

Are high $^3\text{He}/^4\text{He}$ ratios in oceanic basalts an indicator of deep-mantle plume components ?

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Abstract

The existence of a primordial, undegassed lower mantle reservoir characterized by high concentration of ^3He and high $^3\text{He}/^4\text{He}$ ratios is a cornerstone assumption in modern geochemistry. It has become standard practice to interpret high $^3\text{He}/^4\text{He}$ ratios in oceanic basalts as a signature of deep-rooted plumes. The unfiltered He isotope data set for oceanic spreading centers displays a wide, nearly Gaussian, distribution qualitatively similar to the Os isotope ($^{187}\text{Os}/^{188}\text{Os}$) distribution of mantle-derived Os-rich alloys. We propose that both distributions are produced by shallow mantle processes involving mixing between different proportions of recycled, variably aged radiogenic and unradiogenic domains under varying degrees of partial melting. In the case of Re-Os isotopic system, radiogenic (MORB-rich) and unradiogenic (depleted mantle residue) endmembers are constantly produced during partial melting events. In the case of the (U+Th)-He isotope system, effective capture of He-rich bubbles during growth of phenocryst olivine in crystallizing magma chambers provide one mechanism for “freezing in” unradiogenic (i.e. high $^3\text{He}/^4\text{He}$) He isotope ratios, while the higher than chondritic (U+Th)/He elemental ratio in the evolving and partially degassed MORB melt provides the radiogenic (i.e. low $^3\text{He}/^4\text{He}$) endmember. If this scenario is correct, the use of He isotopic signatures as a fingerprint of plume components in oceanic basalts is not justified.

Introduction

An increasingly large body of isotopic and trace-element analyses of MORBs demonstrates that the upper mantle is not homogenous but contains several distinct geochemical domains on a variety of length scales [e.g., 1-7]. However, the physical properties of these domains, i.e. their exact location, size, temperature and dynamics, remain largely unconstrained. This is in part because plate tectonics creates a great variety of heterogeneities including stratified subducted oceanic crust, subducted sediments, continental crust, and the residua from melting beneath arcs, ridges, and oceanic islands.

Perhaps the most important example of an inferred genetic relationship between a geochemical signature and a specific geophysical phenomenon is provided by He isotopes and conjectured deep-rooted mantle plumes. Preponderance of opinion among geochemists is that distinct He isotopic signatures (high $^3\text{He}/^4\text{He}$ ratios) are associated with certain locations, such as Hawaii, Yellowstone and Iceland that, via proposed deep-rooted mantle plumes, are linked to primordial and undegassed, ^3He -rich reservoirs isolated in the lower mantle. In this interpretation, the He isotopic signatures of “hotspot” basalts provide geochemical evidence for large scale stratification of the mantle and the presence of an “undegassed, primordial lower mantle reservoir” [e.g., 8-10].

It has recently been proposed that the He isotopic signatures of Yellowstone and Iceland, and perhaps all hotspots, are acquired in the shallow mantle [11-16]. Here we argue that He isotopic signatures can not be used as a reliable indicator for the presence of a plume or “deep” component in oceanic basalts. Therefore the He isotopic data provide no evidence for an isolated, primordial lower mantle reservoir. This leaves us

without a geochemical signature of the purported “undepleted and undegassed” lower mantle, and perhaps such a reservoir does not exist.

The He isotopic signature of the mantle

The $^3\text{He}/^4\text{He}$ ratio of a closed mantle reservoir continuously evolves toward lower values as ^4He is produced by the decay of U and Th. It therefore depends on several factors, including the He concentration, $(\text{U}+\text{Th})/^3\text{He}$ ratio, time (or age), and mixing between components with different evolution trajectories [17].

Certain volcanic chains are associated with long-lived volcanism that produces Oceanic Island Basalt (OIB). Some OIBs, but certainly not all, are characterized by $^3\text{He}/^4\text{He}$ ratios in the range of 9 to 42 R_A ; where R_A is the present day atmospheric $^3\text{He}/^4\text{He}$ ratio of 1.39×10^{-6} [8, 18]. In contrast, consensus has developed that MORBs have extremely homogeneous $^3\text{He}/^4\text{He}$ ratios in the narrow range of $8 \pm 1 R_A$ [8, 15]. This has created a strong tradition among geochemists for inferring the presence of deep-rooted mantle plumes whenever $^3\text{He}/^4\text{He}$ ratios in excess of ~ 9 or $10 R_A$ are measured in oceanic basalts; the underlying assumption is that high $^3\text{He}/^4\text{He}$ ratios are a signature of an undegassed lower mantle reservoir [8].

However, a number of key observations are inconsistent with this prevailing view of mantle He isotopic signatures.

- i)* Whereas OIBs are characterized by a wide range of $^3\text{He}/^4\text{He}$ ratios from 1 to 42 R_A MORBs are, partly as a result of strict *a priori* data filtering, thought to be characterized by a narrow range from 7-9 R_A [e.g. 2, 8, 14, 15, 19, 20]. However, Anderson [15, 20] has demonstrated that the He isotopic signatures of OIBs and

unfiltered basalts from oceanic spreading centers (Fig. 1) with 95% confidence are drawn from the same statistical population. Based on He isotope systematics of oceanic basalts there is therefore no requirement for the existence of two separate mantle reservoirs; i.e. an upper, degassed *vs.* a lower undegassed mantle.

ii) OIBs have substantially lower He concentrations than MORBs [8, 21]. This is in direct contradiction with the notion that the MORB source regions are thoroughly degassed whereas OIBs derive their noble gas inventory from an undegassed part of the mantle [e.g., 8-10]. This problem has frequently been referred to as one of the “He-paradoxes” [e.g. 14]. However, recent data sets point to an important role for degassing as a function of eruption depth in defining the He concentration of oceanic basalts. For example, Hilton *et al.* [22] demonstrated a positive correlation between He concentration in MORB glasses from the Reykjanes Ridge and sampling depth. These authors also discussed the effect of H₂O content on the solubility of volatiles in basaltic magmas and argued that local variations in water content of the magmas can partly mask the correlation between He concentration and depth of eruption, which might explain the large scatter in their data set (see Supplementary Information). Furthermore, based on analyses of He, Ne, Ar and CO₂ in basaltic glasses from the Southeast Indian Ridge and the Amsterdam-St. Paul Plateau, Burnard *et al.* [23] showed that fractional crystallization plays an important role in determining the fraction of gas lost from an erupting magma. Collectively, these findings indicate that degassing does play an important role in defining the He concentration of basaltic glasses. Perhaps the sampled OIBs, which generally erupted at shallow depths, did originally have He

concentrations similar to average MORBs. However, there is no basis in the existing data sets for concluding that OIBs have generally higher He concentrations than MORBs; the suggested correlations upon which such inferences are based [22] are not statistically robust (see Supplementary Information). The measured He concentrations therefore do not provide evidence for the existence of an undegassed reservoir in the lower mantle.

- iii)* Many hotspot volcanic systems display a distinct temporal evolution of their $^3\text{He}/^4\text{He}$ ratio. An example is Mauna Loa, Hawaii, where drill core samples show $^3\text{He}/^4\text{He}$ ratios decreasing from 18-20 R_A about 250,000 years ago to 8-9 R_A in modern basalts [24]. Such a dramatic temporal evolution in the $^3\text{He}/^4\text{He}$ ratio is not expected if He is funneled to the surface from the deep, undegassed mantle by a stable, long-lived plume.
- iv)* Young ridges, ridges developed in back-arc settings or by expansion of existing ridge systems, and near-axis seamounts tend to have higher and significantly more variable $^3\text{He}/^4\text{He}$ ratios than mature, steady-state ridges. Abandoned ridge systems have even lower $^3\text{He}/^4\text{He}$ ratios [15, 19]. In general, areas with some high $^3\text{He}/^4\text{He}$ samples also exhibit extreme variation in $^3\text{He}/^4\text{He}$ ratios.

Collectively, these observations are not consistent with the standard model, which invokes a deep mantle origin of He characterized by systematically high $^3\text{He}/^4\text{He}$ ratios (tapped by OIBs) and a shallow mantle origin of He characterized by very homogeneous and relatively low $^3\text{He}/^4\text{He}$ ratios (tapped by spreading ridges). The MORB and OIB He

isotope data are better understood in context of a model which involves mixing between radiogenic and unradiogenic components, perhaps in a plum-pudding or marble-cake configuration in the shallow upper mantle. Recently obtained Os isotope data (Fig. 1) [4] and strong petrologic evidence for the capture of He and other volatiles as bubbles in olivine phenocrysts during crystallization of basaltic melts [25] provide an improved basis for this interpretation, as discussed below.

The Os isotopic signature of the upper mantle

The Re-Os isotopic system is ideally suited for tracing the extraction and subduction of basalts to the upper mantle [26]. During partial melting Re is mildly incompatible whereas Os is strongly compatible resulting in high Re/Os elemental ratios in basalts and correspondingly low Re/Os ratios in the refractory, depleted solid residue left behind in the mantle [26]. As ^{187}Re decays to ^{187}Os ($T_{1/2}$ ca. 42 billion years) the $^{187}\text{Os}/^{188}\text{Os}$ ratios of basalt and refractory, depleted mantle residue will diverge. Basalts develop highly radiogenic $^{187}\text{Os}/^{188}\text{Os}$ ratios while the depleted mantle residues develop relatively low, unradiogenic $^{187}\text{Os}/^{188}\text{Os}$ ratios (Fig. 2).

Figure 1 shows the distribution of $^{187}\text{Os}/^{188}\text{Os}$ from more than 700 Os-rich millimeter-sized platinum group element alloys derived from a variety of tectonized peridotite bodies in northwestern California and southwest Oregon [27], USA. These grains are derived from a large number of peridotite bodies and are expected to represent a less biased sample of the upper mantle than samples derived from a single, localized peridotite source [4]. The measured $^{187}\text{Os}/^{188}\text{Os}$ isotope ratios span a wide range from extremely unradiogenic values to radiogenic values, as high as 0.1870 (the most

radiogenic values are not included in Fig. 1). The bulk of these Os isotope data forms a nearly Gaussian distribution, centered around $^{187}\text{Os}/^{188}\text{Os} \sim 0.1245$. As discussed by Meibom et al. [4], the width and Gaussian shape of the Os isotope distribution in Fig. 1 supports the view that the upper mantle is characterized by substantial, long-lived Os isotope heterogeneity [28-32]. Among the more than 700 Os-rich grains the least radiogenic sample has a $^{187}\text{Os}/^{188}\text{Os}$ ratio of 0.1095, corresponding to a Re-depletion age of more than two billion years [4]. This provides an indication of the time scales on which Os isotope heterogeneity can survive in the mantle, in agreement with previous results [e.g. 28-32]. Each Os-rich grain in the distribution (Fig. 1) is homogeneous in its Os isotopic composition and contains Os equivalent to an upper mantle volume on the order of 1 m^3 , assuming an Os concentration of 5 ppb and a density of 3.3 g/cm^3 for the upper mantle [4]. Therefore, homogenization of preexisting Os isotopic heterogeneities has occurred on a minimum length scale of meters, which rules out solid state diffusion as the dominant process [29, 30, 33] (observations of substantial Os isotope heterogeneity in abyssal peridotites on length scales of 10-100 meters [28, 31] might be interpreted as the characteristic length scale for upper mantle Os isotope heterogeneity).

Instead, the Gaussian $^{187}\text{Os}/^{188}\text{Os}$ distribution in Fig. 1 is the expected outcome of random mixing between ancient enriched (high $^{187}\text{Os}/^{188}\text{Os}$) and depleted (low $^{187}\text{Os}/^{188}\text{Os}$) mantle domains, conceivably as a result of metasomatic melt-rock reactions in regions of relatively high degrees of partial melting [e.g., 4, 34, 35]. Where partial melting occurs, for example at mid-ocean ridges, the enriched domains melt first. The melts so produced percolate upwards through an increasingly dense network of grain-scale pores and fractures and react with solid mantle material at shallower depths and

even higher degree partial melt [34-36]. Here ancient, unradiogenic Os-rich mantle sulfides (encapsulated in silicate and spinel crystals), the main carrier of Os in the solid mantle, are capable of equilibrating with more radiogenic Os in the melt [29-31, 33]. A Gaussian Os isotope distribution (Fig. 1) is the expected result of such a random mixing processes. The width of the distribution and the $^{187}\text{Os}/^{188}\text{Os}$ ratios of the most depleted samples (down to 0.1095) indicate that away from mid-ocean ridges, or in regions with low degrees of partial melting, equilibration between enriched and depleted domains is extremely inefficient. In terms of its Re-Os isotopic systematics, the upper mantle can therefore be envisioned as a heterogeneous mixture of endmembers on a characteristic length scale of less than a kilometer [5].

Partial melting of a heterogeneous mantle – implications for He

As shown in Fig. 1, the compiled He isotope data from MORBs form a distribution very similar to the $^{187}\text{Os}/^{188}\text{Os}$ distribution and would be consistent with a qualitatively similar mixing model between unradiogenic (high $^3\text{He}/^4\text{He}$) and radiogenic (low $^3\text{He}/^4\text{He}$) components [13, 15]. In the following we explore such a model and discuss how a mechanism for strongly polarizing the He isotopic composition of different mantle lithologies as a result of partial melting, degassing, and entrapment of He-rich fluids as bubbles filling cavities in phenocryst olivine forming during the early stages of crystallization of basaltic magmas [25].

The He/(U+Th) and Re/Os isotope systems have one important characteristic in common, namely that parent and daughter isotopes easily separate during magmatic

processes such as partial melting and degassing. In this respect these two isotope systems differ from the isotope systems of large-ion lithophile elements (e.g. Sm-Nd and Rb-Sr). Above, we discussed the polarization of Os isotopic evolution of two important mantle lithologies in the upper mantle; depleted residue and MORB-rich domains. Mixing of Os from these two lithologies during partial melting events in the upper mantle will produce a Gaussian distribution such as that observed in Fig. 1. However, there is no evidence from available data that the Os and He isotopic systems in oceanic basalts are coupled (see Supplementary Information). This indicates that the two systems evolve independent of each other, likely as constituents in different mantle lithologies. If, as we suggest, the Gaussian He isotopic distribution in Fig. 1 is the result of a similar mixing between radiogenic and unradiogenic gas components brought together during partial melting in the upper mantle there must be a different mechanism for separating the parent isotopes (U and Th) from the daughter (^4He) to allow an isotopic polarization of the mantle He reservoirs to build up over time.

Measurements of He isotopic compositions in MORBs are done by carefully crushing the samples to release He trapped in vesicles. The parent elements of ^4He (U and Th) are sited in the glassy matrix, from which only minor amounts of radiogenic ^4He is released. Therefore, the measured $^3\text{He}/^4\text{He}$ ratios most likely represent magmatic (source) compositions, with little possibility of post-eruptive contribution of radiogenic ^4He from the glassy matrix [e.g. 22]. In other words, the isotopic composition of the He released from the vesicles reflects the averaged contribution of He from different components, which participated in the melting process and contributed to the generation of the MORB.

During the generation of MORB, several processes are at work. Fractions of the upper mantle melt, and the melt migrates upwards, often to pond in shallow magma chambers and mix with other melts prior to eruption. Here several important processes take place. Because of the generally high U and Th concentrations and the variable loss of He through magma outgassing the $(U+Th)/^3\text{He}$ ratio of the melt becomes significantly higher than that of the starting material. Erupted MORB therefore evolves relatively quickly towards low, radiogenic $^3\text{He}/^4\text{He}$ ratios [e.g., 2, 13, 22, 37]. Outgassing of erupting basalts is, however, far from complete [25]. At shallow depth, magma partly exsolves CO_2 and He, which can become trapped in fluid inclusions or bubbles incorporated into refractory solids traversed, or left behind, by the ascending magma. Natland [25] has documented the effectiveness with which olivine phenocrysts, an early forming solid phase during fractional crystallization of a cooling magma, can overgrow He-rich bubbles trapped in cavities and nucleating on the surfaces of the growing olivine crystals. These volatile-rich inclusions have low $(U+Th)/^3\text{He}$ ratios and will therefore be “frozen in” at relatively high, unradiogenic $^3\text{He}/^4\text{He}$ ratios [37, 38]. This unradiogenic He in the depleted solid residue can not effectively escape because diffusion of He through mantle minerals is sluggish. Estimates of the characteristic diffusion length of He under typical mantle conditions (i.e. ca. 1400 °C) is less than one kilometer for the entire lifetime of the Earth [17, 39]. Hence, on smaller timescales unradiogenic He is easily isolated in sub-kilometer sized domains of the upper mantle [17], and will only be released upon renewed partial melting.

Thus, melting, partial degassing and capture of He and other volatiles during growth of early forming olivine phenocrysts provide an obvious mechanism for strongly

polarizing the He isotopic composition of the upper mantle. MORB with its generally higher (U+Th)/³He elemental ratio will, over time, develop radiogenic (i.e. low ³He/⁴He) ratios. Recycling of MORB and aged MORB-rich domains therefore provide a radiogenic He-isotopic endmember in the upper mantle. At the same time, lithologies rich in early cumulate phases, notably phenocryst olivine, have the potential of preserving unradiogenic ³He/⁴He ratios that provide an unradiogenic He isotopic endmember in the upper mantle (Fig. 2).

Thus, by the same mechanism, i.e. partial melting on small (sub-km) length scales of a heterogeneous mixture of radiogenic and unradiogenic domains of different age in the shallow mantle, a Gaussian distribution is also expected for the global ³He/⁴He data set (Fig. 1). This “dispersed component” model can explain several features of the global He isotope data set that appear inconsistent with the conventional “two-reservoir” model.

In our model, MORBs and OIBs are samples of the qualitatively same population of upper mantle heterogeneities [15, 20]. Under conditions of high degrees of partial melting, such as in mid-ocean ridge settings, relatively large volumes of the upper mantle are blended. The He isotope signatures of MORB samples are therefore expected to show less variation (i.e. have a better defined average value) than OIBs, which sample mantle settings with much smaller degrees of partial melting, and probably less volume of the mantle, and, hence, represent a smaller source volume. This is a consequence of the Central Limit Theorem according to which independent averaging of samples from any given distribution will approach a Gaussian distribution with a variance that becomes smaller as the sampled volume increases [13, 14, 19]. Near-axis seamounts, which are

products of low degrees of partial melting, consistently display high variability in $^3\text{He}/^4\text{He}$ ratios and trace element abundances [15, 19, 40, 41].

Thus, much of the $^3\text{He}/^4\text{He}$ variability in MORBs routinely ascribed to “plume contamination” is in fact the result of different mixing proportions of radiogenic (MORB-rich) and unradiogenic mantle components (e.g. lithologies rich in phenocryst olivines), perhaps in combination with variable degrees of partial melting. The temporal evolution observed among ridge systems, from variable and high $^3\text{He}/^4\text{He}$ ratios in young ridges to less variable and lower $^3\text{He}/^4\text{He}$ ratios in mature ridges, as well as the dramatic temporal evolution in $^3\text{He}/^4\text{He}$ ratios observed in certain OIBs [e.g., 24] are the expected outcome of changing mixing proportions or degrees of melting among the various shallow components with time.

Conclusions and implications

The striking similarity between the He and Os isotopic signatures of samples from the upper mantle (Fig. 1) lead us to suggest a different framework for interpreting He isotopic systematics of oceanic basalts. The generally Gaussian shape of both distributions is not consistent with a stratified, two-reservoir mantle with MORBs derived from “the degassed upper mantle” and OIBs derived from “the deep, primordial and undegassed mantle”. However, they are consistent with a model in which mixing between different proportions of ancient radiogenic and unradiogenic domains, e.g. in a plum pudding configuration, takes place under varying degrees of partial melting in the shallow upper mantle. Extreme isotopic ratios are expected to exist in small scale sampling of a heterogeneous reservoir (OIBs) while large scale sampling or melting of

the same population gives less variance and a tighter Gaussian distribution (MORBs). This is a consequence of the law of large numbers, sampling theory, and, in particular, the Central Limit Theorem. The systematic use of He isotopic signatures as a fingerprint of deep-rooted plume components in oceanic basalts is therefore not justified. This eliminates the only existing geochemical signature of the alleged primordial and undegassed lower mantle. At the same time, there is abundant cosmochemical evidence from meteorites that the Earth accreted in a hot environment in the inner solar system [42] and from partially molten and degassed planetesimals [43]. We therefore hypothesize that a primordial, undegassed lower mantle reservoir does not exist. If our model is correct, drastic revisions in the current views of mantle stratification and evolution are required.

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Figure 1. Compiled He isotopic distribution of unfiltered, global spreading ridge data, the vast majority being MORB samples with a few back-arc basin and near-ridge seamount samples included [15, and references therein] (left) and Os isotope composition of more than 700 mantle-derived detrital Os-rich platinum group element alloys from tectonized peridotite bodies in a variety of tectonic settings along the coast of northern California and southwest Oregon [4] (right). Both distributions display large variability with a nearly Gaussian peak.

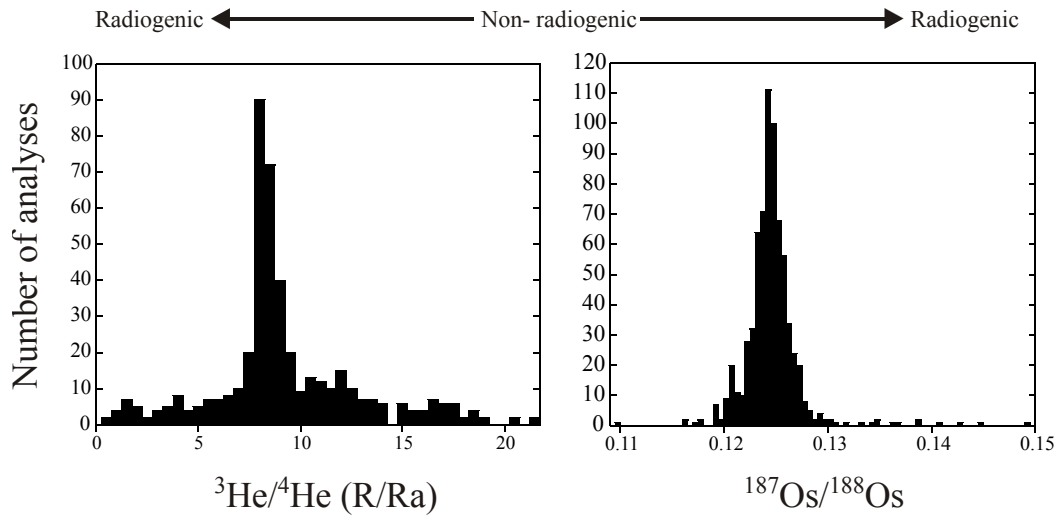


Figure 2. Populations of radiogenic and unradiogenic endmembers contribute to the melt during partial melting of a heterogeneous upper mantle. The resulting mixtures will tend towards a Gaussian distribution, with an average defined by the relative contributions from the endmembers and a width determined by the sampled volume.

