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Helium isotopes in early Iceland plume picrites: Constraints on the composition of high $^3He/^4He$ mantle

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ABSTRACT

A detailed study of the geochemistry of a new suite of early Iceland plume picrites shows that extremely high $^3He/^4He$ ratios (up to 50 R_a) are found in picrites from Baffin Island and West Greenland. High $^3He/^4He$ picrites display a wide range in $^{87}Sr/^{86}Sr$ (0.70288–0.70403), $^{143}Nd/^{144}Nd$ (0.51288–0.51308) and incompatible trace element ratios (e.g. La/Sm_n=0.5–1.6). These overlap the complete range of compositions of mid-ocean ridge basalts and most northern hemisphere ocean island basalts, including Iceland. Crustal contamination modelling in which high-grade Proterozoic crustal basement rocks for the region are mixed with a depleted parent cannot account for the trend displayed by the Baffin Island and West Greenland picrites. This rules out the possibility that the incompatible trace element, Sr and Nd isotope range of the high $^3He/^4He$ picrites is due to crustal contamination. The compositional range at high $^3He/^4He$ is also inconsistent with derivation from a primordial-He-rich reservoir that is a residue of ancient mantle depletion. This implies that the composition of the high $^3He/^4He$ mantle cannot be determined simply by extrapolating ocean island basalt He–Sr–Nd–Pb–Os isotope data.

The apparent decoupling of He from trace element and lithophile radiogenic isotope tracers is difficult to attain by simple mixing of a high-[He], high $^3He/^4He$ reservoir with various depleted and enriched He-poor mantle reservoirs. The possibility that primordial He has diffused into a reservoir with a composition typical of convecting upper mantle cannot be ruled out. If so, the process must have occurred after the development of existing mantle heterogeneity, and requires the existence of a deep, primordial He-rich reservoir.

1. Introduction

The recognition that primordial He trapped during Earth accretion is still degassing to the atmosphere placed fundamental constraints on the evolution of the Earth (e.g. Craig et al., 1975). Subsequent studies demonstrated that basalts from ocean islands such as Iceland, Hawaii and Samoa, have higher time-integrated $^3He/(U+Th)$ than the depleted upper mantle that is sampled by mid-ocean ridge basalt (MORB) volcanism (Kurz et al., 1982; Allègre et al., 1983). This led to the view that the inferred lower mantle source of high $^3He/^4He$ ocean island basalts (OIB) is less degassed, and therefore unprocessed (Kurz et al., 1982; Allègre et al., 1983; Kaneoka et al., 1983; Allègre et al., 1987). While a convectively isolated lower mantle is now difficult to reconcile with geophysical evidence for whole mantle convection (e.g. Grand et al., 1997), the chemical composition of the high-$^3He/^4He$ deep mantle reservoir remains a significant research focus. It has been argued that the incompatible trace element and Sr, Nd and Pb isotope data for high $^3He/^4He$ OIB appear to converge on distinct compositions resulting in a number of postulated mantle "end members" (Farley et al., 1992; Hart et al., 1992; Hilton et al., 1999; Jackson et al., 2007). Incompatible trace element ratios in high $^3He/^4He$ OIB are not primordial, and O, Sr and Os isotopes provide evidence for the presence of recycled material that has undergone low temperature alteration at the Earth's surface (Eiler et al., 1996; Lassiter and Hauri, 1998; Becker, 2000; Macpherson et al., 2005). The correlation between He, O and Os isotopes in Icelandic basalts is best modelled as a mixture of a primordial He-rich mantle reservoir with >1 Ga recycled oceanic crust (Brandon et al., 2007).

To date studies have concentrated largely on OIB with $^3He/^4He$ between 15 and 30 R_a (where R_a is the atmospheric value 1.39×10^{-6}). The highest mantle $^3He/^4He$ (40–50 R_a) recorded in picrites from Baffin Island, northern Canada provide the least diluted primordial He signal (Stuart et al., 2003). The $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$ and incompatible trace element composition of these lavas suggest that the high $^3He/^4He$ source mantle has a composition that is typical of depleted mantle (Stuart et al., 2003). This is counter-intuitive and inconsistent with models that put high $^3He/^4He$ in a more primitive reservoir. A compilation of all $^3He/^4He$ data from the early Iceland plume (Baffin Island, West and East Greenland, Scotland) appears to show a linear array from low $^3He/^4He$,

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low $^{143}Nd/^{144}Nd$ and high $^{87}Sr/^{86}Sr$ to high $^{3}He/^{4}He$, high $^{143}Nd/^{144}Nd$ and low $^{87}Sr/^{86}Sr$, confirming the depleted nature of the high $^{3}He/^{4}He$ source (Ellam and Stuart, 2004). Data arrays from ocean islands never cross this trend, leading Ellam and Stuart (2004) to suggest that the high $^{3}He/^{4}He$ depleted mantle is a discrete reservoir (helium recharged depleted mantle (HRDM)), the result of ancient mixing between primordial gas-rich mantle and depleted mantle.

Contrary to the long-held view that He is more incompatible than U during melting (e.g. Marty and Lussiez, 1993), recent estimates of helium partition coefficients suggests it may actually be more compatible than U during mantle melting (Parman et al., 2005). Class and Goldstein (2005) considered the evolution of He in a convecting whole mantle and concluded that a depleted, high $^{3}He/^{4}He$ mantle reservoir could originate as a residue of ancient melting that has been isolated from the convecting mantle for billions of years. Starting with the assumption that $D_U < D_{He}$, Parman (2007) took this one stage further and argued that apparent peaks in the global OIB $^{3}He/^{4}He$ dataset derive from discrete mantle domains that were depleted episodically over earth history. However, Rudge (2008) shows that only two of the eight peaks apparent in the helium-continental crust correlation of Parman (2007) are likely to be statistically significant. More recently, modelling of the He isotope evolution of the mantle shows that residual mantle formed in the last few Gyr is so strongly depleted in He that the isotopic composition is unlikely to be preserved during subsequent mixing (Porcelli and Elliott, 2008). The recognition that all mantle-derived rocks have $^{142}Nd/^{144}Nd$ ratios that are 20 ppm higher than chondritic values (Boyet and Carlson, 2005) led Boyet and Carlson (2006) to argue that the Earth experienced an early differentiation event that produced a hidden Early Depleted Reservoir with high $^{3}He/^{4}He$ and super-chondritic $^{142}Nd/^{144}Nd$.

In an effort to better constrain the composition of the high $^{3}He/^{4}He$ mantle we carried out a detailed geochemical and isotopic study of a new collection of proto-Iceland plume picrites and magnesian basalts from Baffin Island and West Greenland. The results demonstrate that extremely high $^{3}He/^{4}He$ is a consistent and widespread feature of the earliest lavas erupted by the Iceland plume. In contrast to the apparently specific compositions obtained by extrapolating trends in moderately high $^{3}He/^{4}He$ OIB data, the high $^{3}He/^{4}He$ Baffin Island–West Greenland picrites display a wide range of trace element and radiogenic isotope compositions. This strongly implies that the high $^{3}He/^{4}He$ reservoir in Earth does not have a unique composition for other isotopic tracers.

2. Early Iceland plume volcanic rocks from Baffin Island and West Greenland

Thick sequences of high-Mg basalts were erupted between 62 and 58 Ma on Baffin Island and West Greenland (along with tholeiitic lava flows in south and central east Greenland, the Faeroe Islands and the British Isles; Fig. 1a; Saunders et al., 1997). The major element chemistry of, in particular, the Baffin Island and West Greenland picrites is thought to reflect derivation by high temperature melting (Larsen and Pedersen, 2000; Herzberg et al., 2007) which is consistent with derivation from the start-up head of the Iceland plume.

The Baffin Island and West Greenland lavas are considered to be stratigraphically equivalent. The lavas were erupted through Precambrian crust and lithosphere that was thinned in response to rifting in the Labrador Sea and Baffin Bay (Clarke and Upton, 1971). The onshore part of the Tertiary volcanic province of West Greenland comprises 22,000 km^3 of picritic and basaltic lava flows between the Svartenhuk Peninsula and Disko (Clarke and Pedersen, 1976). Vertical thicknesses of the exposed succession on Disko and Nuussuaq are between 2 and 3 km (Larsen and Pedersen, 2000). From $^{40}Ar/^{39}Ar$ age determinations it appears that the West Greenland succession (Vaigat and Maligat Formations) was erupted in less than 1 million years (Storey et al., 1998). The lowermost lavas (Vaigat Formation) are subdivided into the Anaanaa, Naujánguit and Ordlingassoq members (Larsen and Pedersen, 2000). The start of each phase was characterised by olivine-rich magmas, ending with either a lithospherically and/or crustally contaminated interval in the case of the Anaanaa and Naujánguit members or an evolved interval in the case of Ordlingassoq Member (Larsen and Pedersen, 2000). Except for the very easternmost parts of the region, the lavas lie on Cretaceous and Tertiary sediments of the Nuussuaq Basin.

The lavas on Baffin Island are exposed as a series of outcrops along the eastern coast between Cape Searle and Cape Dyer (Fig. 1b). They lie on Precambrian basement and Tertiary sediments. Total thicknesses never exceed 750 m (Francis, 1985). The lower few hundred metres are dominantly sub-aqueous flows, composed of pillows and hyaloclastite breccias, while the upper 400 m or so comprises thin subaerial flows (Robillard et al., 1992). Our collection also includes a small number of picritic dykes that cut the lavas (samples: BI/CS/7, 8, 11, 12, 13 and BI/PI/16). The Baffin Island lavas have normal magnetic polarity belonging to Chron 27n (Deutsch et al., 1971) which by correlation to the Anaanaa Member (Pedersen et al., 2002) implies an age of 61.7–62.0 Ma.

The Baffin Island and West Greenland lavas contain up to 30 wt.% MgO. Many rocks contain accumulated olivine and very few samples have less than 12 wt.% MgO. The majority of samples are classed as picrites (IUGS classification; Le Bas, 2000) which are dominantly tholeiitic (Larsen et al., 2003). The Baffin Island lavas have been subdivided into N-type and E-type based on their trace element and isotopic composition (Robillard et al., 1992). N-type rocks are dominated by higher MgO composition and have light rare earth element (LREE) depleted patterns typical of N-MORB while E-type lavas have flat to slightly LREE enriched patterns, similar to E-MORB. These N- and E-type groups are not distinguishable by their major element composition. The West Greenland picrites broadly fall into similar N- and E-type groups (Holm et al., 1993). Crustal contamination of the West Greenland lavas is thought to have affected only subordinate units of silica-rich basalts and magnesian andesites (Larsen et al., 2003). The magnesian magmas were erupted through the thick lithosphere very rapidly and are thought to have avoided significant contamination (Gill et al., 1992; Holm et al., 1993; Lightfoot et al., 1997; Larsen and Pedersen, 2000). It is likely that the picrites of Baffin Island were erupted in a very similar manner and therefore also remain unaffected by crustal contamination, something we investigate in Section 5.1.

Olivine phenocrysts in the Baffin Island and West Greenland lavas typically range from Fo_{86} to Fo_{93} (Francis, 1985; Larsen and Pedersen, 2000). Francis (1985) suggested that Fo_{93} olivine crystals in the Baffin Island picrites are xenocrysts. Fo-rich olivines in West Greenland picrites on the other hand were considered to be integral to the cognate mineral assemblage (Larsen and Pedersen, 2000). The high-forsterite crystals have led to controversy regarding the composition and temperature of primary melts. If Fo_{93} olivines represent early-formed phenocrysts rather than xenocrysts then it is possible that Baffin Island picrites require parental magma compositions in excess of 18 wt.% MgO and temperatures of at least 1425 °C (Francis, 1985; Robillard et al., 1992; Herzberg and O'Hara, 2002). Larsen and Pedersen (2000) suggest that $Fo_{92.5}$ crystals in West Greenland picrites can be used to infer parental melts with 20–21 wt.% MgO and liquidus temperatures of 1515–1560 °C. This implies high degrees of melting (~25%) of anomalously hot mantle at depths of 60–120 km (Scarrow and Cox, 1995; Graham et al., 1998; Lightfoot et al., 1997). Disregarding the high-forsterite crystals and assuming that $Fo_{86–89}$ olivines are the most primitive phenocrysts implies parental melts with only 11–13 wt.% MgO (Francis, 1985).

3. Sample description and analytical methods

We present new geochemical data from 108 picrite samples from Baffin Island and West Greenland. Nineteen of the Baffin Island samples were collected in 2004 and 75 were sampled in 1996. The Baffin Island samples were collected from Cape Searle, Padloping Island, Durban Island and Akpat Point. The 14 West Greenland

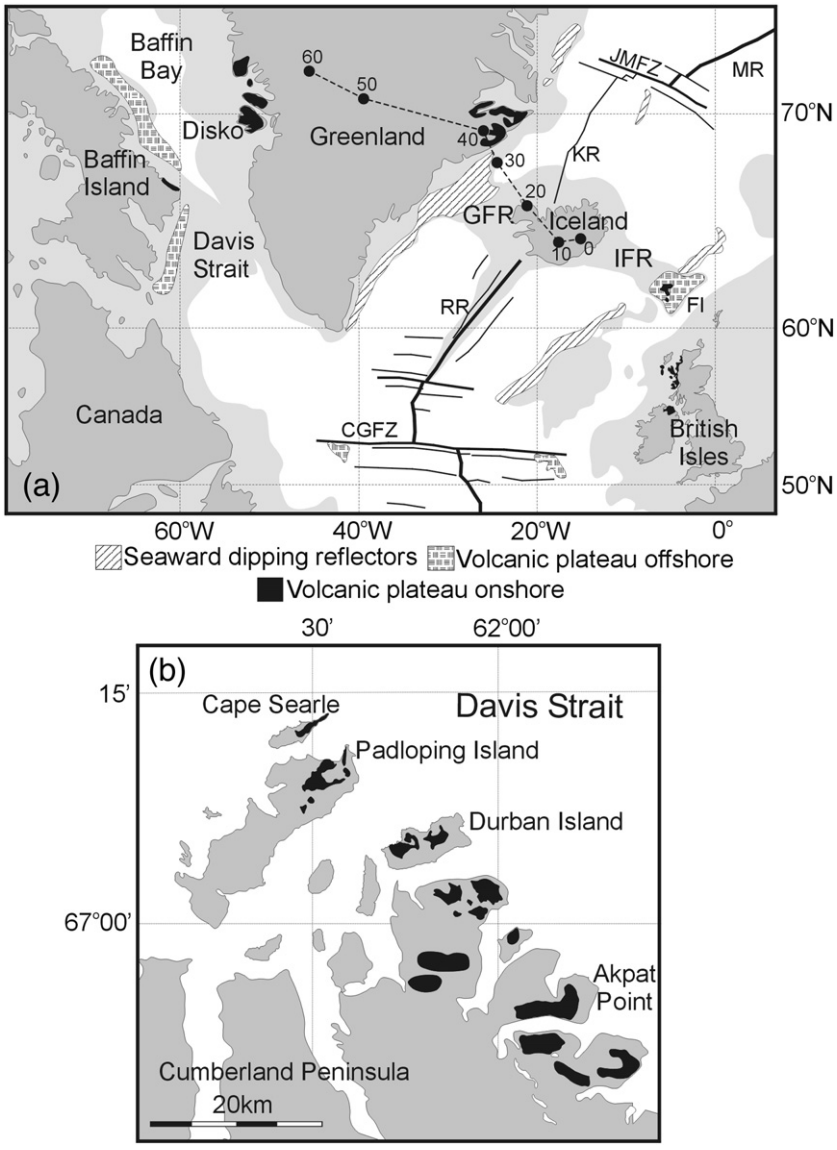

Fig. 1. a) Location of onshore and offshore plateau basalts and seaward dipping reflectors in the North Atlantic Igneous Province (after Saunders et al., 1997). Plume track from 60 Ma to present is shown by dashed line. CGFZ=Charlie Gibbs Fracture Zone, RR=Reykjanes Ridge, GFR=Greenland-Faeroes Ridge, IFR=Iceland-Faeroes Ridge, FI=Faeroe Islands, KR=Kolbeinsey Ridge, JMFZ=Jan Mayen Fracture Zone, MR=Mohns Ridge. b) Location of onshore volcanic outcrops in Baffin Island.

samples form part of an existing collection (Larsen and Pedersen, 2000) and were selected as suitable for He isotope determinations based on their high olivine content. These samples are from all parts of the Vaigat Formation on Disko Island and the Nuussuaq Peninsula: the lowermost Anaanaa Member (4 samples), the Naujánguit Member (5 samples) and the Ordlingassoq Member (5 samples).

Major and selected trace elements (Nb, Y, Zr, Ba, Sr, Rb, Cu, Ni, Cr, V, Sc) were measured on whole-rock powders by X-ray fluorescence spectrometry at the University of Edinburgh following the procedures of Fitton et al. (1998). Trace element precision was estimated by analysing international standards (BHVO1, BCR1, BIR1 and DTS1) repeatedly during analytical runs (see Table 1 in Supplementary Information). Rare earth elements (REE) and selected trace elements (Hf, Ta, Th, U) were measured at Scottish Universities Environmental Research Centre (SUERC) by inductively coupled plasma mass spectrometry (Olive et al., 2001) with a precision of ~2% for all elements (see Table 2 in Supplementary Information). Sr and Nd isotopes were measured at SUERC on separate splits of whole-rock powder from 73 samples by thermal ionisation mass spectrometry (more details provided in Supplementary Information). Helium isotope ratios were measured at SUERC in 19 picrites from Baffin Island and 11 from West Greenland. All samples were collected from near-vertical cliff sections to minimise the potential for *in situ* cosmogenic ^{3}He production. Isotope ratios were measured in gases released by *in vacuo* crushing of ~1 g of olivine phenocrysts following established procedures (Williams et al., 2005). In the earlier study Stuart et al. (2003) demonstrated that there is no significant cosmogenic He present in Baffin Island picrite olivines. Subsequent work confirms that the crushing procedure does not release *in situ* cosmogenic helium from basalts that have been exposed for over 100 kyr (e.g. Foeken et al., 2009). The potential release of post-eruptive radiogenic ^{4}He from the olivine lattice during crushing would only result in measured ratios underestimating the source ^{3}He/^{4}He (Stuart et al., 2003). Full details of analytical techniques can be found in Supplementary Information.

4. Results

^{3}He/^{4}He in the 30 samples ranges from 16.4 to 49.8 R_a (Table 1 presents the representative data with the full dataset in Supplementary Information). These ratios overlap the high values previously obtained

for Baffin Island (Stuart et al., 2003) and are significantly higher than the highest value determined previously for West Greenland picrites (30 R_a; Graham et al., 1998). Sample 400457 reported by Graham et al. (1998) was re-analysed in this study and yielded $^3He/^4He$ of 16.4±2 R_a, within analytical uncertainty of the original value (17.55 R_a). High $^3He/^4He$ lava flows are distributed throughout the entire Vaigat Formation of West Greenland. The high $^3He/^4He$ are clearly characteristic of the early Iceland plume source and are significantly in excess of the highest values measured in recently erupted ocean island basalts (30–34 R_a; Kurz et al., 1982; Macpherson et al., 2005; Jackson et al., 2007).

Major and trace element compositions are presented in Supplementary Information. MgO ranges from 9 to 29 wt.%. Major element oxides define tight linear trends of decreasing SiO_2, CaO, Al_2O_3, Na_2O, TiO_2, MnO, P_2O_5 and increasing FeO, with increasing MgO. The tight linear trends can be modelled as olivine control lines suggesting that the lavas accumulated varying proportions of olivine, accounting for their high MgO contents. No evidence is found for the existence of xenocrystic olivine crystals such as kink banding of large, high forsterite crystals (as documented by Francis, 1985) or rims of crystal overgrowth that would suggest textural or compositional disequilibrium.

At any given MgO content, incompatible trace element concentrations for West Greenland picrites are generally slightly higher than those of Baffin Island but with considerable overlap. When considering both locations Ni and Cr increase with increasing MgO whereas V, Sc and Y decrease. Cu, Sr, Nb and Zr show poor negative correlations with MgO whilst Zn, Rb and Ba are scattered.

Table 1
Nd, Sr and He isotope ratios and selected trace element data for early Tertiary picrites from Baffin Island and West Greenland (Full dataset is in Supplementary Information)

Sample	Nb	Zr	Y	La	Nd	Sm	$^{87}Sr/^{86}Sr_i$	$^{143}Nd/^{144}Nd_i$	$^3He/^4He$ (R/R_a)
Padloping Island									
PAD2	1.40	43.05	15.99	1.874	5.127	1.682	0.703142	0.513000	38.1±3.0
PAD3	0.80	29.91	12.88	1.129	3.465	1.221	0.703050	0.512994	31.1±2.7
PAD4	1.10	39.62	15.49	1.620	4.466	1.536	0.703539		44.4±0.1
PAD5	1.00	39.92	16.89	1.589	4.671	1.642	0.703370	0.512987	40.3±0.4
PAD6	4.19	49.93	17.60	3.385	6.037	1.898	0.703806	0.512876	45.0±1.0
PAD7	1.10	40.53	16.19	1.675	4.815	1.668	0.703058		43.4±0.3
PAD8	1.40	42.85	16.19	1.917	5.041	1.678	0.703109	0.513034	48.0±0.8
PAD9	0.70	22.23	9.67	0.914	2.728	0.963	0.703200	0.513031	43.6±0.6
Durban Island									
DUR3	0.90	38.91	16.99	1.381	4.197	1.549	0.703451		38.5±2.0
DUR5	0.50	27.19	12.38	1.165	3.232	1.152	0.703578	0.512968	40.9±0.6
DUR6	1.00	39.72	17.80	1.681	4.532	1.639			29.7±4.5
DUR8	3.50	41.74	14.08	2.966	5.422	1.636	0.703398	0.512975	49.8±0.7
Akpat Point									
APO1	0.50	20.72	9.37	0.702	2.358	0.871	0.702883	0.513070	36.9±2.1
APO3	1.50	42.75	14.99	1.850	5.129	1.704	0.703243	0.513030	46.2±0.1
APO4	0.80	31.13	13.78	1.079	3.493	1.312	0.703132		38.1±2.0
APO5	1.70	54.58	18.40	2.464	6.792	2.219	0.703225	0.513043	39.2±0.2
APO7	1.20	36.48	13.38	1.675	4.659	1.528	0.703146	0.513058	46.2±0.3
Cape Searle									
BI/CS/22	3.46	44.20	16.20	3.120	5.480	1.740	0.703420	0.512923	31.9±1.0
BI/CS/17	4.17	50.40	18.10	3.500	6.150	1.850	0.703480		27.0±1.0
West Greenland									
138345	1.60	43.25	15.00	2.007	5.297	1.796	0.703652	0.513008	22.7±1.7
340740	5.02	70.25	17.70	4.564	9.162	2.653	0.703287	0.512957	23.4±0.9
354754	2.21	49.02	16.50	2.386	5.904	1.933	0.703336	0.512938	46.7±0.7
400230	3.21	57.31	17.20	2.950	7.035	2.254	0.703384	0.512910	47.6±1.3
410152	6.12	69.54	17.60	5.372	9.680	2.678	0.703469	0.512932	19.3±1.8
264217	1.30	43.46	16.90	1.682	5.365	1.910	0.703052	0.513069	19.1±6.1
332901	4.72	66.61	15.10	4.575	8.961	2.538	1.703461	0.512952	45.8±1.0
362077	0.90	38.00	14.50	1.385	4.333	1.525	0.703042		44.0±0.8
400485	1.00	37.09	12.80	1.435	4.448	1.505	0.703374	0.513075	40.3±3.2
400452	1.10	44.97	15.60	1.702	5.378	1.853	0.704033	0.513053	35.4±2.2
400457	1.20	49.02	16.00	1.806	5.499	1.874	0.703281	0.513056	16.4±2.0

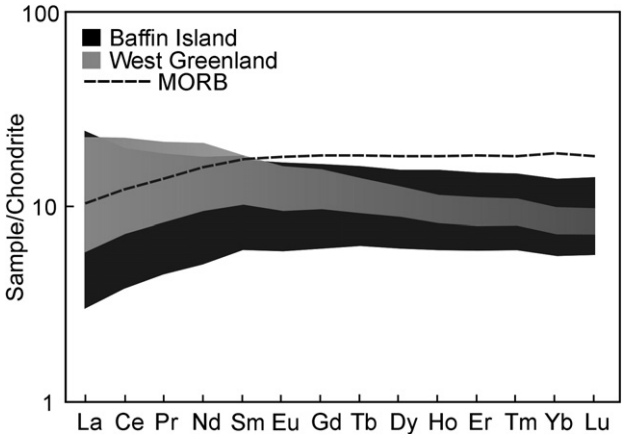

Fig. 2. Chondrite-normalised rare-earth element (REE) diagram for all Baffin Island and West Greenland samples (this study and Stuart et al., 2003). The samples have light-REE-depleted to slightly enriched patterns with West Greenland samples being slightly more LREE-enriched.

REE plots further highlight the higher abundances of incompatible elements in West Greenland compared to Baffin Island (Fig. 2). The chondrite-normalised REE patterns of Baffin Island are flatter than those of West Greenland, which tend to be slightly LREE enriched relative to Baffin Island samples. Comparison of the samples to MORB (Fig. 2) highlights the relative depletion in the HREEs of all the Baffin Island and West Greenland samples, consistent with greater average depth of melting.

All samples plot within the global MORB field on a Nb/Zr–La/Sm(n) plot (Fig. 3a). The samples cluster in two groups with varied degrees of depletion that correspond broadly to the N-MORB and E-MORB classification of Baffin Island picrites (Francis, 1985). The ΔNb parameter (Fitton et al., 1997) is useful as a discriminator of basalts derived from the Iceland-plume (ΔNb >0) and N-MORB (ΔNb <0) mantle sources. The Baffin Island–West Greenland picrites span the transition from N-MORB to Icelandic-type basalts (negative and positive ΔNb) (Fig. 3b) suggesting that both relatively enriched and relatively depleted magma sources were available in the earliest phase of the Iceland plume.

$^{87}Sr/^{86}Sr_i$ and $^{143}Nd/^{144}Nd_i$ (where i denotes the Sr and Nd isotopic value corrected to 60 Ma) for the newly measured samples vary from 0.70288 to 0.70493 and 0.51275 to 0.51308 respectively (Supplementary Information and Fig. 4a,b). The range of Sr isotope ratios extends beyond the Iceland range to both higher and lower values. Nd isotopes cover the range of Iceland values and extend to lower $^{143}Nd/^{144}Nd$. The Sr and Nd isotope values measured here are comparable to those reported in the literature for Vaigat Formation picrites of West Greenland (Holm et al., 1993; Lightfoot et al., 1997; Graham et al., 1998) and Baffin Island picrites (Robillard et al., 1992; Stuart et al., 2003; Kent et al., 2004).

5. Discussion

5.1. Does the high $^3He/^4He$ mantle have a unique composition?

Baffin Island and West Greenland picrite $^3He/^4He$ data (this study; Graham et al., 1998; Stuart et al., 2003) are plotted against Sr and Nd isotopes in Fig. 4. Picrites with $^3He/^4He$ higher than ~35 R_a, the upper limit of contemporary OIB (~34 R_a from Iceland, Macpherson et al., 2005, ~32 R_a from Loihi, Kurz et al., 1982, 27.4 R_a from Galapagos, Kurz and Geist, 1999, and ~33.8 R_a from Samoa, Jackson et al., 2007), display a wide range in $^{87}Sr/^{86}Sr$ (0.70288–0.70388) and $^{143}Nd/^{144}Nd$ (0.51288–0.51308). These values overlap those of lower $^3He/^4He$ (<35 R_a) picrites. Although the association of high $^3He/^4He$ with high $^{143}Nd/^{144}Nd$ and low $^{87}Sr/^{86}Sr$ typical of depleted mantle is still apparent, there are now several high $^3He/^4He$ samples with lower $^{143}Nd/^{144}Nd$ and higher $^{87}Sr/^{86}Sr$ than the range identified for Baffin Island picrites (Stuart et al., 2003). The

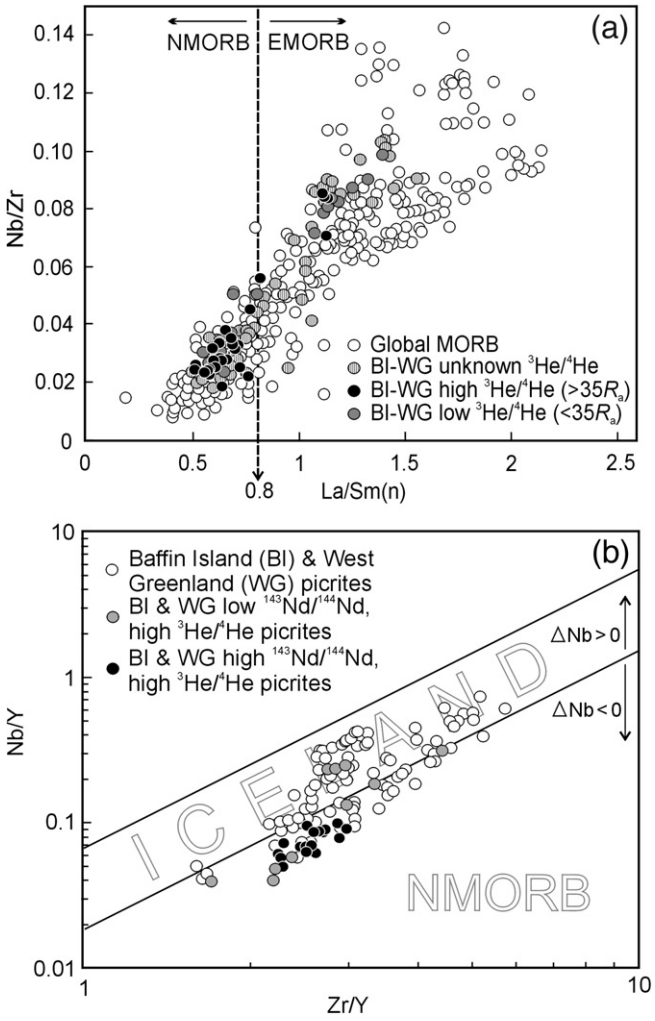

Fig. 3. a. Nb/Zr against La/Sm(n) (La/Sm normalised to primitive mantle values) for global MORB (white circles; data from www.petdb.org), Baffin Island and West Greenland picrites with high ^{3}He/^{4}He (>35 R_a; black circles), low ^{3}He/^{4}He (<35 R_a; grey circles) and unknown ^{3}He/^{4}He (patterned circles) from this and previous studies (Stuart et al., 2003; Graham et al., 1998). Dashed line represents the division between N- and E-MORB at La/Sm(n)=0.8 proposed by Mahoney et al. (2002). Baffin Island and West Greenland samples span the entire MORB range with high ^{3}He/^{4}He samples predominantly falling in the more depleted end but with some high ^{3}He/^{4}He in the less depleted MORB region. b) Nb/Y against Zr/Y for all Baffin Island and West Greenland samples from this and previous studies (Stuart et al., 2003; Graham et al., 1998). High ^{3}He/^{4}He Baffin Island–West Greenland samples are divided at ^{143}Nd/^{144}Nd of 0.51300 to show the relationship between Nd-isotope and incompatible trace element variation. Importantly, some high ^{3}He/^{4}He, relatively enriched (^{143}Nd/^{144}Nd<0.51300) samples plot within the 'Iceland' field (ΔNb>0; Fitton et al., 1997) indicating the availability of relatively enriched and depleted sources in the early Iceland plume and that high ^{3}He/^{4}He was a feature of both.

linear array previously recognised in ^{3}He/^{4}He–^{143}Nd/^{144}Nd space (PIP trend, solid lines Fig. 4b; Stuart et al., 2003) is no longer apparent.

The ΔNb notation places the Baffin Island and West Greenland lavas into both the N-MORB (ΔNb<0) and Icelandic (ΔNb>0) fields (Fig. 3b), corresponding with the N- and E-MORB type relationship long recognised for this region (Francis, 1985). All high ^{3}He/^{4}He samples reported previously had ΔNb<0. However, despite the strong association between high ^{3}He/^{4}He and ΔNb<0 still apparent in the new data, a number of the high ^{3}He/^{4}He samples within this new dataset have ΔNb>0. In addition, a plot of Nb/Zr against La/Sm(n), which was used earlier to highlight the similarity of the Baffin Island–West Greenland lavas to global MORB, further supports the observation that some of the high-^{3}He/^{4}He samples of the new dataset are more enriched than previously recognised. Fig. 5 shows REE profiles for the high ^{3}He/^{4}He samples separated on the basis of ^{143}Nd/^{144}Nd into relatively enriched (<0.51300) and relatively depleted (>0.51300). Although there is overlap between the groups, it is clear that the high-^{3}He/^{4}He, less depleted samples (^{143}Nd/^{143}Nd<0.51300) also have flatter REE profiles than the more depleted samples.

The early Iceland plume magmas were erupted through continental crust. It is therefore critical to assess the extent to which crustal contamination may have influenced the Baffin Island and West Greenland picrites before we can attribute trace element and isotopic variations to heterogeneity within the mantle source. High ^{3}He/^{4}He cannot be explained by crustal contamination because continental crust has extremely low ^{3}He/^{4}He due to radiogenic production of ^{4}He from decay of U and Th. However, it is possible that the latent heat released during olivine crystallisation induces crustal contamination and produces picrites whose early-formed olivines crystallised from an uncontaminated magma but whose groundmass is significantly contaminated. If this were the case then we would expect whole rock ^{143}Nd/^{144}Nd to be lowered, ^{87}Sr/^{86}Sr to be raised and incompatible

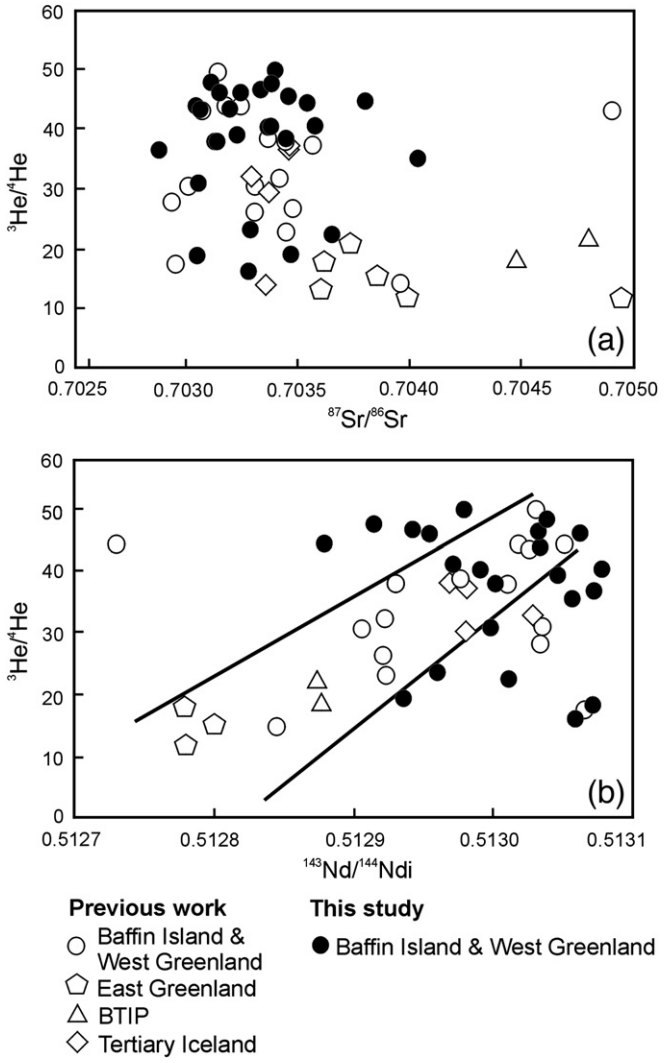

Fig. 4. ^{3}He/^{4}He plotted against a) ^{87}Sr/^{86}Sr and b) ^{143}Nd/^{144}Nd for Baffin Island and West Greenland samples (this and previous studies, Stuart et al., 2003; Graham et al., 1998), East Greenland (Peate et al., 2003; Marty et al., 1998), Tertiary Iceland (Hilton et al., 1999; Ellam and Stuart, 2004) and British Tertiary Igneous Province (Stuart et al., 2000). Proto-Iceland Plume (PIP) trend of Ellam and Stuart (2004) is delineated in Fig. 4b by black lines. The high ^{3}He/^{4}He Baffin Island and West Greenland samples of this study tend to be associated with depleted mantle but, significantly, many samples do not conform to the PIP trend.

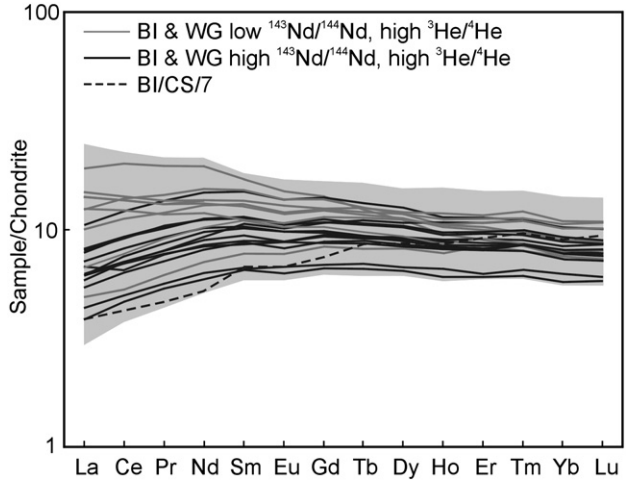

Fig. 5. Chondrite-normalised REE diagram for all Baffin Island and West Greenland samples (grey shading from this and previous study, Stuart et al., 2003). Individual high $^{3}He/^{4}He$ Baffin Island and West Greenland samples (>35 R_a) are shown separated into two groups by $^{143}Nd/^{144}Nd$ (grey <0.51300, black >0.51300). Despite the general overlap it is clear that high $^{3}He/^{4}He$ picrites with $^{143}Nd/^{144}Nd$ <0.51300 have flatter REE patterns than samples with $^{143}Nd/^{144}Nd$ >0.51300. The crustally contaminated sample BI/CS/7, shown by a dashed line, has a REE pattern that cuts across all the others.

element abundances to increase whilst olivine $^{3}He/^{4}He$ remains unaffected (Stuart et al., 2000).

The Baffin Island–West Greenland samples define a negative trend in a plot of Nb/Zr *versus* $^{143}Nd/^{144}Nd$. This trend follows that defined by Icelandic basalts (Fitton et al., 2003; grey field in Fig. 6) suggesting that the Baffin Island–West Greenland trend reflects variations in the mantle source. However, the Baffin Island–West Greenland data appear to scatter to slightly lower $^{143}Nd/^{144}Nd$, outside of the Iceland range, and this might be due to the addition of small amounts of crust. We need to establish whether or not the high $^{3}He/^{4}He$, relatively enriched magmas (high Nb/Zr, low $^{143}Nd/^{144}Nd$) could have been produced by crustal contamination of high $^{3}He/^{4}He$, more depleted melts. Fig. 6 shows curves for simple binary mixing between a typical depleted Baffin Island–West Greenland magma composition and various high-grade Proterozoic crustal basement rocks for this region (Theriault et al., 2001). The range in Nb/Zr of crustal rocks is similar to that of the Baffin Island–West Greenland samples but Nb/Zr is higher in the enriched samples than in any of the crustal rocks. The plot in Fig. 6 is therefore useful in distinguishing between the effects of source variation, crustal contamination, and also partial melting. Crustal contamination trajectories are nearly horizontal and therefore too shallow to account for the steep trend defined by the picrites. It is possible that some of the scatter in $^{143}Nd/^{144}Nd$ at a given Nb/Zr might be due to crustal contamination, but in most cases the amount of crust involved would have to be less than 2%. Furthermore, the $^{143}Nd/^{144}Nd$ of sub-sets of samples with narrow ranges of Nb/Zr does not co-vary with any other index of contamination (e.g. Sr/Nd, K/Nb, Rb/Ba. Rb/Sr, U/Nb or Ba/Th) suggesting that the range in $^{143}Nd/^{144}Nd$ at any given Nb/Zr reflects the natural range in the mantle source (as in Iceland). This plot supports the earlier suggestion (Stuart et al., 2003) that sample BI/CS/7, a high $^{3}He/^{4}He$ dyke from Cape Searle, is crustally contaminated. Our modelling suggests a mix of ~3% of locally available crustal rocks with a depleted parent magma could account for the very low $^{143}Nd/^{144}Nd$ of this sample. We are not able to reconstruct the original source $^{143}Nd/^{144}Nd$ value of BI/CS/7 because the degree of contamination is unknown, so it is not considered in further discussions. In addition, it seems that one of the other dykes (BI/CS/8) and a West Greenland sample (400444) experienced similar levels of contamination.

The main features displayed by the picrites in Fig. 6 are supported by a plot of K/Nb versus $^{143}Nd/^{144}Nd$ (Fig. 7). The main trend, of decreasing K/Nb with decreasing $^{143}Nd/^{144}Nd$, defined by the Baffin Island–West Greenland picrites in Fig. 7 is most likely due to source variation. Crustal rocks have high K/Nb and correspondingly low $^{143}Nd/^{144}Nd$ so crustal contamination can account for the displacement of BI/CS/7, BI/CS/8 and 400444 from the main trend, in agreement with Fig. 6. We also show a vector for the effects of alteration which is expected to increase K/Nb whilst leaving $^{143}Nd/^{144}Nd$ unaffected. Alteration may explain the displacement of five samples to high K/Nb in Fig. 7. The overall trend displayed by the samples in Fig. 7 supports the findings above that the samples with high $^{3}He/^{4}He$ and low $^{143}Nd/^{144}Nd$ (e.g. PAD6, BI/CS/6, 400230 and 332901) have not been affected by crustal contamination, and certainly not to a large enough degree to alter the Nd isotope ratios by a discernable amount. Crustal contamination would act by shifting originally more depleted (high $^{143}Nd/^{144}Nd$) samples away from the main trend on a crustal contamination vector similar to the one shown in Fig. 7. The fact that these important high $^{3}He/^{4}He$, low $^{143}Nd/^{144}Nd$ samples also display low K/Nb indicates that their source $^{143}Nd/^{144}Nd$ has not been affected by crustal contamination.

Crustal contamination cannot account for both the low $^{143}Nd/^{144}Nd$ and the relative enrichment in incompatible elements (e.g. Nb) that

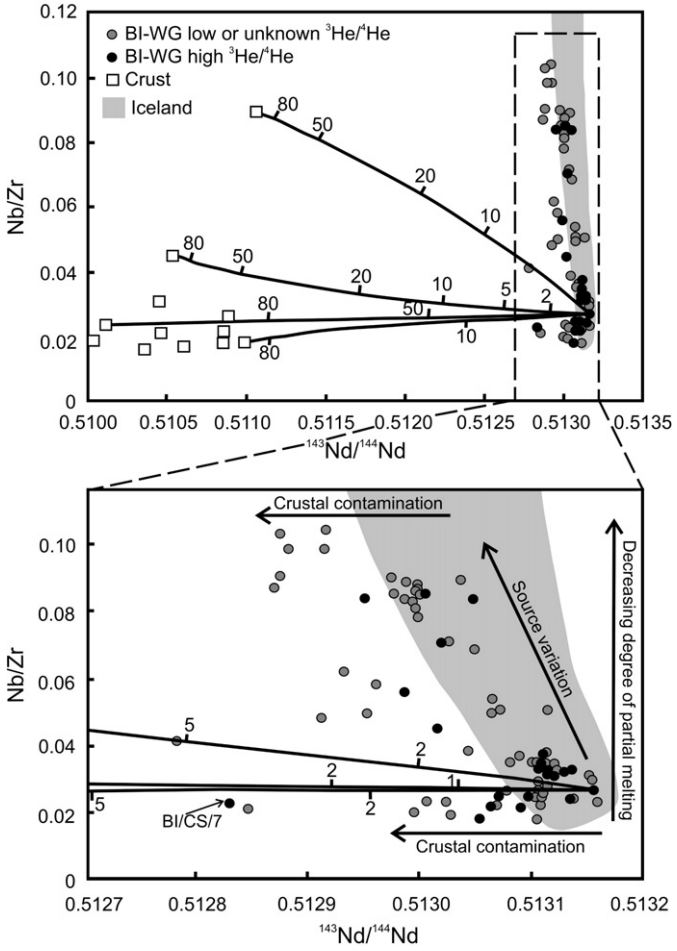

Fig. 6. Nb/Zr versus $^{143}Nd/^{144}Nd$ for all Baffin Island and West Greenland samples with high $^{3}He/^{4}He$ (>35 R_a; black circles) and low or unknown $^{3}He/^{4}He$ (grey circles) from this and previous studies (Stuart et al., 2003; Graham et al., 1998). Grey field represents Icelandic basalts (from Fitton et al., 2003). Simple binary mixing lines between a depleted parent and various crustal end members (white squares; Baffin Island Proterozoic and Archaean basement rocks, Theriault et al., 2001) are shown by solid lines with tick marks indicating percentage of crust in the mixture. Baffin Island–West Greenland samples define a negative trend similar to that for Iceland. The nearly horizontal crustal contamination mixing lines cannot produce the high $^{3}He/^{4}He$, relatively enriched (high Nb/Zr, low $^{143}Nd/^{144}Nd$) samples from more depleted magma compositions. The trend in Baffin Island–West Greenland is more likely to be due to source variations although small variations in $^{143}Nd/^{144}Nd$ within each group may be due to small amounts (<2%) of crustal contamination.

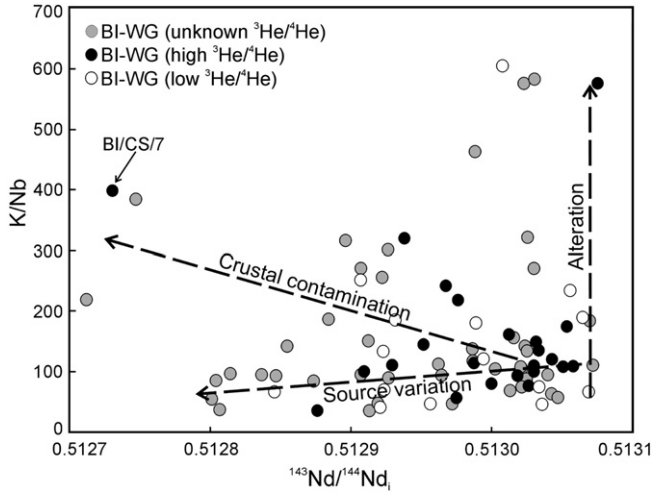

Fig. 7. K/Nb versus ^{143}Nd/^{144}Nd$_i$ ($_i$ denotes ^{143}Nd/^{144}Nd corrected back to 60 Ma). Samples define a positive trend of increasing K/Nb with increasing ^{143}Nd/^{144}Nd which is thought to reflect that of the source. Scatter of some samples to higher K/Nb is either due to alteration or small amounts of crustal contamination (when accompanied by low ^{143}Nd/^{144}Nd). High ^{3}He/^{4}He, low ^{143}Nd/^{144}Nd picrites fall within the main Baffin Island–West Greenland trend and are apparently unaffected by crustal contamination.

characterise the less depleted picrites, suggesting that the range of composition of the high ^{3}He/^{4}He picrites reflects that of their source. We therefore conclude that high ^{3}He/^{4}He is a feature of a range of mantle compositions regardless of the possible effects of small amounts of crustal contamination.

5.2. Relationship between early Iceland plume picrites and OIB sources

The highest ^{3}He/^{4}He recorded by contemporary ocean island volcanism (Iceland, Loihi, Galapagos and Samoa) appear to be rather similar, converging on ^{3}He/^{4}He slightly higher than 30 R_a (Kurz et al., 1982; Kurz and Geist, 1999; Macpherson et al., 2005; Jackson et al., 2007) (Fig. 8). This is considerably lower than the highest ^{3}He/^{4}He (~50R_a) recorded for Baffin Island–West Greenland. Clear correlations between ^{3}He/^{4}He and other isotope systems are observed for moderately high ^{3}He/^{4}He OIB (e.g. Hawaii, Kurz and Kammer, 1991, Iceland, Brandon et al., 2007, and Samoa, Jackson et al., 2007). However, extrapolation of these trends to the high ^{3}He/^{4}He values in the early Iceland plume lavas does not produce reasonable mantle compositions (e.g. Brandon et al., 2007).

Although the ^{143}Nd/^{144}Nd and ^{87}Sr/^{86}Sr range of the Baffin Island–West Greenland picrites overlaps the range recorded by the high ^{3}He/^{4}He northern hemisphere ocean islands (Iceland, Hawaii, Galapagos), they are distinct from the isotopic composition of basalts from Samoa, Heard Island and Kerguelen (Fig. 8). It has been proposed that the compositional difference between high ^{3}He/^{4}He basalts from southern and northern hemisphere islands reflects distinct high ^{3}He/^{4}He reservoirs (Jackson et al., 2007). It appears more likely that this difference is simply due to the addition of helium with high ^{3}He/^{4}He to isotopically distinct 'DUPAL-type' mantle, prevalent in the southern hemisphere.

5.3. Implications for the high ^{3}He/^{4}He mantle reservoir

Demonstrating that the relatively enriched, high ^{3}He/^{4}He picrites cannot be produced by crustal contamination of depleted parental rocks removes the requirement for the high ^{3}He/^{4}He mantle to have a unique depleted composition (Stuart et al., 2003; Class and Goldstein, 2005). Our new data seem inconsistent with recent models (e.g. Class and Goldstein, 2005; Boyet and Carlson, 2006; Parman, 2007) that invoke ancient depleted, and therefore He-poor, mantle as a source of high ^{3}He/^{4}He basaltic rocks. In particular, the observed range of ^{143}Nd/^{144}Nd in the high ^{3}He/^{4}He picrites reduces the likelihood that ancient depleted mantle is the dominant repository of primordial noble gases in Earth. Recent debate has largely focused on whether He is more or less compatible than U and Th during mantle melting (Parman et al., 2005; Heber et al., 2007). In the light of our new data, these discussions are largely irrelevant. While there is little doubt that He is a highly incompatible element, it is more important to consider how He partitions relative to Nd (and other moderately incompatible elements) during mantle melting. Using recent estimates for the partition coefficients for He (1.7×10^{-4}; Heber et al., 2007) and Nd (0.1; McKenzie and O'Nions, 1991) during melting of the most common mantle phases it appears that D_{Nd}/D_{He} is ~600. Consequently, mantle melting will reduce the He concentration in the residue relative to Nd to such an extent that ancient depleted mantle will have extremely low He/Nd. It is therefore difficult to preserve high ^{3}He/^{4}He during subsequent mixing with other mantle components.

The prevailing models for the source of high ^{3}He/^{4}He (and other primordial noble gases) in Earth require a discrete reservoir sampled by upwelling mantle plumes. The composition of this distinct reservoir has been determined based on the isotope and trace element composition of moderately high ^{3}He/^{4}He basalts (10–30 R_a) from ocean islands such as

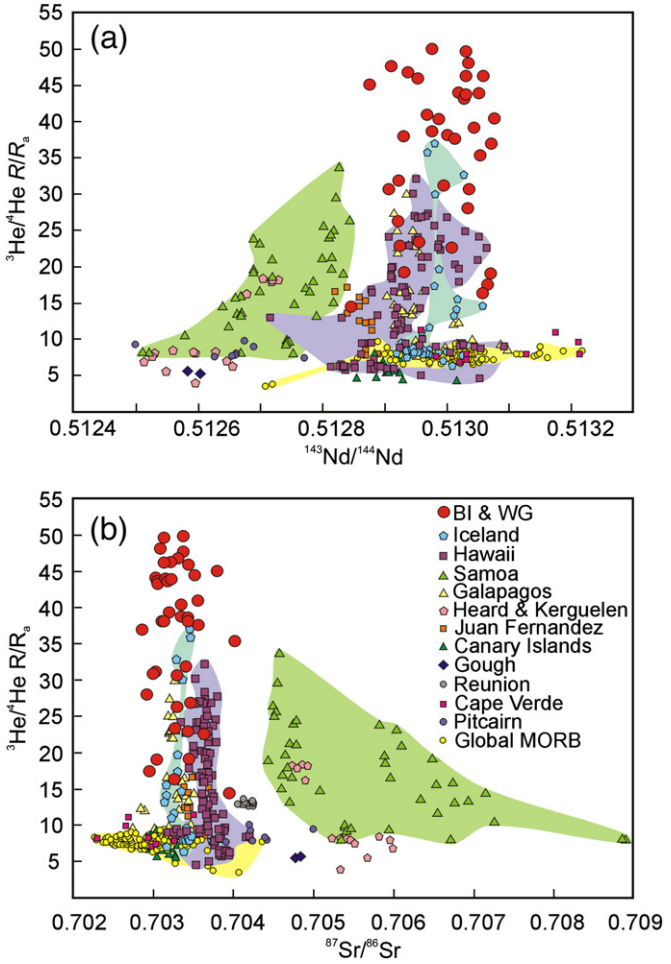

Fig. 8. ^{3}He/^{4}He versus (a) ^{143}Nd/^{144}Nd and (b) ^{87}Sr/^{86}Sr for global OIB (Abedini et al., 2006), MORB (www.petdb.org), Baffin Island (this study; Stuart et al., 2003) and West Greenland (this study; Graham et al., 1998). ^{3}He/^{4}He of OIB reach a maximum of ~30–35 R_a with each island displaying a fairly narrow range in ^{143}Nd/^{144}Nd and ^{87}Sr/^{86}Sr at high ^{3}He/^{4}He. Fields are shown for Samoa (pale green), Hawaii (lilac), Iceland (pale blue) and MORB (pale yellow) to highlight the data amongst the other islands. ^{3}He/^{4}He of Baffin Island and West Greenland (red circles) reach a maximum of ~50 R_a but display the same range in ^{143}Nd/^{144}Nd and ^{87}Sr/^{86}Sr at both high and low ^{3}He/^{4}He. Baffin Island and West Greenland ^{143}Nd/^{144}Nd spans most of the MORB range (yellow circles). Baffin Island and West Greenland represent the earliest starting Iceland plume material.

Hawaii, Galapagos, Iceland and Samoa (Farley et al., 1992; Hart et al., 1992; Hilton et al., 1999; Jackson et al., 2007). Although there is no consensus regarding the precise composition of this reservoir, it is clear that, if it exists, it must contain a significant proportion of depleted peridotite and recycled eclogitic mantle. The Baffin Island–West Greenland picrites should provide a clearer picture of the reservoir's composition since they sample the extreme high $^{3}He/^{4}He$ end-member that is possibly only available in a starting plume. However, these high $^{3}He/^{4}He$ picrites do not converge on a unique radiogenic isotope and trace element composition (Figs. 4 and 8).

The compositional range of the high $^{3}He/^{4}He$ picrites is similar to that of the convecting upper mantle sampled at mid-ocean ridges (i.e. a mixture of both N-MORB (depleted) and E-MORB (relatively enriched) fractions; Fig. 3). MORB have a distinct $^{3}He/^{4}He$ (8 ± 1 R_a) showing no indication of a high $^{3}He/^{4}He$ component. The simplest explanation for the high $^{3}He/^{4}He$ Baffin Island–West Greenland lavas is that their source is convecting upper mantle to which helium with high $^{3}He/^{4}He$ has been added.

It has previously been argued that the depleted high $^{3}He/^{4}He$ picrites from Baffin Island were derived from mantle that was generated by mixing between a primordial He-rich deep reservoir and depleted mantle as sampled by mid-ocean ridges (Ellam and Stuart, 2004). The new He–Nd and He–Sr isotope data are broadly consistent with this model, despite not requiring such a well defined composition at high $^{3}He/^{4}He$ as that proposed by Ellam and Stuart (2004). The main difference is that the He-poor mantle end member must have the full range of compositions available in the upper mantle and sampled by mid-ocean ridges. The observed He–Nd and He–Sr isotope variation (Fig. 4) requires that the He concentration in the high $^{3}He/^{4}He$ end member is high enough to dominate and overwhelm subsequent mixtures with the He-poor mantle end member, irrespective of its degree of depletion or enrichment. The fact that the high $^{3}He/^{4}He$ early Iceland plume picrites record variable mantle compositions places important constraints on the timing and nature of this mixing. Mixing must have occurred after the development of heterogeneity in the He-poor end member. Further, the data require that the He-poor end-members have remained largely unmixed, which would place significant constraints on the scale of mantle heterogeneities.

An alternative explanation is that high $^{3}He/^{4}He$ helium in the early Iceland plume is somehow decoupled from all other mantle components. Diffusion is the easiest way for this to be achieved. The measured diffusion rate of He in olivine suggests that, at mantle temperatures, He from a primordial volatile-rich reservoir could equilibrate with km-scale degassed mantle domains over the lifetime of the Earth (Shuster et al., 2003). O'Nions & Oxburgh (1983) argued that the abnormally high $^{3}He/^{4}He$ of the upper mantle (~8 R_a) results from diffusion of He across a mid-mantle thermal boundary above a hot, convectively-isolated primordial noble gas-rich lower mantle. The importance of diffusive processes has been renewed by a study showing that diffusion of He from unprocessed mantle heterogeneities early in Earth history has the potential to enrich previously depleted mantle layers in primordial He (Albarède, 2008). If the Baffin Island–West Greenland picrite source is similar to that of the lavas erupted more recently in Iceland then it may also show a signature of recycled ocean crust (Thirlwall et al., 2004). Baffin Island–West Greenland picrites with low $^{143}Nd/^{144}Nd$ have a He isotopic range that is indistinguishable from that of the more depleted (high $^{143}Nd/^{144}Nd$) picrites and therefore require that the diffusive equilibration of high $^{3}He/^{4}He$ helium occurred after the heterogeneous mantle structure was established.

Either explanation (mixing or diffusion) require that the mantle sampled by the early Iceland plume gained its high $^{3}He/^{4}He$ signature by contact with a discrete He-rich, high $^{3}He/^{4}He$ reservoir. It is a widely held view that mantle plumes originate at thermal boundary layers within the mantle (Loper and Stacey, 1983). The presence of the highest terrestrial $^{3}He/^{4}He$ in the earliest volcanic products of a major mantle plume suggests that the high $^{3}He/^{4}He$ reservoir is a deep layer rather than dispersed blobs of depleted mantle into which He has diffused. Furthermore, maintaining sufficiently high He concentrations in small-scale mantle heterogeneities over time is difficult to achieve (van Keken and Ballentine, 1999). The ultimate location of the high $^{3}He/^{4}He$ reservoir is unclear, not least because seismic tomography currently does not unequivocally identify the depth of the origin of the Iceland plume (Wolfe et al., 1997; Foulger et al., 2001). A mid-mantle thermal boundary layer, where lower mantle-derived ^{3}He is added to the convecting upper mantle (e.g. Fitton et al., 1997), satisfies the Nd–Sr isotope and trace-element distribution recorded by the Baffin Island–West Greenland picrites. However, this requires a convectively-isolated lower mantle that is difficult to reconcile with geophysical evidence for slab penetration (Bijwaard et al., 1998; Fukao et al., 2001) and numerical models that support whole mantle circulation (Forte and Mitrovica, 2001). The derivation of the Baffin Island–West Greenland picrites from mantle that is 100–200 °C hotter than normal convecting upper mantle (Larsen and Pedersen, 2000; Herzberg et al., 2007) is strong evidence for a deep mantle source especially since high excess temperatures at the core mantle boundary (CMB) can be reduced to 200–250 °C at the surface (Bunge, 2005). The CMB (Samuel and Farnetani, 2003) or the core (Porcelli and Halliday, 2001) are therefore the likeliest candidates for a hidden high $^{3}He/^{4}He$ reservoir.

A convectively isolated layer at the CMB, enriched in primordial volatiles, is a realistic possibility. Crucially this reservoir needs to have maintained a high time-integrated $^{3}He/(U+Th)$. The primordial volatiles may have been derived from subducted meteoritic material early in Earth history prior to convective isolation (Tolstikhin and Hofmann, 2005). However, the likely loss of He during the subduction process makes this difficult to achieve (Hiyagon, 1994). Labrosse et al. (2007) have proposed that the solid phases from a crystallizing early magma ocean might sink to produce a volatile-rich reservoir. Alternatively, if recent estimates of He partition coefficients (Parman et al., 2005) are reliable, this reservoir may have been depleted during a silicate differentiation event in the first few 10s of million years of Earth history (Boyet and Carlson, 2006).

For the outer core to be the source of primordial He requires an exceptionally high initial terrestrial He concentration. Published experimental metal-silicate partitioning data are sparse, but indicate that the core is unlikely to contain a significant proportion of primordial noble gases (e.g. Sudo et al., 1994). The geochemical evidence for a core contribution to plume-derived melts is controversial. For instance, it has been argued that Os isotopes in Hawaiian basalts provide evidence of core addition (Brandon et al., 1999) but they are not accompanied by W isotope anomalies (Scherstén et al., 2004). Os isotopes in high $^{3}He/^{4}He$ basalts from Iceland show no indication of a core signature (Brandon et al., 2007) and the near-chondritic Os isotope signature of Baffin Island–West Greenland basalts tends to rule out a significant contribution of outer core material with super-chondritic Re/Os to the high $^{3}He/^{4}He$ mantle source (Dale et al., in press). This, however, does not rule out diffusive transport of core-derived primordial He across the CMB. Irrespective of the ultimate location of a deep high $^{3}He/^{4}He$ reservoir, the similarity of the isotopic and trace element composition of the Baffin Island–West Greenland picrites with global MORB implies that the whole mantle may have essentially the same composition and degree of heterogeneity as the convecting upper mantle.

6. Conclusions

Extremely high $^{3}He/^{4}He$ ratios are a widespread feature of the very earliest volcanic products of the early Iceland plume. The high $^{3}He/^{4}He$ of picrite lava flows from Baffin Island and West Greenland are associated with a wide range of mantle compositions that span the range exhibited by the global MORB database. These new data strongly suggest that there is no role for a discrete depleted reservoir for high $^{3}He/^{4}He$ in the Earth, and that it is not possible to identify a

compositionally unique high $^3He/^4He$ reservoir. The likeliest explanation is that high $^3He/^4He$ helium has diffused into heterogeneous mantle that is compositionally indistinguishable from the convecting upper mantle. The resultant mixture is only available to starting plumes, but must have a high enough helium concentration to dominate and overwhelm any subsequent mixture so that high $^3He/^4He$ can be imposed upon a significant range of mantle compositions with varied $^3He/(U+Th)$ histories.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.epsl.2008.10.007.

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