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Rare gas systematics and the origin of oceanic islands: the key role of entrainment at the 670 km boundary layer

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Abstract

Using the best estimates of rare gas isotopic ratios for both Oceanic Island Basalts (OIB) and Mid Oceanic Ridge Basalts (MORB) sources, and knowing the incompatible element ratios between these two sources (e.g. K, U), it is therefore possible to determine the ratios between the concentrations of non-radiogenic isotopes (³He, ²²Ne, ³⁶Ar) in the lower mantle and the upper mantle. The calculations suggest that He, Ne and Ar are enriched by 500 in the lower mantle. Therefore, even a small amount of entrainment of lower layer material in a starting mantle plume can change the isotopic ratios of the plume, producing OIB signatures distinct and more variable than MORB, as is actually observed. Unless the lower mantle is strongly heterogeneous for rare gases, the source of OIB is certainly the 670 km thermal boundary layer, rather than the core–mantle boundary. © 2004 Elsevier B.V. All rights reserved.

Keywords: rare gases; helium; neon; argon; mantle plumes

1. Introduction

Among the radiogenic isotopic tracers used to study the evolution of the Earth's reservoirs, the rare gas group is unique. The five rare gases are linked by their common chemical inertness. On the other hand, because of their different masses, they behave differently during physical processes. Helium escapes from the atmosphere, neon probably escaped in the early days [1], whereas Ar, Kr and Xe are retained quantitatively in the atmosphere by the terrestrial gravitational field. Unlike the lithophile isotopic tracers (e.g. Sr, Nd and Pb), which are linked to the complex process of the formation of the continental crust as well as recycling to the mantle by subduction, rare gases are fractionated compared to their radioactive parents by degassing at the surface. Basalts from different environments show a relatively uniform ⁴He/³He ratio for Mid Oceanic Ridge Basalts (MORB) (~90,000±10,000; *R*/*R*_a=8±1 with R=³He/⁴He and *R*_a

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is the atmospheric ratio) [2,3] and a more variable ratio for Oceanic Island Basalts (OIB) (15,000–200,000; $R/R_a=4-50$) [4–6]. The lowest ⁴He/³He ratios are found in OIB with the lowest ratio being measured in the oldest expression of the Iceland hotspot (15,000, $R/R_a\sim50$) [6]. Those results support the ideas of two distinct reservoirs in the Earth's mantle; one being the source of MORB, the other one being the source of OIB [7–9].

For a long time the ${}^{40}\text{Ar}/{}^{36}\text{Ar}$ ratio was complex to interpret because its extreme sensitivity to degassing and atmospheric contamination (there is 1% argon in air). However, since 1985, reliable uncontaminated results have been obtained on basalt glassy margins [10] with very high ${}^{40}\text{Ar}/{}^{36}\text{Ar}$ ratios (>15,000). Because the ⁴⁰Ar is purely radiogenic in the Earth (⁴⁰Ar/³⁶Ar ratio in lunar-solar exposed-soil is <<1 [11], suggesting an initial low argon ratio for the Earth) and because the 40 K content of the whole Earth is relatively well known, it has been possible to compute a simple budget for ⁴⁰Ar [12]. This budget shows clearly that, because the MORB source has a very low content of 40 Ar (~10⁻⁵ ccSTP/g [9,13]), more than half of the argon budget of the Earth has to be in the lower mantle [12].

Recent measurements of neon have shown that it is a useful tracer for chemical geodynamics [1,14–18]. This tracer shows more clearly than helium the differences in isotopic signature between MORB and OIB [18–24] (Fig. 1) where big hotspots such as Hawaii (Loihi seamount), Iceland or Pitcairn form a trend with a very different slope than MORB in the three isotope neon diagram.

Therefore, from the rare gas point of view, the existence of two separated reservoirs in the mantle is clear. We will not discuss again this point; it is the starting point of the present paper. We will focus on absolute primordial rare gas concentrations in the plume source, which appears to be much higher than in the upper mantle by a factor of several hundred.

The starting observation is the fact that the dispersion in the helium isotopic ratios for OIB is far greater than the dispersion for MORB (filtered from the Schilling's effect that consists in on-ridge hotspots) [3]. We also assume that the source of OIB is deeper than the MORB source. One explanation for the large helium isotopic ratio dispersion is to consider the source of OIB as an extremely heteroge-



Fig. 1. Three neon isotope diagram showing MORB data (black symbols) and the OIB data (dots, Loihi, Iceland and Pitcairn). Grey symbols are MORB samples showing a clear Schilling's effect (e.g., plume–ridge interaction). Data from [13,18,19,21,32,46] and unpublished data from IPGP.

neous lower mantle rather than mixing of a homogeneous material with upper mantle material. One argument against this suggestion is given by the ⁴He/³He⁻²¹Ne/²²Ne correlation observed in n-MORB, E-MORB and OIB that suggests a single homogeneous mantle plume source [18–20,22,25,26].

Therefore, assuming that the lower mantle is relatively uniform for rare gas isotopic compositions, we show that mantle plumes cannot derive directly from the lower mantle. Otherwise, due to the large difference in concentrations, the rare gas signatures would be uniform because contamination by upper mantle material would be negligible. The most likely source for the OIB is the boundary layer a 670 km as previously proposed by Allègre et Turcotte [27] and the lower mantle signal is linked to entrainment phenomena [28,29]. Because it is key for the approach, we start by discussing the isotopic geochemistry of neon.

2. Neon systematics

Neon has three stable isotopes ²⁰Ne ²¹Ne ²²Ne. ²¹Ne variations are easily detectable proportion in mantle derived rocks, and are produced by nuclear reactions of alpha particles, emitted by U and Th radioactive chains, on oxygen and magnesium [30]. Therefore, the ²¹Ne/²²Ne ratio varies similarly to the ⁴He/³He [20]. On the other hand, the ²⁰Ne/²²Ne ratio varies in mantle-derived samples by air addition because the atmosphere ratio (9.8) is different from the mantle ratio, which is assumed to be close to the solar wind ratio (~13.8 [31]). Air addition is related to surface processes and contact with seawater, which contains dissolved atmosphere gases.

Neon isotopic results are illustrated in the three isotope diagram ²⁰Ne/²²Ne–²¹Ne/²²Ne. The results for OIB and MORB in such a diagram clearly show two distinct domains (Fig. 1, top). The dispersion of the slopes is larger for OIB than MORB. Note that hotspot source injections into ridge crests (the Schilling effect), such as observed in Discovery or Shona ridge anomalies show clearly the transition from one domain to the other (Fig. 1, bottom) [18,32].

One of the obstacles to using rare gases isotopes in geodynamics is contamination by the atmospheric gases at the surface. This is particularly severe for ${}^{40}\text{Ar}/{}^{36}\text{Ar}$ measurements. In this context, neon has the advantage that the measured ${}^{21}\text{Ne}/{}^{22}\text{Ne}$ can be corrected for atmospheric contamination by assuming that the ${}^{20}\text{Ne}/{}^{22}\text{Ne}$ ratio is solar in the mantle [18,20,23,33]. In the following discussion, only this air corrected ratio will be used. When measured ${}^{40}\text{Ar}/{}^{36}\text{Ar}$ ratios correlate with ${}^{20}\text{Ne}/{}^{22}\text{Ne}$ ratios, during step heating or crushing experiments, the ${}^{40}\text{Ar}/{}^{36}\text{Ar}$ can be corrected from atmospheric contamination by extrapolating to a solar ${}^{20}\text{Ne}/{}^{22}\text{Ne}$ (and the same for xenon isotopes) [13].

3. Calculations of a reference model

A reference model can be built to allow computation of the different parameters. We will then discuss the possible uncertainties of the reference model and the limitations compared to observations. We assume that the mantle is divided into two reservoirs: the upper and the lower mantles and that the boundary is the 670 km seismic discontinuity. Both reservoirs are assumed to have a uniform isotopic composition.

The goal is to determine the concentration ratios for the stable reference isotopes (³He, ²²Ne, ³⁶Ar) between the upper and the lower mantle. We start with argon.

To determine the concentration ratio of the ³⁶Ar between the upper and lower mantle, one can write the evolution with time of the isotopic ratios using the following equations:

$$\begin{bmatrix} \frac{40}{36} \text{Ar} \\ \frac{36}{36} \text{Ar} \end{bmatrix}_{\text{LM}} = \begin{bmatrix} \frac{40}{36} \text{K} \\ \frac{36}{36} \text{Ar} \end{bmatrix}_{\text{LM}} f(T_{\text{L}})$$

$$\begin{bmatrix} \frac{40}{36} \text{Ar} \\ \frac{36}{36} \text{Ar} \end{bmatrix}_{\text{UM}} = \begin{bmatrix} \frac{40}{36} \text{K} \\ \frac{36}{36} \text{Ar} \end{bmatrix}_{\text{UM}} f(T_{\text{U}})$$

where $T_{\rm L}$ and $T_{\rm U}$ are the characteristic times of the lower mantle and upper mantle, respectively, and f(t) is the radioactive production equations for ⁴⁰Ar from ⁴⁰K.

In general, this function includes a loss term (by degassing). However, to simplify the problem, a closed system approximation with a mean-age for each reservoir will be used. This approach has been previously suggested by several authors [34–37].

A residence time for the upper mantle has been estimated at approximately 1 Ga, which is the meanlife for subducted material in the upper mantle [37]. This approximation assumes that the material used to build new oceanic lithosphere is totally outgassed at ridge crest. This assumption is validated by measurements of rare gases on oceanic crust material, which shows almost no residual mantle-derived rare gases [38,39].

For $T_{\rm L}$ we take 4.5 Gy, the "age" of the Earth (the use of younger ages of 4.4 or 4.3 Gy does not change the results). Using this time scale for the lower mantle assumes implicitly a closed system behavior.

We use the following notation:

$$R_{\rm Ar} = \frac{[{}^{36}{\rm Ar}]_{\rm LM}}{[{}^{36}{\rm Ar}]_{\rm UM}} = \frac{\left({}^{40}{\rm Ar}/{}^{36}{\rm Ar}\right)_{\rm UM}}{\left({}^{40}{\rm Ar}/{}^{36}{\rm Ar}\right)_{\rm LM}} \frac{[{}^{40}{\rm K}]_{\rm LM}}{[{}^{40}{\rm K}]_{\rm UM}} \frac{f(T_{\rm L})}{f(T_{\rm U})}$$

The potassium concentrations in the lower and upper mantles are different because the extraction of continental crust has depleted the MORB source in incompatible elements. According to inversion of the mass balance equations, potassium is six times more



Fig. 2. Graphical computations of concentration ratios, noted *R*, between lower and upper mantles, against the isotopic ratios in the lower mantle. For helium, the two curves correspond to two choices for the initial isotopic ratio of the earth (solar or chondritic). For Argon, the tow curves correspond to two different MORB source 40 Ar/ 36 Ar ratios [13,67].

concentrated in the lower mantle than in the upper mantle [40]. However, to include the fact that the mean-age of continental extraction is ~ 2.5 Gy [41], a factor of 4 in the K concentration ratio has to be considered [9,40].

$$R_{\rm Ar} = \frac{\left({}^{40}{\rm Ar}/{}^{36}{\rm Ar}\right)_{\rm UM}}{\left({}^{40}{\rm Ar}/{}^{36}{\rm Ar}\right)_{\rm LM}} 4 \frac{1.11}{0.074} = \frac{\left({}^{40}{\rm Ar}/{}^{36}{\rm Ar}\right)_{\rm UM}}{\left({}^{40}{\rm Ar}/{}^{36}{\rm Ar}\right)_{\rm LM}} 60$$

The 40 Ar/ 36 Ar ratio of the upper mantle is estimated to be ~40,000. This value was determined by an extrapolation to a solar 20 Ne/ 22 Ne ratio of the

correlation observed between neon and argon in the gas rich sample $2\pi D43$, or analyses within single vesicles in this same sample [13,42].

Similar equations can be written for helium and neon considering that the K/U ratio is constant in the mantle (10,000 to 12,000 [43]) and that the Th/U ratios are ≈ 4.2 for the lower mantle and 2.5 for the upper mantle [44,45].

$$R_{\rm He} = \frac{[{}^{3}{\rm He}]_{\rm LM}}{[{}^{3}{\rm He}]_{\rm UM}} = \frac{\left({}^{4}{\rm He}/{}^{3}{\rm He}\right)_{\rm UM}^{*}}{\left({}^{4}{\rm He}/{}^{3}{\rm He}\right)_{\rm LM}^{*}}35$$

$$R_{\rm Ne} = \frac{[^{22}{\rm Ne}]_{\rm LM}}{[^{22}{\rm Ne}]_{\rm UM}} = \frac{\left({}^{21}{\rm Ne}/{}^{22}{\rm Ne}\right)_{\rm UM}^{*}}{\left({}^{21}{\rm Ne}/{}^{22}{\rm Ne}\right)_{\rm LM}^{*}} 35$$

For helium, the calculations are less simple because the initial ⁴He/³He ratio in the lower mantle is not known (40Ar is purely radiogenic in Earth but not 4 He). The meteorite 4 He/ 3 He ratios vary from 2000 to 6800 and since we use a solar value for neon, we also use a solar value of ~ 3000 for the initial ${}^{4}\text{He}/{}^{3}\text{He}$ ratio [11]. However, calculations will also be done with different initial helium isotopic ratios in order to better constrain the $R_{\rm He}$ value. The helium isotopic ratios in n-MORB vary from 90,000 to 120,000 [2,32,46]). A "mean" value of 100,000 will be used in the following. We exclude data from places where hotspot signatures are observed on the ridge, such as the Reykjanes ridge, contaminated by the Iceland hotspot [47,48] or the Shona and Discovery ridge anomalies in the south Atlantic [18,32]. In the same way, the solar corrected ²¹Ne/²²Ne ratio is estimated to be 0.075 in the MORB source [1,13].

The problem is now to estimate the rare gas isotopic ratios for the lower mantle. Most OIB have intermediate rare gas isotopic ratios between the Iceland and MORB values [16,18,32,49–51]. Therefore, the best estimate for the OIB original value seems to be the Icelandic samples, and, in particular, the sub glacial gas-rich glass samples [19,22,52]. Moreover, the argon and xenon isotopic ratios are among the most precise on these samples. The Iceland neon results give a ²¹Ne/²²Ne extrapolated to a solar 20 Ne/ 22 Ne of 0.0355 [19,22,52]. The ${}^{40}\text{Ar}/{}^{36}\text{Ar}_{\text{LM}}$ is difficult to estimate directly because of air contamination. However, based on neon data, the Icelandic study shows clearly that ${}^{40}\text{Ar}/{}^{36}\text{Ar}_{LM}$ \leq 5000 [52]. This is close to the value estimated by Valbracht et al. for Loihi seamount [17] or for carbonatites from Kola by Marty et al. [53]. For the helium isotopic ratio of the lower mantle, ⁴He/³He_{LM}, a value lower than the lowest measurement made on OIB, which is 15,000 $(R/R_a=50)$ [6] has to be used. Taking a set of conservative values such as ⁴He/³He=12,500, ²¹Ne/²²Ne=0.0355 and ⁴⁰Ar/³⁶Ar=4500 for the OIB source, the corresponding values R_{He} , R_{Ne} and R_{Ar} are ~500 for the three ratios (Fig. 2). There are possible sources of possible error for this calculation.

For mean-ages of lower mantle of 3.5 or 4 By, the corresponding R_{Ar} will be 280 and 375, respectively. There is still a huge difference in concentrations between the lower and upper mantles. Taking a [K] difference between the two reservoirs of 3, rather than 4, gives a R_{Ar} of 280, which still suggest a gas-rich lower mantle. Therefore, the interpretation is extremely robust, and a huge difference in rare gas concentrations is required to explain the isotopic ratios.

4. Discussion: the source of mantle plumes

For fluid dynamic reasons, it is generally assumed that mantle plumes are generated as instabilities at a boundary layer. Therefore, two locations have been proposed for their origin. The source can be at the 670 km boundary layer as proposed by Allègre et Turcotte [27]. The source can also be the core–mantle boundary as originally proposed by J. Morgan [54] and Hofmann and White [55] and "apparently" recently supported by seismic tomography (e.g. [56]).

If we assume that the hotspots are derived from the core-mantle boundary, the majority of the mass of the mantle plume would consist of lower mantle material. As shown by Hauri et al. [28] or Whitehead [57], most of the plume material is incorporated near its source. Because of the large contrast in the rare gas concentrations between the lower and upper mantles, the effect of the upper mantle "contamination" on plumes would be completely negligible for rare gases and therefore, the rare gas isotopic compositions of OIB would reflect the isotopic composition of the core-mantle boundary layer. In this case, OIB should be extremely homogeneous for the rare gas isotopic ratios, which is obviously not the case [3,58–60].

If we consider the genesis of a plume at the 670 km discontinuity, the plume will incorporate, by entrainment, a small proportion of lower mantle material, as proposed by Oxburg et O'Nions [61], Allègre et Turcotte [27] or Hauri et al. [28]. This has been clearly shown both by the theoretical and experimental arguments of Davaille [29]. Because of the large difference in rare gas concentrations, 15% to 1% entrainment will create a huge difference in rare gas isotopic ratios, which will be dominated by the lower mantle isotopic signature, but will dilute concentra-

tions. To demonstrate this, one can write the following equations.

$$\left(\frac{{}^{4}\text{He}}{{}^{3}\text{He}}\right)_{\text{plane}} = \alpha \left(\frac{{}^{4}\text{He}}{{}^{3}\text{He}}\right)_{\text{LM}} + (1-\alpha) \left(\frac{{}^{4}\text{He}}{{}^{3}\text{He}}\right)_{\text{UM}}$$

with

$$\alpha = \frac{[{}^{3}\text{He}]_{LM}M_{LM}}{[{}^{3}\text{He}]_{LM}M_{LM} + [{}^{3}\text{He}]_{UM}M_{UM}} = \frac{1}{1 + \frac{[{}^{3}\text{He}]_{LM}M_{UM}}{[{}^{3}\text{He}]_{LM}M_{LM}}}$$
$$= \frac{1}{1 + \frac{M_{UM}}{R_{He}M_{LM}}}$$

If $M_{\rm UM}/M_{\rm LM}=99$ (1% entrainment), and with $R_{\rm He}=500$, we get $\alpha=0.84$ and therefore ${}^{4}{\rm He}/{}^{3}{\rm He}_{\rm plume}=27,000 (R/R_{\rm a}=26)$. Whereas, for the concentrations, $[{}^{3}{\rm He}]_{\rm plume}=(0.01\times500+0.99)[{}^{3}{\rm He}]_{\rm UM}=6[{}^{3}{\rm He}]_{\rm UM}$. This calculation clearly shows that there is a major dilution effect for concentrations.

In addition, if the plumes were coming from a heterogeneous lower mantle, we should observe correlations between ⁴He/³He and ⁸⁷Sr/⁸⁶Sr or ²⁰⁶Pb/²⁰⁴Pb, which is, on a global scale, not observed.

To challenge the deep origin of hot spots/Plumes, Anderson [62] invokes the so-called "helium paradox" which consists of the observation that OIB glasses contain less helium than MORB glasses, whereas it should be the opposite if the upper and lower mantles have very different rare gas concentrations. The present model explains this paradox as we have demonstrated above. This is linked to a dilution effect due to entrainment of only few percents of lower mantle material.

During their ascent to the surface, plumes can lose helium to the ambient mantle by diffusion. At the top of the upper mantle, diffusive loss could result in concentrations few times higher than ambient mantle. As shown by Moreira and Sarda [33], the dynamics of OIB and MORB degassing are very different. Therefore, the final concentrations may not be very different. It is therefore not a paradox but is entirely consistent with the idea of plume generation at 670 km, with entrainment of lower mantle. In fact, the diffusion of rare gases out of the plume during their ascent is probably the mechanism responsible of the transfer of ³He and other primordial gases to the upper mantle [35]. The plumes that do not reach the surface are probably too small in size to have a lot of entrainment during their genesis in the boundary layer of 670 km and therefore do not contribute to the ³He flux.

This model of 670 km discontinuity source for OIB with entrainment of the lower mantle explains also the complete continuity in isotopic composition of Sr, Nd, Pb observed between OIB and MORB [63] as well as the trace element systematics [64].

The only way to escape this conclusion would be to invoke a huge heterogeneity in concentrations and isotopic compositions of the rare gases in the lower mantle. Such a model seems unlikely because the observations made by seismic tomography, as well as fluid dynamic considerations, suggest a convective lower mantle [65,66]. Moreover, if one wants to store the gas rich reservoir within the D" boundary layer, it is difficult to imagine a stagnant, closed system boundary layer surviving for billions of years with the large heat flux from the core. We therefore exclude the D" layer as the primitive rare gas reservoir.

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