High-^{3}He/^{4}He, depleted mantle and low-δ^{18}O recycled oceanic lithosphere in the source of central Iceland magmatism

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Abstract

New helium and oxygen isotope data and trace element concentrations are reported for volcanic rocks from central Iceland. Basalts that are depleted in the most incompatible trace elements possess a wide range in ^{3}He/^{4}He but most ratios are similar to or higher than those of mid-ocean ridge basalt (MORB: ~8R_A [1]). The low concentrations of helium in these rocks suggest that significant degassing has made them susceptible to contamination by low-^{3}He/^{4}He crust, therefore all measured ^{3}He/^{4}He are considered minimum estimates for their sources. Elevated helium isotope ratios in the source of these rocks result from interaction with high-^{3}He/^{4}He mantle. The highest oxygen isotope ratios in the depleted rocks are similar to those in melts from typical depleted upper mantle and the range of δ^{18}O values is consistent with variable, limited amounts of contamination by Icelandic crust.

Most of the incompatible trace element-enriched rocks possess ^{3}He/^{4}He ratios that are similar to or lower than those in MORB. These rocks were erupted close to the postulated centre of the Iceland plume. This observation contradicts models in which high-^{3}He/^{4}He characterises the focus of mantle upwelling, and a source with MORB-like ^{3}He/^{4}He ratios may be common to other parts of the North Atlantic Igneous Province. The highest δ^{18}O values in the enriched rocks are lower than those in MORB and do not appear to have been affected by interaction with low-δ^{18}O Icelandic crust. Recycling of hydrothermally altered oceanic crust that has been subducted into the mantle provides a plausible mechanism for generating an ^{18}O-poor source with the trace element and isotopic characteristics of the enriched lavas.

Keywords: Iceland, helium, oxygen, isotopes, mantle, recycling

1. Introduction

Iceland is a subaerial section of the Mid-Atlantic Ridge, distinguished by anomalously thick crust [2], where volcanism occurs primarily along neovolcanic zones. Lavas from Iceland and the adjacent ridge sections display significant Sr, Nd and Pb isotopic variation which can be explained through contributions from several components that are both incompatible element enriched and depleted compared to normal mid-ocean ridge basalts (MORBs) (e.g. [3-5]). The age of the Icelandic crust limits the degree to which Sr, Nd and Pb isotopic variation can be generated through *in situ* radioactive in-growth: therefore, irrespective of the extent of crustal assimilation, an inescapable conclusion from geochemical studies is that distinct mantle domains exist beneath Iceland.

Geophysical studies also demonstrate the distinctive nature of the Icelandic mantle. For example, seismic wave velocities are slower beneath Iceland than surrounding regions, consistent with the presence of anomalously hot mantle [6-8]. To date, there is no consensus as to the depth at which the heat anomaly originates or whether it is associated with active (plume) or passive mantle upwelling. While some studies have attempted to reconcile geophysical observations of Icelandic mantle and the geochemical diversity revealed in the island's volcanic products [9,10], no consensus has been reached on their relationship.

In this contribution, we concentrate on two isotopic systems (He and O) with proven potential for tracing heterogeneity in mantle-derived lavas [11,12]. We focus on central Iceland – at the convergence of the neovolcanic zones – which is the proposed centre of mantle upwelling in the region [6]. New measurements of helium concentrations and helium and oxygen isotope ratios of hyaloclastic glasses and phenocrysts in subaerial lavas are presented. These demonstrate the variable extent of crust-mantle interaction in central Iceland and the isotopic heterogeneity of He and O in the mantle source region. By identifying samples unaffected by crustal interaction, we show that He and O isotopes reveal information about both the nature of mantle sources contributing to volcanism in central Iceland and the complexity of their spatial distribution.

2. Samples

The neovolcanic zones of Iceland can be subdivided into the western and northern branches, which erupt tholeiitic lavas, and an eastern branch which produces mainly Fe-Ti basalts and alkali basalts. This study focuses on subglacial glasses and subaerial olivine tholeiite lava flows from central Iceland, at the convergence of the northern (NRZ) and western rift zones (WRZ) (Table 1 and Fig. 1), where fissure-fed and central edifice volcanoes occur. Optically unaltered glass was collected from subglacial pillow lavas, some of which contain olivine phenocrysts. The lava flows generally contain abundant olivine phenocrysts set in a fine-grained plagioclase, pyroxene and glass matrix. NAL 625 contains augite phenocrysts, as does NAL 626 which also contains abundant (~30%) plagioclase phenocrysts that are glomerophyric and/or optically zoned. The central Iceland samples are supplemented by glasses and lavas from the NRZ (Th-29) and Eastern Rift Zone (ERZ; SAL-575 and THJOR).

3. Analytical Techniques

Crushing *in vacuo* was used to release helium from inclusions in olivine and clinopyroxene phenocrysts and from vesicles in glasses. Fresh glass and phenocrysts were separated manually from coarsely crushed rock. Separates, free of adhering matrix, were cleaned ultrasonically in ethanol then dried prior to loading into two on-line crushing devices [13]. Short crush times were employed to help avoid the release of extraneous He contained in crystal lattice sites [14]. Gas liberated by crushing was expanded into a stainless steel preparation line. Helium was isolated from other gases by sequential exposure to (i) a Ti getter at 750ºC, decreasing to 400ºC, (ii) a liquid nitrogen-cooled charcoal trap, (iii) a SAES© Zr-Al getter operated at room temperature, and (iv) a liquid He-cooled charcoal trap. Helium was then expanded into a split-tube, dual collection He-isotope mass spectrometer (GAD) for ^{3}He/^{4}He analysis using established protocols [15]. Raw helium isotope ratios were normalised using standard aliquots of SIO air (^{3}He/^{4}He ratio of $1.4 \times 10^{-6} = 1R_A$) and gas from Murdering Mudpots, Yellowstone National Park (= $16.45R_A$). Ne was monitored on every run to correct ^{3}He/^{4}He ratios for the effects of air contamination (<2% for all samples).

Blanks were measured by operating the crusher (~70 beats per minute) for the same duration (150s) as sample analysis, and averaged 0.67 x 10^{-9} cm^3 STP^{4}He (±0.55, 2σ). All unknowns have been corrected for the presence of such a blank.

Oxygen isotope analyses were performed on fresh glass and inclusion-free mafic minerals. These were separated by hand and ultrasonically cleaned in MQ-H$_2$O and methanol. δ^{18}O‡ values were measured at Royal Holloway, University of London on 1-2mg splits using established laser fluorination protocols for glasses [11] and phenocrysts [16]. Yields for all unknowns and standards in this study were 95±5%. The precision and accuracy of analyses was monitored by three internal standards; two different olivines from San Carlos and a garnet. The isotopic composition of these minerals relative to the international standard biotite NBS-30 (δ^{18}O = 5.10‰), and 2σ deviations of replicate analyses over the analytical period were: RHUL SCOL I = +4.84‰ (±0.18, n=23), RHUL SCOL II = +5.22‰ (±0.16, n=19) and UW GMG 2 garnet = +5.71‰ (±0.18, n=22).

Major element analyses were performed at the Nordic Volcanological Institute. Concentrations in glass were measured using an ARL-SEMQ microprobe and whole rocks powders were fused to glass and measured by ICP-AES, with analytical precision of better than ±3% [17]. Trace elements were determined on small (<3mm diameter) disaggregated shards of glass or powdered lava using a Perkin Elmer ELAN 6000 inductively coupled plasma mass spectrometer (ICP-MS) at the University of Durham [18]. The data were calibrated relative to international standards, with reproducibility better than ±2% for all elements except Pb which was better than ±5%. Data for NAL 611 is published in Breddam [17].

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‡ δ^{18}O$_n$ is the per mille (‰) deviation of ^{18}O/^{16}O in n from the international standard (*std*) V-SMOW given by the relationship: δ^{18}O$_n$ = 1000 × (^{18}O/^{16}O$_n$ / ^{18}O/^{16}O$_{std}$ -1)

4. Results

4.1   Major and Trace element results

All samples are tholeiitic in composition. The glasses display a restricted range of compositions with MgO contents varying from 12.5 to 5.0 wt. % (Table 2). Trace elements are enriched relative to primitive mantle by factors up to 40 for the high field strength elements (HFSEs) Nb and Ta, 25 for light rare earth elements (LREEs) and 8 for heavy rare earth elements (HREEs, Table 3). Trace element concentrations and Sr and Nd isotopic ratios can be used to define three groups of glasses (Figs. 2 & 3). Group 1 range from pronounced LREE depletion to mild LREE enrichment ($(La/Yb)_n$ = 0.33-1.77, $(La/Sm)_n$ = 0.44-1.12, $(Sm/Yb)_n$ = 0.75-1.51) and possess lower ^{87}Sr/^{86}Sr and higher ^{143}Nd/^{144}Nd than the other groups. Group 2 glasses have similar HREE contents to Group 1 but greater LREE enrichment ($(La/Yb)_n$ = 2.82-3.36, $(La/Sm)_n$ = 1.44-1.53, $(Sm/Yb)_n$ = 1.96-2.20). Group 3 glasses have the highest incompatible element concentrations but have patterns that are very similar to Group 2 ($(La/Yb)_n$ = 3.58-3.89, $(La/Sm)_n$ = 1.60-1.64, $(Sm/Yb)_n$ = 2.19-2.40). Glasses of Groups 2 and 3 also display very similar Sr and Nd isotopic ratios that are distinct from Group 1 (Fig. 3). All glasses display relative enrichment of Ta and Nb (all have positive ΔNb [19]) and depletion in Pb, with respect to neighbouring elements. Group 1 generally displays positive Ba and Sr anomalies. Positive Ba anomalies also appear in Group 2, while in Group 3 the Ba anomaly is absent and Sr is depleted relative to neighbouring elements (Fig. 2a).

The lava samples all have >7 wt. % MgO. Their normalised trace element patterns are comparable with previously published data (e.g. [9,20]) and can be split into two groups: LREE-enriched (NAL-625 and THJOR) with $(La/Yb)_n$ = 2.36-4.02, $(La/Sm)_n$ = 1.20-1.64 and $(Sm/Yb)_n$ = 1.97-2.34, and LREE-depleted (Th-29, NAL-688) with $(La/Yb)_n$ = 0.27-0.37, $(La/Sm)_n$ = 0.35-0.43 and $(Sm/Yb)_n$ = 0.75-0.77. Depleted lava from Theistareykir (Th-29) resembles the most depleted Group 1 glass, but enriched THJOR also falls mainly within the range of Group 1 (Fig. 2b). While the most enriched lava (NAL-625) has higher La/Yb and

Sm/Yb than the glasses, it more closely resembles Group 1 than Group 2 in having progressive relative depletion of elements more incompatible than Nb.

4.2   He isotope results

Helium isotope ratios (^{3}He/^{4}He) and concentrations ([He]) are reported in Table 1 with sample locations shown in Figure 1. Glass and phenocryst samples from central Iceland are characterised by a wide range in ^{3}He/^{4}He ratios encompassing high-^{3}He hotspot ratios (>> 8R_A), through MORB-like values (8R_A ± 1 [15]) to values considerably lower than MORB indicating a significant contribution of radiogenic He.

The highest ^{3}He/^{4}He of 34.3R_A, for NAL-625 from Vadalda just north of the Vatnajökull Icecap (Fig. 1), is significantly greater than any other He isotope ratio reported for Iceland's neovolcanic zones (≤ 26.2R_A [21,22]). The ^{3}He/^{4}He of Vadalda is comparable with the highest value of older volcanic successions at Selardalur, northwest Iceland (37.7R_A [23]), and is higher than those of olivines from Tertiary lavas throughout the North Atlantic Igneous Province [24-26] except for Baffin Island picrites [27]. The ^{3}He/^{4}He ratio of this sample is also similar to rocks from Loihi Seamount, Hawaii, and the Galapagos islands, the only other modern day ocean islands to register helium isotope ratios greater than 25R_A [28-31]. Vadalda clinopyroxene phenocrysts possess lower ^{3}He/^{4}He than co-existing olivine (Table 1). Such an effect has been observed previously and attributed to preferential capture of radiogenic He in the pyroxene lattice [32]. However, the clinopyroxene ^{3}He/^{4}He is still higher than most of the samples in the present sample suite as well as samples analysed previously from central Iceland [9], corroborating the high-^{3}He/^{4}He source inferred for this particular locality. Lava NAL-625 has the highest La/Yb measured in this work but, as noted above, bears some similarities to Group 1 glasses.

Helium isotope ratios significantly greater than MORB occur in glasses and phenocrysts at five further localities in central Iceland (Fig. 1): Fjallsendi (17.3R_A), Kistufell (16.6R_A), Gaesahjukur (17.6R_A), Ketildyngja (10.6R_A) and Bardarbunga (21.9R_A). These locations lie towards the northern and western parts of the study area. The value obtained for the

Bardarbunga glass compares well with other reported values in the immediate area [21]. We also found elevated ^{3}He/^{4}He for the NRZ sample from Theistareykir (12.2R_A) and the SRZ lavas from Solheimajökull (14.6R_A) and Thjorsardalur (18.3R_A).

MORB-like ^{3}He/^{4}He ratios characterise six localities in central Iceland: Herdubreid, Eggert, Jardfraedingaslod, Upptyppingar, Trolladyngja and Odadahraun (Table 1; Fig. 4). These locations lie immediately north of Vatnajökull and mainly cluster round the most elevated ^{3}He/^{4}He sample from Vadalda (Fig. 1). As with high-^{3}He/^{4}He samples, MORB-like helium isotope ratios occur in both phenocrysts and glass. The glasses contain the greatest He concentrations (up to 127 x 10^{-9} cm^{3}STP/g) but some low He concentration olivines (e.g. TRO-53) also possess MORB-like ^{3}He/^{4}He (Fig. 4). Two glass samples (Herdubreid, Eggert) belong to Group 1, five to Group 2 (Jardfraedingaslod, Upptyppingar and Ketildyngja) and the remaining two are lavas for which trace element data are not available (Table 1).

Helium isotope ratios lower than those in MORB were found for four samples: glasses from Upptyppingar, Hvannalindir and Jardfraedingaslod, and olivine phenocrysts from the centre of the Eggert pillow lava. The three low-^{3}He/^{4}He glasses comprise all of Group 3 and have low He concentrations (Fig. 4). Upptyppingar and Hvannalindir both have [He] < 1 x 10^{-9} cm^{3}STP/g and the Jardfraedingaslod glass has the lowest [He] from this locality. Like Group 2, the Group 3 glasses occur only immediately north of Vatnajökull (Fig. 1). It is notable that olivines from the centre of the Eggert lava (NAL-688) have a lower ^{3}He/^{4}He value (5.9R_A) compared with olivines picked from the rim (8.0R_A) and from the glass phase (8.8R_A). This unusual observation may reflect capture of some xenocrystic olivine by this sample.

4.3   O isotope results

Central Icelandic glasses possess δ^{18}O values in the range +3.31 to +4.82‰ (Table 1). The δ^{18}O values of glasses are lower than basaltic glasses from oceanic and backarc spreading centres [11,33-35] but their upper limit is similar to four values previously obtained from Kistufell [17]. Oxygen isotope ratios in olivine phenocrysts (δ^{18}O$_{ol}$) vary between +3.51 and +5.08‰. The olivine values are lower than those typically observed for MORB [33], ocean

islands [36,37], arc volcanics [38] and mantle peridotites [39]. Results are similar to or lower than values reported for the Reykjanes Peninsula and Theistareykir [20,40,41].

5. Discussion

The new results identify marked He and O isotopic heterogeneity in the magmas erupted in the centre of Iceland. In the following sections, we examine the origin of these isotopic variations in terms of crustal and mantle contributions, and discuss their implications for the evolution of the Icelandic mantle.

5.1   Crustal control on ^{18}O/^{16}O in central Icelandic magmas

Crustal contamination has been shown to modify δ^{18}O values in several previous studies of Icelandic magmatism [20,40,41-43]. The way in which δ^{18}O values of basaltic melt change during interaction with hydrothermally altered, Icelandic crust can be modelled in two ways. First, assimilation of older lavas in the crust may occur during fractional crystallisation (AFC). Alternatively, basaltic magmatism could introduce sufficient heat to generate rhyolitic crustal melts that could then mix with the invading basalt. In reality, there is likely to be a spectrum of processes operating, but these endmember scenarios provide a context for discussing the origin of δ^{18}O variation in the central Icelandic magmas. Crustal contamination models are also sensitive to the δ^{18}O value chosen for the contaminant. This is a contentious issue as there is substantially oxygen isotopic heterogeneity recorded in samples of Icelandic crust [44,45]. Such heterogeneity may be homogenised during crustal melting, so δ^{18}O values of natural rhyolites provide a useful estimate of a generic crustal endmember that might be encountered by batches of basalt erupted in different parts of Iceland. Therefore, we follow [40] in selecting a δ^{18}O value of 0‰ for the crust, as this value is slightly more negative than the lowest values measured in rhyolites from Krafla and Askja [42,46]. This estimate is lower than other estimates of δ^{18}O in Icelandic crust (+2‰ [20,45]) which means that our modelling allows greater isotopic leverage for changing melt δ^{18}O values.

Contamination of a primitive, incompatible trace element-depleted magma (e.g. similar to Th-29) by rhyolite can reproduce the trace element and oxygen isotope characteristics of the central Iceland glasses with 8% to 13% contamination for Group 1, ~ 20% contamination for Group 2 and ~20% to ~35% contamination for Group 3 (curve A, Fig. 5). However, the elevation of SiO_2 and lowering of FeO that would result from addition of these volumes of rhyolitic magma [20,40] are not observed in the central Iceland lavas. The increase in FeO from Group 1, through Group 2 to Group 3 and the limited range in SiO_2 preclude basalt – rhyolite mixing as a mechanism for generating low δ^{18}O values in the central Iceland rocks.

An AFC model in which depleted basaltic magma assimilates "median" crustal andesite [40] can produce a close match to the Group 1 glasses for trace element enrichment and δ^{18}O (curve B, Fig 5). Limited amounts of AFC are consistent with the mafic character of all of the depleted rocks. A similar conclusion has been reached for depleted lavas from Theistareykir in the NRZ [40]. Although the importance of AFC at Theistareykir has been disputed [47], the oxygen isotope data for Group 1 support the conclusions that depleted lavas have interacted with the crust and that depleted Icelandic mantle possesses δ^{18}O which is within the range of the source of MORB [40]. However, more extensive differentiation of depleted magma by AFC cannot lower δ^{18}O and enrich trace elements to the extent observed in Group 2 and 3 glasses (curve B, Fig. 5). Trace element ratios may themselves be fractionated during melt – crust interaction. For example, the trace element concentrations and ratios (e.g. La/Sm) of Krafla rhyolites are significantly different from those of the Krafla basalts from which they are derived [48]. However, such fractionation will also affect the major element composition of the contaminant. The contaminant required to elevate La/Sm sufficiently from a primitive depleted lava through Group 1 to groups 2 and 3 by AFC would possess the trace element composition of a rhyolite (c.f. [20]). The simplest explanation for such a contaminant is that it is a rhyolitic melt but, as discussed above, the major element composition of Group 2 and 3 are inconsistent with addition of a rhyolitic component (curve A, Fig. 5).

A better fit to the Group 2 and 3 data can be achieved for a model in which the invading basalt, with δ^{18}O resembling Group 1, and the crustal contaminant are both enriched in

incompatible trace elements (curve C, Fig. 5). Between 50% to 75% differentiation by AFC can sufficiently lower δ^{18}O and enrich trace elements. While this model does fit the data, it requires that Group 2 melts, erupted at three distinct locations, all experienced very similar amounts of contamination. An alternative interpretation is that the Group 2 lavas retain their primary oxygen isotopic and trace element compositions. In this scenario, Group 3 develops from primary Group 2 melts through more restricted amounts of AFC (curve D, Fig. 5). In fact, the extents of differentiation required for Group 3 in this model are very similar to those modelled for Group 1 (Fig. 5) and for incompatible trace element-depleted lavas from Theistareykir [40].

Irrespective of whether Group 2 rocks retain their primary oxygen isotopic characteristics or have interacted with the crust, the modelling requires that the initial melts were enriched in incompatible trace elements. Furthermore, since differentiation and hydrothermal alteration by meteoric water will have a negligible influence on radiogenic isotope ratios either model can be reconciled with the Sr and Nd isotopic data (Fig. 3). However, the important distinction arises in the implications for δ^{18}O values in the mantle. If Group 2 lavas have retained their primary δ^{18}O then all group 2 and 3 melts are derived from low-δ^{18}O mantle. If, instead, Group 2 have experienced crustal contamination then both groups could be derived from mantle with δ^{18}O values similar to the rest of upper mantle. This issue can be examined further by using helium data.

5.2   Helium - oxygen isotope relationships and early (pre-eruptive) degassing

Helium concentrations lie at the lower end of the range observed in MORB [1,49], probably as a result of melt degassing prior to eruption. He loss renders residual He in melts more susceptible to recording contamination [14,32,49]. Since Icelandic crust is a significant reservoir of radiogenic helium [42] it is important to constrain the effect that degassing/contamination may have had on the primary ^{3}He/^{4}He of melts before determining the origin of ^{3}He/^{4}He variation in the mantle. Figure 6 compares ^{3}He/^{4}He and δ^{18}O for the new dataset. The curvature of binary mixing trajectories between end-members *i* and *j* can be

described by the parameter k (where $k = ([He]/[O]_i)/([He]/[O]_j)$). The abundance of radiogenic He in Icelandic crust depends upon parent element concentration, age of the crust and retention of the He produced. Condomines et al. [42] show that one-million-year-old altered basalt with U = 0.3ppm and Th/U ~3.5, which retains 100% of in-grown He, will contain ~6.5×10^{-8} cm^{3}STP/g of ^{4}He. This value can be reduced or increased proportionally if the crustal component is assumed to be younger or older, respectively.

The range of δ^{18}O and ^{3}He/^{4}He in Group 3 glasses can be modelled by interaction between Group 2 and hydrothermally altered crust. For Icelandic crust with δ^{18}O of 0‰ and ^{3}He/^{4}He of 0.05R_A, a good fit to the Group 3 data is obtained for contamination of a melt containing ~0.22×10^{-8} cm^{3}STP/g He ($k \cong 30$), which is only a factor of three lower than the lowest [He] in any Group 2 glasses (grey, dashed curve, Fig. 6). The amounts of contamination required are similar to those predicted from δ^{18}O values and trace element ratios. This indicates that moderately to extensively degassed magma is susceptible to contamination despite the relatively young age of the Icelandic crust [42]. The lack of ^{3}He/^{4}He variation in Group 2 is more difficult to reconcile with contamination of high-^{3}He/^{4}He melts by radiogenic (crustal) helium. Group 2 glasses display near constant ^{3}He/^{4}He over a wide range of [He] (Fig. 4): a feature that is typical of magma degassing without modification by crustal helium [49,50]. Suites of melts that have interacted with crust tend to display wide variations in ^{3}He/^{4}He at relatively low [He], as is observed in Group 3 (Fig. 4). To generate the recurring ^{3}He/^{4}He signature of Group 2 through melt – crust interaction would require a coincidence of different extents of He contamination for at least three different eruption sites, all producing mixtures with indistinguishable ^{3}He/^{4}He. While we cannot rule out this possibility entirely, we consider it substantially less likely than the alternative hypothesis that Group 2 glasses represent a previously unrecognised source beneath central Iceland with MORB-like ^{3}He/^{4}He (~ 8R_A) and δ^{18}O that is lower than the source of MORB (Section 5.1).

For high-^{3}He/^{4}He magmas we initially assume that a primitive (P) end member samples a less-degassed mantle source, characterised by higher ^{3}He/^{4}He and [He] than crust. Crustal contamination of melts from P should produce convex upwards trajectories to the left of k=1

(Fig. 6); however, all Icelandic glasses (this work and published values) plot with k-values generally between 5 and 50 (i.e. to the right of k=1) implying that $[^3\text{He}]_C \gg [^3\text{He}]_P$. It is highly improbable that Iceland's degassed, basaltic crust will contain more He than primitive mantle melts, which in turn suggests that – as with the model for Group 3 glasses - degassing was able to exert a strong influence on the susceptibility of high-^{3}He/^{4}He melts to crustal contamination. For k-values between 5 and 50 (Fig. 6) the He content of the contaminated melt (P endmember) would fall between 1.36 and 13.6 ($\times$ 10^{-9} cm^{3}STP/g) and, due to the incompatibility of He, the source concentration would be even lower. However, all estimates for mantle [He] are considerably higher than this range. For example, MORB mantle is estimated to contain $\sim$1.5 $\times$ 10^{-5} cm^{3}STP/g [^{4}He] based on measurements of 'popping-rock' from the Mid-Atlantic Ridge [51]. Theoretical estimates of [He] in primitive mantle are even higher: Porcelli and Wasserburg [51] calculated a value of $\sim$6.5$\times$10^{-4} cm^{3}STP/g based upon a steady-state upper mantle fluxed by ^{3}He from a lower mantle reservoir with $\sim$30R_A, whereas Hilton et al. [50] concluded that primitive mantle should contain up to approximately one hundred times more He than MORB-mantle. To explain the He-O isotope relationships (Fig. 6), we conclude that high-^{3}He/^{4}He melts must have undergone extensive magmatic degassing (leading to a reduction in [He] down to 10^{-4} times the initial content) prior to the contamination that introduced radiogenic He and lowered δ^{18}O. We speculate that the enhanced degassing of depleted lavas may be due to higher concentration of intrinsic (source-derived) or extrinsic (crust-derived) water. Icelandic glasses are relatively H$_2$O-rich [52] and water is known to lower the solubility of CO$_2$ in basaltic melt [53]. CO$_2$ is the carrier phase for volatile He and so for any confining pressure (e.g. depth to a magma chamber) a water-rich melt will have lost a greater fraction of its intrinsic helium and is, therefore, more likely to record addition of extraneous (crustal) He [49,54].

5.3   *Helium isotope variations in Central Iceland*

Group 3 glasses have interacted with Icelandic crust so their ^{3}He/^{4}He are not representative of their sources. The helium isotopic characteristics of nearly all other central Iceland rocks fall into two categories: (a) MORB-like values found in trace element-enriched lavas located

immediately north of Vatnajökull, and (b) high-^{3}He/^{4}He values in trace-element depleted rocks found at greater distance from the proposed plume centre. In this respect, the central Iceland data resemble those of Baffin Island which Stuart et al. [27] explained as a result of binary mixing between two mantle components. The first is depleted North Atlantic mantle infiltrated by high-^{3}He/^{4}He material and the second is high-^{87}Sr/^{86}Sr, low ^{143}Nd/^{144}Nd domains in the asthenospheric mantle with ^{3}He/^{4}He $\sim 8R_A$. A similar mixing model can be applied to central Iceland with the lower (MORB-like) ^{3}He/^{4}He mantle concentrated close to Vatnajökull and surrounded by ^{3}He-recharged, depleted mantle (Fig. 1). However, an important difference is that the incompatible trace element-enriched samples from central Iceland have higher ^{143}Nd/^{144}Nd than predicted for the Baffin Island enriched component at ^{3}He/^{4}He of $\sim 8R_A$ (~ 0.51280 [27]; c.f. Fig. 3). This observation suggests that there are differences between the enriched components supplying these two sample suites.

Breddam et al. [9] mapped a "plateau" of high ^{3}He/^{4}He ratios coinciding with the regional gravity and seismic anomalies over central Iceland. These, respective, chemical and physical anomalies were interpreted to represent the focus of mantle upwelling in the region. With our more extensive ^{3}He/^{4}He data set, we are able to recognize additional complexity in the distribution of magma sources in central Iceland. We note the existence of a source located north of Vatnajökull with MORB-like values surrounded by a high-^{3}He/^{4}He source. The close juxtaposition of mantle sources with (i) ^{3}He/^{4}He $\sim 8R_A$ and, (ii) ^{3}He/^{4}He $> 8R_A$ has also been inferred for other OIB localities - such as Heard Island [32] and Hawaii [54]. Big Ben lavas on Heard Island have MORB-like ^{3}He/^{4}He values whereas rocks from the Laurens Peninsula, located ~ 25 km distant, have ^{3}He/^{4}He up to $18R_A$. In the case of Loihi Seamount ($\sim 30R_A$), some nearby edifices have produced magmas with MORB-like ^{3}He/^{4}He e.g. Mauna Kea and Mauna Loa. Kurz et al. [54] attributed this latter observation to radial zonation of ^{3}He/^{4}He in the Hawaiian mantle. The situation appears to be more complex in central Iceland, however, with a core of MORB-like ^{3}He/^{4}He values confined mainly to the area immediately north of Vatnajökull that contains high-^{3}He/^{4}He domains at Vadalda and at Kambsfell (Fig. 1). The MORB-like ^{3}He/^{4}He source is mainly associated with incompatible trace element-enriched

lavas, but at Eggert and Herdubreid trace element depleted rocks have ^{3}He/^{4}He ~ 8R_A. These observations suggest that high-^{3}He/^{4}He material can be introduced to enriched or depleted mantle but in the source of Iceland is more likely to interact with the latter. It is noteworthy that Miocene lavas from Selardalur in NW Iceland, which have the highest ^{3}He/^{4}He of any Icelandic location [23], also possess Sr and Nd isotope ratios similar to Vadalda (Fig. 3).

Breddam et al. [9] also interpreted a steady decrease in ^{3}He/^{4}He along the NRZ to MORB-like values at Theistareykir as reflecting a changing melting regime associated with dispersal of plume material away from the conduit. However, their dataset contained relatively little data for central Iceland and they reported relatively low ^{3}He/^{4}He values of 8 to 9R_A for Theistareykir. Our new Theistareykir data indicate that ^{3}He/^{4}He beneath northernmost Iceland is ≥ 12R_A. Therefore, we suggest that changes in ^{3}He/^{4}He along the strike of the NRZ may reflect (i) a less extreme ^{3}He/^{4}He gradient then envisaged by [9], or (ii) variable interaction between high-^{3}He/^{4}He magmas and the crust. Either scenario is consistent with ^{3}He/^{4}He ratios ≥ 11R_A found offshore to the north of Iceland [55].

Finally, we note that although Vadalda has the highest reported ^{3}He/^{4}He ratio (34.3R_A) from Iceland's neovolcanic zones, there are two reasons to suggest that its ^{3}He/^{4}He ratio may not represent the pure high ^{3}He/^{4}He mantle endmember composition in Icelandic magmatism. First, it falls short of the ratio predicted (~55R_A) from extrapolation of linear He-Pb mixing relationships observed for the Reykjanes Ridge [50] to a ^{206}Pb/^{204}Pb ratio of 19.35. This prediction assumes that (i) central Iceland lies at the core of the present-day Icelandic plume and is characterised by a distinctive enriched source component with ^{206}Pb/^{204}Pb of 19.35 (IE1 of [5]), and (ii) that the Reykjanes Ridge suite also contains a contribution from this component diluted by one or more depleted sources that possess lower ^{206}Pb/^{204}Pb and ^{3}He/^{4}He ratios. Second, if proto-Iceland plume at Baffin Island (~61Ma) is characterised by a ^{3}He/^{4}He ratio of 49.5R_A [27] – itself a minimum estimate - then, for an assumed primitive mantle ^{3}He content of 7.6×10^{10} atoms/g [12], closed-system addition of radiogenic He (Bulk Earth U =21ppb and Th/U = 3.8) should produce a present-day Iceland plume ^{3}He/^{4}He value of ~ 48R_A – considerably higher than the observed value at Vadalda.

5.4 Oxygen isotope heterogeneity of Icelandic mantle

Oxygen isotopic characteristics in central Iceland lavas are different in incompatible trace element enriched and depleted samples. The highest δ^{18}O values of incompatible trace element-depleted rocks overlap with the ranges for oceanic lavas from various tectonic settings, and some lowering of oxygen isotope ratios can be shown to result from interaction with low-δ^{18}O Icelandic crust (Section 5.1). However, incompatible trace-element enriched glasses have lower oxygen isotope ratios that require the presence of a low-δ^{18}O source. Macpherson et al. [11] discussed two possible origins for ^{18}O-poor material in the mantle. The first suggestion - a contribution from ^{18}O-depleted outer core alloy - is highly unlikely in the present case. If the outer core does have low δ^{18}O, it will be lower than the mantle by only a few tenths of a per mille [11] and unable to produce the relatively large δ^{18}O difference required of the enriched source in central Iceland. The alternative possibility is recycling of oceanic crust that was hydrothermally altered (>250°C) prior to subduction. Recycled crustal components have recently been advocated to explain Sr, Nd and Pb isotopic variation throughout Iceland [5], and has previously been proposed to explain low magmatic δ^{18}O in other hotpot localities at Hawaii and the Manus Basin [11,56]. Unfortunately, there is insufficient oxygen isotopic variation in the present suite of glasses to construct meaningful models of this process (c.f. [5]), but simple mass balance calculations can be performed to predict the isotopic composition and quantity of recycled crust that must be added to typical upper mantle (δ^{18}O = +5.5‰ [39]) to produce a source with δ^{18}O of +4‰. Figure 7 demonstrates that ~ 40% recycled crust with δ^{18}O of +2‰, at the lower end of variation recognised in ophiolites or ocean floor drill holes, is required to produce a mixture with δ^{18}O of +4‰ [57,58]. A smaller contribution can be accommodated if the recycled crust possessed lower δ^{18}O (Fig. 7).

5.5   Implications for the He-paradox and statistical interpretation of ^{3}He/^{4}He datasets

Lavas with ^{3}He/^{4}He greater than MORB are often inferred to contain a large contribution from a reservoir that is richer in primordial volatiles than DMM. This reservoir could be the lower mantle [1 and references therein]. However, the observation that high ^{3}He/^{4}He lavas from Loihi Seamount have lower [He] than MORB yields a "helium paradox" [59] between the theory of a He-rich, high-^{3}He/^{4}He source and the reality of He-poor, high-^{3}He/^{4}He lavas. Explanations offered for this paradox include He degassing from ocean island magmas

because of low confining pressures [50], loss of a CO_2-rich carrier phase at the base of the lithosphere beneath ocean islands [29], different gas-loss mechanism for MORB and OIB [60], or recharge of DMM by small volumes of primordial volatile-rich material [27]. The new data indicate that helium concentrations in high-^{3}He/^{4}He magmas from central Iceland have been strongly influenced by degassing, which will lower [He] *prior* to addition of distinctive crustal helium, which will lower ^{3}He/^{4}He. As discussed above, we speculate that this process may be exacerbated by higher water contents in Icelandic lavas. Helium concentrations can be anticipated to have been lowered further during eruption. We suggest that in central Iceland the features typical of the He-paradox result from extensive degassing of magma during their transport through the lithosphere.

6. Summary

Helium and oxygen isotope ratios of basalts from central Iceland show considerable variation. Our samples that are derived from depleted mantle sources predominantly have high ^{3}He/^{4}He but δ^{18}O similar to the upper mantle. Enriched sources tend to produce primary melts with MORB-like ^{3}He/^{4}He (~ $8R_A$) and lower δ^{18}O (~ 4‰). Much of the magmatism in central Iceland is prone to interaction with the crust, which acts to lower both helium and oxygen isotope ratios. For helium, this modification is most apparent in rocks that contained little helium. Helium loss occurs primarily through degassing, which may be exacerbated by high water contents. As a general rule, ^{3}He/^{4}He can be expected to show more variation in suites of magmas that contain high water contents as a result of water-rich sources or interaction with hydrated crust.

High ^{3}He/^{4}He magmatism is concentrated to the north and, particularly, to the west of central Iceland. The elevated ^{3}He/^{4}He in these regions results from interaction between depleted mantle and a helium-rich, high-^{3}He/^{4}He component (c.f. [27]). Despite the effects of the crust some incompatible trace element-enriched lavas from immediately north of Vatnajökull preserve primary MORB-like ^{3}He/^{4}He and low δ^{18}O. The trace element enrichment and low δ^{18}O values most probably reflect a source containing recycled, subducted oceanic crust. High ^{3}He/^{4}He ratios are rare in this area, which is close to the proposed centre of the Icelandic plume. The predominantly MORB-like ^{3}He/^{4}He ratios suggest that the mantle north of Vatnajökull has largely, but not exclusively, escaped the ^{3}He-enrichment observed to the north and west. Since sources with enriched and depleted characteristics can both carry

MORB-like ^{3}He/^{4}He or higher ^{3}He/^{4}He, the origin of high ^{3}He/^{4}He in Icelandic mantle is independent of the processes controlling the evolution of trace elements, and of radiogenic and stable isotope ratios.

Acknowledgements

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Figure Captions

Figure 1. Location map of samples from central Iceland. Solid, open and grey symbols denote glass and lava groups defined in following figures. Crosses are published values for hyaloclastic glasses, phenocrysts and geothermal fluids [9,21,22,61]. Locations are labelled

with ^{3}He/^{4}He ratios (*R/R*$_A$ notation) and the dashed line encloses the area dominated by primary ^{3}He/^{4}He ratios that are not significantly different to MORB. Inset shows location of main map relative to the Western (WRZ), Eastern (ERZ) and Northern (NRZ) rift zones and ^{3}He/^{4}He of samples (this work) outside the area covered in the main panel.

Figure 2. Trace element patterns of (a) subglacial glasses and (b) subaerial lavas normalised to primitive mantle [62]. In (b) small symbols are for samples outside the main panel in Fig. 1. Additional data sources: Th-29 [20] and NAL-611 [9].

Figure 3. ^{143}Nd/^{144}Nd versus ^{87}Sr/^{86}Sr for central Iceland lavas [63]. Symbols as in Fig. 2. Black triangles show endmember mantle sources for Iceland, proposed by Thirlwall et al. [5]. The "enriched" endmember from Baffin Island [27] lies at lower ^{143}Nd/^{144}Nd of ~0.51280.

Figure 4. ^{3}He/^{4}He versus helium concentration ([He]). Central Iceland data are plotted as large symbols and other locations as small symbols. 2σ errors are shown or are smaller than symbols. The range of MORB is shown by dashed lines.

Figure 5. δ^{18}O$_{melt}$ versus La/Sm. Oxygen isotope ratios are plotted either as glass δ^{18}O values or, where olivine was analysed, as melt equivalent based on $\Delta_{melt-ol}$ of +0.4‰. Symbols are as in Fig. 2. The dashed line (curve A) represents mixing between a depleted lava with δ^{18}O = +5.1‰ and La/Sm = 0.67 [40] and rhyolitic magma. The rhyolite has δ^{18}O of 0‰ (see text for discussion) with La = 30ppm and La/Sm = 3.3 (illustrative values based on estimates in the literature [20,40,48]). Tick marks indicate percentage of rhyolite in mixture. Solid curves are models for fractional crystallisation with assimilation of hydrothermally altered crust with the composition proposed by Eiler et al [40]. Curve B is for contamination of depleted lava from Theistareykir [40], curve C is for contamination of a Krafla basalt with La = 6.3 and La/Sm = 2.9 [64] and curve D is for contamination of a Group 2 glass (KVK-169). Tick marks give the fraction of melt remaining. Data for melt compositions calculated from WRZ lavas (inverted triangles) and NRZ lavas (diamonds) are shown for comparison [9,20,40].

Figure 6. Plot of ^{3}He/^{4}He versus δ^{18}O$_{melt}$ for central Iceland glasses and lavas. Symbols as in Fig. 4. Models are shown of mixing between Icelandic crust (C: ^{3}He/^{4}He = 0.05R_A [65]; [He] = 6.5 × 10^{-8} cm^{3}STP/g [42]; δ^{18}O = 0‰ [40]) and two different types of melt. Thin solid lines illustrate various mixtures of this crust with a high-^{3}He/^{4}He melt (P) with ^{3}He/^{4}He = 49.5R_A [27]) and δ^{18}O = 5.1‰ [39,40] for different values of k [= (He/O)$_C$/(He/O)$_P$]. Vertical, dashed lines show the fraction of C in these mixtures. The thicker, dashed, grey line shows crustal contamination of melt with helium and oxygen isotopic characteristics of a Group 2 glass that has been degassed by a factor of 10 (^{3}He/^{4}He = 8.9R_A; [He] = 0.22 × 10^{-8} cm$^{3STP/g}$; δ^{18}O = 4.1‰).

Figure 7. Proportion and oxygen isotopic composition of altered oceanic crust required to generate a source with δ^{18}O of 4‰ by mixing with typical upper mantle [39].
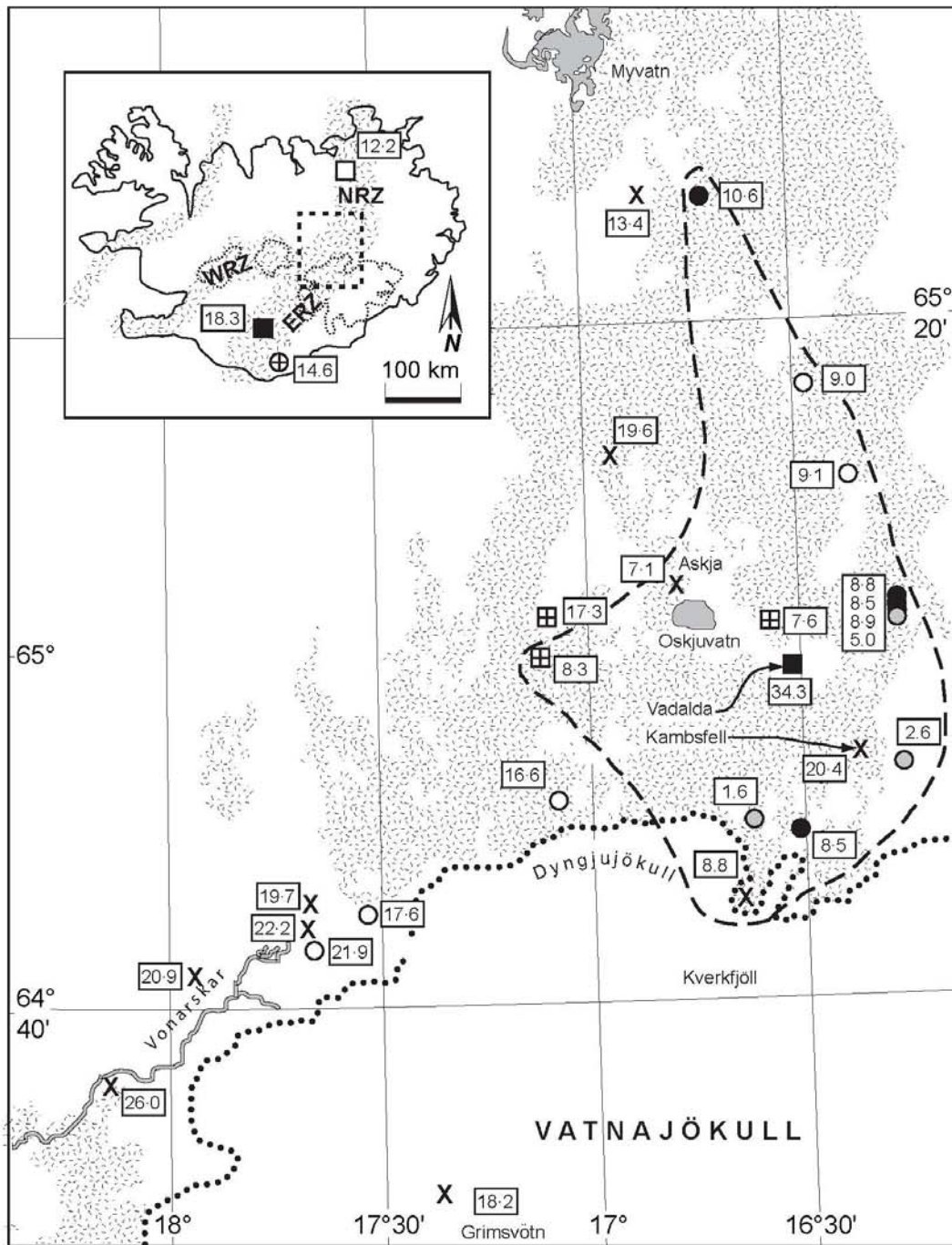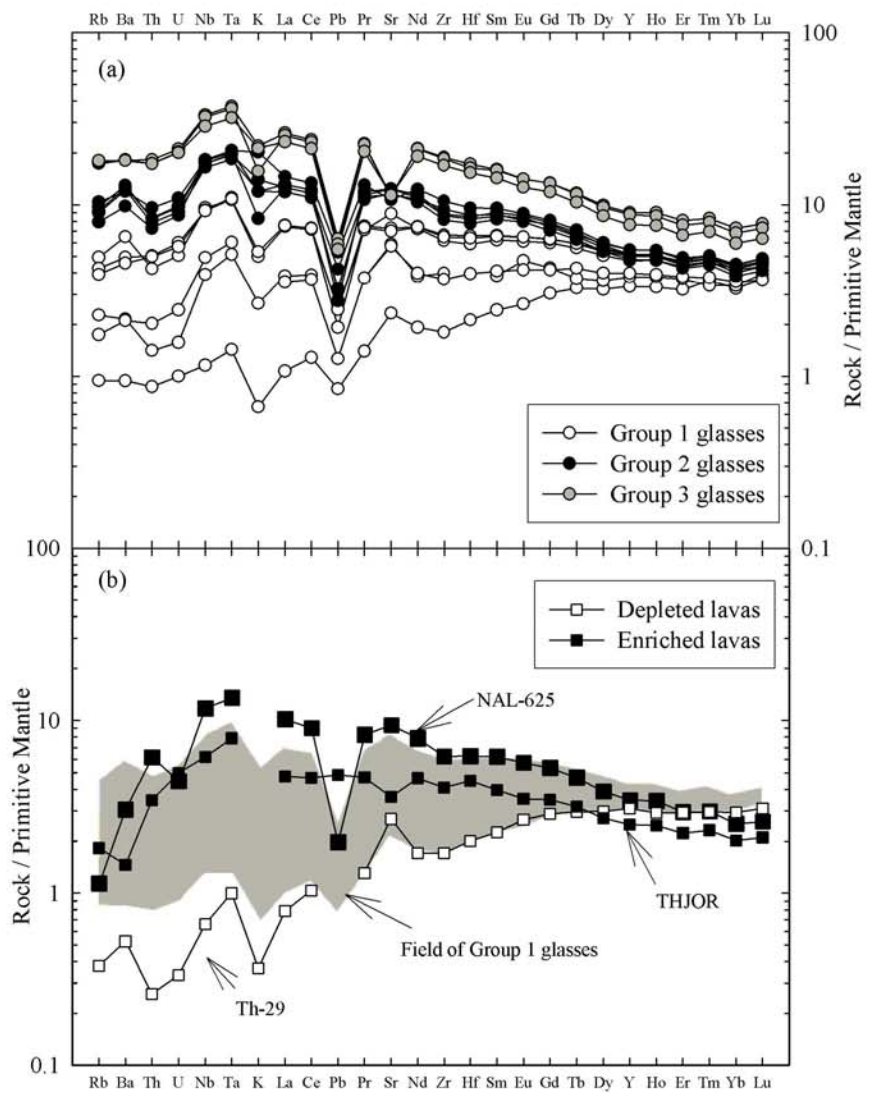
Figure 1

Figure 2

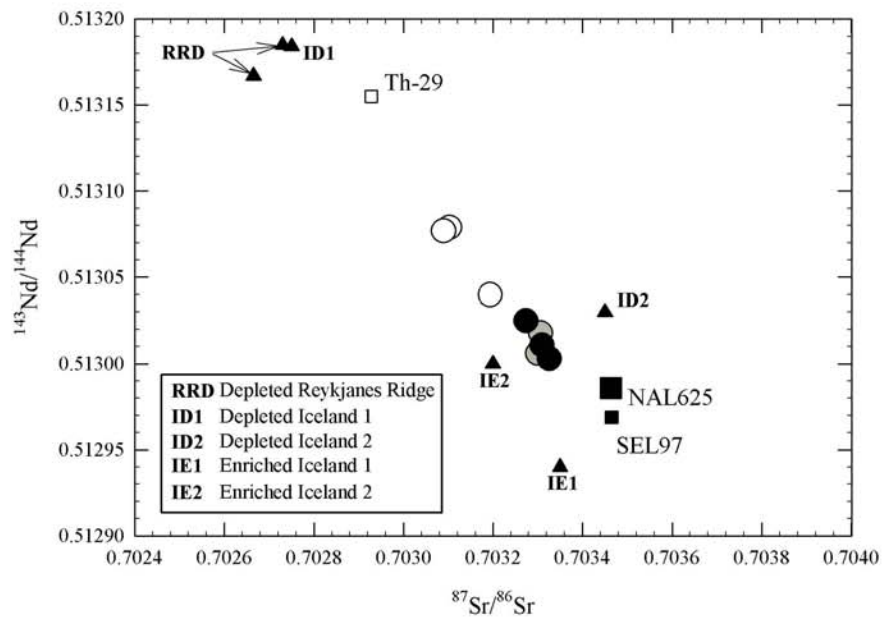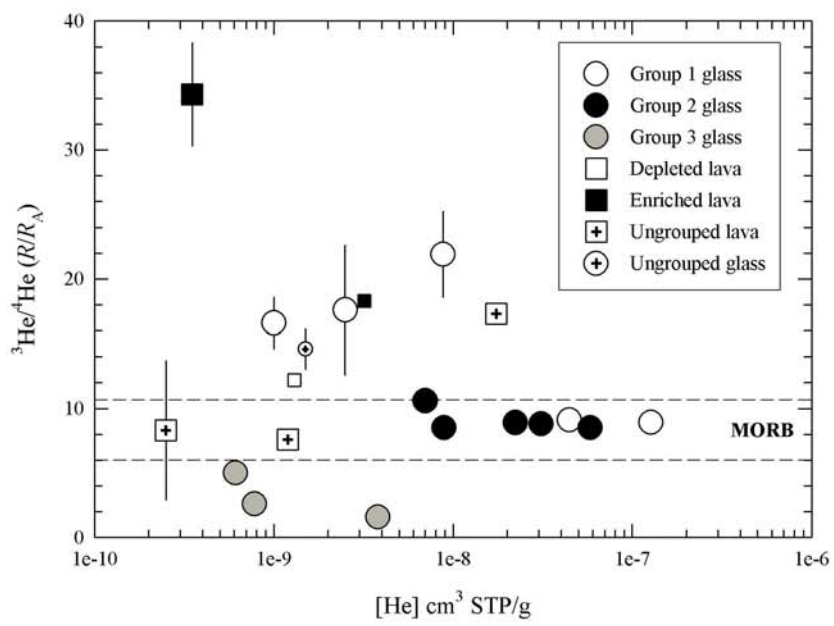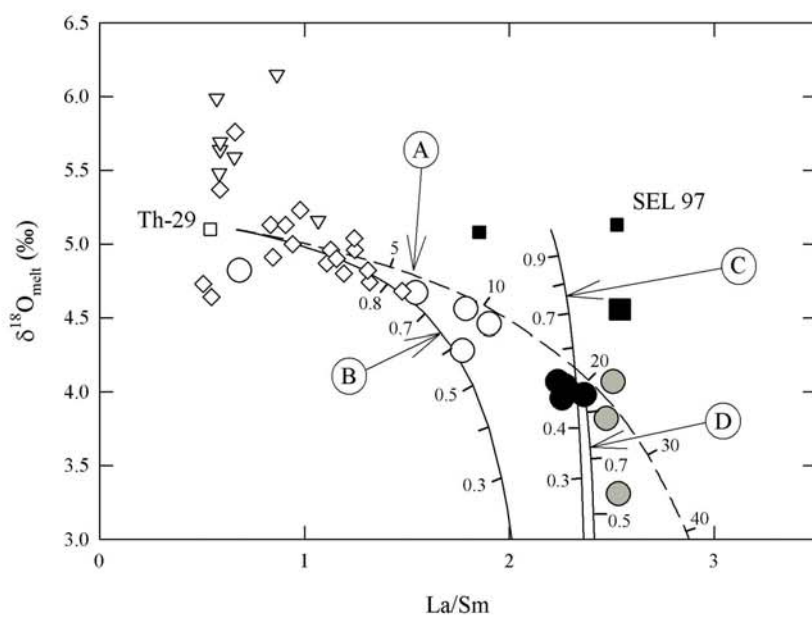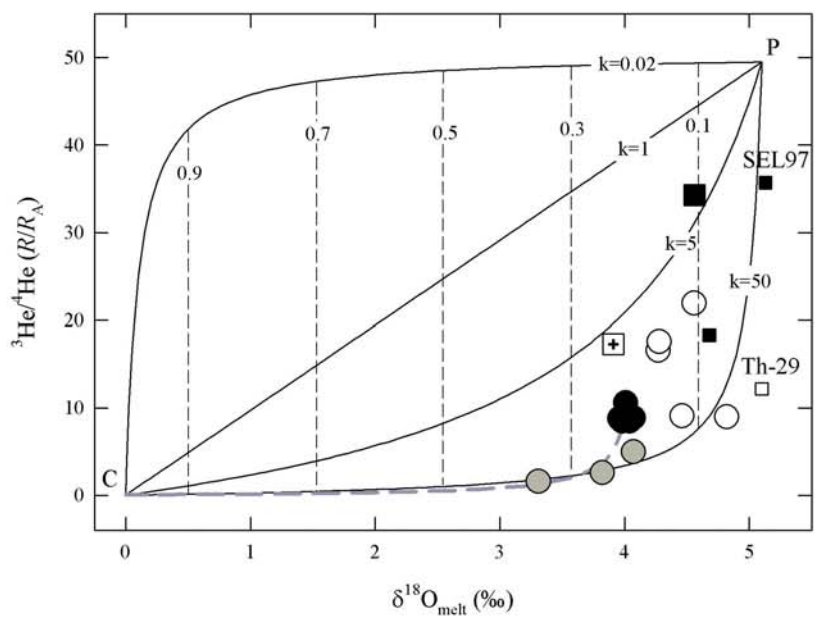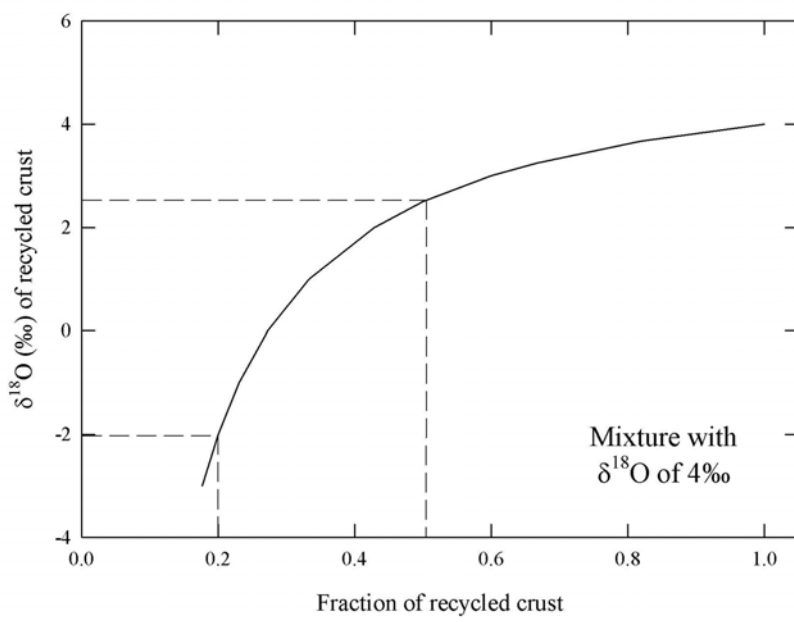**Figure 3**

Figure 4

Figure 5

Figure 6

Figure 7

Table 1. Locations, helium and oxygen isotope data for central Iceland basalts. Helium data obtained by crushing at Scripps Institution of Oceanography and oxygen data obtained at Royal Holloway University of London by laser-fluorination.

Sample	Phase	Location	Latitude / Longitude	wt. (g)	[He][a] ($10^{-9}cm^3$STP/g)	^{3}He/^{4}He (R/R_A)	s.d	δ^{18}O (‰)	s.d.[b]	(La/Yb)$_n$	Group
NRZ											
Th-29	olivine	Theistareykir	65°53'N 16°50W	0.65	1.3	12.2	0.3	4.70[c]	0.03[c]	0.27	D
Central Iceland											
Herdubreid	glass	Herdubreid	65°10'N 16°21'W	0.50	44.5	9.1	0.2	4.46		1.77	1
NAL-611	olivine	Kistufell	64°48'N 16°14'W	2.36	1.0	16.6	2.0	4.27		1.13[d]	1
NAL-496	glass	Gaesahjukur	64°46'N 17°29'W	0.11	2.5	17.6	5.0	4.28		1.72	1
SAL-306	glass	Bardarbunga	64°42'N 17°35'W	0.10	8.8	21.9	3.0	4.56	0.21	1.75	1
NAL-688	glass	Eggert	65°15'N 16°29'W	0.50	127.0	8.9	0.4	4.82	0.06	0.33	1
	repeat			0.12	85.0	8.8	0.2				
	olivine (gr)[e]			1.20	1.2	8.0	0.4	4.38			
	olivine (cc)[e]			1.08	1.3	5.9	0.4	5.08	0.27	0.37	D
NAL-239	olivine	Ketildyngja	65°27'N 16°43'W	0.94	7.0	10.6	0.3	4.01[f]		2.82[f]	2
NAL-355	glass	Upptyppingar	65°02'N 16°14'W	0.11	31.0	8.8	0.5	3.96		3.16	2
NAL-356	glass	Upptyppingar	65°02'N 16°14'W	0.11	58.2	8.5	0.3	4.04		3.14	2
NAL-355/6	glass	Upptyppingar	65°02'N 16°14'W	0.59	22.3	8.9	0.2	4.07		3.1	2
KVK-169	glass	Jardfraedingaslod	64°46'N 16°30'W	0.51	8.9	8.5	0.3	3.98		3.36	2
NAL-357	glass	Upptyppingar	65°02'N 16°14'W	0.59	0.6	5.0	0.7	4.07		3.89	3
KVK-147	glass	Hvannalindir	64°52'N 16°21'W	0.54	0.8	2.6	0.9	3.82		3.71	3
KVK-168	glass	Jardfraedingaslod	64°46'N 16°38'W	0.50	3.8	1.6	0.2	3.31	0.01	3.58	3
NAL-625	olivine	Vadalda	64°56'N 16°28'W	1.10	0.4	34.3	4.0	4.16	0.23	4.02	E
	repeat			3.04	0.2	30.5	3.0				
	clinopyroxene			0.62	0.4	20.2	5.0	4.26			
#9613	olivine	Odadahraun	65°09'N 16°32'W	1.58	1.2	7.6	0.8	n.d.		n.d.	ungrouped
TRO-53	olivine	Trolladyngja	64°54'N 17°06'W	2.77	0.3	8.3	5.0	n.d.		n.d.	ungrouped
NAL-626	olivine	Fjallsendi	65°03'N 17°05'W	0.52	17.4	17.3	0.3	3.51	0.03	n.d.	ungrouped
ERZ											
SAL-575	glass	Solheimajokull	63°32'N 19°23'W	0.87	1.5	14.6	3.1	n.d.		n.d.	ungrouped
THJOR	olivine	Thjorsardalur	64°08'N 20°10'W	1.61	3.2	18.3	0.6	4.68		2.36	E

a. ± 5%

b. standard deviation of replicate analyses.

Table 2. Major element oxide concentrations of central Iceland glasses (gl) and whole rock (wr).

	SAL 306	Herdubreid	NAL 688	NAL 239	NAL 355	NAL 356	KVK169	NAL 357	KVK147	KVK168
	gl	wr	gl	gl	gl	gl	gl	gl	gl	gl
SiO_2	49.61	48.06	49.40	50.71	48.16	47.90	51.43	50.40	47.86	50.50
TiO_2	1.32	1.31	0.57	1.53	1.95	2.19	2.83	2.66	2.64	3.33
Al_2O_3	14.33	16.05	14.30	14.33	15.74	13.40	13.45	12.00	13.24	12.97
FeO	10.68	11.30	9.12	11.19	9.91	11.10	13.59	12.60	14.09	13.94
MnO	0.17	0.18	0.14	0.19	0.16	0.14	0.22	0.17	0.22	0.22
MgO	7.78	12.45	9.73	6.72	8.42	7.17	5.03	5.12	6.43	5.05
CaO	13.50	9.20	14.50	12.34	12.44	13.10	9.44	11.10	11.83	9.87
Na_2O	2.17	1.91	1.46	2.29	2.46	2.30	2.80	2.34	2.69	2.83
K_2O	0.16	0.15	0.02	0.15	0.36	0.42	0.61	0.64	0.47	0.66
P_2O_5	0.17	0.12	0.04	0.27	0.25	0.23	0.32	0.42	0.40	0.42
Total	99.89	100.73	99.10	99.72	99.85	97.95	99.72	97.45	99.87	99.79

Table 3. Trace element concentrations (in ppm) of central Iceland glasses determined at University of Durham by ICP-MS.

	SAL306	NAL496	Herdubreid	NAL688	NAL239	NAL355	NAL355/6	NAL356	KVK-169	NAL357	KVK-147	KVK-168
Group	1	1	1	1	2	2	2	2	2	3	3	3
all ppm												
Rb	2.5	2.7	2.7	0.6	5.0	5.9	5.7	6.2	6.6	11.4	11.2	11.0
Sr	148	154	164	49.2	241	261	247	260	226	239	236	235
Y	22.8	23.4	19.2	15.2	24.9	23.4	21.9	23.1	24.8	34.7	39.8	40.8
Zr	72.6	74.4	61.2	20.1	105.5	98.9	91.3	96.7	117.3	189.2	207.2	210.7
Nb	6.6	6.6	6.2	0.8	12.4	12.8	11.7	12.4	13.0	20.3	23.1	23.7
Ba	32	34	39	6	68	88	85	90	84	126	127	126
La	5.20	5.21	4.60	0.73	8.63	9.01	8.15	8.67	9.99	15.90	17.37	17.96
Ce	12.9	13.0	11.4	2.3	20.7	21.6	19.5	20.7	23.7	37.5	41.0	42.3
Pr	2.0	2.1	1.8	0.4	3.1	3.3	3.0	3.1	3.6	5.6	6.2	6.3
Nd	10.0	10.1	8.6	2.6	14.6	15.5	14.0	14.9	16.7	25.8	28.4	28.6
Sm	2.91	2.94	2.42	1.08	3.90	3.99	3.64	3.82	4.22	6.34	7.02	7.09
Eu	1.09	1.09	0.94	0.44	1.40	1.45	1.34	1.41	1.49	2.12	2.36	2.36
Gd	3.77	3.73	3.11	1.81	4.57	4.67	4.22	4.43	4.82	7.07	7.94	7.97
Tb	0.64	0.64	0.52	0.35	0.72	0.74	0.68	0.70	0.77	1.11	1.24	1.26
Dy	3.93	4.02	3.24	2.37	4.36	4.28	3.92	4.14	4.42	6.33	7.11	7.30
Ho	0.82	0.83	0.68	0.54	0.89	0.84	0.78	0.82	0.89	1.24	1.41	1.47
Er	2.21	2.24	1.84	1.53	2.35	2.19	2.04	2.13	2.29	3.18	3.65	3.87
Tm	0.36	0.36	0.31	0.27	0.37	0.36	0.33	0.35	0.37	0.51	0.58	0.62
Yb	2.14	2.18	1.83	1.60	2.20	2.05	1.88	1.98	2.13	2.93	3.36	3.60
Lu	0.35	0.36	0.30	0.27	0.36	0.32	0.31	0.32	0.34	0.47	0.54	0.57
Hf	1.97	2.04	1.62	0.66	2.68	2.64	2.40	2.56	2.95	4.74	5.04	5.32
Ta	0.44	0.45	0.40	0.06	0.81	0.83	0.75	0.80	0.85	1.31	1.48	1.52
Pb	0.36	0.45	0.28	0.16	0.59	0.51	0.60	0.78	0.98	1.01	1.09	1.17
Th	0.42	0.43	0.32	0.07	0.62	0.71	0.66	0.71	0.81	1.47	1.47	1.55
U	0.12	0.13	0.09	0.02	0.18	0.21	0.19	0.20	0.23	0.42	0.42	0.44