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^{142}Nd/^{144}Nd inferences on the nature and origin of the source of high ^{3}He/^{4}He magmas

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ABSTRACT

High-precision measurements of ^{142}Nd/^{144}Nd in picrites from the Baffin Bay region that contain the highest ^{3}He/^{4}He ratios yet measured in terrestrial mantle-derived rocks are indistinguishable from the value measured in the terrestrial standard and other modern mantle-derived rocks. The Baffin Island lavas are distinguished from other hotspot lavas by their unusually high ^{3}He/^{4}He and ^{182}W/^{184}W ratios, but their Sr, ^{142}Nd, ^{143}Nd, Hf, and Pb isotopic signatures overlap the values measured in North Atlantic MORB. These features imply either that the mantle source region of high ^{3}He/^{4}He magmas carries the lithophile isotopic signatures of incompatible element depletion, or that the He isotope signature of this source is decoupled from the lithophile isotope tracers in the magmas. The coupled ^{142}Nd–^{143}Nd data are consistent with the magma source acquiring the incompatible element depletion during, or shortly after, Earth formation if the bulk-Earth has a ^{142}Nd/^{144}Nd ratio more similar to the average measured for enstatite chondrites than modern terrestrial rocks. If Earth's initial ^{142}Nd/^{144}Nd was higher than the average of enstatite chondrites, the data are consistent with the traditional interpretation that the depleted-mantle reservoir was formed through the extraction of an incompatible-element-rich reservoir, such as continental crust, after the circa 4 Ga extinction of ^{146}Sm. This explanation, however, fails to account for the high ^{3}He/^{4}He. The Nd isotopic composition of the picrites could reflect a dominant contribution from the incompatible element depleted source of North Atlantic MORB, overprinted by a small (10–20%) contribution from a mantle source with He concentrations at least ten times higher than the depleted mantle along with W isotopic compositions substantially higher than typical of mantle-derived rocks.

1. Introduction

The isotopic composition of helium in Earth's interior is an important parameter that must be accounted for by models that seek to explain the differentiation history of the Earth. Mid-ocean ridge basalts (MORB) derived from the convecting upper mantle typically have ^{3}He/^{4}He ratios of approximately 8R_a (where R_a is the atmospheric ^{3}He/^{4}He ratio of 1.39×10^{-6}) (Graham, 2002). Significantly higher ^{3}He/^{4}He ratios are recorded in basalts from several volcanic ocean islands and continental flood basalt provinces (Hilton et al., 1999; Kurz et al., 1982; Starkey et al., 2009; Stuart et al., 2003). In the absence of a credible mechanism by which to generate ^{3}He internally, the mantle ^{3}He inventory must be primordial, originating from high ^{3}He/^{4}He ($>120R_a$) material incorporated into the accreting Earth (Ozima and Nakazawa, 1980). In contrast, ^{4}He is continually produced by radioactive decay of U and Th. Consequently, the source of high ^{3}He/^{4}He in Earth's interior must have evolved in a reservoir with a time-integrated $(U + Th)/^{3}$He ratio, and attendant capacity to generate ^{4}He, that was lower than that of most of the convecting upper mantle. Whether this reflects a mantle reservoir depleted in incompatible elements such as U and Th or one that is so gas-rich as to minimize the contribution of ^{4}He generated by radioactive decay is unclear. For many years, the high ^{3}He/^{4}He ratio of some intra-plate ocean island basalts linked to mantle plumes has been thought to require an undegassed, and by assumption undifferentiated, mantle reservoir deep in the Earth (Kurz et al., 1982).

The highest ^{3}He/^{4}He ratios measured in modern basalts requires the isolation of a mantle reservoir prior to 3 Ga (Porcelli and Elliot, 2008). The view that high ^{3}He/^{4}He basalts are derived from a relatively undegassed, and therefore undifferentiated, man-

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tle reservoir remains a seductive hypothesis. While a convectively-isolated lower mantle source for this reservoir is now difficult to reconcile with geophysical evidence for whole mantle circulation (e.g. Grand et al., 1997), relatively undifferentiated material may be preserved as low-strain pockets in the mantle (Brandenburg et al., 2008) or in the seismically anomalous large-low-shear velocity provinces in the lowermost mantle (e.g. Garnero and McNamara, 2008), which are possibly connected to the source of mantle plumes (Burke et al., 2008; French and Romanowicz, 2015). Of perhaps more concern for the traditional model of an undifferentiated, undegassed, reservoir is that most high $^3He/^4He$ basalts display Nd, Hf, and Sr isotope signatures indicative of a source that is depleted in incompatible elements (Class and Goldstein, 2005; Ellam and Stuart, 2004; Stuart et al., 2003). In addition, the mantle-derived rocks with the highest $^3He/^4He$ ratios have a large range of trace element and Sr, Nd and Os isotopic compositions (Dale et al., 2009; Starkey et al., 2009) that overlap the fields of mid-ocean ridge basalts, not the values expected for undifferentiated mantle. Two options have been proposed to explain the apparent association of high $^3He/^4He$ with the Sr, Nd and Hf isotopic signatures of incompatible element depletion. One suggests that the concentration ratio of He to lithophile elements in the high $^3He/^4He$ source, be it mantle or core (Porcelli and Halliday, 2001), is so high that even a small mass fraction of this source mixed with more "normal" mantle would dominate the He isotopic, but not the lithophile element, composition of the mixture. Another is that the high $^3He/^4He$ reservoir is the product of global differentiation accompanying Earth formation that occurred deep enough in the mantle where crystal–liquid fractionation was efficient, but outgassing was not (Coltice et al., 2011; Jackson et al., 2010).

Traditionally, the incompatible element depletion of the mantle source of MORB has been ascribed to the extraction of continental crust from a primitive portion of the mantle that started with chondritic relative abundances of refractory lithophile elements (Hofmann, 1988; Jacobsen and Wasserburg, 1979). An origin through this mechanism also accounts for the relatively low $^3He/^4He$ ratios seen in MORB because the process of crust formation would have been associated with outgassing of He as the magmas that make up the oceanic and continental crust were erupted onto Earth's surface. The elevated $^{143}Nd/^{144}Nd$ ratio of MORB compared to chondritic meteorites (DePaolo and Wasserburg, 1976; Richard et al., 1976) shows that the incompatible element depletion of MORB is old enough to have allowed ingrowth of ^{143}Nd via the decay of 106 Ga half-life ^{147}Sm. While billion year time scales are required to explain the $^{143}Nd/^{144}Nd$ ratios of the MORB source if it was derived via differentiation of a primitive source with chondritic Sm/Nd ratio, the uncertainty of the average value (Gale et al., 2013; Huang et al., 2013), and the temporal evolution, of the Sm/Nd ratio in the MORB source makes it impossible to define a unique age of differentiation of the MORB source using the ^{147}Sm–^{143}Nd system.

The discovery of a difference in the $^{142}Nd/^{144}Nd$ ratio of all terrestrial rocks compared to most rocky meteorites (Boyet and Carlson, 2005) provided a new look at this question (Boyet and Carlson, 2006; Carlson and Boyet, 2008; Caro, 2011; Caro et al., 2008). Because ^{142}Nd is the product of radioactive decay of 103 Ma half-life ^{146}Sm, the higher $^{142}Nd/^{144}Nd$ ratio of terrestrial rocks compared to most meteorites could indicate that the accessible mantle, including the MORB source, had a superchondritic Sm/Nd ratio within tens of millions of years of Solar System formation. If so, this would imply that the major fraction of the incompatible element depletion of the MORB source was due to a global differentiation event associated with Earth formation, and not primarily due to the gradual extraction of continental crust. While the range in $^{142}Nd/^{144}Nd$ ratios seen between different meteorite groups is now known to reflect nucleosynthetic variability in the materials from which the meteorites and Earth accreted (Bouvier and Boyet, 2016; Burkhardt et al., 2016; Carlson et al., 2007; Gannoun et al., 2011; Render et al., 2017), $^{142}Nd/^{144}Nd$ measured in modern terrestrial mantle-derived rocks lies at, or beyond, the high end of the $^{142}Nd/^{144}Nd$ range defined by meteorites, and overlaps in $^{142}Nd/^{144}Nd$ with only a small fraction of the analyses of one meteorite group, the enstatite chondrites (Burkhardt et al., 2016; Gannoun et al., 2011) and with calcium–aluminum-rich inclusions in primitive carbonaceous chondrites (Bouvier and Boyet, 2016; Marks et al., 2014). This implies either that Earth is made out of materials that are not well represented in the meteorite record (Render et al., 2017), or that the high $^{142}Nd/^{144}Nd$ ratio of all modern Earth rocks compared to the potential meteoritic building blocks of the planet indeed reflects ^{146}Sm decay in an ancient reservoir with superchondritic Sm/Nd ratio. Among their many isotopic similarities to Earth, enstatite chondrites have stable Nd isotopic ratios that overlap terrestrial values (Burkhardt et al., 2016; Gannoun et al., 2011), suggesting that Earth and E-chondrites share similar mixes of Nd nucleosynthetic components. If so, the average $^{142}Nd/^{144}Nd$ measured for E-chondrites, that is about 9 ppm lower than the terrestrial Nd standard (Burkhardt et al., 2016; Gannoun et al., 2011), supports the idea that the accessible mantle's higher $^{142}Nd/^{144}Nd$ is the result of the decay of ^{146}Sm in a reservoir with superchondritic Sm/Nd ratio that formed prior to ∼4 Ga while ^{146}Sm was still extant. Evidence for major Earth differentiation events that occurred within tens of millions of years of Solar System formation is now seen in a number of short-lived isotope systems. Variability in the $^{129}Xe/^{130}Xe$ ratio (due to decay of 15 Ma half-life ^{129}I) between MORB and some ocean island basalts (Mukhopadhyay, 2012), the $^{182}W/^{184}W$ ratio (due to decay of 9 Ma half-life ^{182}Hf) in both ancient crustal rocks (Touboul et al., 2012; Willbold et al., 2011) and modern basalts (Mundle et al., 2017; Rizo et al., 2016), as well as in the $^{142}Nd/^{144}Nd$ ratio of ancient crustal rocks (Bennett et al., 2007; Caro et al., 2006; Morino et al., 2017; O'Neil et al., 2012) all testify to chemical differentiation events that occurred within the first tens to hundreds of million years of Earth history.

In this study, we examine whether the high $^3He/^4He$ mantle reservoir might have differentiated soon after Earth accretion by examining the short-lived ^{146}Sm–^{142}Nd system in picrites from Baffin Island. These lavas, at ∼62 Ma, represent the earliest phase of volcanism that spreads from Baffin Island–West Greenland to the British Isles. They are often interpreted to represent the starting phases of a mantle plume now responsible for recent volcanism in Iceland (Saunders et al., 1997). Many of the mafic lavas from the Baffin Bay area have been shown to have the highest $^3He/^4He$ ratios measured for any mantle-derived rock (Starkey et al., 2009; Stuart et al., 2003). The Baffin Island picrites also have elevated $^{182}W/^{184}W$ ratios compared to the modern mantle (Rizo et al., 2016) and the lowest H/D ratios measured in mantle-derived rocks (Hallis et al., 2015). While we exercise caution that Pb isotope compositions in rocks from this province are particularly sensitive to lithospheric contamination (Dickin, 1981; Larsen and Pedersen, 2009; Lightfoot et al., 1997), the fact that the Pb isotopic composition of the Baffin Island lavas with the highest $^{143}Nd/^{144}Nd$ ratios plot near the 4.5 Ga Pb geochron (Ellam and Stuart, 2004; Graham et al., 1998) has been used to suggest that the source region for these magmas derives from a portion of the mantle formed very early in Earth history that has not experienced the level of outgassing recorded by most of the mantle (Jackson et al., 2010). If the high $^3He/^4He$ source indeed formed in Earth differentiation processes that occurred before ∼4 Ga, while ^{146}Sm was still extant, one might expect to find $^{142}Nd/^{144}Nd$ anomalies in the mantle-derived magmas that have the highest $^3He/^4He$.

Table 1
Nd and He isotope data for Baffin Island picrites.

Sample[a]	^{142}Nd/^{144}Nd ±2σ	μ^{142}Nd ±2σ	^{143}Nd/^{144}Nd ±2σ	ε^{143}Nd ±2σ	^{3}He/^{4}He (R/R_a)[b]
CS6	1.1418305	−4.2	0.513000	+7.22	37.9
	±54	±4.7	±4	±0.08	±1.2
CS7	1.1418267	−7.6	0.512838	+4.05	43.9
	±72	±6.3	±6	±0.12	±0.6
PAD6	1.1418360	+0.6	0.512932	+5.88	45.0
	±84	±7.4	±4	±0.08	±1.0
PAD7	1.1418358	+0.4	0.513103	+9.22	43.4
	±58	±4.4	±5	±0.09	±0.3
PAD8	1.1418354	0.0	0.513091	+9.00	48.0
	±53	±4.6	±5	±0.10	±0.8
PAD9	1.1418374	+1.8	0.513109	+9.34	43.6
	±54	±4.7	±4	±0.08	±0.6
DUR8	1.1418364	+0.9	0.512992	+7.06	49.8
	±52	±4.6	±4	±0.08	±0.7
APO3	1.1418313	−3.5	0.513083	+8.84	46.2
	±54	±4.7	±4	±0.08	±0.1
APO4	1.1418337	−1.4	0.513133	+9.81	38.1
	±52	±4.6	±4	±0.08	±2.0
APO5	1.1418288	−5.7	0.513102	+9.20	39.2
	±58	±5.1	±5	±0.10	±0.2
APO7	1.1418347	−0.6	0.513092	+9.02	46.2
	±49	±4.3	±4	±0.08	±0.3
Average BI	1.1418338	−1.4			
	±72	±6.3			
JNdi	1.1418353	0	0.512106		
	±56	±4.9	±5		

Notes: Average BI (Baffin Island) is the mean of all mass spectrometer runs of all samples. All uncertainties listed are two standard deviations.

[a] Sample names indicate sample locations, with CS = Cape Searle, PAD = Padloping Island, DUR = Durban Island, APO = Akpat Point.

[b] Helium isotope data from Starkey et al. (2009), except for CS6 and CS7 (Stuart et al., 2003).

2. Analytical procedures

For each sample, ~200 mg of whole rock powder was digested at SUERC using successive HF/HNO$_3$–HNO$_3$-6 M HCl treatment in Savillex® vials on a hotplate at 120 °C overnight. Following REE separation on cation exchange columns containing AGW50-X8 resin, Nd was separated by reversed phase extraction chromatography using Ln.Spec resin. The Nd-fraction (containing Ce, but free of Sm) was further purified at DTM using cation exchange columns with AGW50-X8 resin and methylactic acid as eluent. The Nd fractions were dissolved in 3 M HCl and loaded onto the evaporation filament of an outgassed double Re filament assembly that were loaded into the Thermo-Finnigan Triton thermal ionization mass spectrometer at DTM for Nd isotopic measurement. Each dynamic measurement of positive Nd ions consisted of 18 blocks of 30 cycles each, with ~8 s integration time. Baselines were measured before each block, lens focusing every 5th block and peak centering every 10th block. All ratios were corrected for mass fractionation using the exponential law and ^{146}Nd/^{144}Nd = 0.7219. Corrections were applied for Ce and Sm interferences, but are insignificant due to low Ce and Sm signals. Target ion beam intensity was 3×10^{-11} A on ^{142}Nd. Some samples were run twice whenever possible to allow measurement of more cycles which were combined with the previous run to obtain overall higher precision. The measured Nd isotopic data, including data for all the stable Nd isotopes and for potential interferences, are reported in the supplementary file, while Table 1 summarizes the results for ^{142}Nd/^{144}Nd and ^{143}Nd/^{144}Nd.

3. Results

For this study, eleven high ^{3}He/^{4}He ratio picrites from Baffin Island and surrounding areas were chosen for analysis of ^{142}Nd/^{144}Nd. The samples selected for this study have ^{3}He/^{4}He between 37.9 and 49.8R$_a$ with ε^{143}Nd from +4.0 to +9.8 (Table 1). The new data complement ^{142}Nd/^{144}Nd analyses of two Baffin Island picrites presented by Rizo et al. (2016) that averaged 3 to 5 ppm above the value measured for the terrestrial standard. The ^{142}Nd/^{144}Nd results for the samples measured in this study range from identical to the JNdi standard to as much as 8 ppm lower (Table 1). The average μ^{142}Nd for all samples is -1.4 ± 6.3 ppm (2σ of 15 analyses of 11 samples including 4 repeat analyses – see supplemental data file for complete data) and thus is not resolved from the value of the JNdi Nd standard. μ^{142}Nd is defined as:

$$\mu^{142}\text{Nd} = \left(\frac{\frac{^{142}\text{Nd}}{^{144}\text{Nd}}_{Sample}}{\frac{^{142}\text{Nd}}{^{144}\text{Nd}}_{Standard}} - 1 \right) \times 10^6$$

These values are indistinguishable within uncertainty from all published high-precision ^{142}Nd/^{144}Nd data for modern mantle-derived basalts (e.g. Jackson and Carlson, 2012). The new ^{142}Nd/^{144}Nd data show no correlation with ^{3}He/^{4}He (Fig. 1a) or ^{143}Nd/^{144}Nd (Fig. 1b) and thus fail to distinguish them from any other Phanerozoic mantle-derived melt, including MORB.

4. Discussion

4.1. Ancient, but incompatible element depleted, source?

Whether or not the results reported here support or argue against an ancient formation of the high ^{3}He/^{4}He source depends in part on whether or not the ^{142}Nd/^{144}Nd offset between Earth and meteorites is entirely due to nucleosynthetic causes. If due solely to nucleosynthetic differences, Fig. 2a shows the Nd data for the Baffin Island picrites to overlap or scatter below the two-stage Nd-geochron. This would imply that the superchondritic ^{143}Nd/^{144}Nd of the samples reflects a source whose Sm/Nd ratio became superchondritic after the extinction of ^{146}Sm; essentially any time after about 4 Ga. Such a possibility would be consistent with the traditional explanation that the incompatible element depleted mantle became so because of the extraction of incompatible element rich continental crust over Earth history. The problem is then how to explain the apparent coupling of high ^{3}He/^{4}He with the Sr, Nd and Hf isotopic signatures of lithophile element depletion. On the other hand, if the Earth started with a ^{142}Nd/^{144}Nd ratio similar to that of enstatite chondrites, the new data lie within the two-stage Nd-geochron and provide poorly defined source differentiation ages between 4.3 and 4.5 Ga (Fig. 2b). This could imply that the high ^{3}He/^{4}He mantle reservoir is an incompatible element depleted material created during early Earth differentiation. This interpretation, however, also would apply to modern MORB, which, with the limited data currently available (Boyet and Carlson, 2006; Caro et al., 2006; Jackson and Carlson, 2012), overlap in ^{142}Nd/^{144}Nd with the picrite data reported here. To explain the relatively low ^{3}He/^{4}He of MORB would then require the addition of another mechanism, such as the extraction of Earth's crust, to effectively outgas the MORB source. The degree to which the mantle residue left behind by continent formation is depleted in incompatible elements depends on the volume of mantle whose composition was affected by continent extraction. To explain all of the incompatible element depletion of the modern MORB-source mantle by continent extraction requires that only roughly a third of the mantle have been affected by continent extraction (Jacobsen and Wasserburg, 1979). In contrast, if the continental crust was extracted from the whole mantle, the degree of incompatible element depletion of the residual mantle is less significant, resulting in present day ε^{143}Nd in the residual mantle that would range from +4.7 to +1.9 for mean crust extraction ages of 4.57 Ga and 1.8 Ga, respectively. This result presents the possibility that the modern depleted mantle was derived by

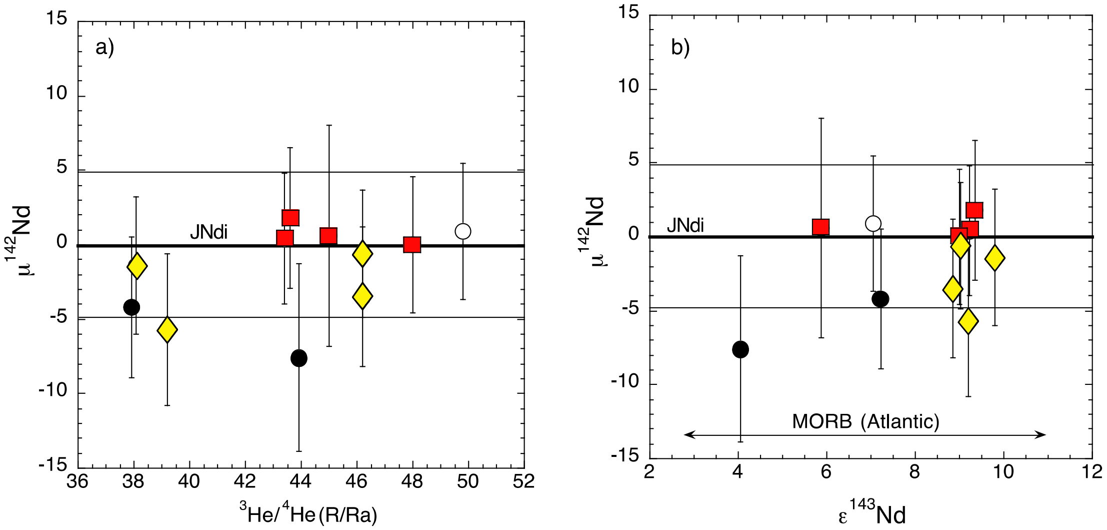

Fig. 1. a. ^{142}Nd/^{144}Nd versus ^{3}He/^{4}He indicating constant ^{142}Nd/^{144}Nd for a range of ^{3}He/^{4}He in the Baffin Island picrites. Average ^{142}Nd/^{144}Nd value for the high ^{3}He/^{4}He Baffin Island picrites is 1.141834 ± 7 (2σ of 15 measurements of 11 samples) compared to 1.141835 ± 6 (2σ of 7 runs: horizontal lines) for the terrestrial Nd standard JNdi measured at the time of the sample runs. Sample locations: Cape Searle (black circles), Padloping Island (red squares), Durban Island (open circle), Akpat Point (yellow diamonds). b. ^{142}Nd/^{144}Nd versus ^{143}Nd/^{144}Nd indicating constant ^{142}Nd/^{144}Nd for a range of ^{143}Nd/^{144}Nd in the Baffin Island picrites. Arrow indicates ^{143}Nd/^{144}Nd range in Atlantic MORB where very limited data (Jackson and Carlson, 2012) indicate these MORB have μ^{142}Nd = 0.

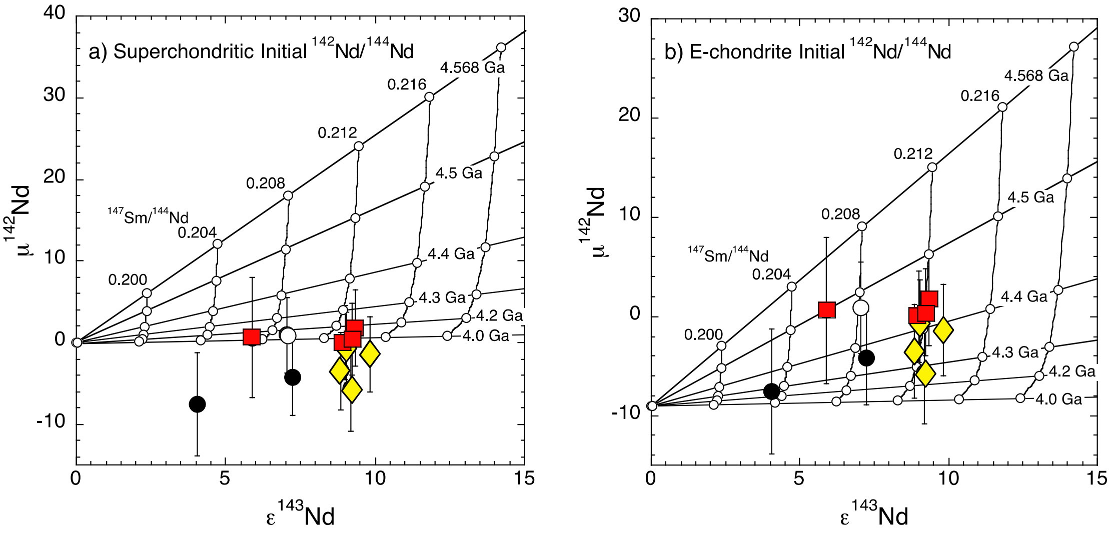

Fig. 2. Two-stage model isotopic evolution for the coupled ^{146}Sm–^{142}Nd and ^{147}Sm–^{143}Nd systems. Stage one involves evolution with chondritic ^{147}Sm/^{144}Nd (0.1960) (Bouvier et al., 2008) until the times marked on the lines to the right of each figure at which point the ^{147}Sm/^{144}Nd ratios are increased to the values denoted by the near-vertical lines connecting the open circles. The data points are the Baffin picrite data using the same point code as in Fig. 1. In part "a", the Earth is assumed to start with a ^{142}Nd/^{144}Nd ratio that would evolve to the present day value measured for the JNdi Nd standard after 4.568 Ga of evolution with chondritic Sm/Nd ratio. Part "b" assumes Earth starts from the 9 ppm lower ^{142}Nd/^{144}Nd average value measured for enstatite chondrites (Burkhardt et al., 2016; Gannoun et al., 2011).

continent extraction not from a primitive reservoir with chondritic relative abundances of the refractory lithophile elements, but instead one that was already incompatible element depleted as a result of early Earth differentiation events (Boyet and Carlson, 2006; Caro, 2011). In this case, the higher ^{143}Nd/^{144}Nd, lower ^{3}He/^{4}He, but similar ^{142}Nd/^{144}Nd of MORB compared to the Baffin Island picrites would reflect the additional incompatible element depletion of the MORB source caused by continent extraction that occurred after ^{146}Sm was extinct.

4.2. Consequences of crustal contamination for estimating source composition

The question that remains is whether the high ^{3}He/^{4}He that distinguishes the Baffin Island picrites from most other modern mantle-derived melts can be tied to a source that has the chemical and isotopic signatures of incompatible-element-depletion. This topic has seen considerable previous discussion, much of which centers on the role of mixing of a high ^{3}He/^{4}He source with either the ancient continental crust through which these magmas erupted (Dale et al., 2009; Day, 2016; Starkey et al., 2009; Stuart et al., 2003) or the incompatible-element depleted upper mantle source of MORB (Starkey et al., 2009). Such mixing scenarios are hard to quantify because the potential end member compositions are so variable, but some generalities can be drawn. Fig. 3 shows the results of two types of mixing scenarios, one involving mixing between a primitive picritic magma and Archean continental crust, and the other between the incompatible element depleted mantle (Workman and Hart, 2005) and a hypothetical primitive mantle reservoir with chondritic relative abundances of the refractory lithophile elements (McDonough and Sun, 1995). Contamination with ancient continental crust will drive the mag-

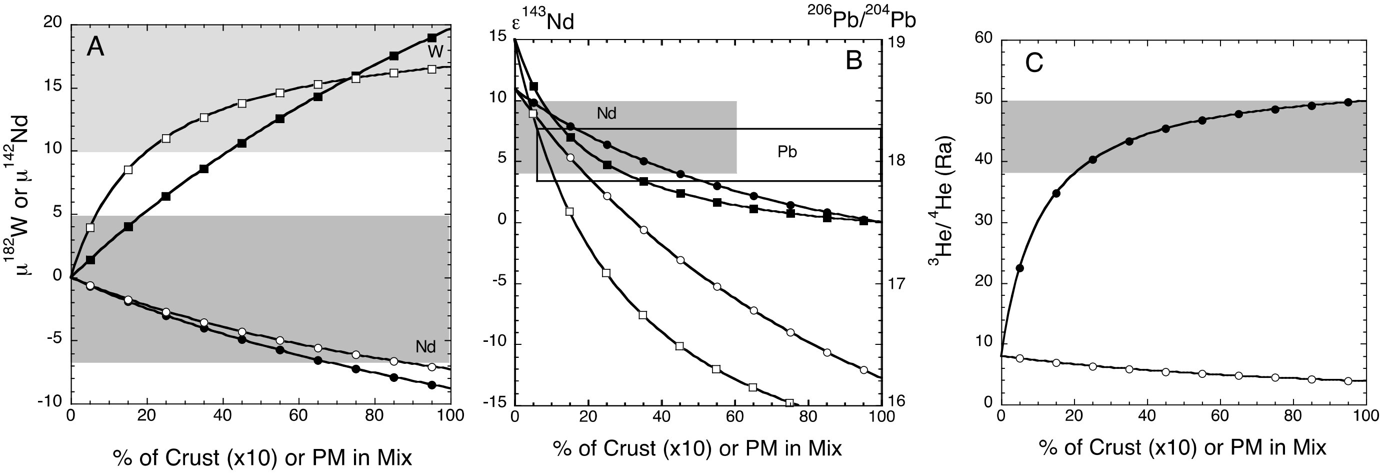

Fig. 3. Mixing curves between the mantle source of MORB (DMM) and primitive mantle (PM) (lines marked with solid symbols) where the *x*-axis ranges from 0 to 100% PM in the mixture, and between a primitive Baffin Island picritic magma and Archean continental crust (lines marked with open symbols) with the *x*-axis range from 0 to 10% crust. Figure A shows μ^{182}W (squares) and μ^{142}Nd (circles), B shows ε^{143}Nd (circles) and ^{206}Pb/^{204}Pb (squares), and C shows He isotopic composition expressed as the ratio to air (Ra). The composition of the end members is given in Supplemental Table 2. The boxes in each figure show the range in isotopic composition measured for the Baffin Island picrites.

mas to lower ^{143}Nd/^{144}Nd, ^{206}Pb/^{204}Pb, and ^{3}He/^{4}He, which would imply that the pre-contamination magmas had Nd and Pb isotopic compositions that trend towards values typical of North Atlantic MORB, but with higher ^{3}He/^{4}He than seen for any MORB. As seen in Fig. 3B, only 1–2% crustal contamination would be needed to lower the ε^{143}Nd and ^{206}Pb/^{204}Pb from values typical of North Atlantic MORB into the range measured for the Baffin Island picrites. The ability of crustal contamination to overprint ^{142}Nd/^{144}Nd is minimal simply because the total range in ^{142}Nd/^{144}Nd in terrestrial rocks is of the order of 30 ppm, only some 6 times typical measurement precisions for this ratio. Contamination by crust with μ^{142}Nd in the range of the lowest measured for any terrestrial rock (Caro et al., 2006; O'Neil et al., 2012) does not change the magma μ^{142}Nd beyond analytical uncertainty until the amount of crust in the mixture is some 8–10%. At this point the ε^{143}Nd is well below the values measured in these samples (Fig. 3A, B). Given the large W concentration difference between crust and mantle, the W isotopic composition of the Baffin Island magmas might be susceptible to substantial modification by crustal contamination. For example, the mixing calculations illustrated in Fig. 3A show that the μ^{182}W can be increased into the low end of the range measured for the Baffin Island picrites (Rizo et al., 2016) if the W isotopic composition of the crustal contaminant is in the range measured for Archean rocks from Greenland (Touboul et al., 2012; Willbold et al., 2011). Fig. 3C shows that He is among the elements that are not likely to be dramatically affected by crustal contamination. This is consistent with the observation that extremely high ^{3}He/^{4}He is recorded by olivines from crustally-contaminated basalts (e.g. Stuart et al., 2000; Stuart et al., 2003), likely reflecting trapping of melt inclusions in olivine in the deep crust prior to shallow contamination (Starkey et al., 2012). Helium isotopic compositions of relatively old (~62 Ma) basalt olivines can be lowered by radiogenic ingrowth of ^{4}He (Jackson et al., 2010; Zindler and Hart, 1986). While this could explain the range in ^{3}He/^{4}He seen in the picrites measured here, and hence the lack of correlation with ^{142}Nd/^{144}Nd, all the samples studied here have among the highest ^{3}He/^{4}He measured in terrestrial basalts, so the ingrowth of ^{4}He since eruption is of limited relevance in the interpretation of this dataset. Given the expected consequences of crustal contamination on the various isotope systems, the samples with the highest ^{143}Nd/^{144}Nd, ^{206}Pb/^{204}Pb and ^{3}He/^{4}He are the least crustally contaminated. The key question is whether even these samples have assimilated crust. If so, then the primary magma had Sr, Nd and Pb isotopic compositions indistinguishable from North Atlantic MORB, but much higher ^{3}He/^{4}He.

4.3. Isotopic expression of mixing of incompatible element depleted and primitive mantle

Whether or not any of the Baffin Island picrites have escaped a crustal overprint, most of the high ^{3}He/^{4}He ratio basalts appear to be associated with Sr and Nd isotopic compositions indicative of an incompatible element depleted source. Starkey et al. (2009) argued that this reflects decoupling of He from other chemical and isotopic tracers. One way to accomplish this decoupling is through mixing between reservoirs with similar incompatible element abundances, but very different He concentrations. Fig. 3 explores the isotopic consequences of such a mixing scenario. Given concentration estimates for the primitive mantle (PM) (McDonough and Sun, 1995) and the depleted MORB mantle (DMM) (Workman and Hart, 2005), Fig. 3B shows that mixtures of these two mantle reservoirs can produce a source with Nd and Pb isotopic composition that overlaps the picrites at DMM:PM ratios from 85:15 to 70:30. Any crustal contamination experienced by these magmas would drive these ratios in the direction of an increased abundance of DMM in the mixture. If the PM has He concentrations at least ten times higher than the DMM, then similar DMM-PM mixing ratios can also explain the high ^{3}He/^{4}He ratios measured in the Baffin Island picrites (Fig. 3C). Fig. 3A indicates that the PM would have to have μ^{142}Nd similar to, or lower than, DMM, and μ^{182}W substantially higher than DMM in order to match the isotopic composition of the Baffin picrites. The association of high ^{3}He/^{4}He with high μ^{182}W (Rizo et al., 2016) is opposite of the trend to lower μ^{182}W with increasing ^{3}He/^{4}He reported by Mundle et al. (2017) in a variety of ocean island basalts, so the relationship of high ^{3}He/^{4}He mantle sources with the information recorded in short-lived isotope systems requires more data to be fully understood.

5. Conclusions

The high ^{3}He/^{4}He and lithophile element isotope systematics of the Baffin Island picrites thus can be explained in at least two ways:

1) The picrite melts are a mixture of DMM and PM. In this case, the PM has He concentrations at least ten times higher than DMM,

μ^{142}Nd either the same as, or as much as 30 ppm lower than, DMM, with μ^{182}W substantially higher than DMM.

2) The picrite source is an incompatible element depleted region of the mantle formed in an early-Earth differentiation event that did not allow effective outgassing of He. This model is supported if the difference in ^{142}Nd/^{144}Nd between the modern Earth and chondrites is not solely the result of a different mixture of the various nucleosynthetic contributions to Nd. The overlapping μ^{142}Nd, but very different ^{3}He/^{4}He of the Baffin picrites and MORB in this model would imply that DMM was developed by continent extraction from a portion of the early-formed mantle reservoir, increasing the incompatible element depletion and causing a dramatic reduction in noble gas concentrations.

Formation of the incompatible element depletion of the MORB source early in Earth history is supported by the difference in the ^{129}Xe/^{130}Xe ratio between some MORB and some OIB (Mukhopadhyay, 2012), which was generated by the decay of 15 Ma half-life ^{129}I. Similarly, the elevated μ^{182}W of Baffin Island picrites also supports the development of a mantle reservoir very early in Earth history while ^{182}Hf was still extant. The lack of an obvious signature of a fractionated Sm/Nd ratio as reflected in the ^{142}Nd/^{144}Nd isotopic composition of the Baffin Island picrites suggests that the early differentiation was more effective at fractionating siderophile and atmophile, than lithophile, elements. The mechanism of origin of the high ^{3}He/^{4}He source thus may reflect core formation and volatile loss from other portions of the mantle, but not necessarily intra-mantle differentiation.

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Appendix A. Supplementary material

Supplementary material related to this article can be found online at http://dx.doi.org/10.1016/j.epsl.2017.05.005.

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