A model to explain the various paradoxes associated with mantle noble gas geochemistry

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ABSTRACT As a result of an energetic accretion, the Earth is a volatile-poor and strongly differentiated planet. The volatile elements can be accounted for by a late veneer (~1% of total mass of the Earth). The incompatible elements are strongly concentrated into the exosphere (atmosphere, oceans, sediments, and crust) and upper mantle. Recent geochemical models invoke a large primordial undegassed reservoir with chondritic abundances of uranium and helium, which is clearly at odds with mass and energy balance calculations. The basic assumption behind these models is that excess "primordial" 3He is responsible for 4He/4He ratios higher than the average for midocean ridge basalt. The evidence however favors depletion of 4He and excessive depletion of 4He and, therefore, favors a refractory, residual (low U, Th) source. Petrological processes such as melt-crystal and melt-gas separation fractionate helium from U and Th and, with time, generate inhomogeneities in the 4He/4He ratio. A self-consistent model for noble gases involves a gas-poor planet with trapping of CO2 and noble gases in the shallow mantle. Such trapped gases are released by later tectonic and magmatic processes. Most of the mantle was depleted and degassed during the accretion process. High 4He/4He gases are viewed as products of ancient gas exsolution stored in low U environments, rather than products of primordial reservoirs.

The Earth accreted energetically in a high temperature part of the solar nebula (1, 2). It therefore is depleted in the volatile elements, and the large-ion elements are strongly concentrated toward the surface (3). The Earth is cooling down and this accounts for approximately one-half of the heat flow (4). These physical constraints are violated by recent geochemical models that imply a volatile-rich planet (5) and a balance between radioactive heat productivity and present heat flow (3). The Earth is cooling down and this accounts for approximately one-half of the heat flow (4). Volatile elements such as U and Th are excluded from mantle minerals and are concentrated in melts. Helium is a gas, is neutral, and escapes degassing and has primordial abundances of the elements. Hotspots tap this deep primordial reservoir via narrow plumes and provide high 3He/4He basalts.

There are various problems with this model:
1. High 3He/4He ratio basalts have very little 3He, often two orders of magnitude less than MORB; 2. the He/Ar, He/Ne, etc., ratios in high R basalts do not indicate that they have lost more He than MORB; 3. the inferred He content of the primordial reservoir requires a gas-rich planet; 4. many high R environments have average R values similar to MORB; 5. basalts from the conjectured primordial mantle do not have other primordial characteristics, such as primitive Pb-isotopic compositions; 6. the 3He gas flux from hotspot volcanoes is orders of magnitude less than from mid-ocean ridges and island arcs (10), contrary to predictions of the Standard Model; 7. the present 4He flux from the mantle is an order of magnitude less than predicted from primordial U and Th abundances and the observed heat flow; and 8. the main carrier of He, CO2, is depleted in the outer parts of the Earth by an order of magnitude compared with other volatiles.

These problems are known as "paradoxes" in the noble gas literature.

Alternative Model. High R also may represent low 4He or low time integrated U, Th abundances. In this case, the high R reservoir is not primitive; it can be a shallow refractory reservoir such as residual peridotite, harzburgite, or magma chamber cumulates. There are various mechanisms for separating U, Th from He and, therefore, for generating an inhomogeneous 3He/4He distribution. Large-ion lithophile elements such as U and Th are excluded from mantle minerals and are concentrated in melts. Helium is a gas, is neutral, and has a small radius. It therefore has quite a different melt-solid partition coefficient than U and Th, and the U/He ratio in residual crystals is likely to be lower than in silicate melts. Even if He is expelled from the crystal during melting, some is reincorporated, as fluid-filled inclusions or vugs, during shal-

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Abbreviations: MORB, midocean ridge basalt; OIB, ocean island basalt; MM, MORB mantle; HIMU, high values of 238U/204Pb; LONU, low values of 238U/232Th.
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low crystallization of the melt. This process need not be efficient because high $^{3}$He/$^{4}$He melts and gases account for a very small fraction of the total $^{3}$He brought to the Earth’s surface. Melt-gas separation also separates U, Th from He and melt can escape from the mantle easier than can a free-gas phase that does not wet grain boundaries.

Fig. 2 illustrates a mechanism whereby a magma degasses at shallow depth and stores some of the gases in refractory (U, Th-poor) mantle. In this case, the gas is separated from MORB at or near a ridge and released at midplate environments at a much later time. This model explains the low $^{3}$He content and low He/Ne and He/Ar ratios of high R basalts because the noble gases in these basalts are contaminants derived from ancient MORB-degassing events. CO$_{2}$ is the main carrier phase.

The parameters controlling $^{3}$He/$^{4}$He variations in the mantle are $^{238}$U/$^{3}$He (NU) and the Th/U ratio. The production of $^{4}$He is low in low $^{238}$U/$^{3}$He (LONU) environments, such as pelitic rocks residual after melt (i.e., U, Th) extraction. With time, such environments have high $^{3}$He/$^{4}$He ratios compared with basalts, fertile mantle or recycled material, or the MORB reservoir.

**Tests of the Model.** Noble gas ratios in midplate volcanics suggest relative depletion in those elements most soluble in melts and contamination with seawater infiltrated crust (11, 12). CO$_{2}$ and noble gases exsolved from magmas are partially trapped in fluid-filled inclusions and vugs in the shallow mantle and carry with them the $^{4}$He/$^{3}$He ratio of MORB, at the time of degassing. If these gases are stored in refractory, U-poor, mantle, the R values of the inclusions and the parent magma reservoir will diverge. R in old inclusions can be much greater than in modern basalts from the same reservoir, and these can contaminate midplate magmas. Fig. 2 illustrates the process.

Trapping of CO$_{2}$-He in lower crustal or upper mantle cumulates provides a potentially high $^{3}$He/$^{4}$He component for future magmas to inherit, especially at the onset of magmatism. An estimate of the dimensions of the source, if it is in the current lithosphere, is provided by the deep seismicity under Kilauea and Loihi (13). $^{3}$He abundances in deep xenoliths (14, 15) yield an estimate of $2 \times 10^{9}$ cm$^{3}$ of $^{3}$He in the seismically active portion of the lithosphere under Hawaii by using the seismic zone as an estimate of degassing volume ($2 \times 10^{7}$ km$^{3}$).

This represents 2,000 years of $^{3}$He available at the current eruption rates. $^{3}$He/$^{4}$He ratios in Hawaiian basalts approach MORB values on time scales of thousands of years (16). The temporal decrease of R in Hawaiian volcanoes, generally attributed to drifting off of a deep mantle plume, may instead be due to exhaustion of CO$_{2}$-rich pockets in the shallow mantle. A new conduit will restart a high R pathway. The source of helium in this model is very local. Such a model explains why there are no global correlations of helium with other isotopes but sometimes very good local correlations and why there are large local variations in R.

As volcanoes grow and erode, new cracks open up and the lithosphere is exposed to ascending magmas. I attribute the temporal change of noble gas chemistry at Hawaiian volcanoes to lithospheric or shallow mantle characteristics (new cracks and new conduits) rather than to motion of the plate over a static plume.

The $^{3}$He/$^{4}$He ratio in Réunion Island basalts has remained high for approximately 360,000 years, ~10 times longer than in Hawaiian volcanoes (17). Réunion is much less active than Hawaii, and the eruption rates are an order of magnitude lower. Any helium stored in the shallow mantle should therefore survive much longer.

Midplate volcanoes initiate with cracks, caused by plate-tectonic stresses and volcanic loading. Magma, in fact, can
fracture its way to the surface in lithosphere exposed to tensile stresses. Generally, lithosphere is under compression and magma pressure is not high enough to fracture it. The initial magmas interact with CO$_2$-rich fluid-filled inclusions with $^{3}$He/$^{4}$He ratios frozen in at the time of stabilization of the lithosphere (Fig. 2). Virgin lithosphere, or recycled slab, with fluid-filled inclusions is a potential low-$\nu$ (LONU) reservoir (low $^{238}$U, high CO$_2$-“high” He). As magmas ascend, they interact with shallow mantle, altered oceanic crust and seawater infiltrated rocks, and sediments. As magma conduits get established, the nature and extent of contamination change. In particular, the high $^{3}$He/$^{4}$He fluid inclusions get used up and low $^{3}$He/$^{4}$He crustal xenoliths (in a HINU environment) may lower the helium ratio. The crust may have lost most of its helium so lithospheric contamination may be more important than crustal contamination. When a conduit gets plugged, or a new crack opens, a new transient stage of high $^{3}$He/$^{4}$He magmas can start. This evolutionary history explains why hotspot magmas are diverse and why atmospheric components are intimately related to “primordial” components (11). It also explains the range of R found on oceanic islands and the temporal evolution toward MORB values (16). Ocean island basalts should have lower helium abundances than MORB or than gas-rich mantle xenoliths, as observed.

**DISCUSSION**

There are several possible refractory LONU reservoirs: (1) ancient lithosphere, (2) deep crustal cumulates, and (3) the lithosphere beneath the island.

The latter is the most interesting case, but the question arises, can the $^{3}$He/$^{4}$He ratio in the reservoir from which the oceanic crust and lithosphere formed have decreased by a factor of three since the fluid inclusions were emplaced? This depends on $\nu$ of the MORB mantle (MM). Fig. 3 shows the tradeoffs between U/He ratios and time. MORB-like R values can be generated from Loihi-like values in $10^{6}$--$10^{8}$ years if $\nu$ of MM is similar to MORB. High R reservoirs can be preserved if $\nu$ is lower than in MM by 1--2 orders of magnitude. Refractory mantle with CO$_2$-rich inclusions, can have very low $\nu$. Although we do not have a good estimate of $\nu$ of MM, or how it changes with time, high values of $\nu$ lead to difficulties when extrapolated too far into the past. There is some evidence for rapid increases of U in the upper mantle (18, 19). Because the value and history of the $^{238}$U/$^{204}$Pb ratio ($\mu$) in the upper mantle are uncertain, it is premature to rule out a high or increasing $^{238}$U/$^{3}$He ratio ($\nu$). It also is not certain that the age of the mantle under Hawaii is the same age as the crust. Swells such as the Hawaiian swell may be partially held up by buoyant refractory residue and may be unsubductible. Young crust can form on old lithosphere. There is, in fact, evidence that the Hawaiian swell is not a thermal swell but may be a thick section of depletions peridotite (20). It may be older than the overlying crust. Other oceanic swells or plateaus are suspected of having an ancient, or continental, component.

An alternative low $\nu$ reservoir for high $^{3}$He/$^{4}$He inclusions is ancient lithosphere that has been recycled and stored for longer periods of time. This would be similar to the recycling of ancient oceanic crust, which is the conventional explanation for some high $^{238}$U/$^{204}$Pb (HIMU) plume components (19, 21, 22, 23).

The range in $\nu$ for MORB is approximately a factor of 6 (23). Because He is a gaseous species and recycling and degassing both increase the U/He ratio, a time integrated two-order of magnitude variation in $\nu$ (5, 6) seems plausible (Fig. 3), even in the upper mantle.

**Change of $^{3}$He/$^{4}$He with Time.** If high R locales, such as Loihi, represent old MM values of R, we can estimate the $^{238}$U/$^{3}$He and $^{3}$He/$^{2}$Ne ratios of MM. I adopt R for Loihi as the value for MM at the time of formation of the oceanic crust at Hawaii ($\sim 100$ Ma) and solve for $\nu$ of MM, which yields current MORB values for $^{3}$He/$^{4}$He. Because this is a minimum age for the LONU reservoir, it yields a maximum NU for MM. The result, assuming Th/U = 3.9, is $2.2 \times 10^{6}$ for $^{238}$U/$^{3}$He (atomic ratio), or $2.6 \times 10^{9}$ ppm/g/cc. This is in the range of MORB (24). If NU (MM) is much lower than this, then the degassing event must be much older than the age of the crust under Hawaii.

If noble gases presently at Loihi were exsolved from the MORB reservoir at some time in the past, and subsequently stored in a refractory (LONU) environment, one can calculate the $^{21}$Ne/$^{22}$Ne and $^{3}$He/$^{2}$Ne ratios of MM, from the difference in $^{3}$He/$^{4}$He between MORB and Loihi and the $^{3}$He/$^{2}$Ne production ratio.

The growth of $^{21}$Ne, calculated from the growth of $^{4}$He via the production ratio, yields $3.4$--$4.6$ for the atomic ratio $^{3}$He/$^{2}$Ne. The lower values are close to the solar ratio, consistent with the solar ratios for $^{20}$Ne/$^{22}$Ne found in MORB. Ratios involving the radiogenic isotope $^{4}$He and the nucleogenic isotope $^{21}$Ne depend on $\nu$. The production ratio $^{3}$He/$^{21}$Ne is approximately $1$--$3.5$ ($\times 10^{6}$). If the Loihi $^{3}$He/$^{4}$He ratio represents the MORB value at some time in the past, the $^{20}$Ne/$^{22}$Ne ratio in MORB can be calculated from this ratio in Loihi basalts and the difference in R values. For a MM $^{20}$Ne/$^{22}$Ne ratio of 0.24, a typical MORB value (11), a decrease of R from 30 R$_{o}$ to 8 R$_{a}$ (MORB) is accompanied by an increase in $^{21}$Ne/$^{22}$Ne of 0.03, which agrees with the increase.
from Loihi basalts to MORB. This calculation has been used in the past to support the undegassed primitive mantle model. Furthermore, if gases inherited by Loihi basalts and xenoliths, and other OIB, were in equilibrium with MORB, they should have lower $^{3}$He/$^{22}$Ne ratios than MORB (as well as high $^{3}$He/$^{4}$He ratio) (Figs. 2 and 4).

Loihi basalts have much lower $^{3}$He/$^{22}$Ne ratios than MORB (11, 25, 26), consistent with vesicles (and air) being the source of Loihi gases. Estimates of $^{4}$He/$^{22}$Ne ratios of MORB (9, 4) and Loihi (6, 9) are both above the solar ratio (27). These high ratios can be explained by degassing of the MORB precursor, at depth, raising the He/Ne ratio in the residual melt and then contaminating this melt with shallow mantle vesicles and seawater-infiltrated crust, both serving to decrease He/Ne (Fig. 4A). In the steady-state primordial mantle models with noble gases leaking into the upper mantle, the $^{4}$He/$^{22}$Ne ratios in EM and DUM must be identical. Porcelli and Wasserburg (5) predict a ratio of 4.5 for both reservoirs, much lower than observed.

Degassing of magma increases the He/Ne and He/Ar ratios of the residual magma because of the high solubility of He in the melt. MORB have higher ratios than OIB (Fig. 4). Mixing of degassed MORB with air or seawater gases or with gases expelled from previous magmatic episodes gives trends of $^{3}$He/$^{22}$Ne vs. $^{4}$He/$^{22}$Ne (Fig. 4A) and $^{4}$He/$^{40}$Ar and $^{3}$He/$^{22}$Ne vs. $^{4}$He or $^{3}$He (Fig. 4B), similar to those observed.

A decrease in $^{3}$He/$^{4}$He over 10$^{7}$ years from $\approx 30 R_{\odot}$ to 8 $R_{\odot}$ requires a $v$ of $\approx 2 \times 10^{9}$. If MM contains 10 ppb of $^{238}$U, the implied $^{3}$He content is $10^{6}$ atoms/g, an order of magnitude lower than MORB that concentrates U and He, relative to MM, by the melting process. For an undegassed Earth, $v$ has been estimated to be $\approx 10^{5}$ (5, 11, 26). Degassing increases this ratio. The He abundances and U/He ratios implied by the shallow mantle contamination model are therefore reasonable. One implication is that the MORB reservoir (MM) is depleted in noble gases, compared with chondrites, by $\approx$two orders of magnitude relative to uranium. Other volatile elements are depleted in MM by factors of $>10$ (3), and more extreme depletion of He is expected. The bulk of the mantle was degassed during accretion and should have even lower volatile contents. Part of the mantle may be volatile-rich because of recycling. However, it is the top of the slab that contains these volatiles, and they enter the mantle wedge at shallow depths; noble gases may not be efficiently recycled. Massive deep (>200 km) recycling of upper slab volatiles is unlikely (28). The shallow mantle is the main depository of the incompatible and volatile recycled components, even if the demetasomatised slab dives deeply into the mantle. The conclusion that the Earth is extensively depleted in very volatile elements relative to chondritic meteorites is not new but is different from the implications of current gas-rich models (5, 6, 9, 26). If the Earth differentiates during accretion (3), or if a late veneer is involved in the volatile budget of the Earth (29), the upper mantle should be more volatile-rich than the bulk of the mantle. The present model resolves one other minor paradox. R of HIMU basalts are slightly lower than MORB but $^{206}$Pb/$^{204}$Pb ratios are substantially higher. Mixing calculations with low $^{3}$He contents in MORB do not satisfy the data (44). If the MORB source has higher He/Pb ratios than the endmember HIMU source, the basalt data for HIMU islands can be satisfied.

Ridges and subduction zones migrate about the surface of the Earth. These processes introduce U (22) into the mantle and remove He, giving a progressive increase in $v$ and $u$ of the upper mantle. The upper mantle is processed about once every 1 or 2 Ga (30). The present high value for $v$ cannot be extrapolated far into the past and excessively large $^{3}$He/$^{4}$He ratios in the distant past are not expected. Multiple stage evolutionary models are required to explain Pb and other isotopes. Nevertheless, some MORB have $^{230}$U/$^{206}$Pb ratios close to the geochron and therefore are consistent with a closed system evolution for 4.5 Ga. The most gas-rich MORB have He-U ages of $\approx$1–2 Ga (31), a minimum age assuming undegassed magma. The relatively high $^{129}$Xe content of MORB also is consistent with an ancient reservoir. If the difference between MORB and Loihi helium (and neon) isotopic ratios evolved over $\approx 10^{7}$ years, instead of the age of the crust under Hawaii, then the LONU source of Loihi simply may be the mantle complement to the crustal HIMU component.

The $^{4}$He-Heat Flow Paradox. Only $\approx 10\%$ of the $^{4}$He flux predicted from heat flow is observed to degas from the Earth (32). In contrast, $\approx 70\%$ of the $^{40}$Ar produced by $^{40}$K decay is in the atmosphere, indicating efficient degassing of Ar in early Earth history (3, 33). Part of the discrepancy may be due to the relatively high solubility of He in magma and the retention of CO$_{2}$ and helium in the shallow mantle. The gas content of average MORB vs. popping rock implies that most MORB have lost 90% or more of their gas, at depth, before extrusion.
and eruption (34). The He-heat flow paradox indicates that He is not easily or promptly degassed. Helium in undegassed magmas, or trapped in the upper mantle, may be recycled and retained in the upper mantle for a considerable period. With usual estimates of U abundances in MM, it takes the age of the Earth to build up the observed levels of 4He and 21Ne in a closed system (30, 31). With 200–300 ppb of U, the observed 4He levels (both concentrations referring to the basalt, not the source) can be built up during periods of time comparable with the isolation time [(1 to 2) × 10^9 years] of a given portion of the upper mantle (30).

There is no support for the hypotheses that hotspots dominate the 4He-degassing budget of the Earth and that high R magmas are more extensively degassed than MORB. The relatively high 4He/21Ne, 4He/22Ne, and 4He/CO2 ratios in MORB suggest that MORB is helium-rich compared with high R basalts and, therefore, that the high 4He/3He ratio in MORB is due to excess 4He, not a deficit in the “primordial” isotope 3He. The 238U/206Pb ratio in some MORB imply closed system evolution for nearly the age of the Earth. If MORB are 80% degassed just before or during eruption, then the helium age of the MORB reservoir (inferred from U/4He) also is comparable to the age of the Earth, and MM can be viewed as an ancient, relatively undegassed reservoir, the opposite of the usual interpretation.

As magmas ascend to the surface, or crystallize, CO2 is released, some of which is trapped in refractory crystals and in magma chambers. The trapping temperature is ~600°C (35), roughly the temperature at which the lithosphere forms (36). These CO2–He-rich fluid inclusions represent time capsules, which retain the high 4He/21Ne ratios of a previous generation of ocean ridge magmatism. These time capsules are isolated from mantle convection by the buoyancy and strength of the lithosphere and perisphere. The upper mantle may contain large reservoirs of free fluid CO2 in fluid-filled cavities. Up to 3 vol percent fluid (CO2, He, Ar ...) has been found in cavities up to 1.5 cm in length in mantle xenoliths from Australia (42). Decompressing or crystallizing magma releases volatiles to the wall rock, and these volatiles may be picked up by magmas passing through this vesicular upper mantle (42). A CO2-rich shallow mantle may be at least a partial explanation for the missing CO2 in the atmosphere, oceans, sediments, and crust.

The heavy noble gases in Loih basaltic reservoirs are the result of contamination with atmosphere derived noble gases dissolved in seawater (25). Other characteristics of hotspot basalts and xenoliths with high R, include carbonates and other contamination indicators. Iodine is a tracer of pelagic sediments, and I is also high in Loih basaltic (37). Interplanetary dust particles (IPD), abundant in pelagic sediments, have high 4He contents and 4He/He ratios. It has been a paradox that midplate basalts and xenoliths with the most “primitive” character (high R) also show extensive contamination with material of atmospheric, seawater and sedimentary heritage.

The standard models of noble gas geochemistry involve two large hypothetical reservoirs, the upper (degassed) and lower (undegassed) mantles (5, 6, 9). The present model also involves two reservoirs, the MORB and the shallow (regassed) mantles, but the total fertile and gas-rich mantle is a small fraction of the Earth. Helium and CO2 are trapped in the shallow mantle and are released by midplate tectonic and magmatic processes, rather than by deep-seated convective processes. Melt-vapor fractionation has been invoked before (33, 38) but usually as related to early processes and atmospheric formation, not the final stages of midplate magmatism.

The present paper interprets the spread in R as being due to differing ages and υ of the reservoirs. It is also possible that high R sources have a different origin than MORB gases; for example, a late veneer (30).

Most of the characteristics of “hotspot” magmas such as isotopic chemistry (Sr, Nd, Pb, O, and C), oxidation state, and noble gases show extensive contamination with material of atmospheric, seawater and sedimentary heritage. These CO2–He-rich fluid inclusions represent time capsules, roughly the temperature at which the lithosphere forms (36). Decompressing or crystallizing magma releases volatiles to the wall rock, and these volatiles may be picked up by magmas passing through this vesicular upper mantle (42). A CO2-rich shallow mantle may be at least a partial explanation for the missing CO2 in the atmosphere, oceans, sediments, and crust.

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