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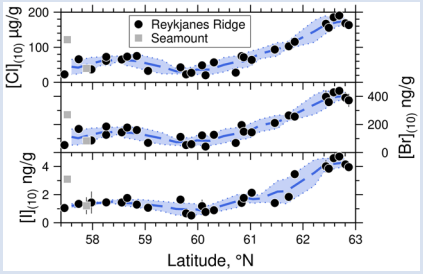
Halogens track mantle enrichment along the Reykjanes Ridge towards Iceland

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Abstract



Halogens enter the mantle by subduction of altered oceanic crust, and return to the exosphere through magmatic degassing. Submarine basaltic glasses from the Reykjanes Ridge sample a 700 km transect towards the Iceland plume, capturing a transition from depleted towards strongly plume-influenced mantle. Chlorine, Br and I concentrations increase sharply as the mantle becomes more enriched, matching the behaviour of lithophile trace elements and radiogenic isotopes. Halogen enrichment does not correlate with $^{3}He/^{4}He$, suggesting that the halogen budget of Reykjanes Ridge magmas approaching Iceland is controlled by an enriched upper mantle component, not by the high-$^{3}He/^{4}He$ plume source. This enriched mantle likely contains 33–61 µg/g Cl, 77–149 ng/g Br and 0.8–1.6 ng/g I, with most of this halogen inventory derived from recycled oceanic crust.

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Introduction

The flux of volatile elements into the mantle *via* subduction has created significant geochemical heterogeneity in the deep Earth over geological time (*e.g.*, Bekaert *et al.*, 2024). The fluid-mobile and highly incompatible halogens (Cl, Br, I) are significantly enriched in seawater, sediments, altered oceanic crust and serpentinised lithosphere compared to depleted upper mantle (Barnes *et al.*, 2018). Although halogens from subducting slabs are lost to the mantle wedge and arc magmatism, primarily *via* slab dehydration, exhumed high-pressure antigorite serpentinites can retain significant halogen concentrations (John *et al.*, 2011). The stability of antigorite to depths of 150–200 km (Ulmer and Trommsdorff, 1995) suggests that slab residues have capacity to return a significant flux of halogens to the deep mantle (Barnes *et al.*, 2018), making halogens potentially powerful tracers of recycled subducted lithologies.

Diverse halogen enrichments and $\delta^{37}Cl$ values have been observed among plume settings (*e.g.*, John *et al.*, 2010; Kendrick *et al.*, 2015; Halldórsson *et al.*, 2016a), implying variability in the efficiency of the solid Earth halogen cycle in space and/or time. The Iceland mantle plume, as the largest modern expression of a deep mantle upwelling (Hoggard *et al.*, 2016) and known to return recycled material to the surface (Shorttle and Maclennan, 2011), provides an excellent opportunity to evaluate the solid Earth halogen cycle.

We report halogen (Cl, Br, I) compositions in 29 basalt glasses dredged from the Reykjanes Ridge between 57.4° N and 63° N during the RSS Charles Darwin cruise CD80 (Murton *et al.*, 2002) (Fig. S-1). The Reykjanes Ridge lies in a region of the North Atlantic with positive residual depth and V-shaped ridges manifest, preserving an indirect record of thermal pulsing of the Iceland plume over time (Parnell-Turner *et al.*, 2017). It samples along-ridge mantle heterogeneity associated with the Iceland plume, as well as local heterogeneity in adjacent seamounts. Lithophile trace element and isotopic signatures (*e.g.*, Murton *et al.*, 2002) indicate the progressive melting out of enriched domains as the mantle flows radially outwards from the Iceland plume and through the melting region of the Reykjanes Ridge (Shorttle *et al.*, 2010), until by 61° N the fingerprint of trace element and isotopic enrichment is gone. A broad peak in $^{3}He/^{4}He$ (16.0 R/R_A) at 59–62° N coincides with the first V-shaped ridge, while elevated $^{3}He/^{4}He$ above 11 R/R_A persists as far south as 57° N (Hilton *et al.*, 2000). To a first order, these observations suggest a heterogeneous plume in which lithophile element and helium isotopic signatures are carried by different mantle components, with the high-$^{3}He/^{4}He$ component becoming visible once the enriched component(s) is exhausted and/or at very high temperatures of melting (*e.g.*, Rasmussen *et al.*, 2020). Icelandic basalts are overall geochemically depleted and exhibit neither extreme EM nor HIMU compositions (*e.g.*, Harðardóttir *et al.*, 2022); however, their trace element and isotopic heterogeneity requires at least two depleted and three

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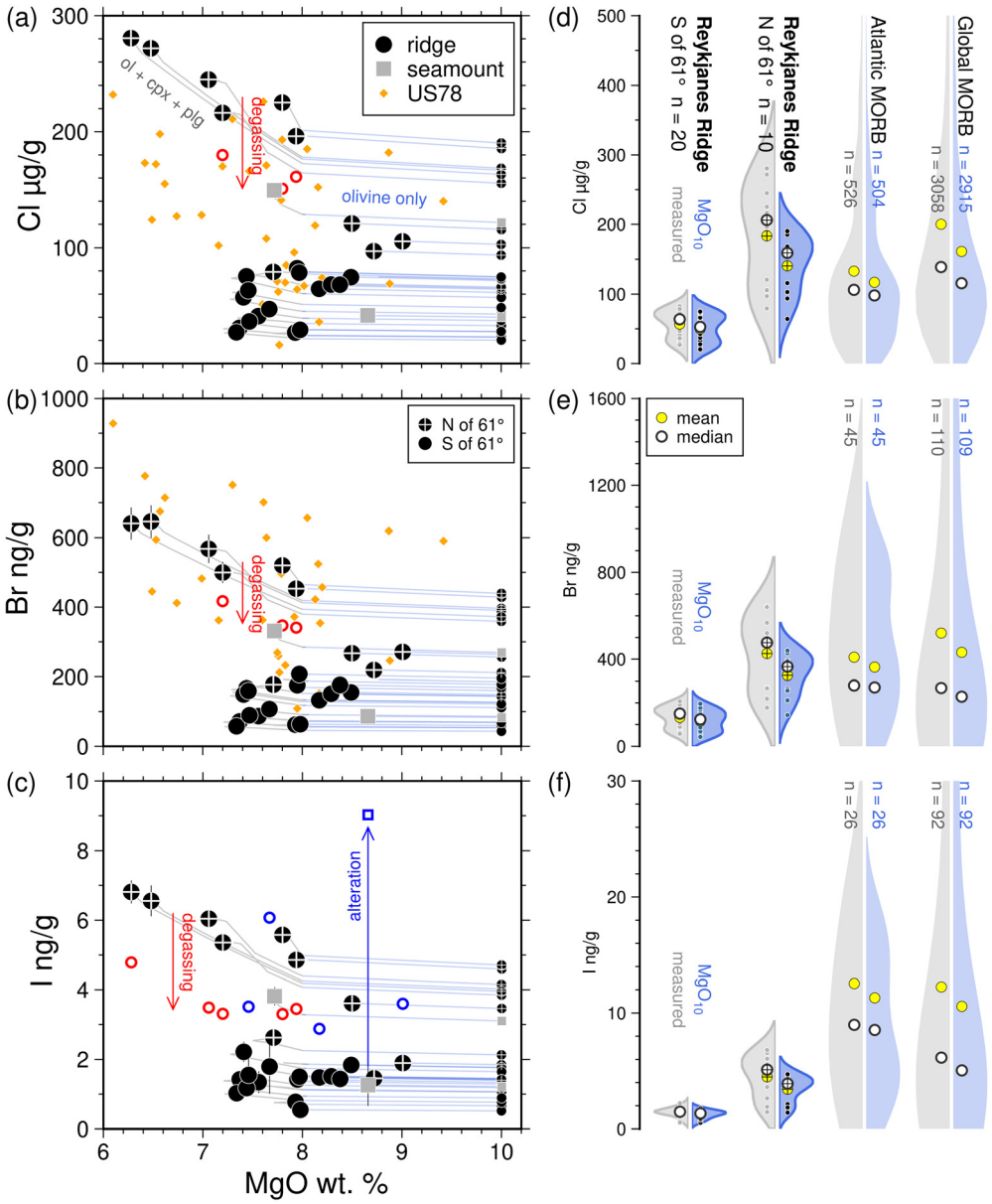


Figure 1 **(a–c)** Halogen contents in Reykjanes Ridge glasses from 57.4–63° N *vs.* MgO, with reverse fractional crystallisation paths to 10 wt. % MgO. Crossed symbols indicate samples from the northern Reykjanes Ridge ≥61° N. Open symbols show glass compositions without correction for degassing (red) or alteration (blue). Orange diamonds are Reykjanes Ridge whole rocks from 57.7–63.8° N (US78; Unni and Schilling, 1978). **(d–f)** Kernel density distributions of glass compositions as measured (left, grey; note change of *y* axis scale), and after correction to 10 wt. % MgO (right, blue). Southern (<61° N) and northern segments of the Reykjanes Ridge are compared with Atlantic and global MORB (data compiled from PetDB, www.earthchem.org/petdb).

enriched local mantle endmembers with weak but variable EM1- or HIMU-like affinities (*e.g.*, Peate *et al.*, 2010). We aim to constrain the relative contributions of enriched, depleted and high-$^{3}He/^{4}He$ mantle endmembers to the halogen budget of Reykjanes Ridge magmas.

Halogen Abundances in Reykjanes Ridge Glasses

We measured Cl, Br, I and K in fresh glass chips by neutron irradiation noble gas mass spectrometry (Ruzié-Hamilton *et al.*, 2016). Analytical and data reduction procedures, and complete datasets, are provided as Supplementary Information. Typical 1σ uncertainties are ±1–3 µg/g for Cl, ±5–40 ng/g for Br, ±0.05–0.30 ng/g for I, and ±1–10 µg/g for K. From a mantle geochemical perspective, it is important to ascertain whether halogen contents were modified by magma degassing or assimilation of seawater or brines. We corrected five samples for degassing and five for alteration. Following corrections, pristine Reykjanes Ridge glasses contain 26–281 µg/g Cl, 57–646 ng/g Br, 0.5–9.0 ng/g I, and 270–1580 µg/g K. Negative correlations between MgO and halogens in glasses from the northern Reykjanes Ridge ≥61° N (Fig. 1a–c) are well explained as incompatible element enrichment by exclusion during fractional crystallisation, arguing strongly against an assimilation origin for the elemental halogen variability in these samples.

To explore mantle-derived variability in primary melt halogen contents we correct all Reykjanes Ridge glasses to 10 wt. % MgO. This yields primary melt halogen abundances of 20–190 µg/g Cl, 43–440 ng/g Br and 0.5–4.7 ng/g I (Fig. 1).

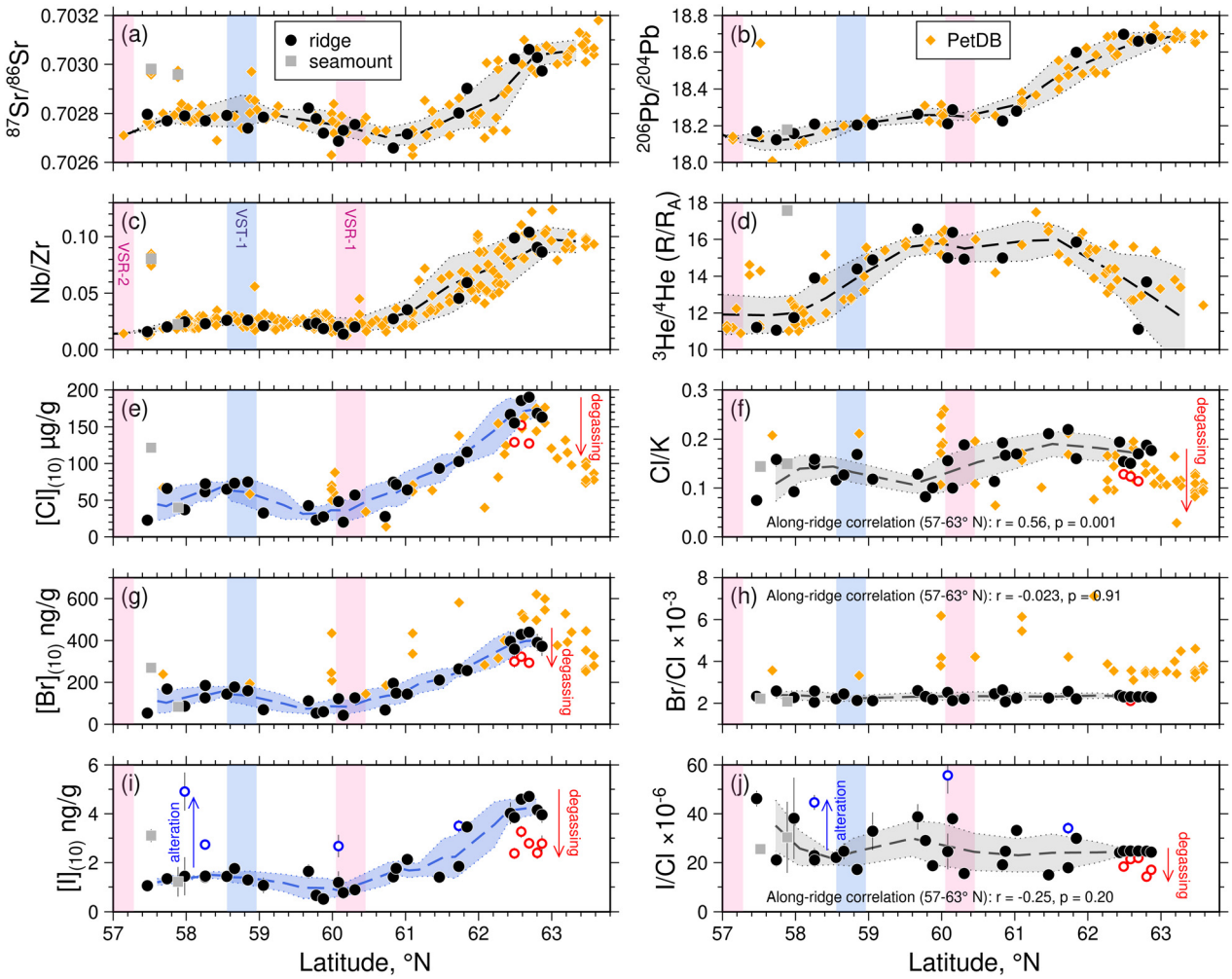


Figure 2 Along-ridge compositional variation in Reykjanes Ridge glasses. **(a)** $^{87}Sr/^{86}Sr$, **(b)** $^{206}Pb/^{204}Pb$, **(c)** Nb/Zr, **(d)** $^{3}He/^{4}He$ (R/R_A notation), **(e, g, h)** halogen concentrations after fractionation correction to 10 wt. % MgO (halogen contents without fractionation correction are shown in Fig. S-7), and **(f)** Cl/K, **(h)** Br/Cl, **(j)** I/Cl ratios (no fractionation correction). Vertical bands: V-shaped ridges (VSR, pink) and troughs (VST, blue). Open red and blue symbols as for Figure 1. Dashed lines and shaded areas show the along-ridge running average and 1σ standard deviation, calculated using a boxcar filter with bandwidth 2° latitude. Reykjanes Ridge glass and whole-rock compositions (orange diamonds) are compiled from PetDB; Br and Cl (Unni and Schilling, 1978) are not corrected for the effects of degassing or alteration.

Samples from <61° N have lower mean and median halogen contents than Atlantic and global MORB (Fig. 1d–f). All Reykjanes Ridge basalts have lower I contents than Atlantic or global MORB, although the broad MORB composition distributions extending to 30 ng/g I (Fig. 1f) likely include samples affected by alteration.

Along-Ridge Variations in Halogen Geochemistry

Fractionation-corrected Cl, Br and I contents show significant along-ridge enrichments north of 61° N (Figs. 2e,g,i, S-7), and correlate positively with radiogenic isotopic signatures (Figs. 2a,b, S-8); incompatible trace element abundances; and ratios of similarly incompatible trace elements such as Nb/Zr (Figs. 2c; S-8). South of 61° N, halogens and Cl/K co-vary with Sr isotopes and Nb/Zr in a broad swell centred around 58.7° N (Fig. 2). This region includes two enriched seamounts, one of which has high halogen abundances (Fig. 2).

South of 61° N, the mean Cl/K = 0.13 ± 0.04 and mean Br/K = (0.31 ± 0.09) × 10^{-3} are higher than reported for global MORB (Kendrick, 2024) but overlap within 1σ; the mean I/K = (3.4 ± 0.7) × 10^{-6} lies towards the lower end of the global MORB range. Halogen/K ratios show statistically significant along-ridge increases between 57.4° N and 63° N (*e.g.*, Cl/K rises from ~0.1 to ~0.19; Fig. 2f), but halogen/halogen ratios remain constant (Fig. 2h,j). The near doubling of halogen abundances with respect to K cannot be explained by simple fractionation control. These along-ridge geochemical associations indicate that the high halogen budgets of basalts from ≥61° N are strongly controlled by the influence of an enriched upper mantle component that is inherently halogen-rich compared to local depleted mantle.

Reykjanes Ridge Halogen Ratios are Mantle-Like

Potassium, Cl, Br and I have similar partition coefficients during mantle melting (Kendrick *et al.*, 2012); thus, melt halogen/K and halogen/halogen ratios should reflect their mantle source. Reykjanes Ridge glasses show mantle-like Br/Cl and H_2O/Cl, and their I/Cl and K/Cl overlap global MORB and the lower bounds of mantle ranges (Fig. 3).

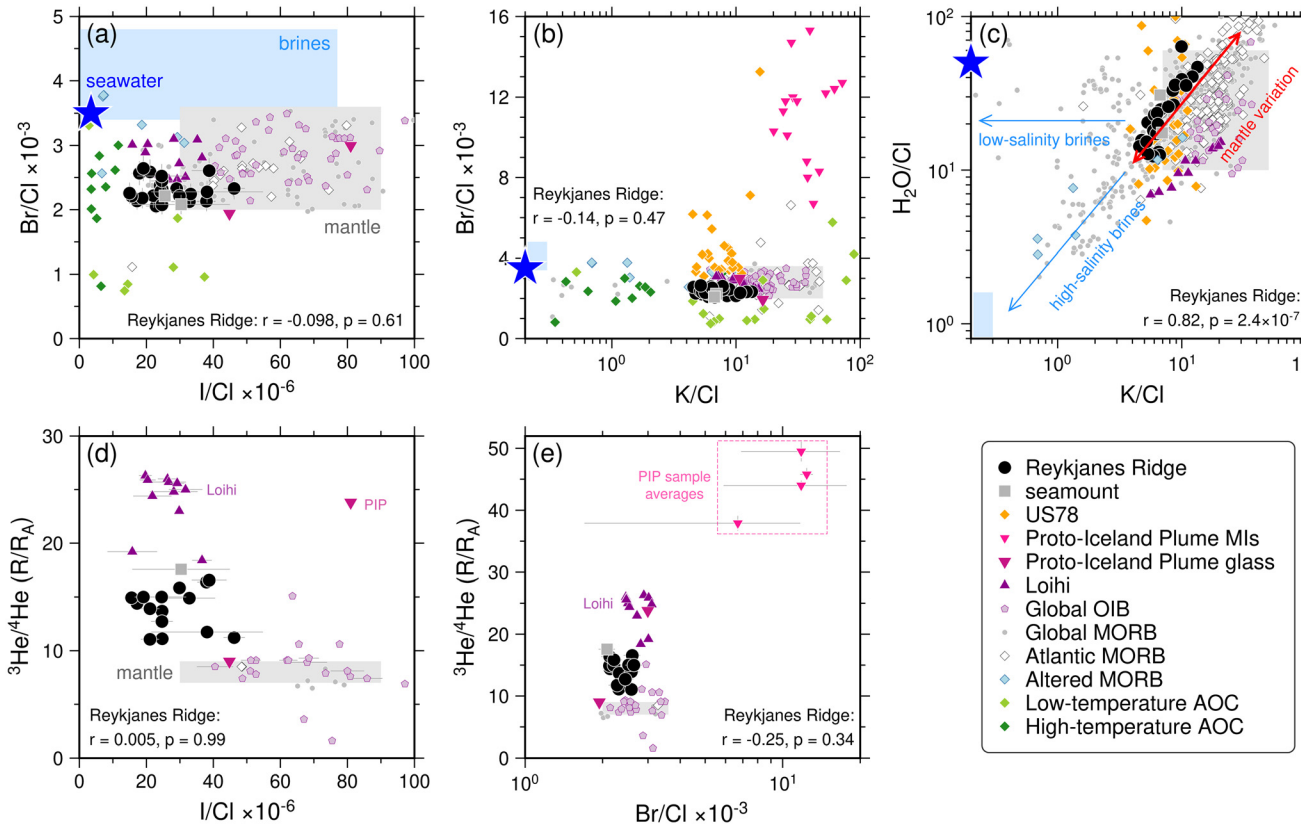


Figure 3 Compositions of Reykjanes Ridge glasses (this study) and whole rocks (US78; Unni and Schilling, 1978) compared with proto-Iceland plume melt inclusions (MI; Tomlinson *et al.*, 2025), glasses from Lō'ihi (Kendrick *et al.*, 2025), global MORB and OIB, and altered oceanic crust (AOC). Data sources listed in Table S-2.

High Cl/K in Reykjanes Ridge glasses has previously been interpreted as indicating seawater-derived Cl enrichment *via* assimilation of hydrothermally altered material (Michael and Cornell, 1998). High-temperature alteration of basalt and dolerite in the upper oceanic crust may lead to K and I leaching coupled with crystallisation of Cl-rich amphiboles, producing high bulk Cl/K and low I/Cl compared to fresh MORB (Chavrit *et al.*, 2016). However, Reykjanes Ridge Br/Cl, I/Cl and K/Cl are not correlated, and are not similar to altered oceanic crust (Fig. 3). Likewise, systematic covariation of halogens, Sr and Pb isotopes, and Nb/Zr (Fig. 2) indicates no role for progressive seawater assimilation with proximity to Iceland. We therefore interpret our Reykjanes Ridge halogen/halogen and halogen/K ratios as primary and mantle-derived. The high Cl/K suggests that the enriched upper mantle component sampled by the northern Reykjanes Ridge tends toward weak HIMU affinity (*e.g.*, Kendrick, 2024 and references therein).

Helium isotopes are decoupled from trace elements and halogen enrichments (Fig. 2) and show no relationship with halogen/halogen or halogen/K ratios (Figs. 3d,e, S-8 and S-9). Helium concentrations decrease due to pre-eruptive magmatic degassing as eruption depth shallows from ~2 km at 57.4° N to ~300 m at 63° N (Hilton *et al.*, 2000), but degassing is not expected to fractionate $^{3}He/^{4}He$. Lower $^{3}He/^{4}He$ in the northern ridge segment ≥61° N can be attributed to the addition of radiogenic crustal helium to already-degassed magmas (Hilton *et al.*, 2000), obscuring mantle-derived $^{3}He/^{4}He$ signatures. However, the absence of correlation between halogen abundances and $^{3}He/^{4}He$ persists even if samples from ≥61° N are excluded (Fig. S-8). Olivine-hosted melt inclusions in high-$^{3}He/^{4}He$ proto-Iceland plume (PIP) basalts from Baffin Island and West Greenland exhibit high Br/Cl (Tomlinson *et al.*, 2025), but this signature is absent from the moderate-$^{3}He/^{4}He$ Reykjanes Ridge glasses (Figs. 3e, S-8). Reykjanes Ridge glasses have low I/Cl similar to glasses from Lō'ihi (Kendrick *et al.*, 2025), but are unlike high-$^{3}He/^{4}He$ PIP glass which has high I/Cl (Fig. 3d). Taken together, these observations suggest that Iceland's high-$^{3}He/^{4}He$ mantle contributes minimally to the halogen budget of Reykjanes Ridge magmas, and consequently that low I/Cl is intrinsic to the Reykjanes Ridge mantle rather than associated with the high-$^{3}He/^{4}He$ component.

Halogen Abundances of Enriched and Depleted Primary Melts along the Reykjanes Ridge

Our data suggest that the halogen contents of Reykjanes Ridge basalts are relatively insensitive to the complex multi-component heterogeneity of the Iceland plume and can, to a first order, be modelled as binary mixtures of depleted and enriched endmember melts. This affords an opportunity to calculate a predicted halogen budget for Iceland's most enriched basalts. To do this, we take the average composition of Reykjanes Ridge basalts south of 61° N as the depleted melt endmember, and for the enriched endmember we select the onland Stapafell subglacial formation (Fig. S-1) because it continues the towards-Iceland geochemical enrichment trends set by the Reykjanes Ridge (Novella *et al.*, 2020). Stapafell basalts (8.3 wt. % MgO) represent some of the most enriched compositions in Iceland's neovolcanic rift zones: they have $^{3}He/^{4}He \approx 14\ R/R_A$ (Halldórsson *et al.*, 2016b) and high incompatible element contents, low $^{143}Nd/^{144}Nd \approx 0.5013$ and high $^{206}Pb/^{204}Pb \approx 18.9$

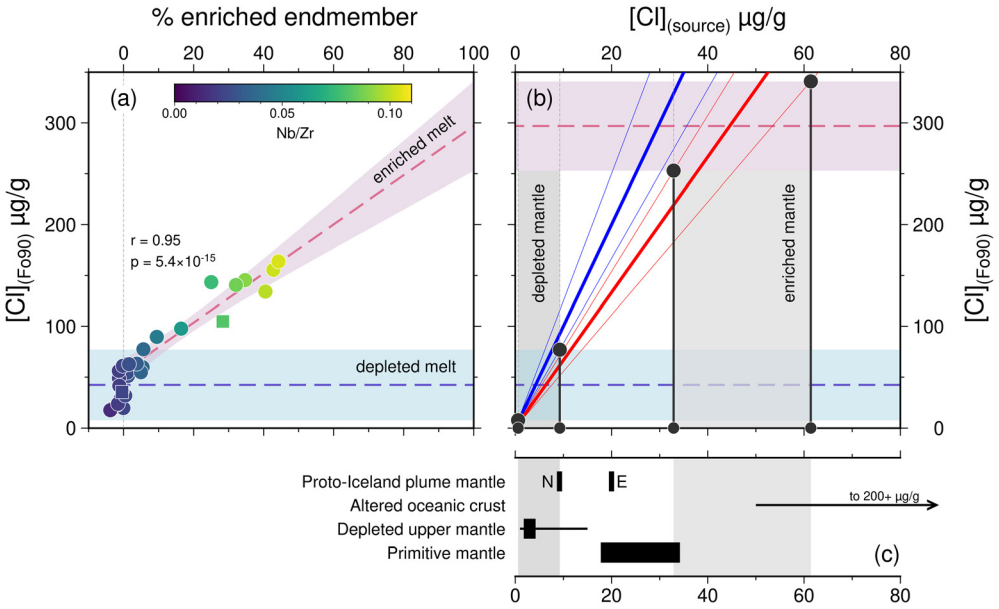


Figure 4 **(a)** The proportion of enriched melt contributing to Reykjanes Ridge basalts is regressed against the fractionation-corrected $[Cl]_{(Fo90)}$. Shaded area shows 95 % confidence interval on the best-fit regression. Right-most edge of the plot indicates the expected Cl content of enriched Icelandic basalts as represented by Stapafell. **(b)** An aggregate fractional melting model is used to link depleted and enriched primary melt compositions with plausible mantle source compositions, $[Cl]_{(source)}$. Blue lines show aggregate melt compositions for spinel lherzolite, calculated for melt fractions $0.08 < F < 0.12$ with $D_{Cl}^{bulk} = 0.0015$. Red lines show aggregate melt compositions for an enriched pyroxenite ($0.13 < F < 0.18$; $D_{Cl}^{bulk} = 0.0197$). Vertical grey bars mark viable mantle source compositions that, for the stated assumptions regarding F and D_{Cl}^{bulk}, could produce the depleted and enriched primary melt compositions. **(c)** Viable mantle source Cl contents are compared with published mantle compositions and altered oceanic crust. Data sources listed in Table S-7. Equivalent figures for Br and I are provided as Supplementary Information.

(Peate *et al.*, 2009) that could be interpreted as consistent with either very weak EM1 or HIMU affinity.

We use the incompatible trace element ratio Nb/Zr as a proxy for enrichment (Shorttle and Maclennan, 2011). Reykjanes Ridge basalts south of 61° N have mean Nb/Zr = 0.02 (Murton *et al.*, 2002); Stapafell basalts have Nb/Zr = 0.159 (Peate *et al.*, 2009). We use mass balance to constrain the mass fraction of enriched melt that must be added to ambient Reykjanes Ridge depleted melt to produce the observed Nb/Zr for each Reykjanes Ridge sample. Calculated contributions from the enriched endmember are then plotted against sample halogen contents that are fractionation-corrected to be in equilibrium with Fo_{90} mantle olivine (Fig. 4a), and best-fit regression is used to constrain the halogen content of the enriched primary melt. Depleted Reykjanes Ridge primary melts are constrained by our samples to contain 7.5–75 µg/g Cl (Fig. 4a), 55–140 ng/g Br and 0.75–1.35 ng/g I. Enriched Stapafell-like primary melts are projected to contain 215–340 µg/g Cl (Fig. 4a), 590–830 ng/g Br and 6.5–9.1 ng/g I.

Halogen Abundances of Enriched and Depleted Mantle Sources Supplying the Reykjanes Ridge

We next project the depleted and enriched primary melt compositions into plausible estimates of the halogen contents of their respective mantle sources. We constructed an aggregate fractional melting model to constrain the mantle lithology, composition and melting degree that best reproduce the trace element systematics of Reykjanes Ridge and Stapafell basalts (Supplementary Information). Halogens are incorporated into the melting model by calculating suitable partition coefficients based on mineral-melt partitioning experiments (Joachim *et al.*, 2015), and assuming that $D_{Cl}^{bulk} = D_{Br}^{bulk} = D_{I}^{bulk}$.

Trace element systematics of Reykjanes Ridge basalts south of 61° N are reproduced by 8–13 % melting of depleted spinel peridotite (Fig. S-10), consistent with previous estimates (Murton *et al.*, 2002). Taking this mantle lithology and range of melting degrees, and using our estimated halogen contents for depleted primary melts along the Reykjanes Ridge as input liquid compositions, we calculate that the depleted mantle sourcing these melts contains 1–10 µg/g Cl (Fig. 4b), 4–17 ng/g Br and 0.06–0.16 ng/g I; all are within published estimates for the upper mantle (Kendrick, 2024). Setting melting degree to $F = 0.1$ we obtain preferred $Br/Cl = 2.3 \times 10^{-3}$ and $I/Cl = 25 \times 10^{-6}$ (Table S-6).

Trace element systematics of enriched Stapafell-like magmas are best reproduced by a more complex melting model in which instantaneous melts from a Reykjanes Ridge-like depleted mantle mix with melts of a more enriched upper mantle component (Novella *et al.*, 2020) – a scenario similar to that proposed for the 2021 Fagradalsfjall eruption (Marshall *et al.*, 2024). To bracket plausible halogen contents for the enriched mantle, we require reasonable assumptions regarding its lithology, composition and melting degree. To calculate lower-bound halogen abundances we consider 13 % melting of primitive spinel pyroxenite, and for the upper bound we consider 18 % melting of primitive garnet pyroxenite. These lithologies and melting degrees closely match those chosen by Shorttle *et al.* (2015) as reasonable estimates given the requirement for melting model results to match along-ridge crustal thickness variations. Using these assumptions, we calculate that the enriched Icelandic mantle component, as represented by Stapafell, could contain 33–61 µg/g Cl (Fig. 4b), 77–149 ng/g Br and 0.8–1.6 ng/g I (Table S-6). These Cl and Br contents overlap upper estimates for primitive mantle

and lowermost estimates for altered oceanic crust (Figs. 4c, S-11c); the I content is lower than primitive mantle (7 ± 4 ng/g; Kendrick, 2024). Setting melting degree to $F = 0.15$ we obtain preferred Br/Cl = 2.4×10^{-3} and I/Cl = 26×10^{-6}, indistinguishable from depleted Reykjanes Ridge mantle. While our calculations cannot uniquely constrain the halogen budget of the enriched Icelandic mantle component, they illustrate the key parameters that must be considered in order to arrive at plausible estimates.

Despite the limitations of our modelling approach, our results strongly indicate that the enriched Icelandic mantle component is halogen-rich compared to depleted Reykjanes Ridge mantle. This enriched component is well matched by a refertilised pyroxenite containing up to 50 % recycled subducted oceanic crust, *i.e.* the KG1 lithology (Shorttle and Maclennan, 2011). Thus, we propose that the high halogen inventory of enriched Icelandic melts can be attributed to recycled oceanic crust in the mantle source. This recycled component is constrained to be as young as Phanerozoic in age (Halldórsson *et al.*, 2016b, and references therein). Strong positive correlations between halogens, radiogenic isotopes and the proportion of enriched melt (Figs. 3, S-8) reveal that high halogen abundances are characteristic fingerprints of recycled oceanic crust. In contrast, enriched Icelandic and depleted Reykjanes Ridge mantle have near-identical Br/Cl and I/Cl, despite their differing halogen contents. This is consistent with halogens being minimally fractionated from one another during subduction processing and/or in the upper mantle during mixing and dilution of subducted materials with halogen-poor depleted peridotite over geological time (*e.g.*, Kendrick, 2024). Our preferred I/Cl lies slightly below mantle estimates, suggesting that at least some global-scale heterogeneity of halogen ratios is retained in the upper mantle and recovered at the surface in erupted basalt compositions.

Conclusions and Implications

We have constrained the halogen budget of an enriched upper mantle component that contributes significantly to trace element, radiogenic isotope and halogen enrichment in Reykjanes Ridge basalts approaching Iceland. The halogen signature of Iceland's high-^{3}He/^{4}He plume source is not resolved in our data, suggesting that Iceland's high-^{3}He/^{4}He mantle may be halogen-poor, similar to the Cl- and strongly I-depleted Lō'ihi source (Kendrick *et al.*, 2025), and that low I/Cl is intrinsic to local Reykjanes Ridge mantle. The I/Cl signature of Iceland's high-^{3}He/^{4}He plume remains to be characterised, leaving open the possibility that halogen depletions and low I/Cl are characteristic of global high-^{3}He/^{4}He mantle sources.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2640.

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