strongly divergent from the ambient mantle (*33*), so the broadly chondritic $^{187}Os/^{188}Os$ of the lavas with high $^3He/^4He$ does not provide an additional constraint. Further, although data are limited, there appears to be no correlation between $\mu^{182}W$ and $^{186}Os/^{188}Os$ for the Hawaiian suite (fig. S4). This suggests that the ancient fractionations in Pt/Os causing variations in $^{186}Os/^{188}Os$ are not related to the Hf/W fractionations that led to isotopic heterogeneity in ^{182}W.

There are other possible sources of metal-derived primordial W and He in the mantle that may better account for the observations. Earth's modern hot spots preferentially overlie a pair of large, low-shear-velocity provinces (LLSVPs) (*34*), whose unusual seismic characteristics—including anticorrelation of bulk sound and shear wave speed variations, and laterally abrupt boundaries—suggest chemical heterogeneity (*35*). Potentially, LLSVPs represent a deep undegassed reservoir formed from dense silicate melts (*36*), crystallization of a basal magma ocean (*37*), or accumulation of subducted oceanic crust (*38*) or mantle lithosphere (*39*). However, none of these processes, if they solely involved silicates, would predictably result in the observed negative correlation between $\mu^{182}W$ and $^3He/^4He$. Cumulates from an early magma ocean might, however, account for flood basalts characterized by high $^3He/^4He$ and positive $\mu^{182}W$, as observed in lavas from Baffin Bay and Ontong Java (*9*).

Zhang *et al.* (*40*) proposed that the seismic properties of LLSVPs might be explained by the presence of suspended droplets of Fe-Ni-S liquids. These authors suggested that such liquids could potentially store substantial amounts of noble gases, such as He, and would likely have very low Hf/W. If formed within the first 60 million years of solar system history, such droplets today would be characterized by negative ^{182}W anomalies and high $^3He/^4He$. Addition of varying amounts of early formed Fe-Ni-S liquids into an OIB source could, therefore, result in a negative correlation between $\mu^{182}W$ and $^3He/^4He$. In contrast to a contribution from the core, it is difficult to predict the effect of the addition of LLSVP-derived metal to HSE abundances in an OIB source, given the unknown abundances of HSE in this putative metal. Hence, HSE data neither support nor refute this possibility. However, if LLSVPs are the source of the isotopic signatures reported here, then their large spatial extent, encompassing up to 8% of the mantle by volume (*41*), would imply that the signatures might also be seen in other OIB that we analyzed, rather than just Hawaii, Samoa, and Iceland. For example, prominent seismic low-velocity conduits appear to connect La Palma and Pitcairn to the African and Pacific LLSVPs, respectively (*42*); yet, anomalous ^{182}W isotopic signatures are not observed in samples from either hot spot.

Ultralow-velocity zones (ULVZ), which are characterized by much smaller spatial scales but much greater velocity reductions as compared with those of LLSVPs, represent an alternative source domain for the anomalous OIB isotopic signatures. The extreme seismic signatures of ULVZs and their very low aspect ratios have been attributed to the presence of iron-rich silicate melt (*43*) or melt-free but iron-rich ferropericlase and/or bridgmanite (*44*, *45*). If ULVZs contain mantle partial melt, then enrichment in other incompatible elements, including U and Th, would be expected, and compatibility with observed He and W isotopic signatures would be unlikely. As with LLSVPs, however, the seismic properties of ULVZs might also be explained by a combination of metal and silicate (*46*). Metallic iron droplets could have been formed by disproportionation reactions ($3Fe^{2+} \rightarrow 2Fe^{3+} + Fe^0$) during the crystallization of one or more magma oceans early in Earth history (*47*). Tungsten is a moderately siderophile element, so unlike the highly siderophile elements, its abundance in the mantle at any stage in the growth history of Earth was not dependent on prior late accretion. Hence, metal generated by disproportionation may be characterized by moderate W abundances but very low HSE abundances, which is consistent with the lack of HSE enrichment in the lavas with low $\mu^{182}W$. For the same reasons discussed above for LLSVPs, such domains, if formed early in Earth history, could also be characterized by negative ^{182}W anomalies and high $^3He/^4He$. Intriguingly, a new class of large-scale ULVZs was recently discovered beneath Hawaii (*48*), and only two other examples of this type of "mega-ULVZ" have since been detected, beneath Iceland and Samoa (*49*). It might be expected that entrainment is more likely above mega-ULVZs, which may be hotter than ambient lower mantle, explaining why the signatures are only found in Hawaiian, Samoan, and Icelandic OIB.

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SUPPLEMENTARY MATERIALS

www.sciencemag.org/content/356/6333/66/suppl/DC1
Materials and Methods
Supplementary Text
Figures S1 to S4
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Tungsten-182 heterogeneity in modern ocean island basalts
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Editor's Summary

A mantle story told with metal and gas

Differences in isotopic compositions of trace elements can help identify how regions of Earth's mantle have evolved over time. Mundl *et al.* identified several ancient domains that have been isolated from mantle homogenization and thus contain signatures of primordial material. Tungsten and helium isotope values indicate fractionation and isolation of these mantle domains just after Earth's formation. The findings help constrain ancient processes such as core formation, but also provide insight into unexplained structures in the lower mantle today.

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