

Evolution of helium isotopes in the Earth's mantle

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Degassing of the Earth's mantle through magmatism results in the irreversible loss of helium to space, and high $^3\text{He}/^4\text{He}$ ratios observed in oceanic basalts have been considered the main evidence for a 'primordial' undegassed deep mantle reservoir. Here we present a new global data compilation of ocean island basalts, representing upwelling 'plumes' from the deep mantle, and show that island groups with the highest primordial signal (high $^3\text{He}/^4\text{He}$ ratios) have striking chemical and isotopic similarities to mid-ocean-ridge basalts. We interpret this as indicating a common history of mantle trace element depletion through magmatism. The high $^3\text{He}/^4\text{He}$ in plumes may thus reflect incomplete degassing of the deep Earth during continent and ocean crust formation. We infer that differences between plumes and the upper-mantle source of ocean-ridge basalts reflect isolation of plume sources from the convecting mantle for $\sim 1\text{--}2$ Gyr. An undegassed, primordial reservoir in the mantle would therefore not be required, thus reconciling a long-standing contradiction in mantle dynamics.

The presence of primordial ^3He in mantle gases has been considered the primary evidence for the contribution of undegassed, primordial lower mantle to upwelling plumes that generate ocean island basalts (OIB)^{1–4}, and forms a strong basis for models of two-layer mantle convection^{2,5,6}. On the basis of their helium isotope ratios relative to mid-ocean-ridge basalts (MORB, with $^3\text{He}/^4\text{He} = 8 \pm 1R_A$, where R_A is the atmospheric ratio of 1.4×10^{-6})⁷, so-called 'high- $^3\text{He}/^4\text{He}$ ' and 'low- $^3\text{He}/^4\text{He}$ ' mantle plumes have been distinguished². It has been argued that low- $^3\text{He}/^4\text{He}$ plumes reflect degassed, recycled material from the surface of the Earth dominated by radiogenic ^4He , whereas high- $^3\text{He}/^4\text{He}$ plumes reflect contributions from undegassed mantle^{2,4,8–12}. However, it has been long recognized from trace element and Sr–Nd–Pb–Hf isotope ratios in high- $^3\text{He}/^4\text{He}$ OIB that mantle plume sources do not represent undegassed primitive material^{4,13–16}. In addition, geophysical evidence for whole-mantle convection^{17–20} makes the isolation of a primordial, undegassed high- $^3\text{He}/^4\text{He}$ component in the lower mantle problematic. Thus, the preservation of significant primordial ^3He in OIB remains difficult to explain.

Here we present a global compilation of OIB and show that primordial ^3He in plume sources is represented by a common component, which shows striking chemical and Pb–Sr–Nd isotopic similarities to MORB mantle. The data indicate that high- $^3\text{He}/^4\text{He}$ plume sources, like the upper-mantle source of MORB, chemically complement the continental crust and have been depleted in incompatible elements through Earth history through melt extraction. The observations are explained by means of a model of mantle helium evolution that does not require the existence of a primordial, undegassed high- $^3\text{He}/^4\text{He}$ reservoir.

Helium isotope systematics of oceanic basalts

Our new data compilation of Th, U and La abundances and Sr–Nd–Pb isotope ratios of OIB and MORB are based on global data from the online GEOROC (<http://georoc.mpch-mainz.gwdg.de/>) and PetDB (www.petdb.org) databases, and the USGS noble gas database²¹. To obtain a data set representative of the upper-mantle sources of MORB, only analyses on fresh glasses from a water

depth of greater than 2000 m are used, thus filtering out plume influenced samples. To obtain a representative data set of mantle plume sources, only basaltic rocks from the constructional phase of ocean islands are used (details in Supplementary Information). Basalts from individual islands often show a range in $^3\text{He}/^4\text{He}$ ratios, which often does not covary with Sr–Nd–Pb isotope ratios. This 'decoupling' is not well understood, and it has been suggested that helium as a noble gas behaves differently from lithophile trace elements within the deep mantle or during partial melting processes, magma transport and storage^{14,22}. In addition, Hawaii, Iceland and Galapagos basalts show a range in $^3\text{He}/^4\text{He}$ ratios, from high values to MORB-like values, which partly seem to reflect entrainment of ambient MORB-type mantle, but also require heterogeneity in the plumes (recently reviewed for Hawaii in ref. 23). This contribution uses the highest helium isotope ratios in individual oceanic islands ($^3\text{He}/^4\text{He}_{\text{max}}$) to constrain the nature of the high- $^3\text{He}/^4\text{He}$ component in the associated mantle plumes. We divide the islands into four groups on the basis of the highest $^3\text{He}/^4\text{He}$ ratios from mineral separates and glass: first, 'low $^3\text{He}/^4\text{He}$ ' ($^3\text{He}/^4\text{He} < 7R_A$); second, 'MORB-like $^3\text{He}/^4\text{He}$ ' ($8 \pm 1R_A$); third, 'moderately high $^3\text{He}/^4\text{He}$ ' ($9\text{--}15R_A$); and fourth, 'high $^3\text{He}/^4\text{He}$ ' ($^3\text{He}/^4\text{He} > 15R_A$). These designations enable us to evaluate ocean islands and their plume sources on the basis of their $^3\text{He}/^4\text{He}$ ratios, despite the paucity of combined He–Sr–Nd–Pb–Hf isotope data on individual samples.

The four OIB groups show strong relationships to their Pb–Nd isotope ratios (Fig. 1). Lower $^{143}\text{Nd}/^{144}\text{Nd}$ ratios are associated with lower $^3\text{He}/^4\text{He}$ (Fig. 1). For a constant $^{143}\text{Nd}/^{144}\text{Nd}$ ratio, higher $^{206}\text{Pb}/^{204}\text{Pb}$ ratios are associated with groups with lower $^3\text{He}/^4\text{He}$. Thus, once Nd isotopes are 'normalized' out, high $^3\text{He}/^4\text{He}$ ratios in ocean islands are associated with low $^{206}\text{Pb}/^{204}\text{Pb}$ ratios (Fig. 1, dark blue symbols). The only known exception, Samoa, is a 'high $^3\text{He}/^4\text{He}$ ' plume with low $^{143}\text{Nd}/^{144}\text{Nd}$ ratios and is discussed below. Because ^{206}Pb , ^{207}Pb , ^{208}Pb and ^4He are generated by the radioactive decay of U and Th, the global systematics indicate a direct relationship between $^3\text{He}/^4\text{He}$ and either the plume source formation age or their Th and U content (with older plume sources

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or those with higher Th + U having lower $^3\text{He}/^4\text{He}$ ratios). Here we constrain the Th and U contents of plume sources.

Thorium and lanthanum are both highly incompatible elements and therefore Th/La ratios of basaltic rocks are expected to closely approximate the melt sources. High Th/La therefore indicates Th

enrichment relative to La in the mantle sources. With a few exceptions, mostly from the Society Islands (Fig. 2a, plus signs), Th/La and $^{208}\text{Pb}/^{204}\text{Pb}$ covary positively in OIB, which is consistent with radiogenic ingrowth of ^{208}Pb due to Th decay. The same is true for U/La (not shown), but the data are more scattered owing to the susceptibility of U to alteration by weathering. The Th contents in OIB vary by more than 10^2 and are positively correlated with Th/La (Fig. 2b). Whereas the high-Th end of the OIB array is dominated by relatively

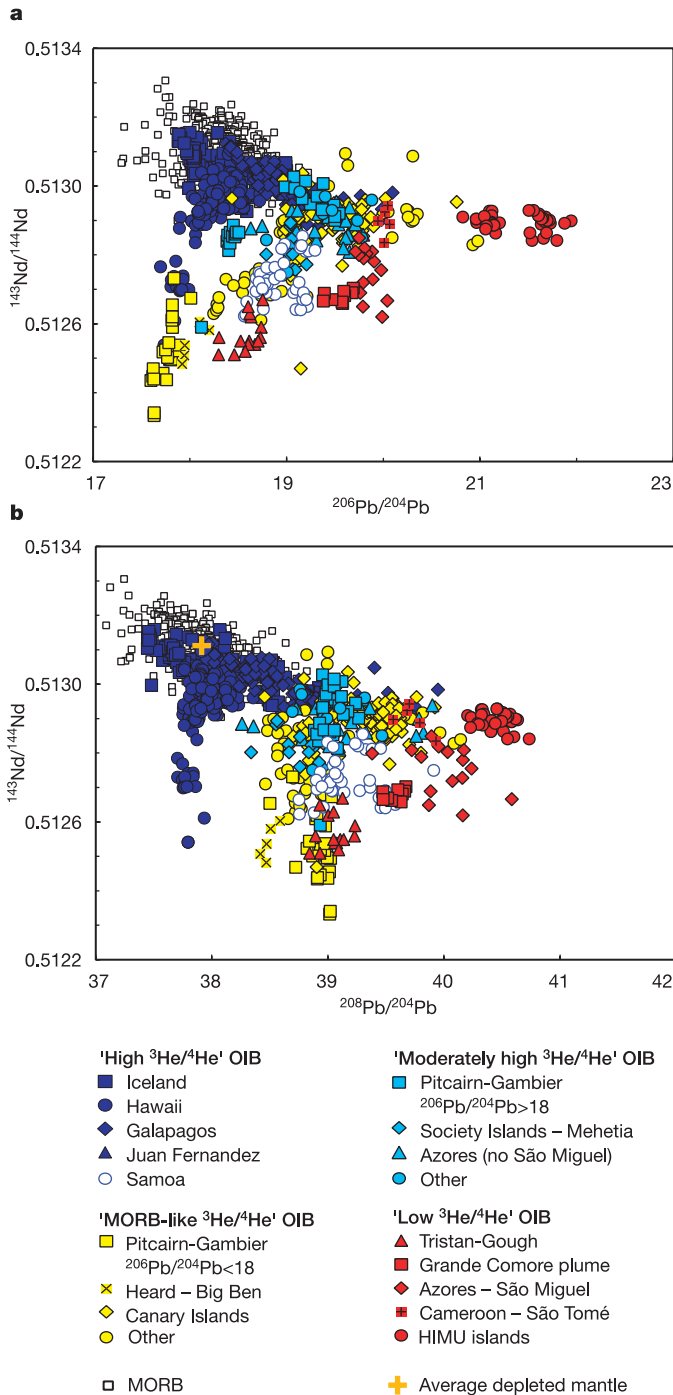


Figure 1 | Nd-Pb-He isotopes of OIB and MORB. High $^3\text{He}/^4\text{He}$ OIB have the most MORB-like Nd and the lowest Pb isotope ratios. Plots of $^{206}\text{Pb}/^{204}\text{Pb}$ (a) and $^{208}\text{Pb}/^{204}\text{Pb}$ (b) against $^{143}\text{Nd}/^{144}\text{Nd}$ are shown. OIB and MORB data are from the GEOROC and PetDB databases⁴⁹, respectively. Symbol colours correspond to the helium isotope groups of global plumes (see the text and Supplementary Information). Orange plus sign, estimate of average depleted (MORB) mantle⁴⁴. Other 'moderately high $^3\text{He}/^4\text{He}$ ' OIB include Cape Verde northern Islands, Réunion. Other 'MORB-like $^3\text{He}/^4\text{He}$ ' OIB include Austral-Cook Islands Rarotonga and Rurutu younger series, Cape Verde southern Islands, Society seamounts Tehetia, Rocard.

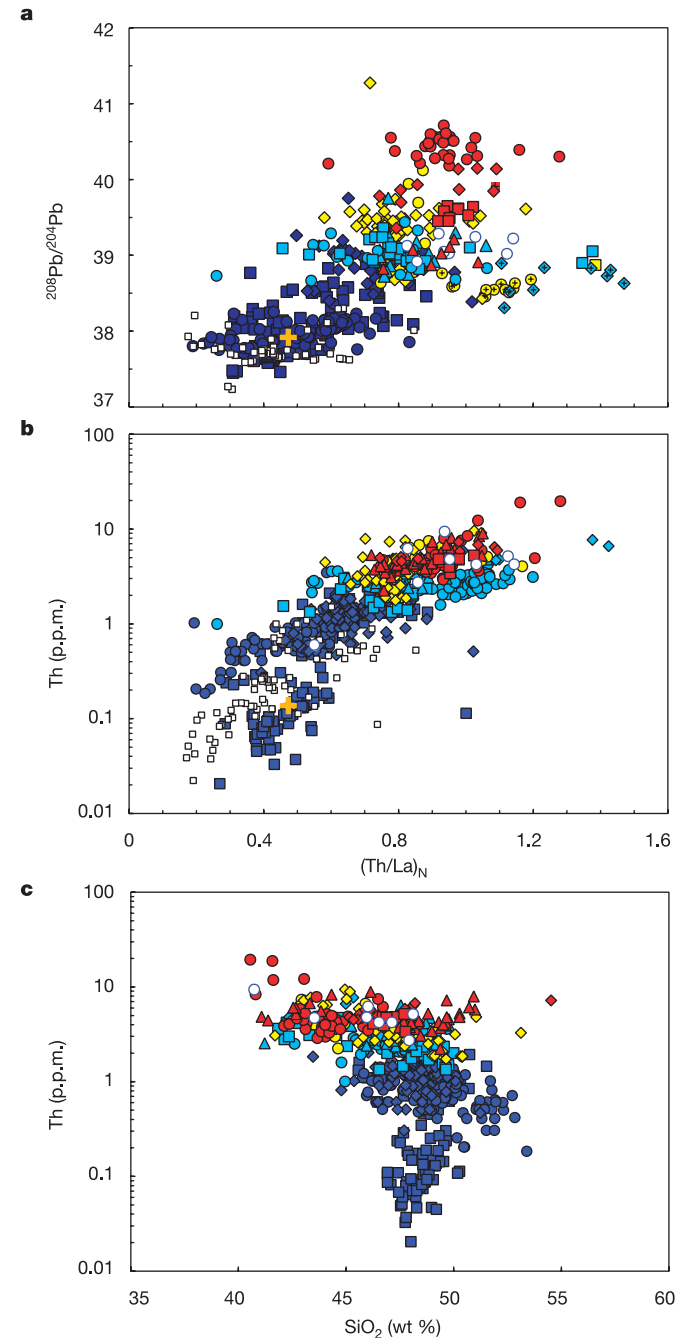


Figure 2 | Th, Th/La, SiO_2 and helium isotope ratios of OIB and MORB. High $^3\text{He}/^4\text{He}$ OIB have MORB-like Th contents and $(\text{Th}/\text{La})_N$ ratios (normalized to primitive mantle⁴⁵). **a, b**, Plots of $^{208}\text{Pb}/^{204}\text{Pb}$ (a) and Th content (b) against $(\text{Th}/\text{La})_N$ in OIB and MORB. **c**, Plot of Th against SiO_2 in OIB. For Th, only samples with 5wt% < MgO < 15wt% are shown. Symbols and data sources are as in Fig. 1. Society Island samples fall off the main trend in **a** (black plus signs) but not in **b** and **c**. 'Average depleted MORB' (orange plus sign) is estimated to be tenfold the depleted MORB mantle composition⁴⁴ for Th.

low-degree alkali basaltic partial melts, and the low-Th end by high-degree tholeiitic partial melts, the huge range in Th content cannot be solely the result of variable degrees of melting. For example, if global plume sources had similar Th abundances, and tholeiites with 0.5 p.p.m. Th were formed by 6% partial melting, then alkali basalts with about 10 p.p.m. Th would be formed by only about 0.3% partial melting, which is unreasonably low for such rocks. Moreover, lower-degree melts generally have lower silica contents than higher-degree melts, and the absence of a correlation between the SiO₂ and Th content among the helium groups (Fig. 2c) shows that the global range of Th contents in OIB is not the result of different degrees of partial melting. Rather, the large range reflects variable enrichment of Th in the mantle sources of plumes.

A compilation of samples containing both ³He/⁴He and Th data clearly confirms that plumes with high ³He/⁴He_{max} erupt lavas with lower Th contents (Fig. 3a). However, this figure shows that only a small fraction of samples have both ³He/⁴He and Th analyses. There are no Th data on the samples with the highest ³He/⁴He ratios from

the ‘high ³He/⁴He’ group, and the ‘MORB-like ³He/⁴He’ group is not represented at all (the range of ³He/⁴He for each island is illustrated by bars). In Fig. 3b, all measured ³He/⁴He ratios are plotted against average Th contents of individual islands or plumes. With the exception of Samoa (blue open circles) and Mehetia in the Society Islands (light blue diamonds), ³He/⁴He_{max} ratios from individual islands show an inverse relation with Th abundance (and U; compare ref. 16).

Samoa is an outlier to these global systematics, erupting lavas with high Pb and He isotope ratios, and high Th and U. Samoa is also an exceptional plume in its unusual tectonic setting at the northern terminus of the Tonga subduction zone. Farley²⁴ noted the unusual combination of high ³He/⁴He and Pb–Sr–Nd isotope ratios indicating long-term incompatible element enrichment, and suggested that sediment from the subduction zone became rapidly cycled through the mantle, produced only a small amount of radiogenic ⁴He over this short timescale, and became mixed into the rising ‘high ³He/⁴He’ Samoa plume. Because Samoa is such a clear exception to the general rule, we exclude it from the discussion below.

Recycled components and ³He/⁴He of plumes

There is ample evidence that plume sources contain recycled oceanic crust and sediment that affect their Sr–Nd–Pb–Hf isotope ratios and contribute trace elements such as Th and U^{15,25}. Because helium is degassed during ocean crust formation, alteration and subduction²⁶, the recycled component cannot provide significant primordial ³He, but it would contain radiogenic ⁴He produced after degassing by Th + U decay⁷. The inverse global relationship between ³He/⁴He_{max} and Th abundances (Fig. 3) indicates that plumes may contain both a primordial helium component and extra ⁴He from Th + U decay in recycled oceanic crust. This is illustrated by the curves in Fig. 3b, where the ³He/⁴He ratios reflect combinations of radiogenic ⁴He from old (1.5 or 0.8 Gyr old) recycled oceanic crust with variable Th plus primordial helium from small amounts (1% or 2%) of ‘bulk silicate Earth’. The curves fit the inverse relationship between ³He/⁴He_{max} and Th abundances for all helium groups (with Samoa as an exception), which requires similar ³He abundances in the plume sources (within a factor of about 2). The Th abundances of the ‘MORB-like ³He/⁴He’ and ‘low ³He/⁴He’ groups overlap (Figs 2b and 3b), and this might be explained by differences in the age of formation of the plume sources.

Thus, although some of the variability probably reflects differences in plume source formation age, the global variation of ³He/⁴He_{max} and Th can be explained by a small but similar amount of primordial helium plus radiogenic production of ⁴He as a function of variable Th + U in global plume sources. Neon data are consistent with this conclusion (see Supplementary Information).

Mantle depletion and high ³He/⁴He in plumes

The OIB–MORB data compilation shows that the high-³He/⁴He component of mantle plumes is a global endmember with Th and U abundances and Sr–Nd isotope ratios similar to MORB (Figs 1–3), indicating that the high-³He/⁴He endmember is ‘depleted mantle’. This observation is strongly supported by the depleted-mantle composition of the highest ³He/⁴He endmember for mantle plumes as measured in Baffin Island basalts, associated with the early Iceland plume¹². These have ³He/⁴He ratios up to 50R_A and Sr–Nd isotope ratios and La/Sm in the range of those of depleted MORB. The Baffin data are not shown in Figs 1 and 2 because trace-element abundances and Pb isotope ratios have not yet been published¹². The ‘depleted mantle’ character of the high-³He/⁴He endmember in OIB is also consistent with previous interpretations of Sr–Nd–Hf isotope arrays from individual ocean islands, which seem to converge at isotope ratios reflecting long-term depletion of lithophile elements, which has been termed the ‘focus zone’ or FOZO³ and is similar in concept and composition to ‘primitive He mantle’ or PHEM⁴. FOZO also has a low ¹⁸⁷Os/¹⁸⁸Os, indicative of peridotitic mantle²⁷. The global data

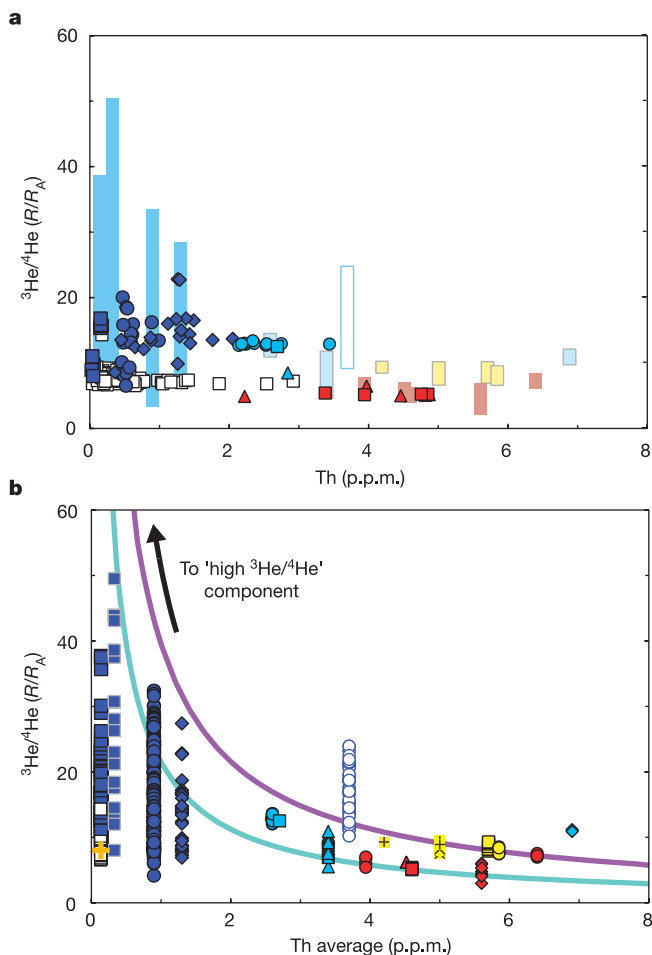


Figure 3 | ³He/⁴He – Th of OIB and MORB. **a**, Symbols show available combined Th–³He/⁴He data. Bars show data range from **b**. **b**, Plot of measured ³He/⁴He against average Th of different plumes and MORB. ³He/⁴He_{max} reflect ⁴He production from Th + U in recycled components plus a uniform amount of high-³He/⁴He component as shown by curves with variable plume source ages plus primitive mantle contribution: blue, 1.5 Gyr (1%); purple, 1.5 Gyr (2%) or 0.8 Gyr (1%). OIBs are assumed to reflect 6% melts with average Th/U = 3.45; primitive mantle ³He/⁴He_{4.55 Gyr} = 230R_A; ⁴He_{4.55 Gyr} = 1.2 nmol g⁻¹; Th and U from ref. 45. Data sources, filters and symbols as in Figs 1 and 2 plus additional data from the USGS noble gas database²¹; blue squares with grey outline, Baffin Island (early Iceland plume); yellow squares containing plus sign, Rurutu Recent Series and Rarotonga.

are fully consistent with FOZO's representing depleted MORB-like peridotitic mantle with some recycled oceanic crust.

Whereas the upper-mantle source of MORB has lower $^3\text{He}/^4\text{He}$ ratios than the high- $^3\text{He}/^4\text{He}$ endmember of mantle plumes, the MORB $^3\text{He}/^4\text{He}$ value of $8 \pm 1R_A$ (ref. 7), shows that the MORB mantle still contains a significant component of primordial ^3He . Most attempts to explain $^3\text{He}/^4\text{He}$ in MORB have argued for an addition of ^3He to the degassed MORB mantle through a plume flux^{6,28,29} or an independent flux of primordial helium from the lower mantle²². We suggest that the striking chemical and isotopic similarities between depleted MORB mantle and the high- $^3\text{He}/^4\text{He}$ endmember in OIB strongly indicates that their formation is directly related.

A role for high $^3\text{He}/(\text{U} + \text{Th})$ OIB reservoirs has previously been suggested^{12,16,30,31}. Anderson³⁰ assumes that $^3\text{He}/^4\text{He}$ does not covary with other chemical tracers, whereas Figs 1 and 2 show that strong systematic relationships do indeed exist. It has been suggested¹² that depleted upper mantle is re-gassed near the boundary between upper and lower mantle. We suggest that this is problematic because some plumes appear to originate from the core/mantle boundary³² and because plumes have similar concentrations of primordial ^3He , as shown above. The small range of ^3He abundances globally also poses problems for models of recycling of oceanic lithosphere stripped of rare gases into the lower mantle^{16,31}. A well-mixed mantle 'plum pudding' based on two-stage melting of primitive mantle³³ could result in a small range of ^3He abundances, but if this model were correct the high- $^3\text{He}/^4\text{He}$ endmember of OIB should trend towards primitive rather than depleted-mantle compositions (Figs 1 and 2). In addition, all these models^{16,31,34} require primordial, undegassed regions within the lower mantle to preserve primordial ^3He . The 'missing Ar' argument³⁵ concludes that a gas-rich reservoir in the Earth's mantle represents about 50% of its mass and has been used as strong support for a layered mantle, but this is obviated if recent estimates of lower K abundances for the silicate Earth are correct^{30,36,37}. Given the chemical and isotopic similarities between MORB and the high- $^3\text{He}/^4\text{He}$ endmember in OIB, and geophysical evidence for whole-mantle convection^{17–20}, we suggest that high $^3\text{He}/^4\text{He}$ in plumes can be explained by whole-mantle convection and the formation of continents and oceanic crust.

Recent work on noble-gas partition coefficients indicate that they are not extracted with nearly 100% efficiency but instead behave like highly incompatible elements during melting, with a small fraction remaining in melt residues^{38,39}. This allows for some primordial ^3He to be retained in mantle volumes that have been depleted by melt events such as the formation of oceanic crust. We propose that the similarity between MORB and the high- $^3\text{He}/^4\text{He}$ OIB in terms of Th and U contents, Th/La ratios and Sr-Nd-Pb isotope ratios (Figs 1–3), as well as the elevated $^3\text{He}/^4\text{He}$ of MORB (even if lower than 'high $^3\text{He}/^4\text{He}$ ' OIB), could be related through incomplete degassing during ocean and continental crust formation by a mantle undergoing whole-mantle convection. We present a model illustrating how this might work. An initial degassing stage during accretion of the Earth is constrained by xenon isotopes⁴⁰; we consider it to be the event that set up the initial conditions. In this model, the mantle is depleted in incompatible elements (including Th and U) by continent formation over an interval of 4.4–2.7 Gyr, and $^3\text{He}/^4\text{He}$ ratios in this 'depleted mantle' evolve through the decay of Th and U and helium degassing associated with ocean crust formation. The mean age of present-day continental crust is probably closer to 2 Gyr (see ref. 41, for example) but this younger age partly reflects recycling of continental crust to the mantle through sediment subduction and tectonic erosion of continental margins. The higher $^3\text{He}/^4\text{He}$ ratios in plumes than in MORB reflect isolation from the convecting mantle for significant geological intervals. The isolation allows plume sources to retain more ^3He relative to the upper-mantle MORB source, which continually undergoes degassing as it forms oceanic crust. Thus, in this model both MORB and OIB sources have been

significantly degassed through magmatism due to ocean crust and continent formation, but the high- $^3\text{He}/^4\text{He}$ endmember in mantle plumes represents depleted mantle that was 'less degassed' through isolation most probably somewhere in the lower mantle.

The helium evolution model is illustrated in Fig. 4 (calculations and model parameters in Supplementary Methods). The $^3\text{He}/^4\text{He}$ ratio of primitive mantle decreases owing to radioactive production of ^4He (dark blue curve), which would reach about $95R_A$ were it not for the formation of continents and ocean crust. Degassing rates through geological time are under-constrained (details in Supplementary Methods). The most solid constraint is the degassing rate of the oceanic mantle today⁴², which in our model needs to be adjusted with more vigorous degassing in the past. In the example shown, the degassing rate, which reflects both the helium abundance and the rate of ocean crust formation, was 34-fold higher 4.4 Gyr ago. A curve illustrating the effects of degassing on the upper-mantle source of MORB is shown in Fig. 4 (red curve). The evolution of a 'high- $^3\text{He}/^4\text{He}$ component' in plumes is also shown (yellow curve), which is derived from the 'depleted' mantle but becomes isolated from degassing in a boundary layer. In this example the isolation occurred about 1.5 Gyr ago. In contrast to the continuously degassing upper mantle, the $^3\text{He}/^4\text{He}$ ratio of this 'high- $^3\text{He}/^4\text{He}$ component' declines much less and maintains a high present-day value of about $72R_A$.

Figure 4 thus shows that in general terms the high $^3\text{He}/^4\text{He}$ ratios in OIB can be explained through normal mantle differentiation processes. Continent formation results in the generation of a mantle depleted in incompatible elements. The mantle evolves to lower $^3\text{He}/^4\text{He}$ values (as observed in MORB today) through melting and degassing at ocean ridges, re-mixing of degassed mantle by convection and continuous formation of ^4He by radioactive decay. If a portion of this mantle is isolated from degassing, the $^3\text{He}/^4\text{He}$ ratio will remain high, as observed in plume sources, with a value between those of primitive mantle and MORB.

On the basis of this general degassing model, ages of plume sources can be made a dependent variable and calculated (Fig. 5). We calculate Th and U contents of OIB sources assuming that lavas reflect 6% partial melting of peridotite. Plume sources contain variable amounts of recycled components of variable compositions¹⁵.

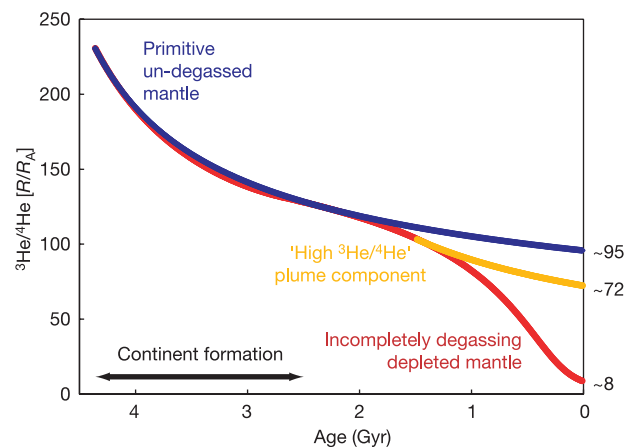


Figure 4 | Helium isotope evolution of mantle reservoirs. Illustration of the incomplete degassing model. Blue line, primitive, undegassed Earth's mantle: $^3\text{He}/^4\text{He} = 230R_A$ (ref. 46), $[^3\text{He}] = 1.5 \times 10^{11}$ atoms g^{-1} based on refs 47, 48. Th and U abundances: primitive mantle⁴⁵, depleted mantle⁴⁴. The depleted-mantle degassing rate today is $^3\text{He} = 1,060$ mol yr^{-1} from refs 1, 42, and increases in the past. Red line, incompletely degassing depleted mantle. The endpoints of the yellow and red lines represent the 'high- $^3\text{He}/^4\text{He}$ ' plume components and present-day MORB, respectively. Details of model parameters and the calculation are in Supplementary Information.

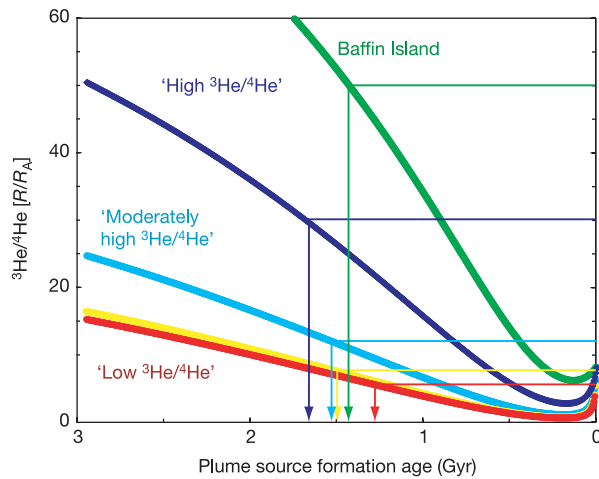


Figure 5 | Plume source formation ages from the incomplete degassing model. Th and U in plume sources are calculated on the assumption that the average Th and Th/U ratio of OIB He groups are formed by 6% partial melting; Baffin endmember with Th = 0.02 p.p.m. and Th/U = 3.31. OIB sources are assumed to represent mixtures of depleted mantle and a constant volume of a recycled component with variable Th + U content (see text). The $^3\text{He}/^4\text{He}$ ratio of a plume source as a function of its formation age is calculated on the basis of the He evolution of the depleted mantle plus the variable contributions of radiogenic ^4He for each He group.

For simplicity, we assume that the amounts of recycled component in all plume sources are constant, and allow the composition (the Th + U content) to vary to calculate the ^4He production rate of the recycled components. For each He-isotope group of OIB, the production rate of ^4He and the $^3\text{He}/^4\text{He}$ ratios can be calculated as a function of the plume source formation age. For the input parameters illustrated in Fig. 5, the four $^3\text{He}/^4\text{He}$ groups and the Baffin Island endmember yield formation ages of between about 1.3 and 1.7 Gyr, which is consistent with previously suggested plume source ages (see ref. 43, for example). Of course, the derived ages vary depending on the choice of model parameters, but the small range of formation ages for all of the $^3\text{He}/^4\text{He}$ groups of OIB is a robust result for any set of inputs. The mantle $^3\text{He}/^4\text{He}$ evolution as presented here is consistent with the whole mantle being 'depleted' by continent and ocean crust formation. However, the addition of recycled crustal components to plume sources, combined with their ancient formation ages, results in incompatible element 'enriched' plume sources, isolated from the depleted mantle, some of which evolve to low $^3\text{He}/^4\text{He}$ and Nd isotope ratios and high Sr and Pb isotope ratios.

Thus, the new global compilation of oceanic basalt data reveals striking similarities between high- $^3\text{He}/^4\text{He}$ OIB and MORB in terms of their trace-element and Pb-Nd-Sr isotope ratios. In addition, global OIB show systematic variability of incompatible element abundances and He-Pb-Sr-Nd isotope ratios, which reflect variable contributions from recycled crust. A model of incomplete degassing for the mantle that allows for the isolation of plume sources reproduces the $^3\text{He}/^4\text{He}$ ratios of MORB and OIB. We conclude that the helium isotope evolution of the mantle does not require the existence of a primordial, undegassed high- $^3\text{He}/^4\text{He}$ mantle reservoir but rather the high- $^3\text{He}/^4\text{He}$ component of OIB represents mantle that has been subject to depletion in incompatible elements by the formation of continents and ocean crust throughout Earth history.

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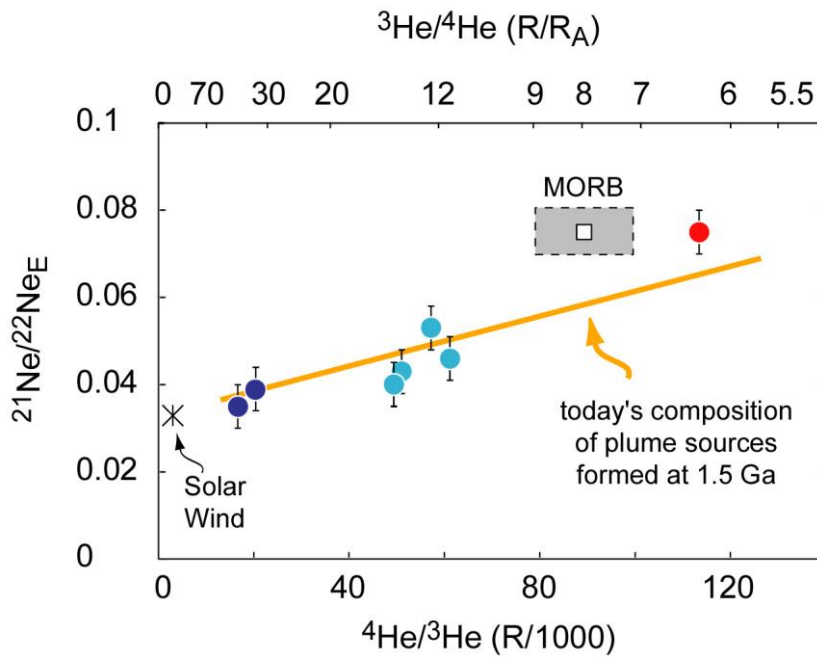
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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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Supplementary Figure 1: Neon-helium isotope relationships in OIB and MORB. $^{21}\text{Ne}/^{22}\text{Ne}_E$ ratios (E: extrapolated; i.e. the $^{21}\text{Ne}/^{22}\text{Ne}$ ratio of a sample suite after correction for air contamination) for Iceland^{40,41} and Loihi⁴⁴ of the “high $^3\text{He}/^4\text{He}$ ” group (dark blue circles), Réunion^{45,46}, Discovery^{38,47}, Shona^{38,47} and Kerguelen³⁹ of the “moderately high $^3\text{He}/^4\text{He}$ ” group (light blue circles), the Cameroon Line¹² of the “low $^3\text{He}/^4\text{He}$ ” group of OIB (red circle), and MORB (open square and grey field)^{48,49}. The Ne-He isotope correlation of 1.5 Ga-old plume sources derived from the helium evolution model of the mantle (orange line, see text) is consistent with the Ne-He isotopic compositions of OIB. Thus, neon isotope data are consistent with the proposed helium evolution of the mantle. Discovery and Shona plume influenced MORB samples show large range in $^{21}\text{Ne}/^{22}\text{Ne}_E$ ratios but consistent values for samples with high $^3\text{He}/^4\text{He}$ ratios, which is plotted here. This figure follows previous presentations (for example fig.18 of ref.29) with the modification that only the $^3\text{He}/^4\text{He}_{\text{max}}$ for individual plumes (as used in this paper) are plotted in order to clarify the correlation between ^{21}Ne and ^4He based on radiogenic production from Th and U decay in plume sources.

Supplementary Information

Global data compilation. Only alkali basalts, basalts, tholeiites, picrites and equivalents from the shield-building stage of ocean islands are included. This omits effects from fractional crystallization in evolved rocks and from very low degrees of partial melting in basanites, nephelinites and equivalents. In addition, samples with low precision Th concentration data, mostly by XRF were omitted, as well as those with strong negative or positive Eu anomalies. Ocean islands are grouped based on their highest $^3\text{He}/^4\text{He}$ ratio as measured on olivine mineral separates obtained by crushing. Samples with very low He $\ll 40$ nano ccSTP/g, which have been documented to be prone to crustal contamination ¹, were excluded. The grouping of OIB as “high $^3\text{He}/^4\text{He}$ ” and “low $^3\text{He}/^4\text{He}$ ” has been discussed over the last 20-plus years primarily in terms of $^3\text{He}/^4\text{He}$ as R/R_A ratios ², which is the notation adopted in this paper, although radioactive decay products are logically discussed as daughter/reference isotope ratios (i.e. $^4\text{He}/^3\text{He}$ ³). OIB are from the GEOROC database (<http://georoc.mpch-mainz.gwdg.de/>) and the helium database of Abedini and Hurwitz ⁴, and MORB are from the PetDB database (<http://www.petdb.org>). For MORB, only analyses on fresh glasses were considered. Like Salters and Stracke ⁵ we use only samples erupted in water depths of greater than 2000 m, which filters out strongly plume influenced samples. Shona and Discovery plume anomalies ⁶ and southern Chile ridge data ⁷ were excluded. For diagrams including Th concentrations (main text, Figures 2b, 2c, 3) only samples with 5wt.% < MgO < 15wt.% were plotted to exclude samples where Th contents might have been concentrated through fractional crystallization, or diluted through crystal accumulation. Low degree melts from Tristan da Cunha and Cape Verde islands were not included.

Data groups: The following references include the helium data forming the basis of the OIB groupings. “Low $^3\text{He}/^4\text{He}$ ” OIB (Fig. 1-2, text, red symbols) include Austral-Cook Islands Tubuaii and Mangaia ⁸, Tristan da Cunha and Gough islands ^{2,9} (basanites from Tristan da Cunha are thought to represent the plume source compositions, as supported by similar

compositions of alkali basalts from Inaccessible ¹⁰), the Grande Comore plume endmember ^{9,11}, Cameroon Line island São Tomé ¹², Azores island São Miguel samples with ²⁰⁶Pb/²⁰⁴Pb ≥ 19.7 ¹³. “MORB-like ³He/⁴He” OIB (yellow symbols) include Austral-Cook islands Rarotonga and Rurutu younger series ⁸, Heard Island Big Ben volcano ¹⁴, Pitcairn-Gambier chain with ²⁰⁶Pb/²⁰⁴Pb < 18 ¹⁵, Canary Islands ¹⁶, Cape Verde southern islands ¹⁷, Society seamounts Tehetia, Rocard ¹⁸. “Moderately high ³He/⁴He” OIB (light blue symbols) include Pitcairn-Gambier chain with ²⁰⁶Pb/²⁰⁴Pb > 18 ¹⁵, Cape Verde northern islands ¹⁷, Society seamount Mehetia ¹⁸, Réunion ¹⁹, Azores including São Miguel with ²⁰⁶Pb/²⁰⁴Pb < 19.7 ¹³. “High ³He/⁴He” OIB (dark blue symbols) include Hawaii, ²⁰, Iceland ²¹, Galapagos ²², Juan Fernandez ²³. In this group Samoa ²⁴ is an exception and marked with different symbols (Fig. 1-2, text, blue open circles).

Model parameters and calculation. We use ${}^3\text{He}/{}^4\text{He} = 230 \cdot R_A$ ²⁵ for the initial “un-degassed” (i.e. primitive, never melted) Earth’s mantle, which is between the inferred initial solar nebula ratio of ${}^3\text{He}/{}^4\text{He} = 120 \cdot R_A$ and the solar wind value of ${}^3\text{He}/{}^4\text{He} = 330 \cdot R_A$, and is relevant if terrestrial He was derived from solar wind implantations on accreting materials ²⁶. The helium abundance in the un-degassed mantle is poorly constrained. It has been estimated based on the assumption that the highest helium isotope ratio measured in OIB reflects the ratio in the present-day un-degassed part of the Earth’s mantle, which then allows calculation of the ³He concentration from the well-constrained U (0.021 ppm) and Th (0.085 ppm) contents of the primitive mantle ²⁷, and which yields ${}^3\text{He} \sim 8 \times 10^{10}$ atoms/g for an initial ${}^3\text{He}/{}^4\text{He} = 120 \cdot R_A$ and a maximum OIB ${}^3\text{He}/{}^4\text{He} = 37 R_A$ ²⁶. Higher ³He abundances of $\sim 10^{12}$ atoms/g have been inferred for a D” layer formed from accreted smaller bodies with high concentrations of helium through implantation of solar wind ²⁸. Thus, the concentration used for this model calculation, with ${}^3\text{He} = 1.1 \times 10^{11}$ atoms/g, lies well within the range of previously used abundances. The ${}^3\text{He}/{}^4\text{He}$ ratio of the Earth’s mantle evolves to lower values through the continuous production of radiogenic ⁴He(rad,t) following the equation ²⁹:

$${}^4\text{He}(\text{rad},t) = {}^{238}\text{U} \cdot \{ 8 \cdot (\exp(\lambda_{238} \cdot T) - \exp(\lambda_{238} \cdot t)) + (7/137.88) \cdot (\exp(\lambda_{235} \cdot T) - \exp(\lambda_{235} \cdot t)) \}$$

$$\exp(\lambda_{235} * t) + 6 * (^{232}\text{Th} / ^{238}\text{U}) * (\exp(\lambda_{232} * T) - \exp(\lambda_{232} * t)) \quad (1),$$

where λ_{238} , λ_{235} and λ_{232} are the decay constants for ^{238}U , ^{235}U , and ^{232}Th respectively. The model starts at time $T = 4.4$ Ga after the first initial stage of degassing during accretion, which lasts ~ 100 Ma as constrained by xenon isotopes²⁶. “ t ” is the time before present day (Figure 4 in text). For our calculation, continent formation is assumed to follow a linear growth curve from 4.4-2.7 Ga, at which time U and Th of the depleted mantle are assumed to reach present-day MORB mantle values⁵. This is clearly an oversimplification of the history of mantle differentiation. As discussed in the text, the mean age of the continental crust is probably closer to 2 Ga, but the lower age reflects recycling of continent to the mantle and the older one allows for sequestering of significant U and Th in the continental crust over the first half of Earth history. In the model the depleted mantle is assumed to degas continuously through the formation of oceanic crust. U and Th concentrations are maintained through mixing of recycled ocean crust and sediment back into the mantle. We assume that at present day the depleted mantle degasses ^3He at a rate of 1060 mol/a as measured in the oceans^{30,31} and that the degassing of helium has decreased linearly since 4.4 Ga.

This simple model, which illustrates that whole mantle incomplete degassing can explain the helium isotope signature in both OIB and MORB, reflects our understanding that two main factors govern the helium degassing rate and isotopic evolution in a mantle undergoing whole mantle convection: (1) the ocean crust formation rate, and (2) the concentration of helium in the mantle. The present-day degassing rate is well constrained through measurements on ^3He in the oceans^{30,31}. This degassing rate is a function of the helium abundance in the present-day upper mantle and today’s ocean crust formation rate. Ideally the degassing rate in the past should be modelled as a function of the past ocean crust formation rate as well as the changing helium concentration due to degassing and radioactive ingrowth. However, the initial mantle helium concentration is poorly constrained, and the present-day concentration in the MORB mantle is uncertain due to the degassed nature of basaltic samples, unknown partition coefficients and possibly fast diffusion of helium³². The ocean crust formation rate is constrained over the past ~ 180 Ma and varies by some 15-30%

³³. Past formation rates may be higher or lower than today. For example, during the early Earth a thick basaltic crust could have dominated due to hotter temperatures and higher degrees of melting, with accordingly low ocean crust formation rates and degassing rates (recently reviewed in ref. ³⁴). Thus, degassing rates through geological time are under-constrained. The most solid constraint is the degassing rate of the oceanic mantle today ³¹, which in our model needs to be adjusted with more vigorous degassing 4.4 Ga ago by a factor of 34 (combined effect of past ocean crust formation rates and higher helium contents) to generate a present-day $^3\text{He}/^4\text{He} \approx 8R_A$ in MORB.

In order to model degassing in the past we use the flux of ^4He , which we derive for the present-day from the flux of ^3He into the oceans and its $^3\text{He}/^4\text{He} \approx 8R_A$. Because $^4\text{He}(\text{rad})$ is continuously formed through radioactive decay, the $^3\text{He}/^4\text{He}$ of the depleted mantle is changing and therefore $^3\text{He}(\text{degas}, t)$ is changing, such that

$$^3\text{He}(\text{degas}, t) = (^3\text{He}/^4\text{He})_t * ^4\text{He}(t) \quad (2),$$

with

$$(^3\text{He}/^4\text{He})_t = \frac{^3\text{He}(T) - ^3\text{He}(\text{degas}, t)}{^4\text{He}(T) + ^4\text{He}(\text{rad}, t) - ^4\text{He}(\text{degas}, t)} \quad (3),$$

with $T=4.4$ Ga. Because $(^3\text{He}/^4\text{He})_t$ is on both sides of the equation, $(^3\text{He}/^4\text{He})_{t+\Delta t}$ was used to calculate $^3\text{He}(\text{degas}, t)$ with $\Delta t=5\text{Ma}$. The solution to the model is found through adjusting f using a classical shooting method.

Plume source formation ages. Based on this general degassing model, ages of plume sources

can be made a dependent variable and calculated. Average Th contents and Th/U ratios of the He isotope groups of OIB were calculated based on the global data compilation and give:

“Low $^3\text{He}/^4\text{He}$ ” – Th=5.6 ppm, Th/U=4.4; “MORB-like $^3\text{He}/^4\text{He}$ ” – Th=4.7 ppm, Th/U=3.7;

“moderately high $^3\text{He}/^4\text{He}$ ” – Th=3 ppm, Th/U=4.0; “high $^3\text{He}/^4\text{He}$ ” – Th=1 ppm, Th/U=3.3;

for the Baffin Island endmember Th and U data are not published yet and we assumed

Th=0.02 ppm and Th/U=3.3 (same Th/U as in the “high $^3\text{He}/^4\text{He}$ ” group). We calculate Th

and U contents of OIB sources assuming that lavas reflect 6% melts of peridotite. Plume sources contain variable amounts of recycled components of variable compositions³⁵. For simplicity a constant volume approach is used here, because it fits the global variability of present-day subducting oceanic crust plus variable sediments³⁶ and allows calculation of the Th+U content and ⁴He production rate of the recycled components. Reasonable Th,U abundances for the recycled component indicate that the recycled component constitutes 40% of the source by mass, 60% are formed by depleted mantle. For each He isotope group of OIB, the production rate of ⁴He and the ³He/⁴He ratios can be calculated as a function of the formation age.

Neon isotopes. Neon isotope ratios provide a test for our hypothesis that ³He/⁴He ratios in OIB reflect different production rates of radiogenic ⁴He. Radiogenic production of ²¹Ne is directly proportional to the production of ⁴He (α particles), as it is driven by neutrons produced by α,n reactions (i.e. ¹⁸O(α,n)²¹Ne, and to a much lesser extent ²⁴Mg(n,α)²¹Ne). The production ratio has been estimated as $^{21}\text{Ne}/^4\text{He} = 4.5 \times 10^{-8}$ ³⁷. Thus, ²¹Ne/²²Ne is proportional to the inverse of the ³He/⁴He ratio after correcting for atmospheric contamination of Ne in oceanic basalts, using the so-called extrapolated ²¹Ne/²²Ne_E (where E – extrapolated; that is, ²¹Ne/²²Ne_E is determined through linear extrapolation through the data and air to a solar ²⁰Ne/²²Ne ratio³⁸, which gives one value of ²¹Ne/²²Ne_E for a sample suite from an individual mantle plume). The oceanic basalt Ne data set is still very limited. We include mantle xenolith data from Kerguelen and the Cameroon Line^{12,39}, which is problematic because it is not established whether those data represent the plume compositions, however, neon data on “MORB-like” and “low” ³He/⁴He group OIB have not been measured yet. Shona and Discovery plume-influenced MORB show contributions of ambient depleted upper mantle, and ²¹Ne/²²Ne_E were calculated for the samples with the highest ³He/⁴He and lowest ²¹Ne/²²Ne_{measured} ratios to get the best estimate for these plumes. Available data show that ²¹Ne/²²Ne_E increases in OIB with decreasing ³He/⁴He (Supplementary Figure 1). Thus, ²¹Ne/²²Ne ratios follow the same general systematics as helium isotopes in OIB, supporting our conclusion that variations in these ratios reflect different production rates of radiogenic

^4He and ^{21}Ne as a direct function of the Th and U content of plume sources. In contrast to helium, $^{21}\text{Ne}/^{22}\text{Ne}_E$ in “high $^3\text{He}/^4\text{He}$ ” group OIB from Iceland reach the solar value^{40,41}, which suggests $\text{Ne}/(\text{Th}+\text{U})$ in this group of OIB is sufficiently high that radiogenic ^{21}Ne does not change the $^{21}\text{Ne}/^{22}\text{Ne}_E$ relative to the solar value.

If we assume a solar $^{21}\text{Ne}/^{22}\text{Ne}_E$ for the high $^3\text{He}/^4\text{He}$ endmember of OIB, we can apply the helium evolution model (Figures 3 and 4 in the main text) to neon isotopes by using the average plume source formation age of 1.5 Ga, which defines the $^3\text{He}/^4\text{He}$ ratio of the high $^3\text{He}/^4\text{He}$ endmember of OIB. The $^{21}\text{Ne}/^{22}\text{Ne}_E$ as a function of the He isotope ratio of OIB is then calculated based on the $^{21}\text{Ne}/^4\text{He}$ production ratio. The modelled Ne-He isotope correlation of plume sources formed at 1.5 Ga is generally consistent with measured Ne-He isotope ratios of OIB (Suppl. Fig. 1, orange line). The present-day $^{21}\text{Ne}/^{22}\text{Ne}_E$ ratios of the depleted mantle isolated from convection 1.5 Ga ago is given by the low end of the line. As expected for a continuously degassing mantle, present-day MORB has a higher $^{21}\text{Ne}/^{22}\text{Ne}_E$ ratios and lower $^3\text{He}/^4\text{He}$ ratios than the high $^3\text{He}/^4\text{He}$ endmember of plume sources. In principle, neon isotopes could be modelled as described for helium isotopes in the main text, however, due to unknown degassing rates and indications for a decoupling of neon and helium isotopes^{42,43} we did not yet attempt to model neon in detail. Nevertheless, the data show that the neon isotope systematics is generally consistent with the helium evolution of the mantle.

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