# $^{186}$Os and $^{187}$Os enrichments and high-$^{3}$He/$^{4}$He sources in the Earth's mantle: evidence from Icelandic picrites

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## Abstract

Picrites from the neovolcanic zones in Iceland display a range in $^{187}Os/^{188}Os$ from 0.1297 to 0.1381 ($\gamma_{Os}$ = 0.0 to 6.5) and uniform $^{186}Os/^{188}Os$ of 0.1198375±32 (2σ). The value for $^{186}Os/^{188}Os$ is within uncertainty of the present-day value for the primitive upper mantle of 0.1198398±16. These Os isotope systematics are best explained by ancient recycled crust or melt enrichment in the mantle source region. If so, then the coupled enrichments displayed in $^{186}Os/^{188}Os$ and $^{187}Os/^{188}Os$ from lavas of other plume systems must result from an independent process, the most viable candidate at present remains core-mantle interaction. While some plumes with high $^{3}He/^{4}He$, such as Hawaii, appear to have been subjected to detectable addition of Os (and possibly He) from the outer core, others such as Iceland do not.

A positive correlation between $^{187}Os/^{188}Os$ and $^{3}He/^{4}He$ from 9.6 to 19 $R_A$ in Iceland picrites is best modeled as mixtures of 500 Ma or older ancient recycled crust mixed with primitive mantle, creating a hybrid source region that subsequently mixes with the convecting MORB mantle during ascent and melting. This multistage mechanism to explain these isotope systematics is consistent with ancient recycled crust juxtaposed with more primitive, relatively He-rich mantle, in convective isolation from the upper mantle, most likely in the lowermost mantle. This is inconsistent with models that propose random mixing between heterogeneities in the convecting upper mantle as a mechanism to explain the observed isotopic variation in oceanic lavas or models that produce a high $^{3}He/^{4}He$ signature in melt depleted and strongly outgassed, He-poor mantle. Instead these systematics require a deep mantle source to explain the $^{3}He/^{4}He$ signature in Iceland lavas. The $^{3}He/^{4}He$ of lavas derived from the Iceland plume changed over time, from a maximum of 50 $R_A$ at 60 Ma, to approximately 25-27 $R_A$ at present. The changes are coupled with distinct compositional gaps between the different aged lavas when $^{3}He/^{4}He$ is plotted versus various geochemical parameters such as $^{143}Nd/^{144}Nd$ and La/Sm. These relationships can be interpreted as an increase in the proportion of ancient recycled crust in the upwelling plume over this time period.

## 1. Introduction

The $^{187}$Re-$^{187}$Os and $^{190}$Pt-$^{186}$Os isotopic systems have been valuable for tracing depths of origins of plumes and for constraining sources for mantle-derived magmas (Walker et al., l995; l997; Brandon et al., l998; l999; 2003; Puchtel et al., 2004; 2005; Brandon and Walker, 2005). The isotope $^{187}$Re decays via beta emission to $^{187}$Os with $\lambda = 1.67E^{-11}yr^{-1}$, and $^{190}$Pt decays via alpha emission to $^{186}$Os with $\lambda = 1.48E^{-12}yr^{-1}$ (Smoliar et al., 1996; Begemann et al., 2001). Because $^{190}$Pt is a minor isotope of Pt in combination with the small decay constant, this system is very insensitive to perturbations by most geochemical processes. Long-term and relatively large fractionations of Pt from Os are required to produce measurable differences in $^{186}$Os/$^{188}$Os ratios. The outer core may have developed higher Pt/Os and Re/Os relative to chondrites and the Earth's mantle, owing to the crystallization of the inner core and leading to elevated $^{186}$Os/$^{188}$Os and $^{187}$Os/$^{188}$Os over time (Morgan et al., l995; Walker et al., l995). This is based on the assumption that these elements partition between solid metal and liquid metal in the Earth’s core in a similar manner to their partitioning behavior in asteroidal cores where $D_{Os} > D_{Re} > D_{Pt}$ (D = solid metal/liquid metal bulk distribution coefficient). Recent experimental evidence on solid metal/liquid metal partitioning of Re, Os and Pt supports the notion that the outer core evolves with elevated Re/Os and Pt/Os (Lauer and Jones, l998; Walker, 2000; Chabot and Jones, 2003). Coupled enrichments in $^{186}$Os/$^{188}$Os and $^{187}$Os/$^{188}$Os have been found for some mantle-derived materials from three plume systems, the 251 Ma Siberian Traps, 89 Ma Gorgona Island, and present-day Hawaiian lavas , consistent with a small addition (~0.5 Wt.%) of outer core material into localized portions of the sources for these plumes (Brandon et al., l998; l999; 2003; Brandon and Walker, 2005).

The Hawaiian system in particular has provided important constraints on the origins of coupled $^{186}$Os/$^{188}$Os and $^{187}$Os/$^{188}$Os enrichments in some plume-derived magmas potentially resulting from core-mantle interaction and related to deep mantle sources. Elevated Fe/Mn ratios for the same samples measured for $^{186}$Os/$^{188}$Os and $^{187}$Os/$^{188}$Os is consistent with addition of Fe metal into the source of the Hawaiian plume (Humayun et al., 2004). Alternatively, the incorporation of 10 to 20% mafic materials such as pyroxenite, or 1 to 3% Fe-Mn-rich sediment in the Hawaiian plume system could explain these Fe and Os isotope data (Schersten et al., 2004; Baker and Jensen, 2004; Smith, 2003). Thallium isotopes for these same samples indicate that admixture of ~20 ppm Fe-Mn sediments with the ambient Hawaiian mantle may have occurred (Nielsen et al., 2006). These proportions of Fe-Mn sediments are several orders of magnitude too low to explain the elevated $^{186}Os/^{188}Os$ in some of the Hawaiian samples, and preclude them as a source for the coupled Os isotope enrichments (Brandon et al., 2003; Nielsen et al., 2006; Brandon and Walker, 2005). However, the issue of ancient mafic material, or other sediments that may be present in ancient recycled slabs, contributing to the coupled Os isotope enrichments remains unresolved. Brandon et al. (l998; 1999; 2003) showed that such materials had Pt/Re ratios that were ≤ 15, too low to evolve over time to be the high $^{186}Os/^{188}Os$ and $^{187}Os/^{188}Os$ end-member needed to explain the linear mixing arrays for the Gorgonan, Siberian, and Hawaiian data. The Pt/Re ratios of the radiogenic Os isotope end-member must be ≥ 80 to be consistent with the known geochemical behavior of Pt and Re during fractionation of solid metal leaving an evolved liquid, that could be in the outer core. Nevertheless, samples from additional plume systems need to be measured where geochemical evidence for ancient mafic and crustal materials in their mantle sources has been well established, in order to provide a test for the effects such material may have on the Os isotope budgets of plume systems.

Combined studies of Os and He isotopes have the potential to further unravel the source contributions to oceanic basalts and the origins of high $^{3}He/^{4}He$ in their mantle sources. A positive correlation between $^{186}Os/^{188}Os$ and $^{3}He/^{4}He$ previously observed in Hawaiian picritic lavas is consistent with a deep mantle origin of the coupled $^{187}Os$-$^{186}Os$ isotope signatures, and provides a potential link between Os and noble gas compositions in plume systems (Brandon et al., l999). One possible source for primordial noble gas may be the outer core (Porcelli and Halliday, 2001). If this is the case, then samples from other high $^{3}He/^{4}He$ plume systems should also show correlated enrichments in $^{186}Os/^{188}Os$ and $^{187}Os/^{188}Os$. Alternatively, the high $^{3}He/^{4}He$ signature could result from partial outgassing of a lower mantle source that was originally primordial with respect to noble gases, but which was convectively isolated from the upper mantle during later stages of Earth history (Class and Goldstein, 2005; Ballentine et al., 2005). Such a high $^{3}He/^{4}He$ source may or may not produce a correlation with Os isotopes in plume-derived magmas, because the extent of correlation would depend on mixing between different domains within the plume source and during partial melting in the upper mantle. In another case, if high $^{3}He/^{4}He$ results for retention of He relative to U and Th during partial melting early in Earth history (Meibom and Anderson, 2004; Meibom et al., 2005), a negative correlation between $^{3}He/^{4}He$ and $^{187}Os/^{188}Os$ could be expected in the ambient upper mantle because of a corresponding lowering of Re/Os in the peridotite residue. This does not appear to be manifested in the sources for the Hawaiian picrites, but may be present in other magmatic systems. Finally, no correlation would be expected between He and Os isotopes if the source of coupled enrichments in $^{186}$Os/$^{188}$Os and $^{187}$Os/$^{188}$Os is from ancient mafic or crustal materials.

To provide additional tests for these issues, Os and He isotopic relationships are examined here for a suite of picrites that represent the range of geochemical variability from the neovolcanic zones in Iceland. These data are then compared with those from older magmatic rocks of the Iceland plume system in order to assess possible changes over time. The Iceland plume system is ideal for this examination because of the well-established high $^3$He/$^4$He and the near-solar Ne isotopic compositions in many Iceland lavas. These noble gas characteristics likely require a deep mantle source that is distinct from the convecting upper mantle source for MORB (Kurz et al., 1982; 1985; Graham et al., 1998; Harrison et al., 1999; Hilton et al., 1999; Breddam et al., 2000; Dixon et al., 2000; Moreira and Sarda, 2000; Moreira et al., 2001; Stuart et al., 2003; Graham, 2005; Ballentine et al., 2005; Macpherson et al., 2005). In addition, there is evidence in some Icelandic lavas, from both noble gases and lithophile elements, for varying contributions from the upper mantle MORB source as well as from several ancient crustal and/or mafic components within the mantle (Hemond et al., 1993; Hanan and Schilling, 1997; Fitton et al., 1997; Taylor et al., 1997; Stecher et al., 1999; Hanan et al., 2000; Breddam et al., 2000; Chauvel and Hemond, 2000; Kempton et al., 2000; Thirlwall et al., 2004; Macpherson et al., 2005; Thirlwall et al., 2006; Kokfelt et al., 2006). These relationships are consistent with melting and mixing that accompanies the physical setting of the present-day Iceland plume beneath the spreading Mid-Atlantic Ridge system. This locality thereby provides a unique opportunity for systematic study of Os and He source contributions from both plume and convecting MORB mantle sources.

## 2. Analytical Techniques

Bulk-rock powders were analyzed by x-ray fluorescence for major and minor elements at the Geochemical Laboratories at McGill University on a Phillips PW2440 4kW automated XRF spectrometer system with a Rhodium 60kV end window x-ray tube (Table 1, Appendix 1). The analyses were done on fused beads prepared from ignited samples. Analytical procedures are listed elsewhere (http://www.eps.mcgill.ca/~geochem/XRF_analytical_procedure2005.html).

Bulk-rock powders were analyzed for trace elements at the University of Tasmania using a inductively coupled plasma mass spectrometer. Calibration was done with the USGS BHVO-1 standard, with other USGS rock standards run as quality control. Relevant data for this study are reported as trace element ratios in Table 1, and the remainder will be reported elsewhere.

The Nd isotopic analyses (Table 1) were carried out on a bulk rare earth element cut collected from conventional cation exchange columns. Analyses were run on the Axiom MC-ICPMS at the Danish Lithosphere Center (DLC) using methods similar to those described by Luais et al. (1997). The $^{143}Nd/^{144}Nd$ for the LaJolla standard was 0.511843±10 (n=2) and for the DLC/AMES standard was 0.512132±11 ($2\sigma$, n=23) during the analytical campaign where the Iceland picrites were analyzed.

Details of Re-Os analytical procedures have been previously documented (Brandon et al., l999; 2000; 2003; Brandon et al., 2005a; 2005b) and only a brief description is provided here. Rhenium and osmium concentration data (Table 2) were obtained on 2 gram aliquots via isotope dilution. The sample powders were combined with a multi Re-PGE (platinum group element) spike and loaded into Carius tubes with aqua regia and heated for 48 hours at 250°C. This insured sample-spike equilibrium for Os. Osmium was extracted from the aqua regia with $CCl_4$, and then back extracted into HBr. Osmium was then purified by microdistillation using chromic acid as an oxidant and HBr as a trap. The aqua regia was divided in half. One half was processed for PGE isotope dilution measurements. These data have been reported elsewhere (Humayun et al., 2002). The other half was processed for Re using anion exchange chemistry. Procedural blanks ran with the samples were 3.3±0.5 pg for Os, with $^{187}Os/^{188}Os = 0.175\pm0.005$, and 46±10 pg for Re. Blank corrections applied to Os concentrations were 0.12 to 2.0%, and to Re concentrations were 3 to 12%. The corrections for blank contribution on the $^{187}Os/^{188}Os$ ratio ranged from 0.00007 to 0.0009, depending on concentration of Os in each sample.

The spiked aliquots for Re and Os were measured on the Thermo Finnigan Triton thermal ionization mass spectrometer in negative ion mode (NTIMS) with the exception of data for sample DMF 9101. The latter data was obtained at the University of Maryland Isotope Geochemistry Laboratory in l999 using NTIMS and following procedures described elsewhere (Brandon et al., l999). The Os cuts were loaded on to ultra-pure Pt filaments, with $Ba(OH)_2$. The spiked Os aliquots were measured on the SEM as $OsO_3^-$ ions, in dynamic, peak-hopping mode. Ultra-pure, dry, oxygen pressure in the source was maintained at 1-3 x $10^{-7}$ mbar for all measurements. Oxygen corrections were made using the oxygen isotopic composition measured on 2-ng loads of $ReO_4^-$ on the Faraday cups. Instrumental mass fractionation corrections were applied using $^{192}$Os/$^{188}$Os = 3.083 and the exponential law. Four replicates of the Johnson Matthey Os standard during the analytical campaign gave $^{187}$Os/$^{188}$Os = 0.11393±0.00026 (2σ), within the range of accepted values (e.g. 0.1138067±21, Brandon et al., l999) and those for the high-precision, static Faraday cup runs described below. The Re cuts were loaded onto high purity Ni filaments with $Ba(NO)_3$ and measured in static mode on the faraday cups. Rhenium instrumental mass fractionation was estimated to be negligible based on the data for the in-house Re standard ($^{187}$Re/$^{185}$Re = 1.6733±19, 2σ). The concentration data for Re and Os, and $^{187}$Os/$^{188}$Os for the spiked runs are reported in Table 1.

The unspiked aliquots were processed following the procedures described in Brandon et al. (l999; 2000; 2003) and briefly reported here. The goal was to obtain from 100 to 200 ng of Os to analyze for each measured. This required 100 to 1000 grams of sample powder, depending on the Os concentration. The Ni-S fire assay technique was employed to purify Os. The NiS blebs containing concentrated Os are dissolved in 12N HCl, and insoluble Os sulfide is trapped on cellulose paper by filtering the HCl solution. The Os-bearing cellulose is dissolved in a Carius tube using reverse aqua regia, and Os is purified using the procedures listed above for the spiked samples.

High-precision Os isotope data for unspiked aliquots of DMF9101 was obtained via NTIMS at the University of Maryland following the measurement protocol outlined previously (Brandon et al., l998; l999; 2000, 2003). The remainder of the high-precision Os isotope data were obtained by NTIMS on the Thermo Finnigan Triton (Table 2). The data were obtained in static mode using 7 Faraday collectors. Signal intensities of 120-180 mV on mass 234 ($^{186}Os^{16}O_3^-$) and 235 ($^{187}Os^{16}O_3^-$) were generated for ≥180 ratios to reach run precision of ±0.0000026 or better ($2\sigma_{mean}$) for the $^{186}$Os/$^{188}$Os ratio. Each cycle had an integration time of 17s followed by a 4s settling time. The isobaric interference of $^{186}W^{16}O_3^-$ on $^{186}Os^{16}O_3^-$ was monitored by measuring $^{184}Os^{16}O_3^-$ ($^{184}W^{16}O_3^-$). This interference has not been observed on this Triton during Os measurements which is consistent with the very different ionization behaviors of W and Os negative oxide species. (Brandon et al; 2003; Puchtel et al., 2004, 2005; Walker et al, 2005; Brandon et al. 2005a, 2005b). Oxygen corrections were made using the oxygen isotopic composition obtained for 2 ng loads of $ReO_4$ on the Faraday cups as listed above. Only one bottle of oxygen was used during the analytical campaign that included all samples listed in Table 1. Repeated measurements for the oxygen isotopic composition using Re indicated no change over time. The oxygen pressure for all runs was maintained in the range of 2-3x$10^{-7}$ mbar. After oxygen corrections were made on the raw data, instrumental mass fractionation corrections were performed using the exponential law and $^{192}Os/^{188}Os = 3.083$. The mean of 6 runs of the Johnson-Matthey Os standard during the analytical campaign was 0.0013136±38 for $^{184}Os/^{188}Os$, 0.1198518±23 for $^{186}Os/^{188}Os$ and 0.1137999±64 for $^{187}Os/^{188}Os$ (±2σ). The average for $^{186}Os/^{188}Os$ on the Johnson-Matthey standard during this analytical campaign was 49 ppm higher than previously measured at the University of Maryland and on this Triton of $^{186}Os/^{188}Os = 0.1198475$ (Walker et al., 1997; Brandon et al., 1998; Brandon et al., 1999; Brandon et al., 2000; Brandon et al, 2003; Puchtel et al., 2004; 2005; Walker et al, 2005; Brandon et al., 2005a; 2005b). Accordingly, for direct comparison to earlier data, the Iceland picrite and basalt data in Table 1 were corrected to the $^{186}Os/^{188}Os$ ratio of 0.1198475 of the Johnson Matthey standard obtained in the earlier studies.

For He analyses, details of the procedures are presented elsewhere (Graham et al., 1998). Olivine and glass separates were analyzed for He trapped in melt/fluid inclusions by in vacuo crushing using approximately 200 high impact strokes. The reported 2σ errors (Table 4) include in-run analytical uncertainties, plus those associated with air standards and blank corrections.

## 3. Results

The Iceland samples studied here are primarily picrites with olivine phenocrysts. The samples come from widely dispersed locales within the neovolcanic zones (Table 1, Figure 1), ranging from Theistareykir and Hruthalsar in the Eastern Rift Zone (ERZ), to Skridufell in the Southern Extensional Zone (SEZ), and 13 locales in the Western Extensional Zone (WEZ). The MgO contents range from 9.5% wt.% to 28.5% wt.% (Table 1). The Mg#'s (calculated with all Fe as FeO) range from 0.61 to 0.86. The samples have from 4 to 32 vol.% olivine phenocrysts. The samples range from light rare earth element (LREE) depleted with $(La/Sm)_n$ of 0.31 to enriched with $(La/Sm)_n$ of 1.82 (normalized to CI chondrites, Anders and Grevesse, 1989). The $(Th/Sm)_n$ ratios also range from depleted with 0.11 to enriched with 1.82 (Table 1). All of the samples have ΔNb values of > 0, consistent with their occurrence from Iceland and distinct from samples erupted in surrounding regions that have ΔNb values of < 0 (Fitton et al., 1997). The Nd isotopic compositions range from 0.512999 to 0.513147 ($\varepsilon_{Nd}$ from 7.0 to 9.9), typical for neovolcanic zone Iceland lavas (Taylor et al., l997; Kempton et al., 2000; Thirlwall et al, 2004).

Samples ICE 5 and DMF 9101 were obtained during separate field campaigns from the same quarry at Dagmalafell, Midfell. Samples ICE 4a and 4b are from adjacent outcrops at Maelifell, while ICE 8a and 8b are from the same flow at Skrudufell. The different samples from each locale display differing major element compositions relative to each other as a result of variable amounts of olivine phenocrysts and potentially other fractionating phases, that were the result of differentiation subsequent to extraction from the mantle (e.g. MgO and Mg#, Table 1; Brandon et al., 2001). The reproducibilities of lithophile trace element ratios and isotopic data for these duplicate samples from the same locales show that these compositional parameters are independent of shallow level processes that may affect these magmas, such as crustal assimilation and concomitant or subsequent crystal fractionation (Tables 1-3).

Concentrations of Re and Os for the Iceland picrites and basalts analyzed in this study are comparable to those for other picrite suites such as Hawaii. Bennett et al. (l996) and Brandon et al. (l999) reported a range of Re 181 to 1093 ppt, and Os concentrations from 465 to 1083 ppt, for a suite of Hawaiian picrites. The Iceland samples studied here have Re from 152 to 712 ppt and Os from 63 to 1065 ppt (Table 2), similar to the range of Os concentrations previously reported for 9 Iceland picrites (Skovgaard et al., 2001). Osmium concentrations correlate inversely with $^{187}$Re/$^{188}$Os but scatter with $^{187}$Os/$^{188}$Os (Figure 2). The $^{187}$Re/$^{188}$Os ratios range from 0.7 to 44.9, and show a rough correlation with decreasing MgO (Figure 2; and Mg#, not shown). The $^{187}$Os/$^{188}$Os ratios range from 0.1297 to 0.1381 and scatter with MgO (Figure 3). These relationships are consistent with the compatible and incompatible behavior of Os and Re, respectively, during fractionation and olivine accumulation. The lack of correlation of $^{187}$Os/$^{188}$Os with any differentiation indices implies that the Os isotopic composition of these magmas was set prior to any residence in the Icelandic crust and thus cannot be a function of crustal assimilation. This is consistent with additional evidence presented in the preceeding paragraph and below, that the geochemical features of these lavas reflect their mantle source regions.

High precision Os isotopic measurements for picrites and basalts from the Neovolcanic zones in Iceland, ranging from southern locales in the Rekjyanes Peninsula in the WEZ to one sample in the ERZ in the Theistareykir area are listed in Table 2. The $^{186}$Os/$^{188}$Os ratios range from 0.1198350±48 (2σ) for DMF 9101 measured at UMd, and from 0.1198359±21 to 0.1198410±17 for the 13 samples measured on the Triton. This range of values is slightly greater than the external precision based on replicate measurements of the standard, which for this study, was ±0.0000023 (2σ).

Helium concentrations and $^{3}$He/$^{4}$He ratios for olivine and glass from the samples measured in this study are reported in Table 3. He isotope data are reported relative to the atmospheric $^{3}$He/$^{4}$He ratio of $1.39 \times 10^{-6}$ ($R_A$), using the $R/R_A$ notation. In addition, He data published elsewhere, for samples that come from the same outcrops or lava flows as those in the present study, are also listed. Samples ICE 5 and DMF 9101 come from a quarry at Dagmalafell, Midfell, and have $^{3}$He/$^{4}$He values of 16.4±0.51 and 17.20±0.09 $R_A$ for olivine and glass, respectively (Table 1). These data compare well with values of 16.5 and 17.5 $R_A$ measured in glass samples from this quarry and reported elsewhere (Harrison et al. l999).

For the samples measured in this study, $^{3}$He/$^{4}$He ranges from 9.6 $R_A$ for ICE 0 in the ERZ, to 17.2 $R_A$ for ICE 8b in the SEZ. The Eldborg basalt flow studied here has a previously reported $^{3}$He/$^{4}$He = 19±2 $R_A$ (Dixon et al., 2003). Published data for Iceland lavas within the neovolcanic zones have $^{3}$He/$^{4}$He ranging from 8.0 to 21 $R_A$ (Breddam et al., 2000; Macpherson et al., 2005, and references therein), with one glass sample having a value as high as 26 $R_A$ (Kurz et al., l985) . A single sample containing both clinopyroxene and olivine shows values of 20.2±10 and 34.3±8 $R_A$ (2σ, Macpherson et al., 2005), but given the low He concentrations and large analytical uncertainties, the significance of the high $^{3}$He/$^{4}$He in this sample is unclear. Hence, the samples in this study have $^{3}$He/$^{4}$He that spans the range reported previously for samples from the neovolcanic zones in Iceland, with the exception of two samples having $^{3}$He/$^{4}$He > 21 $R_A$, as noted.

## 4. Discussion

### *4.1 Mantle Sources for Os*

The average $^{186}$Os/$^{188}$Os for the 13 Iceland samples measured on the Triton is 0.1198375±32 (2σ). This value is within uncertainty of the estimated present-day primitive mantle (PM) value for $^{186}$Os/$^{188}$Os of 0.1198398±16 (2σ), recently established by Brandon et al.

(2006) on new measurements of chondrites, komatiites, and mantle-derived Os-rich alloys. The $^{187}Os/^{188}Os$ ratios for the Iceland samples in this study range from 0.1297 to 0.1381. This range extends from the present-day estimated value for PM of 0.1296±0.0008 (Meisel et al., 2001) to more radiogenic values. These Os isotope systematics for the Iceland picrites are in contrast with data for other Phanerozoic plume systems, namely the 251 Ma Siberian, 89 Ma Gorgonan, and the recent Hawaiian systems, where samples showing coupled enrichments in $^{186}Os/^{188}Os$ and γOs (where $\gamma Os = [(^{187}Os/^{188}Os_{sample@t}/^{187}Os/^{188}Os_{chondrite@t} - 1) * 100]$, present-day $^{187}Os/^{188}Os_{chondrite} = 0.1270$, and $^{187}Re/^{188}Os_{chondrite} = 0.4083$, Shirey and Walker, l998) relative to PM (Figure 4). Hence, the coupled radiogenic $^{186}Os/^{188}Os$ and $^{187}Os/^{188}Os$ component that is manifested in the samples from the other 3 plume systems is absent in the Iceland samples.

The enrichment in $^{187}Os/^{188}Os$ with no variation in $^{186}Os/^{188}Os$ relative to a present day chondritic upper mantle has important implications for the origins of Os isotopic variation in mantle-derived materials. Unlike the samples from Siberia, Gorgona, and Hawaii that require a radiogenic end-member with long-term superchondritic Pt/Os, Re/Os, and Pt/Re, the Iceland picrites show that a radiogenic end-member with long-term superchondritic Re/Os is present in their source, but no long-term superchondritic Pt/Os or Pt/Re is manifested in the $^{186}Os/^{188}Os$ ratios. For example, ancient basaltic components that were formed at 2 and 1 Ga from a primitive upper mantle, evolve over time with shallow vectors in $^{186}Os/^{188}Os$ versus $^{187}Os/^{188}Os$, as a result of strong Re enrichment with only minor Pt enrichment (Figure 4). Such a radiogenic component can explain not only the $^{187}Os/^{188}Os$ in the Iceland picrite data, but also that needed for other mantle-derived materials that display Re-enrichments in their sources. Other types of crustal materials thought to be present in ancient subducted slabs plot with similar shallow vectors in this diagram (Brandon et al. 1999). This is consistent with observations on Os-rich alloys found in mantle samples in ophiolites associated with subduction zone settings (Brandon et al., 2006). Given the revised estimate for the PUM for $^{186}Os/^{188}Os$ of 0.1198398±16, a source having long-term Re-enrichment with no Pt-enrichment is also necessary to explain the Hawaiian end-member represented by Mauna Kea lavas (with $^{186}Os/^{188}Os = 0.119835$ and $\gamma Os = 2$) as well as for the Koolau end-member (with $^{186}Os/^{188}Os = 0.119835$ to 0.119840 and $\gamma Os \geq 8$) (Brandon et al. 1999; Figure 4). Trace element ratios, and lithophile isotopic compositions for the Kea compositional end-member indicate that its source underwent melt depletion in the past, followed by an enrichment overprint consistent with the constraints from Os isotopes in Figure 4 (Hauri, l996; Hoffman and Jochum, l996; Eiler et al., l996).

These Os isotope systematics for the Iceland picrites are consistent with additional geochemical evidence for ancient recycled slab components in the mantle sources for Iceland magmas (Hemond et al., l993; Hanan and Schilling, l997; Taylor et al., l997; Stecher et al., l999; Hanan et al., 2000; Chauvel and Hemond, 2000; Kempton et al., 2000; Thirlwall et al., 2004; Macpherson et al., 2005; Thirlwall et al., 2006). Ancient melt enrichment processes and/or additions of recycled crust to the mantle source generate radiogenic $^{187}$Os/$^{188}$Os in lavas and mantle peridotites with no resolvable effects on $^{186}$Os/$^{188}$Os (Figure 4). The absence of $^{186}$Os/$^{188}$Os enrichments in Iceland is, in turn, strong evidence that some other process produces the coupled enrichments of $^{186}$Os/$^{188}$Os and $^{187}$Os/$^{188}$Os observed at other mantle plume localities. The observation that these coupled Os isotope enrichments occur independently of both melt enrichment processes or the presence of ancient recycled crust in their mantle sources is further support for an origin from core-mantle interaction. These new data for the Iceland picrites suggest that core-mantle interaction is a localized process and unlikely to be prevalent in all plume source regions (Brandon et al., 2003; Brandon and Walker, 2005). While some plumes with high $^{3}$He/$^{4}$He, such as Hawaii, appear to have been subjected to detectable addition of Os from the outer core, others such as Iceland do not.

### *4.2 Origins of Os-He variation and the role of ancient recycled lithosphere*

One of the most interesting geochemical features of this suite of Iceland picrites is the strong positive correlation of $^{187}$Os/$^{188}$Os with $^{3}$He/$^{4}$He (Figure 5A). This correlation ranges from $^{187}$Os/$^{188}$Os = 0.1297 and $^{3}$He/$^{4}$He = 9.6 $R_A$, typical for upper mantle material, to $^{187}$Os/$^{188}$Os ~ 0.1381 and $^{3}$He/$^{4}$He ~ 19 $R_A$. This correlation defies standard explanation because as shown above, the radiogenic $^{187}$Os/$^{188}$Os indicates a source with a high time-integrated Re/Os ratio, typical of ancient recycled crust or melt-enriched lithospheric mantle. The presence of such material in the mantle source should also not lead to high $^{3}$He/$^{4}$He a priori. Recent experiments for olivine/melt pairs suggest that He may sometimes behave as a slightly more compatible element than U or Th during partial melting (Parman et al. 2005), but the relative behaviors are only crudely understood and it seems just as likely, if not more so, that He behaves overall more incompatibly that U and Th during partial melting of peridotite or pyroxenite. The high

$^{187}Os/^{188}Os$ lavas in Iceland also have superchondritic La/Sm (Figure 5B) and Th/Sm, with chondritic Th/La (Figure 6) consistent with a lithophile trace element-enriched source. The Pb isotope variation for Iceland lavas from the neovolcanic zones are consistent with superchondritic time-integrated U/Sm and Th/Sm (Hanan and Schilling, l997; Taylor et al., l997; Chauvel and Hemond, 2000; Hanan et al., 2000; Kempton et al., 2000; Thirlwall et al., 2004; 2006; Kokfelt et al., 2006). This should lead to $^{4}He$ accumulation in the source resulting from decay of $^{238}U$, $^{235}U$, and $^{232}Th$, with consequent low $^{3}He/^{4}He$ that is in contrast to the high $^{3}He/^{4}He$ observed for the lavas that have the strongest signals for enrichment (Figure 5, 6). In addition to the correlation between $^{187}Os/^{188}Os$, $^{3}He/^{4}He$ and the trace element ratios observed here, the high $^{3}He/^{4}He$ source for neovolcanic zone Iceland lavas may also have a low $\delta^{18}O$ relative to MORB mantle (Thirlwall et al., 2006).

In the following sections, models are formulated, based on the Os-He isotope systematics, to account for these features. The goal is to reproduce the positive trend between $^{187}Os/^{188}Os$ - $^{3}He/^{4}He$ with realistic source parameters that are input to a mixing model (Table 4). In the first step, a hybrid mantle source is constructed as an end-member composition having elevated $^{3}He/^{4}He \geq 25\ R_A$ (the maximum value established for Iceland neovolcanic zone lavas) and $^{187}Os/^{188}Os \geq 0.1381$ (the maximum value measured in this study, Table 2) that represents the component with high $^{3}He/^{4}He$ and $^{187}Os/^{188}Os$ (Figure 5A). The approach taken is to mix ancient recycled crust with hypothetical primitive mantle compositions as the most straightforward mechanism. The primitive mantle end-member is strictly defined here only with respect to He and Os systematics, and has no quantifiable implications to other elemental systems. One of the key parameters considered is to assess the possible age of ancient recycled material. In order to account for the Pb isotopic systematics of Iceland neovolcanic zone lavas that trend with a negative $\Delta^{207}Pb$ (deviation of $^{207}Pb/^{204}Pb$ for a given $^{206}Pb/^{204}Pb$ relative to the Northern Hemisphere Reference Line), Thirlwall et al. (2004) suggested that such material in the Iceland mantle would form with a higher U/Pb ratio relative to the upper mantle MORB source at a time as young as 170 Ma ago. In contrast, Kokfelt et al. (2006) suggests that recycled lithosphere of ~1.5 Ga can account for the enriched Pb isotopic signatures of neovolcanic zone lavas. Hence, constraining the age of the recycled material or time of melt-enrichment to account for geochemical features of Iceland neovolcanic zone lavas is crucial for any viable model. The parameters chosen for the models are constrained in the following two sections, followed by evaluation of the models in the third section.

### *4.2.1. He Constraints*

Values of $^3He/^4He$ for the high $R_A$ Icelandic mantle source range from the maximum $^3He/^4He$ measured in mantle-derived volcanic rocks from the Iceland plume at 60 Ma ($^3He/^4He$ = 50 $R_A$, Stuart et al., 2003), to a lower value of 37 $R_A$, based on a measurement of a Neogene lava from Iceland (Hilton et al., l999). In order to minimize the amount of ancient recycled crust necessary to create a hybrid component having 25-33 $R_A$, the lower value of 37 $R_A$ is used in the models presented in the following sections. This approach is taken because constraining the minimum amount of crust necessary at any given time of formation will evaluate $^{187}Os/^{188}Os$ - $^3He/^4He$ in the context of having a relatively young recycled component of several hundred Ma and possibly as low as 170 Ma as noted above (Thirlwall et al., 2004). The He concentrations for a source with 37 $R_A$ are modeled for two end-member scenarios. In the first scenario, estimates of a strongly outgassed, initially primordial He source follows Class and Goldstein (2005), where this source has an initial $^3He$ = 3.54 x$10^{-13}$ mol/g and a corresponding $^4He$ of 1.11x$10^{-9}$ mol/g at 230 $R_A$. This equates to 24.8 μcc(STP)/g He. If this source remained closed for He over earth history, it would evolve from 230 $R_A$ at 4.4 Ga to 95 $R_A$ at present as a result of U and Th decay producing $^4He$. Partial melting of this primordial source over earth history to create continental crust and complementary depleted mantle will result in loss of U and Th and outgassing of He. In the model of Class and Goldstein (2005) used here, a 37 $R_A$ source from such a depleted mantle would have been isolated at 415 Ma with a strongly reduced He concentration of 0.402 μcc(STP)/g (Depleted Plume Source (DPS), Table 4). This source will evolve to present-day composition of the depleted mantle with 8 $R_A$ (Class and Goldstein, 2005). In a second scenario, a primordial source evolves from 4.4 Ga with 230 $R_A$ to 37 $R_A$ at present, with no fractionation between U, Th, and He. For example, a source with 230 $R_A$ at 4.4 Ga and bulk silicate Earth U and Th/U of 21 ppb and 4.05, respectively, would require an initial amount of $^3He$ of 9.58x$10^{-14}$ mol/g and a corresponding $^4He$ content of 3.00x$10^{-10}$ mol/g (6.72 μcc(STP/g) to evolve to 37 $R_A$ at present and having 41.7 μcc(STP)/g (Primitive Mantle (PM), Table 4). This source had an initial inventory of He that is 60% less than the estimate of a primordial source of Class and Goldstein (2005) but because of largely reduced outgassing of He and removal of U and Th, evolves to the present to contain a factor of 100 more times He in a present-day source when compared to the depleted mantle model source with 37 $R_A$ of Class and Goldstein (2005). While there are enormous uncertainties associated with both model 37 $R_A$ sources and hence the He concentrations are considered here as time-integrated averaged approximations, these two end-member scenarios, having significantly different He concentrations resulting from strong outgassing and melt depletion in one case, and less outgassing and melt depletion in a second case, serve to constrain possible origins of the high $R_A$ signature in the Iceland lavas. Does the high $R_A$ reflect isolation in the past from an evolving depleted mantle, presumably in the upper portions of the mantle, or does it reflect a source that is closer to primordial, possibly deeper in the mantle, and hence has much larger concentrations of He? The outcomes of the models presented below will address these questions.

The MORB mantle source is estimated to contain 1.2-4.6 x $10^9$ atoms $^3$He/g (Pepin and Porcelli, 2002; Graham, 2002; and references therein), which equates to [He] = 4 to 15.4 μcc(STP)/g at $^3$He/$^4$He= 8 $R_A$. The depleted mantle outgassing model of Class and Goldstein (2005), results in a source with 8.15 $R_A$ and 0.27 μcc(STP)/g at present and reflects large outgassing of the depleted mantle source than the high He concentration estimates. These are considered within the models presented below.

The He concentrations for ancient recycled crustal components depend on a number of factors, including the amount of degassing at or near the time of formation, the concentrations of U and Th, and time for $^4$He ingrowth. This is complicated further by the potential loss of He, U, and Th during dehydration and heating of subducted slabs under volcanic arcs (Dodson and Brandon, l999; Becker et al., 2000; Plank et al., 2002; Kelley et al., 2005; Kessel et al., 2005). The basaltic portions of fresh oceanic crust have U and Th concentrations ranging from 50 to 120 ppb, and 70 to 190 ppb, respectively, with a Th/U of approximately 3, while altered basaltic crust is estimated to have U and Th concentrations of 300 and 187 ppb, respectively (Hofmann, l988; Wedephol and Hartmann, l994; Staudigel et al, l996; Hart et al. l999). The U and Th concentrations for dehydrated eclogites (T>900°C) have been estimated to be 117 and 168 ppb, representing approximately 60% and 10% removal, respectively, of these two elements during subduction processes from altered basaltic portions of slabs (Becker et al., 2000). Hence, for calculating $^4$He production over time in basaltic portions of slabs, a U concentration of 100 ppb and Th/U of 3 is used, that approximates the average of both unaltered and dehydrated lithologies. The calculations are performed for 0, 50, and 90% initial outgassing of He, and an initial $^3He/^4He$ of 8 $R_A$.

Ocean sediments have U and Th concentrations ranging from ~0.05 to 7 and ~0.07 to 18 ppm, respectively, with widely varying Th/U from 0.2 to 10 (Klinkhammer and Palmer, l99l; Plank and Langmuir, l998; Plank et al., 2002). These concentrations are a function of the amount of detrital and Fe-Mn hydrogenous components (Th), and redox state (U), in the sediment (Plank and Langmuir, l998), and greatly vary in any given location and individual layer of sediment. It is not possible to uniquely quantify $^4He$ ingrowth over time for these wide variations. Instead, the global subducting sediment (GLOSS) average of 6.91 ppm Th and 1.68 ppm U, with Th/U = 4.1 (Plank and Langmuir, l998) is considered here to represent the effects of $^4He$ accumulation in ancient subducted sediment. Dehydration and heating during subduction is thought to remove some portion of U and Th from sediments from the slab although the exact amounts of removal continue to be debated (Elliot et al., 1997; Kelley et al., 2005; Kessel et al., 2005). In the models presented here, the effects of 50% and 90% removal of U and Th are considered. It is also assumed that all of the He in sediment, irrespective of a meteoritic or endogenous origin, is outgassed during subduction (Hiyagon, l994; Porcelli and Ballentine, 2002), such that the $^3He/^4He$ of the residual slab sediment is initially very low - a value of 0.01 $R_A$ is assumed here because it adequately approximates the radiogenic $^3He/^4He$ production ratio.

Using the parameters for He concentrations and $^3He/^4He$ defined above, the effects of $^4He$ production and mixing of ancient recycled crust into a plume source having different He concentrations can be evaluated. For recycled crust of about 0.5 Ga and older, initial He degassing has only a small effect on the amount of $^4He$ in the basaltic units (Figure 7). The effect of initial He degassing producing large differences in $^4He$ concentration and $^3He/^4He$ becomes more significant for recycled basaltic material younger than 0.5 Ga. In the case of mixing with a relatively undegassed plume, the amount of recycled basaltic material mixed with a plume source of 25 to 27 $R_A$ (the maximum measured in Iceland lavas from the neovolcanic zones and the minimum needed to generate the hybrid component) can range from as little as 25% for 2 Ga slab to as much as 80% for 0.25 Ga slabs (Figure 8A) depending on the extent of degassing of the recycled material. Because of the generally larger concentrations of U and Th in sediments, they can contribute proportionally larger amounts of per unit mass compared to recycled basalt, except where ≥90% of the U and Th are removed under arcs during dehydration and heating, or if the sediment is initially poor in these elements relative to GLOSS (Figure 7). This in turn results in correspondingly less sediment necessary to create a hybrid component with 25 to 27 $R_A$, except for a young slab (i.e. 0.25 Ga) that has lost 90% or more of its U and Th during subduction (Figure 8B). In the case of mixing of ancient recycled basalt and sediment into a plume source with 37 $R_A$ that represents strong outgassing and much lower He concentrations than a source that has undergone much less degassing, much less than 5% of ancient recycled crust material is necessary to create a hybrid component with 25-27 $R_A$ (Figure 8). The implications of these mixing requirements for a hybrid component will be further considered in the context of combined Os-He isotope systematics below.

### *4.2.2. Os Constraints*

The Os concentrations and isotopic compositions for present-day PM are well constrained and are used for the primitive mantle component present in the plume (Table 4; Meisel et al., 2001; Morgan et al., 2001; Brandon et al., 2006). The DPS component represents a source with a slightly lower long-term Re/Os resulting from melt removal and loss of slightly incompatible Re relative to Os in the convecting mantle, consistent with the outgassing He model of Class and Goldstein (2005). The isotopic compositions for variably aged basalt and sediment components used in the models are based on mafic granulites and eclogites, representing dehydrated mafic slab material in subduction zones (Becker, 2000), and average reduced ocean floor sediment (Ravizza and Pyle, 1997; Brandon et al., 1999). These are evolved to present-day values using the average published Re and Os concentrations (Becker, 2000; Ravizza and Pyle, 1997; Brandon et al., 1999). Platinum, Re and Os are likely removed from slabs during subduction processes (Brandon et al., 1996; Brandon et al., 1999; McInnes et al., 1999; Becker, 2000; Becker et al. 2001; Sun et al., 2004; Becker et al., 2004). Therefore, the amount of ancient recycled crust of different ages required in a mixture with PM to create a hybrid component will increase proportionally to the amount of removal of Re and Os. In the case of the basalt portion of the slab, the concentrations of mafic granulites and eclogites are considered to be those following removal during subduction processes. In the case of sediments in slabs, the effects of subduction zone processing on these elements is at present unknown. The composition of reduced sediments were chosen because of their relatively large concentrations of Re and Os (Table 4) compared to other ocean sediments, such that very small amounts of this material produce a strong increase in $^{187}Os/^{188}Os$ when added to PM to create the hybrid component.

The $^{187}Os/^{188}Os$ isotopic composition of the MORB upper mantle has been postulated to range from the present-day PM composition of 0.1296 (Meisel et al., 2001; Walker et al., 2002) to that having a slightly depleted long-term Re/Os relative to PM with $^{187}Os/^{188}Os$ ~0.125 (Snow and Reisberg 1995; Brandon et al., 2000). As such, a MORB mantle encompassing both of these scenarios is considered in the models. Although ancient recycled crust can have present-day strongly radiogenic $^{186}Os/^{188}Os$, the amounts of such materials necessary to explain the $^{187}Os/^{188}Os$ of the Iceland picrites do not result in detectable increases in $^{186}Os/^{188}Os$ in the mixing models (Table 4). This is because of the low Pt/Re in crustal materials relative to chondrites and PUM (Brandon et al., l999; 2003; Brandon and Walker, 2005). These observations are consistent with the lack of detectable elevation of $^{186}Os/^{188}Os$ in the picrites relative to upper mantle materials as discussed earlier (Figure 4).

### *4.2.3. Os-He Isotope Models*

The results of the models for mixing of primitive mantle (PM) and a depleted plume source (DPS) components both having 37 $R_A$ (Table 4), with ancient recycled crust (ARC) are displayed in Figure 9. The initial aim of this modeling is to produce a hybrid source having $^3He/^4He$ of $\geq 25$ $R_A$ and $^{187}Os/^{188}Os \geq 0.1381$, the maximum values for the Iceland neovolcanic zones lavas. The range of compositions for the hybrid source, when mixed with an upper mantle source having $^3He/^4He$ of $\leq 9$ $R_A$ and $^{187}Os/^{188}Os \leq 0.1297$, should produce compositions that fall within the shaded area that extends beyond the upper end of the positive correlation of the Iceland picrites (Figure 9). The He compositions used for the crustal end-members in these models are for a basalt that has undergone 50% outgassing during subduction, and for sediment that has 50% U and Th contents relative to GLOSS as a result of loss during subduction processes.

Mixing curves between DPS and 3 different ARC compositions, consisting of 2 Ga and 250 Ma sediment, and a 2 Ga slab with 95 basalt:5 sediment, are shown (Figure 9). Because of the low concentrations of He in DPS compared to the different aged recycled materials (Table 4), the mixing curves show rapid decreases from 37 $R_A$ (DPS) with little change in $^{187}Os/^{188}Os$ until $^3He/^4He$ values of 10 $R_A$ or less are reached. These results are independent of the age or composition of the ancient recycled material. Hence, hypothetical compositions resulting from long-term protracted outgassing and melt removal over earth history to create a depleted mantle source with high $^{3}$He/$^{4}$He and consequent low He concentrations (i.e. Class and Goldstein, 2005) cannot be used to mix with ancient recycled materials to explain the coupled enrichments in $^{187}$Os/$^{188}$Os and $^{3}$He/$^{4}$He observed in the Iceland picrites. The implications from this to the source of high $R_A$ in the Iceland plume are considered in more detail below.

Mixing curves between PM with much greater He concentrations than DPS (Table 4) and 3 different ARC compositions, consisting of basalt, sediment, and 95 basalt:5 sediment, for 2 Ga and 250 Ma crust are shown (Figure 9). The shapes of the curves for different aged ARC of different compositions are generally the same. This is because the amounts of $^{4}$He and $^{187}$Os produced, which control the concentrations of He and Os in this crust, are proportional to the age of the crust having the same (U+Th)/$^{3}$He and Re/Os ratios (Table 4). Hence, the curves are independent of the age. This conclusion also holds for other crustal materials unless the concentrations of U and Th, which affect the $^{4}$He production rate, and Re/Os, are strongly different. In each case however, the mixing curves employing younger crust require a greater amount of that crust to fall within the shaded region that represents a potentially viable hybrid end-member.

Because of the very low concentration of Os in basalt crust relative to the primitive mantle (Table 4), the mixing curves between these two end-members are hyperbolic (Figure 9). A minimum of 70% and 30% of 250 Ma and 2 Ga basalt crust, respectively, is required to mix with PM to obtain values within the shaded region (Figure 9). Mixtures between sediment and primitive mantle produce near-vertical curves (Figure 9). This is because of the very high $^{187}$Os/$^{188}$Os of the ancient sediment (Table 4). At least 5% of 250 Ma, and < 5% of 2 Ga sediment, is required to mix with PM to produce hybrid source regions with He-Os isotopic systematics necessary to be the end-member on the high side of the picrite array (Figure 9). These amounts would decrease proportionally if the sediment did not lose U and Th during subduction. These mixing scenarios indicate that only very small amounts of sediment are necessary to mix with PM to create a viable hybrid component, even in the case of U and Th loss during subduction. Composite crustal sources (for example, consisting of 95% basalt and 5% sediment) mixed with primitive mantle result in intermediate curves relative to those for 100%

basalt or sediment and corresponding intermediate values for the amount required to produce a viable hybrid source - ≥ 45% of a 250 Ma ARC, and ≥ 8% of 2 Ga ARC (Figure 9).

All of these hybrid sources, where PM is mixed with variable amounts of basalt and sediment of different ages, could be potential end-members that when mixed with MORB mantle could produce the positive correlation between $^{187}$Os/$^{188}$Os and $^{3}$He/$^{4}$He. However, for all ARC compositions, younger ages require large amounts of this component. Also, for a basalt ARC of any age, the amounts necessary are significantly higher compared to those where a small amount of sediment is involved. This is because of the larger Re/Os, Os and U and Th concentrations of sediment relative to basalt that will produce a correspondingly greater $^{187}$Os/$^{188}$Os and $^{4}$He concentration (Figure 7, Table 4). Hence, for the mixing models between a hybrid end-member and the MORB mantle, ARC compositions of 95 basalt:5 sediment were used in order to minimize the amount of crust necessary in the hybrid component to produce the $^{187}$Os/$^{188}$Os and $^{3}$He/$^{4}$He correlation. Except for the case where ≥90% of the U and Th were lost from the sediment during subduction, or when the sediment is less than about 500 Ma (Figure 7), any larger proportion of sediment in the mix results in a hybrid component that will have excessively high He concentrations requiring correspondingly high He concentrations in the MORB mantle (see below). Any lesser amount of sediment in ARC will result in very large quantities of basalt in the hybrid component.

Mixing lines between MORB mantle having variable $^{3}$He contents (and constant $^{3}$He/$^{4}$He = 8 $R_A$) and hybrid Iceland plume mantle sources are shown in Figure 10. The MORB mantle was modeled as having a He concentration of 10 μcc(STP)/g (‘Normal He’), which is an intermediate value to various estimates noted earlier (Pepin and Porcelli, 2002; Graham, 2002; and references therein), and one with a He concentration ~3 times higher than the maximum estimates with 50 μcc(STP)/g (STP)/g (‘High He’). The MORB mantle was modeled with $^{187}$Os/$^{188}$Os of 0.1296 (‘M1’) identical to PUM, and one with $^{187}$Os/$^{188}$Os of 0.125 (‘M2’) similar to the estimate from abyssal peridotites (Snow and Reisberg, 1995; Brandon et al., 2000). These compositions were mixed with two different hybrid plume components having 250 Ma and 2 Ga ARC. In one case, the hybrid component has a present-day $^{187}$Os/$^{188}$Os of 0.1435, with 30.5 and 31.3 $R_A$, for 250 Ma (‘H1’) and 2 Ga (‘H3’) ARC added in, respectively (Table 4). In the other case, the hybrid component has $^{187}$Os/$^{188}$Os of 0.1610, with 25.1 and 26.3 $R_A$, for 250 Ma (‘H2’) and 2 Ga ('H4') ARC added in, respectively. These compositions span the range of viable solutions for the hybrid component within the defined parameter space outlined earlier.

Several conclusions can be drawn from the mixing lines generated between these end-members (Figure 10.). First, using a depleted (MORB source) mantle $^{187}Os/^{188}Os$ of 0.125 (M2) cannot produce mixing lines that follow the positive correlation between $^{187}Os/^{188}Os$ and $^{3}He/^{4}He$. Instead, they fall to the right of the data when mixing with H1 (or H3), and cross-cut the data for mixtures with H2 (or H4) (Figure 10C,D). The only trajectories using M2 that match the Iceland picrite correlation are via mixing with either H1 or H3. In order to generate such mixing lines, the He concentration of the MORB mantle must be increased to values well in excess of those estimated, or the He concentration of the hybrid component much be decreased by an order of magnitude. Hence, unless new He concentration estimates are proposed for these sources that can accommodate these changes, it is unlikely that a depleted MORB mantle with $^{187}Os/^{188}Os$ significantly less (i.e. ≥3%) than the PUM value of 0.1296 can be an end-member for the Iceland picrites. Decreasing the He concentration in the MORB mantle to the value obtained by Class and Goldstein (2005), of 0.27 μcc(STP)/g does not provide a solution. This is because such a source should have lost He contemporaneous with Re and consequent reduction in long-term Re/Os during partial melting, and should therefore result in a mixing line that is even more hyperbolic and concave to the right than when mixing with the 'Normal He' MORB mantle end-member.

Second, mixing lines that match the positive correlation between $^{187}Os/^{188}Os$ and $^{3}He/^{4}He$ for the Iceland picrites are those generated between a less depleted MORB mantle for both Re/Os ratio and He concentration. The trend can be generated using M1 with a He concentration of ~3 times those of the MORB mantle estimates ('High He' end-member) and H1 (or H3) or with a 'Normal He' concentration in the MORB mantle and H2 (or H4). As in the case for mixing between depleted MORB mantle (M2), significantly lower or higher He concentrations than those estimated for the MORB mantle will produce mixing lines that have greater concavity or are convex, respectively, and do not match the trend. Hence, taking reasonable parameters for ARC (of variable age), for primitive mantle, and for MORB mantle with no discernable depletion in long-term Re/Os (i.e. lower $^{187}Os/^{188}Os$ than PUM), and with He concentrations within the range of those estimated for the MORB mantle reservoir, can produce the trend for the Iceland picrites via three-component mixing.

Third, for the mixing lines that match the trend for the Iceland picrites, greater proportions of younger crust than older crust are necessary. For example, it is possible to estimate the amount of crust of a given age that is required for the mixture of MORB mantle plus primitive mantle to generate the maximum $^{187}Os/^{188}Os$ of 0.1381 in the Iceland picrites (Table 2) (Figure 11). In the case of a wholly basalt crust, the amount necessary within this mixture would range from ~30% for 2 Ga ARC, to ~82% for 250 Ma ARC in the mix. Such large amounts of crust present in the mantle during partial melting are unlikely to produce primitive high MgO melts that are parental to picrites (Norman and Garcia, l999). When a small amount of sediment is present in the mix in the proportion of 95% basalt:5% sediment, the amount of crust required is significantly reduced, to 7% for 2 Ga ARC and to ~37% for 250 Ma ARC (Figure 11). Younger ARC requires even more added into the mix and becomes unviable from a petrologic standpoint. Hence, if the enrichments observed in the Iceland picrites result from having ARC in the mantle source, then these models show that the Os-He isotope relationships are not consistent with recycled crust as young as 170Ma as previously proposed (Thirlwall et al., 2004). These enrichments become most easily generated from a mantle source with 500 Ma or older ARC, consistent with the constraints from Pb isotopic compositions of Iceland neovolcanic zone lavas modeled by Kokfelt et al. (2006), where less than 20% of a crust composed largely of basalt plus a small amount of sediment is present (Figure 11). Such proportions of crust in the mantle source of oceanic island basalts, or mixing proportions between melts derived from such sources, is consistent with a wide spectrum of geochemical data (e.g. Lassiter and Hauri, 1998; Kogiso et al., 2004).

Fourth, an alternative to having ARC in the source of the Iceland magmas is melt enrichment in the past, whereby Re, U, and Th were enriched. This leads to increased production of $^{187}Os$ and $^{4}He$ and a source that evolves away from primitive mantle Os and He isotopic compositions to those estimated for the hybrid components depicted in Figures 9 and 10. One advantage to this approach over adding ARC to primitive mantle, is that for a young trace element enrichment event (where it is assumed that the primary changes are in trace elements) the source remains peridotitic and therefore high MgO magmas are petrologically realistic products of partial melting. Hence, such a mechanism may be a solution to generating the trace element-enrichment signature between 500 and 170 Ma predicted for the Pb isotopic compositions of neovolcanic zone lavas (Thirlwall et al., 2004).

If the calculated $^{187}Os/^{188}Os$, $^{3}He/^{4}He$, Re/Os, Th/U, and U and He concentrations for hybrid sources 1 and 2 at 250 Ma (Table 4) are applicable to mixing between melt-enriched sources and plume mantle, then the relative amounts of these sources necessary to create the hybrid end-member effectively remains the same as calculated in the recycling model. Such parameters may be relevant for the case of cryptically metasomatized harzburgitic lithosphere, where small degree partial melts under mid-ocean ridges infiltrate the oceanic lithosphere and either result in cryptic metasomatism or deposition of veins of pyroxene-rich cumulates within the depleted harzburgitic layer (e.g., Niu et al. 2002). The small degree melts likely have very high amounts of incompatible elements when compared to ocean floor basalt. When this lithosphere is subducted, Niu et al. (2002) note that melt veins or the cryptically metasomatized harzburgitic lower (colder) portions of the oceanic lithosphere would not be affected by subuduction zone processes, and could thus be preserved for recycling and ultimate entrainment into upwelling plumes.

For pyroxenite veins produced at 250 Ma, if these have Re/Os, U, and Th similar to the basalt estimates in Table 4, then unreasonably high amounts are necessary in the hyrbid component (≥70%), making it difficult to produce high-MgO parental magmas to the picrites. However, if the veins are more-enriched in Re/Os, U, and Th, then the amount of material necessary would scale down accordingly. Thirlwall et al. (2004) calculated that in order to get a constant $\Delta^{207}Pb$ of -2.2 from $^{206}Pb/^{204}Pb$ of 18.2 to 19.2 displayed by the neovolcanic zone lavas, the averaged $\mu$ ($^{238}U/^{204}Pb$) of the source was increased at 250 Ma to about 20. If this source had a primitive mantle $\mu$ of 8-9 (McDonough and Sun l995), then the enrichment resulted in an increase in $\mu$ to about 28-30. High U/Pb ratios are predicted to result from small degree melt infiltration and entrapment because $D_U<D_{Pb}$ during partial melting (Niu et al., 2002). Hence, melt-enriched lithosphere in the plume source may be favored over basaltic ocean crust for generating the Os-He relationships of the Iceland picrites if the recyled material is geologically young. Additional constraints are required in to order to further test the viability of such a model.

In summary, these models demonstrate that the mechanisms that could produce the Os-He systematics for the Iceland picrites are twofold. First, a hybrid mantle must exist from the juxtaposition of 500 Ma or older recycled crust and primitive mantle, or from melts derived from these sources. This hybrid mantle has radiogenic $^{187}Os/^{188}Os$ relative to depleted MORB or primitive mantle, but elevated $^{3}$He/$^{4}$He relative to depleted MORB mantle. The relationships presented above indicate that mixing models incorporating a range of reasonable parameters for these materials can produce such a hybrid mantle source. Importantly, the most favorable models to explain Os-He systematics are those where both mafic material and sediment from an ancient recycled slab are present in the Iceland mantle source. This model is particularly attractive for Proterozoic-aged slabs, but for younger slabs, the amount of material necessary to admix with plume mantle becomes large and petrologically difficult to reconcile with the generation of high MgO parental magmas to picrites. If the trace-element enriched material in the plume is younger, then models involving melt-enrichment of recycled oceanic lithosphere are a more likely explanation. Second, this hybrid source must be physically separated from the depleted MORB upper mantle in order to maintain its distinct geochemical characteristics over time, and when mixed with MORB mantle, produces a quasi-linear array for $^{187}$Os/$^{188}$Os-$^{3}$He/$^{4}$He as observed in the Iceland picrites (Figure 5A).

### *4.3 He isotope evidence for a compositionally evolving Iceland plume over 60 Ma?*

The origin of the high $^{3}$He/$^{4}$He signature observed in oceanic lavas is under considerable debate. Most proposed models for high $^{3}$He/$^{4}$He range from an undegassed primordial source to a largely outgassed source that has been isolated relative to the convecting upper mantle during the later stages Earth history (Class and Goldstein, 2005 and references therein). This $^{3}$He source may be the outer core (Porcelli and Halliday, 2001) or the lower mantle (Kurz et al., 1982). Alternative models propose that isolated materials of high $^{3}$He/$^{4}$He are present throughout the convecting upper mantle (Anderson, 2000; Meibom and Anderson, 2004; Meibom et al., 2005; Coltice and Ricard, 2002). These materials could be strongly melt depleted mantle blobs on a sub-kilometer scale that consist primarily of olivine (Meibom et al., 2005). A similar explanation involves harzburgitic ancient subcontinental lithosphere residing under cratons. Both of these materials would have lost U and Th through melt depletion in the past resulting in less $^{4}$He ingrowth relative to the MORB (upper mantle) source. Helium has a moderately high solubility in olivine; melt-depleted olivine-rich mantle might preserve a high $^{3}$He/$^{4}$He signal if He behaves more compatibly than U and Th during partial melting (Graham et al., 1990; Parman et al., 2005).

The data for lavas from the Iceland magmatic system help constrain the origin of the high $^3He/^4He$ source in the context of such proposed models. The high $^3He/^4He$ component for Iceland's neovolcanic zone lavas does not appear to be associated with the MORB source mantle. As shown above, in order to explain the Os-He isotope systematics, the source of high $^3He/^4He$ first mixes with mantle that has either undergone melt enrichment in the past or contains ancient recycled crust to create a hybrid source. Only after such a process has occurred, can the hybrid source then mix with the MORB mantle source. These conditions are inconsistent with models where all observed geochemical heterogeneities are juxtaposed in the upper mantle (i.e. a statistical upper mantle assemblage - SUMA, Meibom and Anderson, 2004; Meibom et al., 2005). Given the widely variable parameters of Os and He concentrations and isotopic compositions in mantle and crustal materials, SUMA-type models instead predict a wider array of scattering that is not observed, rather than the well-defined array for the Iceland picrite data (Figure 5A). The high $^3He/^4He$ source therefore cannot have a direct physical relationship with the convecting MORB mantle over extended timescales and must exist in a region where recycled lithosphere/crustal material can be mixed in without contribution from the MORB mantle source. A similar inference was made based on the negative correlation of low $\delta^{18}O$ with high $^3He/^4He$ observed in lavas from the neovocalanic zones in Iceland and the Reykjanes ridge (Thirlwall et al., 2006). Further support that the geochemical variability of Iceland magmatism reflects an origin from a deep mantle plume may be inferred from the primordial Ne isotopic compositions in neovolcanic zone lavas (Dixon et al., 2000; Moreira et al., 2001). Finally, the source of the high $^3He/^4He$ in the hybrid component cannot be the result of a largely outgassed (i.e. factor of $\geq$100 reduction in He concentration, DPS) source evolving into depleted mantle to present day that was isolated only several hundred Ma in the past (i.e. Class and Goldstein, 2005). Such a source with very small He concentrations cannot viably mix with ancient recycled crust of any age to produce the high $^{187}Os/^{188}Os$-high $^3He/^4He$ hybrid component necessary to explain the Iceland picrite data (Figure 9). This is because of the rapid ingrowth of $^4He$ in crustal materials. Instead, the high $^3He/^4He$ (PM) source evolved to the present-day must initially contain at 4.4 Ga about a factor of 4 less He (6.72 μcc(STP/g) than the primordial high $^3He/^4He$ source (24.8 μcc(STP)/g) used by Class and Goldstein (2005) in order explain viable hybrid end-member compositions. These arguments appear to eliminate models for volcanism in Iceland solely involving the upper mantle or lithosphere with no involvement of a deep, convectively isolated lower mantle source (Anderson, 2000; Foulger and Pearson, 2001; Meibom and Anderson, 2004, Foulger et al., 2005; Meibom et al., 2005). Whether the high $^{3}He/^{4}He$ originates from the lowermost mantle or from the core is uncertain. Coupled enrichments for $^{186}Os/^{188}Os$ and $^{187}Os/^{188}Os$ correlated with $^{3}He/^{4}He$ in Hawaiian picrites (Brandon et al. 1999) could be explained by primordial He derived from the core (e.g. Porcelli and Halliday, 2001). However, the Iceland picrites in this study show no enrichments in $^{186}Os/^{188}Os$ and do no support an origin of primordial He from the core unless the mechanisms that impart outer core Os and He signatures to the lowermost mantle are not always coupled. Additional work is obviously required to further explore the origin of the deep high $^{3}He/^{4}He$ source.

A high $^{3}He/^{4}He$ source has been identified in the Iceland magmatic system since 60 Ma. A late Tertiary lava from Selardalur (≤ 14.9 Ma) in NW Iceland has $^{3}He/^{4}He = 37\ R_A$, 61 Ma picrites from the Vaigat Formation in West Greenland have $^{3}He/^{4}He$ of up to 31 $R_A$, and 61 Ma picrites from Baffin Island that are correlative with those from West Greenland have $^{3}He/^{4}He$ of up to 49.5 $R_A$ (Graham et al., l998; Hilton et al., l999; Stuart et al., 2003). East Greenland picrites erupted from the Iceland plume at 58 Ma have $^{3}He/^{4}He$ of up to 20 $R_A$, but likely have been lowered via crustal contamination (Marty et al., l998).

The high $^{3}He/^{4}He$ in these older lavas is not reflected to the same degree in the neovolcanic zones of Iceland. This is exemplified by the Iceland picrites in this study that have distinct isotope and trace element characteristics relative to those for the earlier lavas (Figures 12,13). For example the combined $^{3}He/^{4}He$, $\varepsilon_{Nd}$ (i.e. $^{143}Nd/^{144}Nd$ normalized to chondrite uniform reservoir - CHUR), ΔNb, and $(La/Sm)_n$ systematics for these lavas show that a strong compositional gap exists between the samples with high $^{3}He/^{4}He$ in the older lavas and the neovolcanic zone lavas. The Nd-He isotope systematics in particular, show different relationships for the older Iceland plume lavas compared to lavas from the neovolcanic zones (Figure 10A). The older lavas have experienced some mixing between Precambrian Greenland crust and a mantle plume component of high $^{3}He/^{4}He$ (Marty et al., l998). These observations, combined with the fact that all neovolcanic zone lavas have $^{3}He/^{4}He$ <27 $R_A$, suggest that a fundamental change occurred in the character of the Iceland plume source during the past 60 million years. The $^{3}He/^{4}He$ of ~50 $R_A$ in the older picrites with high $\varepsilon_{Nd}$ and subchondritic $(La/Sm)_n$ suggests a source more depleted in U+Th and light rare earth elements. At the onset of plume head impingement under Greenland, recycled crust/lithosphere components were therefore either not present in significant amounts, or were not effectivley sampled within the upwelling plume. In more recent times and with further upwelling, recycled lithosphere entrained within the plume was more prevalent, as reflected by the mixture of the hybrid component plus MORB mantle that produces the Os-He isotope systematics of the Iceland picrites. Therefore, the distinct high $^{3}$He/$^{4}$He compositions of the different aged Iceland magmatic rocks are interpreted here as a manifestation of the same, deep-mantle source of He, and the observed compositional variations are related to dynamics and melting of the plume system over time.

## Conclusions

The Os and He isotopes of Iceland picrites provide important insights into the Iceland plume system from 60 Ma to present. High-precision $^{186}$Os/$^{188}$Os-$^{187}$Os/$^{188}$Os data for these picrites do not display coupled enrichments observed in other suites including Hawaiian picrites, Gorgona komatiites, and a sample from the Siberian plume system. Instead the Iceland picrites show uniform $^{186}$Os/$^{188}$Os with an average of 0.1198375±32, similar to the estimated present-day primitive mantle value of 0.1198398±16. The lack of variation in $^{186}$Os/$^{188}$Os is coupled with a range of variation in $^{187}$Os/$^{188}$Os from 0.1297 to 0.1381. These systematics are reasonably explained by addition of 1 to 2 Ga recycled crust into the source of the Iceland plume. This is consistent with lithophile isotope and elemental evidence from these picrites and other neovolcanic zone lavas for a recycled crust contribution to the Iceland mantle plume. These inferences demonstrate that addition of ancient recycled crust into the source regions of plumes does not result in coupled enrichments of $^{186}$Os/$^{188}$Os and $^{187}$Os/$^{188}$Os found in other plume systems studied previously. If so, the coupled enrichments result by another mechanism, such as injection of evolved outer core metal into mantle plume sources at the core-mantle boundary. This process may be localized to such an extent that not all plume systems will show evidence of core-mantle interaction.

The Iceland picrites show a positive correlation between $^{187}$Os/$^{188}$Os from 0.1297 to 0.1381 and $^{3}$He/$^{4}$He from 9.6 to 19 $R_A$, respectively. The most favorable models that explain the origin of this variation show the following key features. Recycled crust older than 500 Ma is initially mixed with primitive mantle (either as solids or as melts derived from these sources), generating a hybrid mantle source with radiogenic $^{187}$Os/$^{188}$Os but with elevated $^{3}$He/$^{4}$He. The ancient recycled crust must be juxtaposed with a high $^3He/^4He$ mantle and both are physically separated from the depleted MORB upper mantle. Mixing with MORB mantle subsequently produces the quasi-linear array for $^{187}Os/^{188}Os$-$^3He/^4He$ observed in the Iceland picrites. The successful models that generate the high $^{187}Os/^{188}Os$ and $^3He/^4He$ end-member as a hybrid mixture between ancient recycled crust and a high $^3He/^4He$ mantle have concentrations of He that appear to result from less outgassing over time than the strongly degassed high $^3He/^4He$, low [He], hypothetical mantle component of Class and Goldstein (2005). These relationships between $^{187}Os/^{188}Os$, $^3He/^4He$, and He concentration with solar Ne isotopic compositions of some Iceland lavas, are consistent with a deep mantle origin of the $^3He/^4He$ mantle component, where juxtaposition of an early formed layer with high $^3He/^4He$ and younger recycled material may occur at the core mantle boundary over Earth history (Toltskihin and Hofmann, 2005).

The relationships between He-Nd-Os isotopes and lithophile element concentrations have changed during the eruptive history of the Iceland plume. At 60 Ma, lavas erupted from the ancestral plume head which impinged upon the Greenland craton appear to have sampled little or no recycled crust/lithosphere within the plume. Upon continued upwelling from the deep mantle, ancient recycled material was brought into the zone of melting under Iceland, probably during the last 15 Ma.

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**Figure Captions**

Figure 1. Map of Iceland and showing the sample locations for this study as well as zones of active volcanism as shaded regions. Abbreviations are as follows: RR = Reykjanes Ridge, KR = Kolbeinsey Ridge, TFZ = Tjornes Fracture Zone, WEZ = western Extensional Zone, ERZ = east Rift Zone, SEZ = southern Extensional Zone.

Figure 2. (A) $^{187}Os/^{188}Os$-Os for Iceland picrites. The half filled square is sample ICE 6 from Theistareykir, all other samples are solid squares. (B) $^{187}Re/^{188}Os$-Os.

Figure 3. (A) MgO-$^{187}Os/^{188}Os$ for Iceland picrites. Sample symbols as in Figure 2. (B) MgO-$^{187}Re/^{188}Os$.

Figure 4. $\gamma Os$ versus $^{186}Os/^{188}Os$ for Iceland picrites, Hawaiian picrites, 89 Ma Gorgona komatiites, and 251 Ma Siberian sample, where $\gamma Os = [(^{187}Os/^{188}Os_{sample@t}/^{187}Os/^{188}Os_{chondrite@t} - 1) * 100]$, present-day $^{187}Os/^{188}Os_{chondrite} = 0.1270$, and $^{187}Re/^{188}Os_{chondrite} = 0.4083$, Shirey and Walker, l998. The field for global upper mantle samples is shown as a shaded region, the primitive mantle (PM) evolution curve is shown with 500 Ma increments to the present-day value, dashed lines extending to the right from the PUM curve are for mafic material evolution trends formed from a PM reservoir at 1 Ga and 2 Ga (Brandon et al., 2006). Regression lines through the Gorgonan and Hawaiian-Siberian trends are shown with calculated $r^2$'s (Brandon et al., 1999; 2003). Typical $\pm 2\sigma$ uncertainty for the data is shown in the upper right of the diagram.

Figure 5. (A) $^{3}He/^{4}He$-$^{187}Os/^{188}Os$ for Iceland picrites. Solid symbols are for samples where He was measured in the present study, open symbols for those taken from the literature from the same locales (Table 3). Error bars for $^{3}He/^{4}He$ are $\pm 2\sigma$. (B) $(La/Sm)_n$-$^{187}Os/^{188}Os$.

Figure 6. (A) $(La/Sm)_n$ -$(Th/La)_n$ for Iceland picrites. Sample symbols as in Figure 2. (B) $(La/Sm)_n$-$(Th/Sm)_n$.

Figure 7. The He isotopic composition (A) and $^{4}He$ concentration cc(STP)/g (B) of ancient recycled basalt and sediment that is aged from 0 to 2 Ga using initial $^{3}He/^{4}He$, U abundances, and Th/U listed in Table 4. For the basalt, the curves represent 0, 50, and 90% of outgassing of endogenous He closely following the time of formation of ocean crust. For the sediment, the curves represent 0, 50 and 90% loss of U and Th during subduction processes. Subsequent closed-system ingrowth of $^{4}He$ is calculated following these events.

Figure. 8. Mixing between different aged recycled components and a mantle sources having $^{3}He/^{4}He=37\ R_A$. One set of lines are for mixing with an undegassed primtive mantle source (PM), and another set of lines are for mixing with a degassed plume source (DPS) as labeled, with mixing parameters listed in Table 4. The shaded bands are the maximum $^3$He/$^4$He values measured in Iceland neovolcanic zone lavas. (A) Mixing between high $^3$He/$^4$He mantle and recycled basalt. The upper dashed lines for 250 Ma are for 0, 50, and 90% outgassing of endogenous He closely following the time of formation of ocean crust. (B) Mixing between high $^3$He/$^4$He mantle and recycled sediment. The upper dashed curves at 250 Ma are for 50 and 90% loss of U and Th relative to GLOSS during subduction processes (Table 4).

Figure 9. $^3$He/$^4$He-$^{187}$Os/$^{188}$Os mixing models for a primitive mantle (PM) with $^{187}$Os/$^{188}$Os = 0.1296 and $^3$He/$^4$He = 37 $R_A$, and a depleted plume source (DPS) with $^{187}$Os/$^{188}$Os = 0.1250 and $^3$He/$^4$He = 37 $R_A$, with 250 Ma (dashed lines) and 2 Ga crustal (solid lines) compositions evolved to the present where Bas = basalt, Sed = sediment, Slab = 95 basalt:5 sediment. The Iceland picrite data are shown with symbols as in Figure 5. See Table 4 for end-member compositions modeled and for hybrid slab-mantle components derived from the mixes. The shaded area is the region where mixing lines between a hybrid component with high $^{187}$Os/$^{188}$Os and $^3$He/$^4$He that is itself a mixture between PM and slab, and an upper mantle with low $^{187}$Os/$^{188}$Os and $^3$He/$^4$He will lie in order to plot along the trend for the Iceland picrite data. Note that no mixture between crustal compositions of any age and DPS will result in a high $^{187}$Os/$^{188}$Os and $^3$He/$^4$He hybrid component. See text for additional discussion.

Figure 10. Mixing models between 250 Ma and 2 Ga hybrid components (consisting of recycled slab and primitive mantle), and MORB source mantle for $^3$He/$^4$He -$^{187}$Os/$^{188}$Os. The Iceland picrite data are shown with symbols as in Figure 5. End-member compositions are listed in Table 4. Symbols on all of the curves in the 4 diagrams are for 10 increments of mixing between the hybrid and MORB mantle components. (A) Mixtures of 250 Ma Hybrid 1 (H1) and Hybrid 2 (H2) isotopic components, with MORB mantle components having High He (curves with X symbols) and Normal He (curves with + symbols) concentrations and $^{187}$Os/$^{188}$Os = 0.1296 (M1). (B) Mixtures of 2 Ga Hybrid 3 (H3) and Hybrid 4 (H4) isotopic components, with MORB mantle components having High He (curves with X symbols) and Normal He (curves with + symbols) concentrations and $^{187}$Os/$^{188}$Os = 0.1296 (M1). (C) Mixtures of 250 Ma Hybrid 1 (H1) and Hybrid 2 (H2) isotopic components, with MORB mantle components having High He (curves with X symbols) and Normal He (curves with + symbols) concentrations and $^{187}$Os/$^{188}$Os = 0.1250 (M2). (D) Mixtures of 2 Ga Hybrid 3 (H3) and Hybrid 4 (H4) isotopic components,

with MORB mantle components having High He (curves with X symbols) and Normal He (curves with + symbols) concentrations and $^{187}Os/^{188}Os = 0.1250$ (M2).

Figure 11. The percent crust versus time of formation of crust, necessary in a mixture with primitive mantle and MORB upper mantle to obtain the maximum value of $^{187}Os/^{188}Os$ for the Iceland picrites of 0.1381. End-member compositions used to model the evolution of the crustal sources and the mixtures are listed in Table 4.

Figure 12. $(La/Sm)_n$-$^3He/^4He$ for lavas from the Iceland plume from 61 Ma to present. Iceland picrite symbols as in Figure 5. The 61 Ma Baffin Island picrites are from Stuart et al. (2003). The $(La/Sm)_n$ ratio of the least crustally contaminated picrite sample from the 61 Ma Vaigat Formation of West Greenland (Lightfoot et al., 1997) is combined with the highest $^3He/^4He$ value from a picrite from the same locale on Nuussuaq Peninsula (Graham et al., 1998). A late Tertiary lava from Selardalur (≤ 14.9 Ma) in NW Iceland is from Harrison et al. (1999).

Figure 13. (A) $\varepsilon_{Nd}$ -$^3He/^4He$ for lavas from the Iceland plume from 61 Ma to present. Additional picrite samples from the 58 Ma East Greenland are from Marty et al. (1998). The $^{143}Nd/^{144}Nd$ of the samples older than present have been corrected to the present by first calculating their Sm/Nd ratios for their sources relative to the CHUR value for each age (present-day CHUR $^{143}Nd/^{144}Nd = 0.512638$, $^{147}Sm/^{144}Nd = 0.1967$) evolving the $^{143}Nd/^{144}Nd$ to present using the calculated Sm/Nd of the sources. $\varepsilon_{Nd} = [(^{143}Nd/^{144}Nd_{sample}/^{143}Nd/^{144}Nd_{CHUR}) - 1] \times 10^4$. (B) $\Delta Nb$-$^3He/^4He$ for lavas from the Iceland plume from 61 Ma to present. $\Delta Nb = 1/75 + \log(Nb/Y) - 1.92\log(Zr/Y)$ (Fitton et al., 1997). Iceland picrite symbols as in Figure 5.

Table 2. Iceland Re-Os isotopic data. Concentrations of Re and Os are in parts per trillion. Reported uncertainties in parentheses are ±2σ for the last decimal place.

| **Sample** | **Re** | **Os** | **$^{187}$Os/$^{188}$Os[1]** | **$^{187}$Re/$^{188}$Os** | **$^{184}$Os/$^{188}$Os[2]** | **$^{186}$Os/$^{188}$Os[2]** | **$^{187}$Os/$^{188}$Os[2]** |
|---|---|---|---|---|---|---|---|
| ICE 0 Hruthalsar | 612.3 | 863.2 | 0.12941(15) | 3.42 | 0.0013109(15) | 0.1198382(18) | 0.1297040(20) |
| ICE 2 Lagafell | 305.7 | 1053.0 | 0.13109(24) | 1.40 | 0.0013088(12) | 0.1198386(14) | 0.1323100(14) |
| ICE 3 Stapafell | 355.5 | 123.6 | 0.13168(26) | 13.86 | | | |
| ICE 4a Maelifell | 493.9 | 457.1 | 0.13335(20) | 5.21 | 0.0013266(24) | 0.1198376(24) | 0.1326225(26) |
| ICE 4b Maelifell | 615.5 | 881.6 | 0.13315(14) | 3.37 | 0.0013101(17) | 0.1198369(17) | 0.1327022(17) |
| ICE 5 Dagmalafell | 459.8 | 410.9 | 0.13305(20) | 5.40 | | | |
| DMF 9101 Dagmalafell[3] | 352.6 | 746.3 | 0.13208(19) | 2.28 | 0.001313(1) | 0.1198350(48) | 0.1324049(12) |
| ICE 6 Theistareykir | 711.8 | 139.9 | 0.13467(21) | 24.54 | 0.0013139(19) | 0.1198359(21) | 0.1352774(27) |
| ICE 8a Skridufell | 487.3 | 273.5 | 0.13716(24) | 8.59 | 0.0013076(13) | 0.1198404(16) | 0.1371644(16) |
| ICE 8b Skridufell | 279.7 | 516.4 | 0.13750(22) | 2.61 | 0.0013089(23) | 0.1198369(23) | 0.1376726(23) |
| ICE 9a Skridufell | 476.1 | 148.2 | 0.13784(22) | 15.49 | 0.0013084(14) | 0.1198367(18) | 0.1381292(18) |
| ICE 10 Hrudurkarlarnir | 432.4 | 183.8 | 0.13140(20) | 11.34 | | | |
| ICE 11 Grindarik | 390.7 | 877.1 | 0.13116(24) | 2.15 | 0.0013109(23) | 0.1198390(24) | 0.1318350(19) |
| 9805 Haleyjabunga | 152.3 | 1064.9 | 0.13151(21) | 0.689 | 0.0013122(19) | 0.1198365(19) | 0.1319716(22) |
| 9806 Vatnsheidi | 357.8 | 674.2 | 0.13278(22) | 2.56 | | | |
| 9809 Fagradalshraun | 336.6 | 1028.3 | 0.13081(31) | 1.58 | 0.0013157(16) | 0.1198410(17) | 0.1308974(17) |
| 9810 Eldborg | 585.9 | 62.9 | 0.13518(31) | 44.92 | | | |
| 9812 Asar | 496.0 | 424.2 | 0.13406(19) | 5.64 | 0.0013108(19) | 0.1198381(21) | 0.1340553(26) |
| 9815 Lyngfell | 573.5 | 203.7 | 0.13586(21) | 13.58 | 0.0013065(24) | 0.1198371(26) | 0.1344091(31) |
| | | | | | | | |
| Johnson-Matthey Std. | | | | | | | |
| 1 | | | | | 0.0013194(25) | 0.1198520(29) | 0.1137986(33) |
| 2 | | | | | 0.0013125(17) | 0.1198513(19) | 0.1138026(19) |
| 3 | | | | | 0.0013088(19) | 0.1198522(22) | 0.1137990(22) |
| 4 | | | | | 0.0013113(23) | 0.1198518(22) | 0.1138000(20) |
| 5 | | | | | 0.0013102(24) | 0.1198503(27) | 0.1137952(29) |
| 6 | | | | | 0.0013192(20) | 0.1198535(24) | 0.1138042(22) |

1 – Isotopic compositions obtained from spiked aliquots.

2 – Isotopic compositions obtained from unspiked aliquots.

3 - Measuremens for DMF 9101 performed at University of Maryland.

Table 3. He isotope and concentration data for Iceland picrites. New results from this study, with the weight of olivine or glass analyzed, are for helium extractions by in vaco crushing. Previous results from the literature are reported in italics.

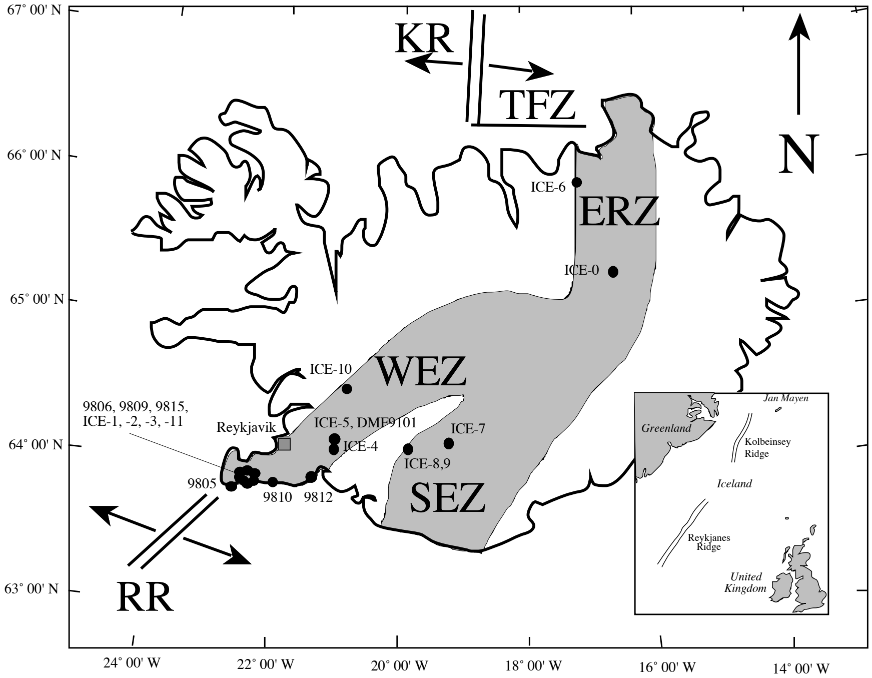
| **Sample** | **Wt. (mg)** | **Phase** | **$^3$He/$^4$He (R/R$_A$)** | **±2σ** | **[He]** (nccSTP/g)[4] |
|---|---|---|---|---|---|
| ICE 0 Hruthalsar | 535.0 | olivine | 9.62 | 0.63 | 1.14 |
| ICE 2 Lagafell | 452.1 | olivine | 11.18 | 1.63 | 3.90 |
| | | *olivine* | *14.09*[1] | *0.14* | *1.53* |
| ICE 3 Stapafell | | *glass* | *13.9*[1] | | *1430* |
| ICE 4a Maelifell | 650.6 | olivine | 11.46 | 0.32 | 5.36 |
| ICE 5 Dagmalafell | 548.5 | olivine | 16.62 | 0.26 | 12.2 |
| | 362.5 | glass | 17.20 | 0.09 | 2560 |
| DMF 9101 Dagmalafell | 530.4 | olivine | 16.40 | 0.51 | 11.7 |
| | | *glass* | *16.50*[2] | *0.97* | *512* |
| | | *glass* | *17.53*[2] | *1.25* | *258* |
| ICE 8b Skridufell | 619.0 | olivine | 17.30 | 0.25 | 37.3 |
| ICE 10 Hrudurkarlarnir | 500.7 | olivine | 12.76 | 0.74 | 1.16 |
| 9805 Haleyjabunga | | *olivine* | *10.89*[1] | *0.60* | *0.52* |
| 9806 Vatnsheidi | | *olivine* | *11.65*[1] | *0.22* | *4.26* |
| 9809 Fagradalshraun | 629.5 | olivine | 12.74 | 0.46 | 1.92 |
| 9810 Eldborg | | *olivine* | *19.0*[3] | *2* | *13.0* |
| 9812 Asar | 225.9 | olivine | 16.80 | 2.15 | 0.61 |
| 9815 Lyngfell | 496.1 | olivine | 15.83 | 0.59 | 3.48 |

1 - Breddam et al. (2000)

2 – Harrison et al. (l999), average of n=11, and n=5

3 - Dixon et al. (2003)

4 - nccSTP/g = $10^{-9}$ ccSTP/g


67° 00' N
66° 00' N
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KR
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ICE-6
ERZ
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ICE-10
WEZ
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ICE-1, -2, -3, -11
Reykjavik
ICE-5, DMF9101
ICE-4
ICE-7
ICE-8,9
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Jan Mayen
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Kolbeinsey Ridge
Iceland
Reykjanes Ridge
United Kingdom

Figure 1

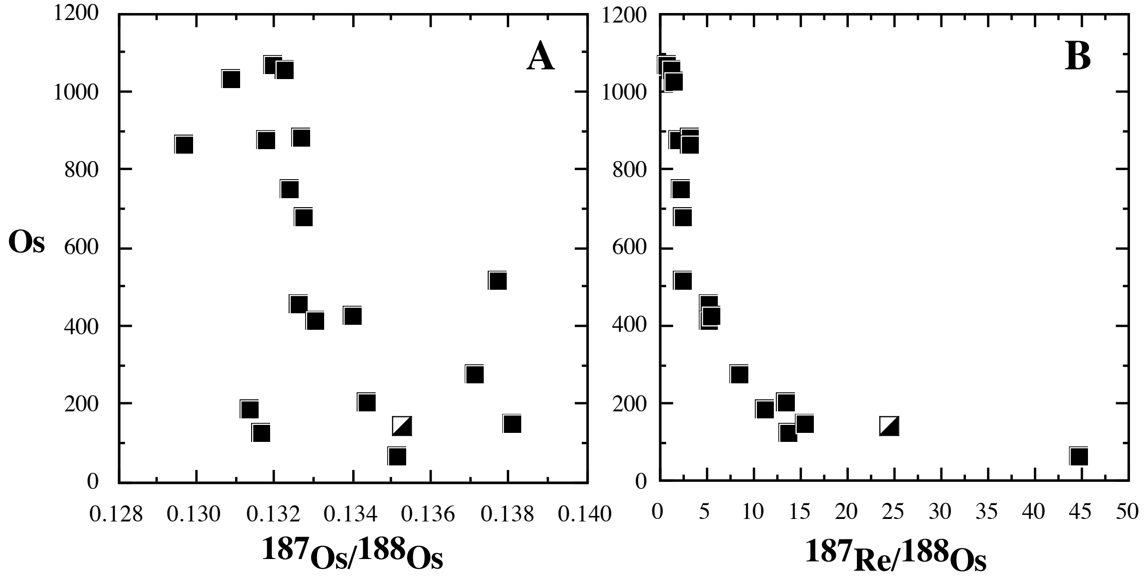


Figure 2

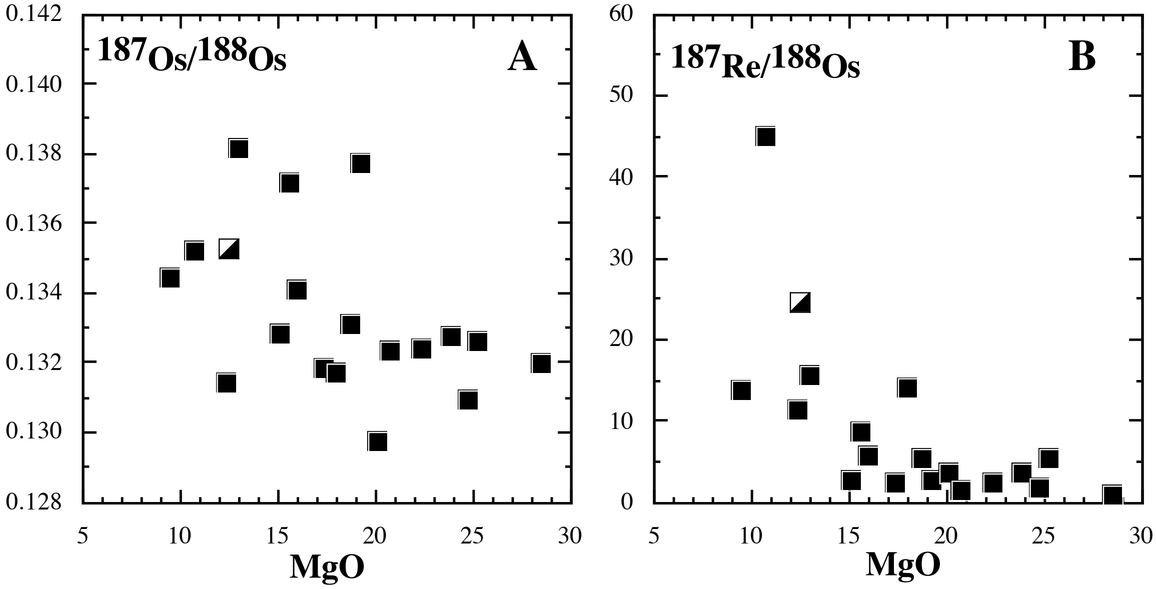


Figure 3

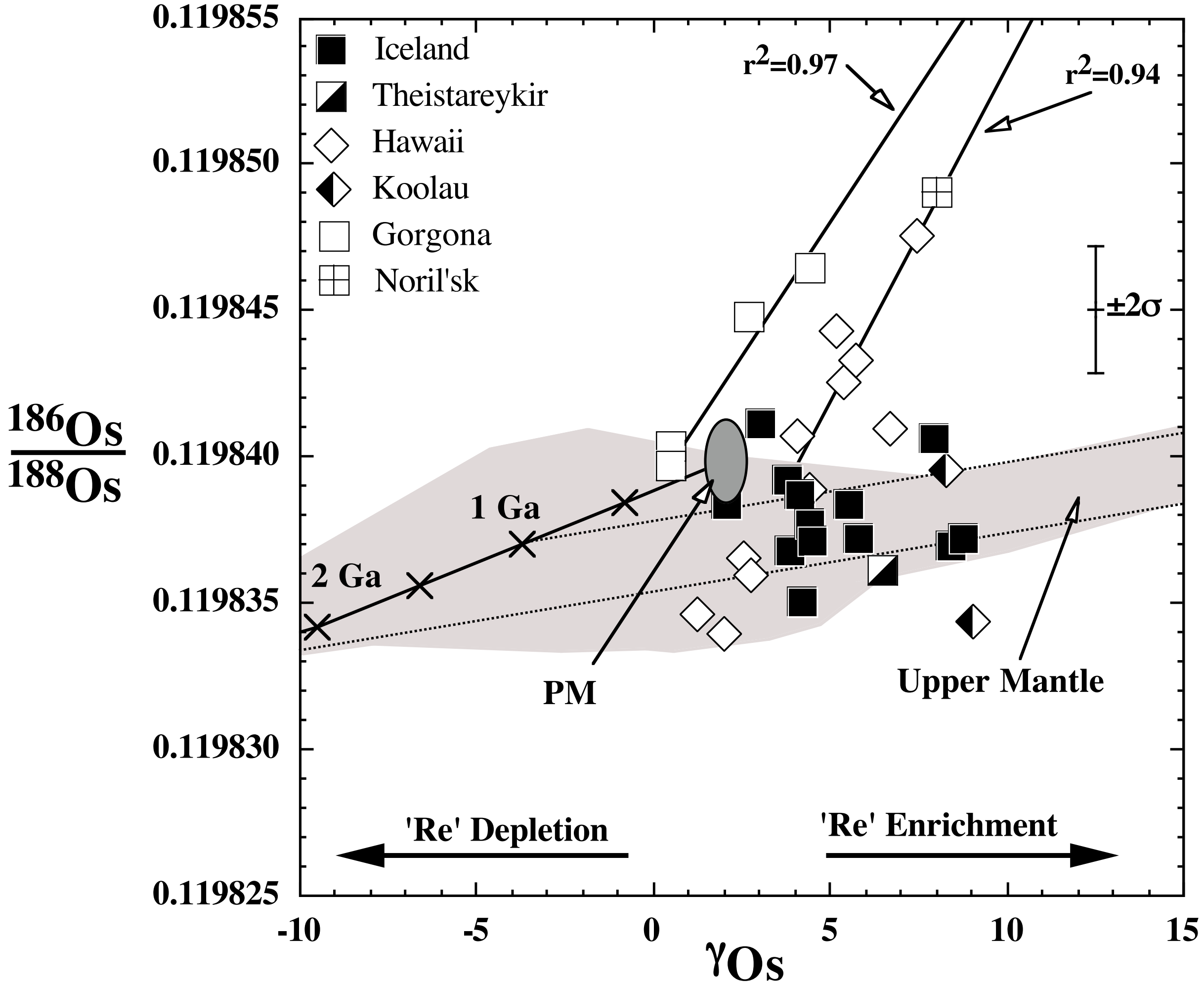

■ Iceland
◪ Theistareykir
◇ Hawaii
◆ Koolau
□ Gorgona
⊞ Noril'sk
$r^2$=0.97
$r^2$=0.94
±2σ
1 Ga
2 Ga
PM
Upper Mantle
'Re' Depletion
'Re' Enrichment
$^{186}Os/^{188}Os$
0.119855
0.119850
0.119845
0.119840
0.119835
0.119830
0.119825
-10
-5
0
5
10
15
$\gamma Os$

Figure 4

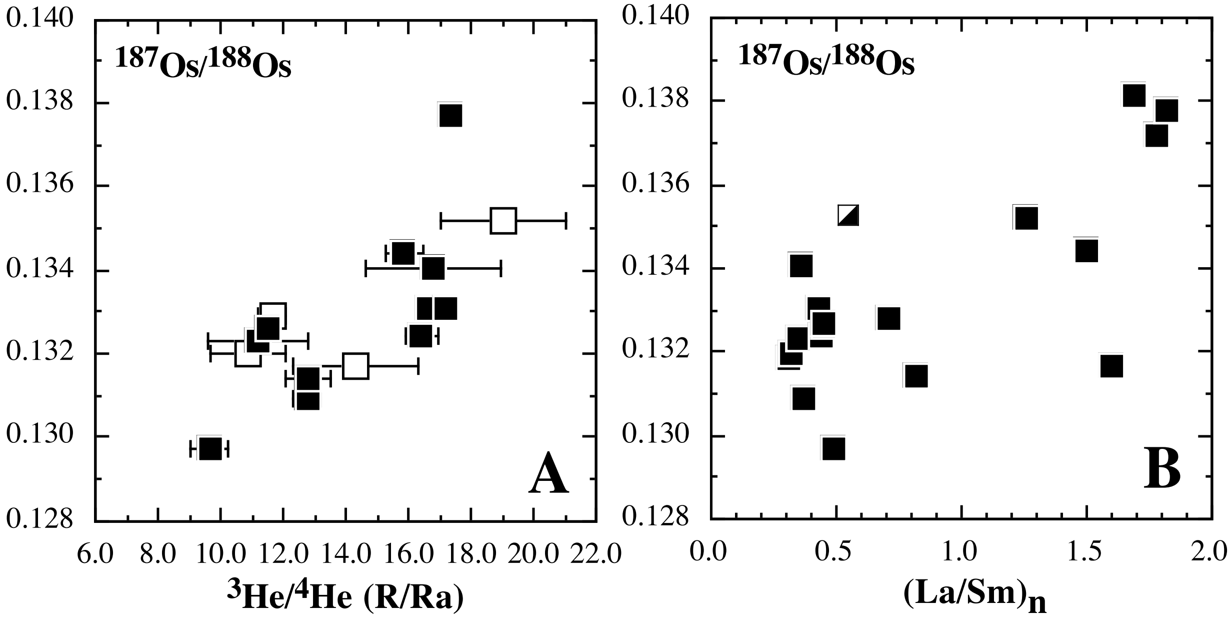
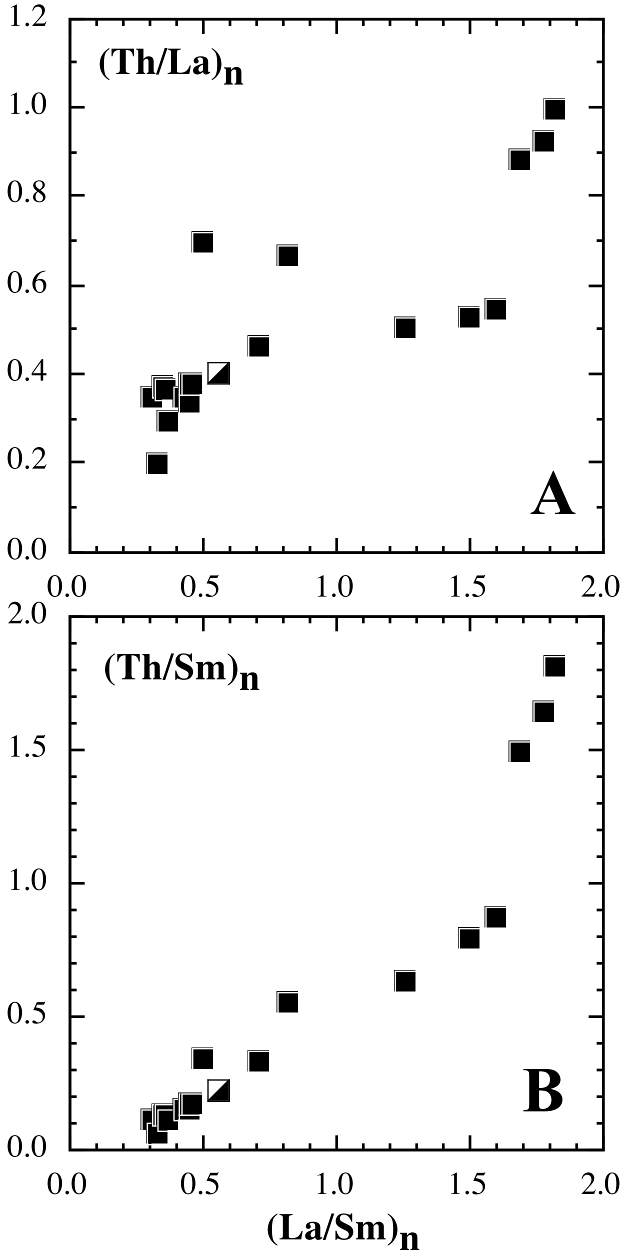

$^{187}$Os/$^{188}$Os
0.140
0.138
0.136
0.134
0.132
0.130
0.128
6.0
8.0
10.0
12.0
14.0
16.0
18.0
20.0
22.0
$^{3}$He/$^{4}$He (R/Ra)
A
$^{187}$Os/$^{188}$Os
0.0
0.5
1.0
1.5
2.0
(La/Sm)$_n$
B

Figure 5


1.2
1.0
0.8
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0.2
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(Th/La)n
A
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2.0
(Th/Sm)n
1.5
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B
0.0
0.5
1.0
1.5
2.0
(La/Sm)n

Figure 6.

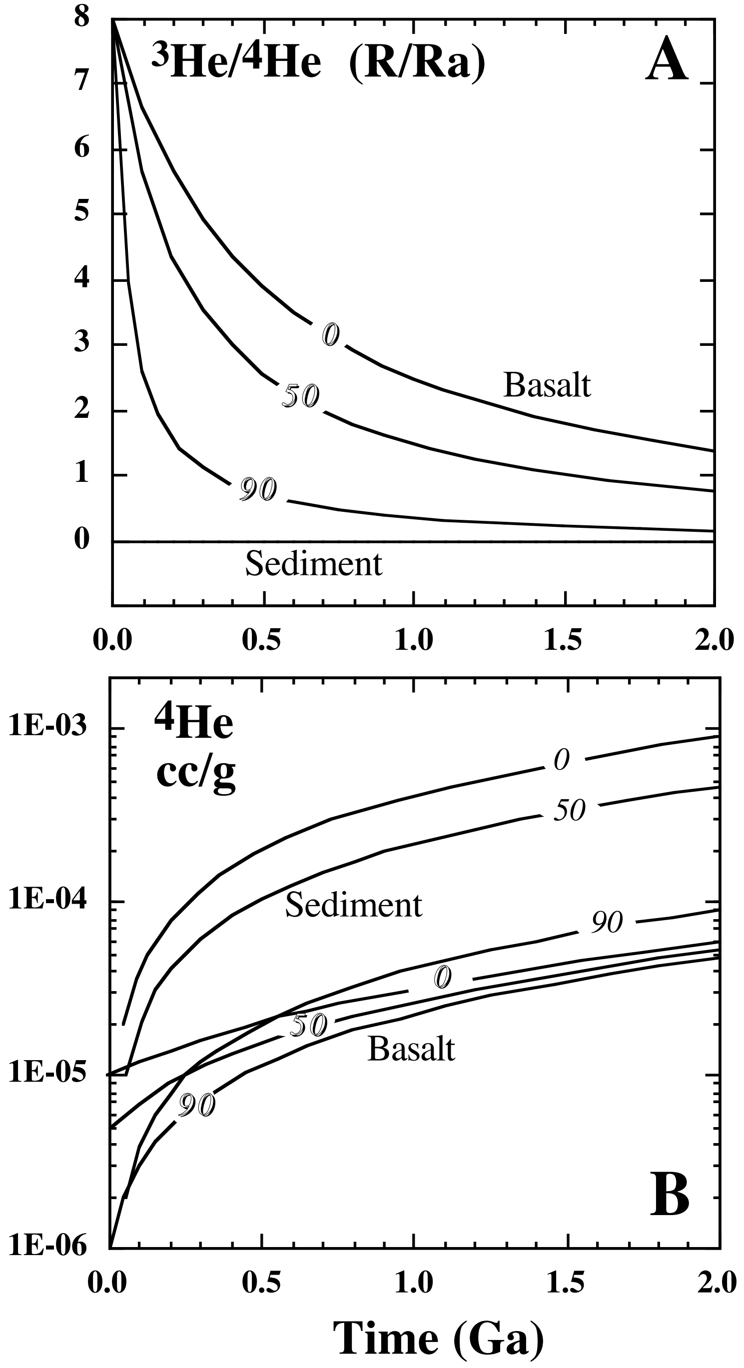

3He/4He (R/Ra)
A
8
7
6
5
4
3
2
1
0
0
50
90
Basalt
Sediment
0.0
0.5
1.0
1.5
2.0
4He
cc/g
1E-03
1E-04
1E-05
1E-06
0
50
Sediment
90
0
50
Basalt
90
B
0.0
0.5
1.0
1.5
2.0
Time (Ga)

Figure 7.

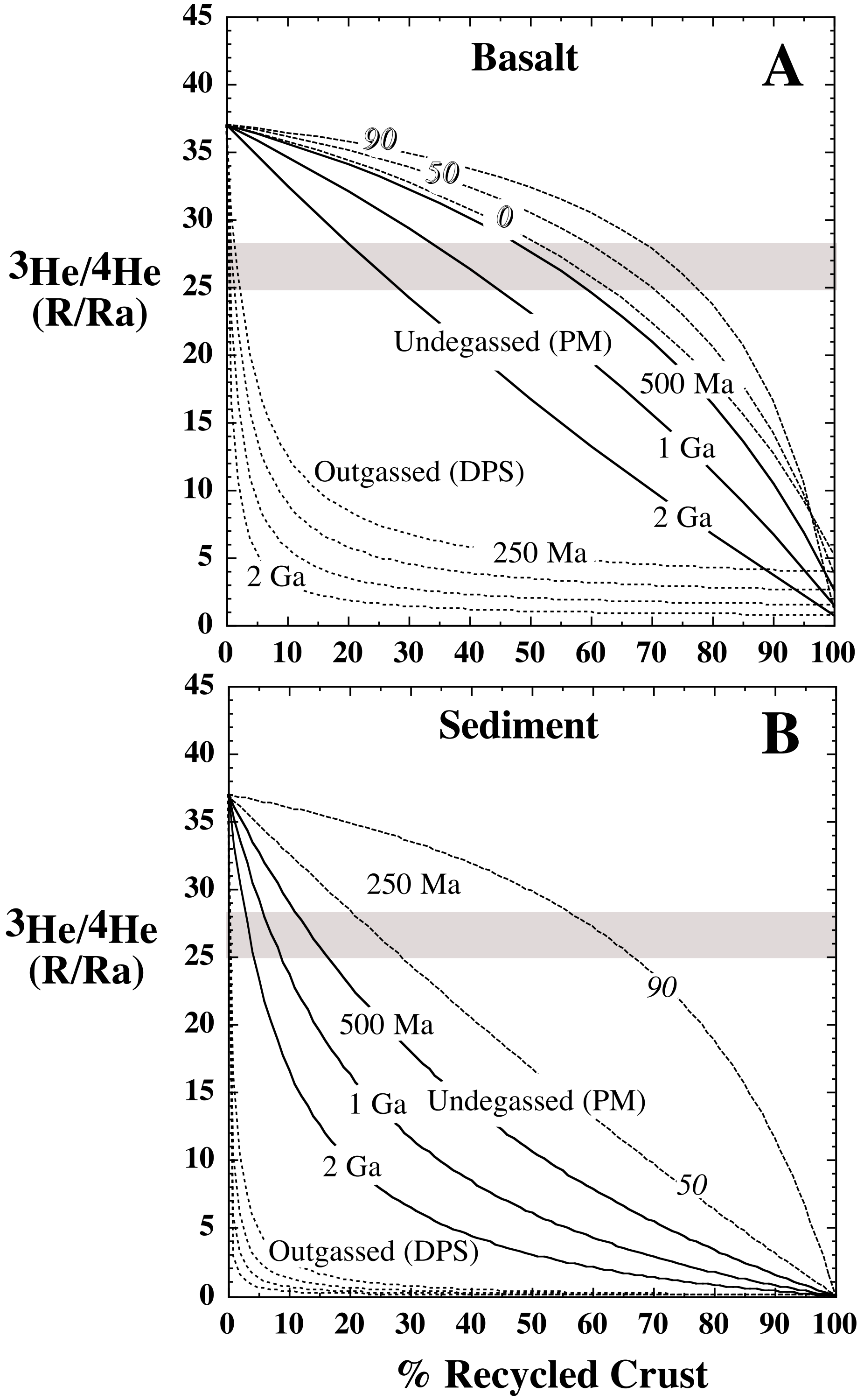

Basalt
A
3He/4He (R/Ra)
90
50
0
Undegassed (PM)
500 Ma
1 Ga
2 Ga
Outgassed (DPS)
250 Ma
2 Ga
0
5
10
15
20
25
30
35
40
45
0 10 20 30 40 50 60 70 80 90 100
Sediment
B
3He/4He (R/Ra)
250 Ma
90
500 Ma
1 Ga
Undegassed (PM)
2 Ga
50
Outgassed (DPS)
0
5
10
15
20
25
30
35
40
45
0 10 20 30 40 50 60 70 80 90 100
% Recycled Crust

Figure 8

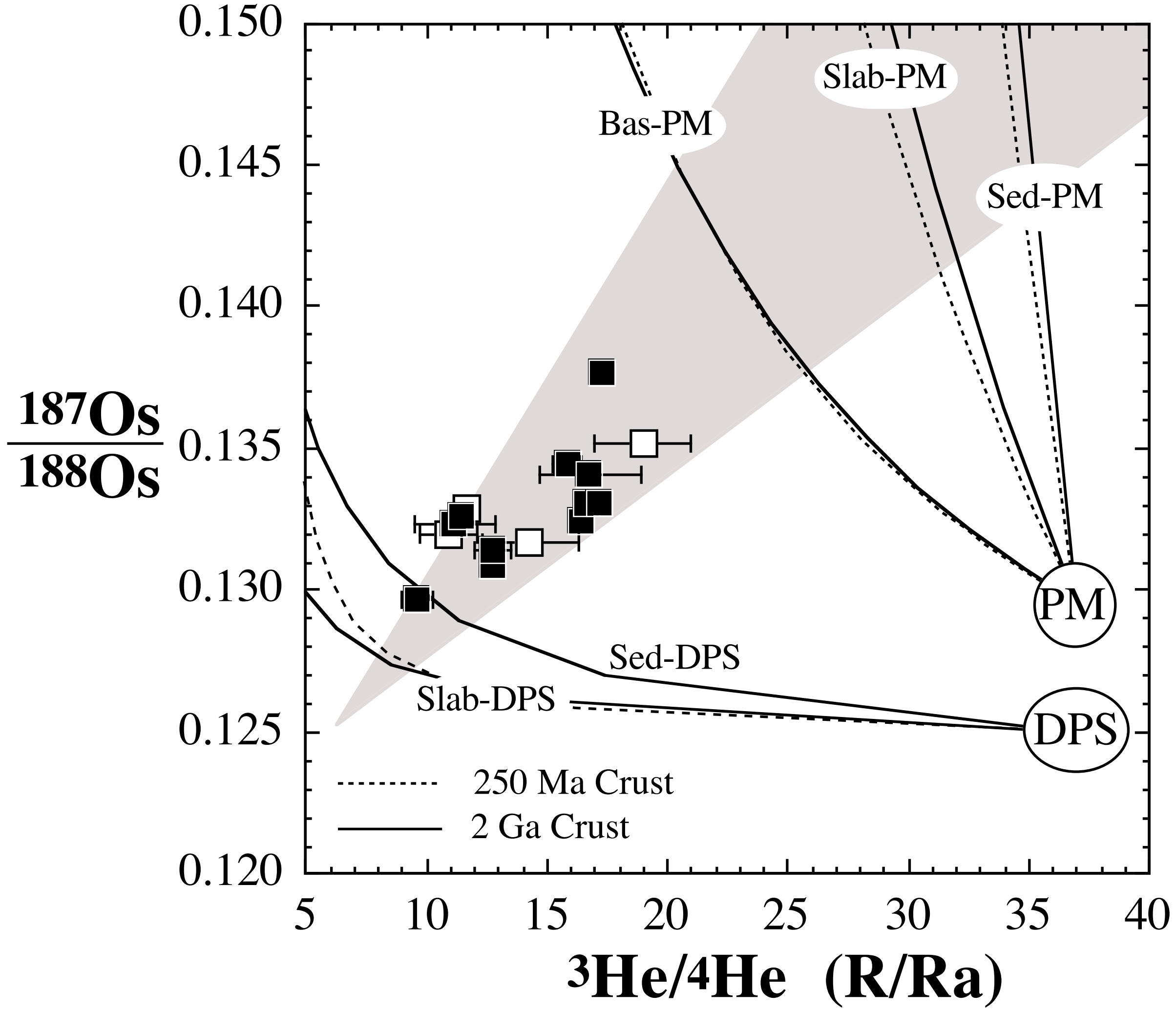
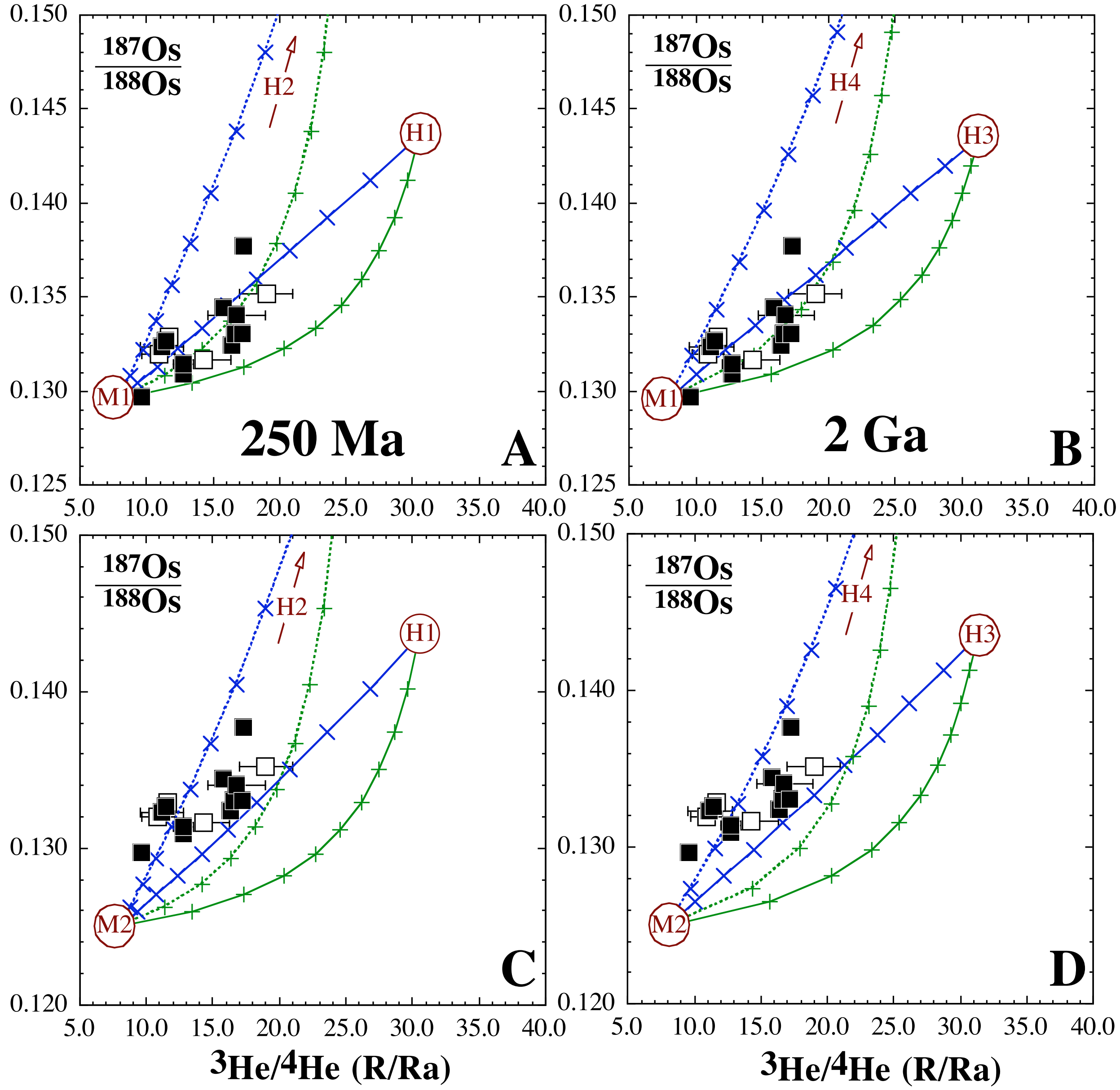

0.150
0.145
0.140
0.135
0.130
0.125
0.120
$\frac{^{187}Os}{^{188}Os}$
5
10
15
20
25
30
35
40
$^{3}He/^{4}He$ (R/Ra)
Bas-PM
Slab-PM
Sed-PM
PM
Sed-DPS
Slab-DPS
DPS
250 Ma Crust
2 Ga Crust

Figure 9.


0.150
0.145
0.140
0.135
0.130
0.125
0.120
187Os
188Os
H1
H2
H3
H4
M1
M2
250 Ma
2 Ga
A
B
C
D
5.0
10.0
15.0
20.0
25.0
30.0
35.0
40.0
3He/4He (R/Ra)

`

Figure 10.

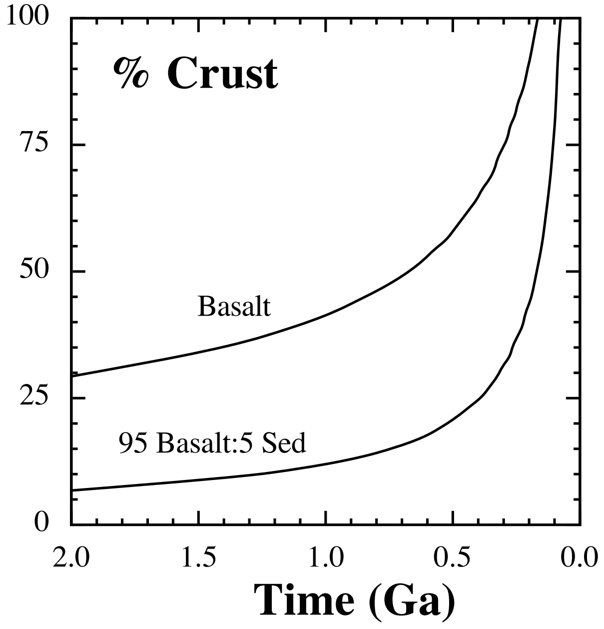


Figure 11

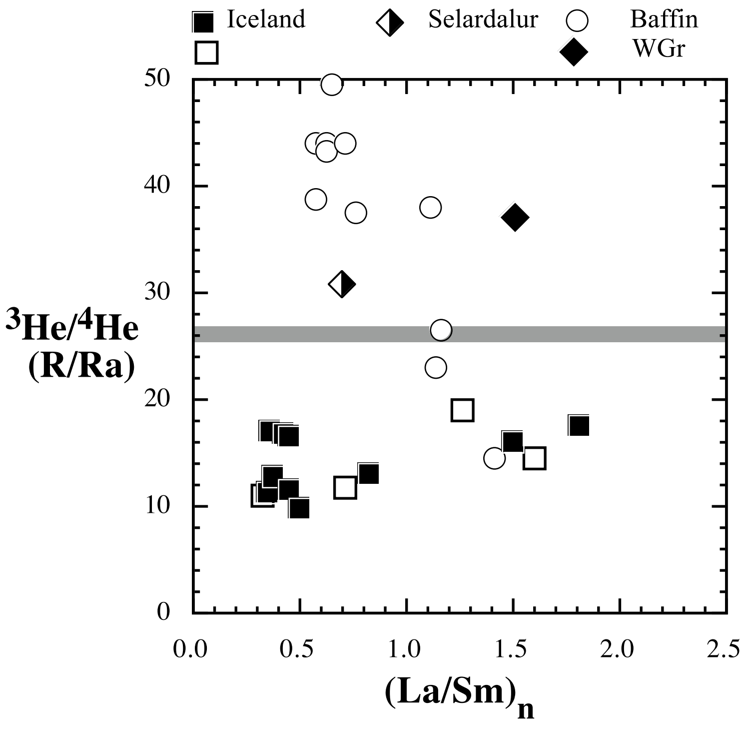


Figure 12

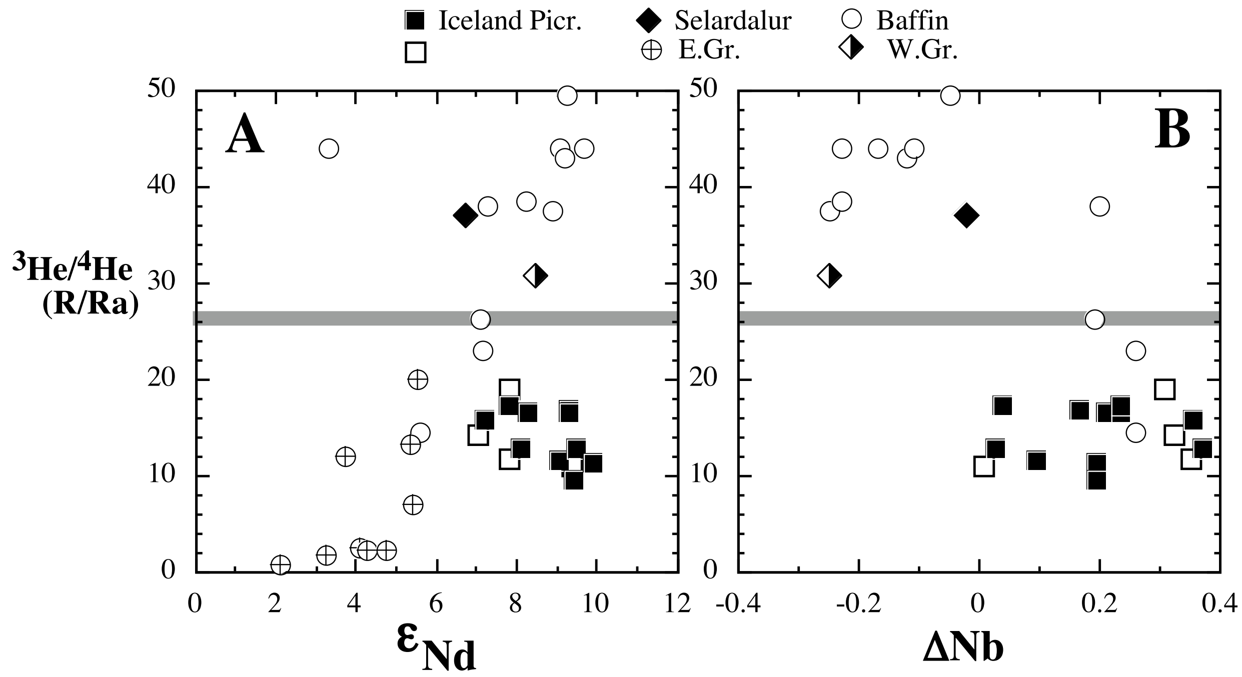

■ Iceland Picr.
□
◆ Selardalur
⊕ E.Gr.
○ Baffin
W.Gr.
A
B
$^3$He/$^4$He (R/Ra)
0
10
20
30
40
50
0
2
4
6
8
10
12
-0.4
-0.2
0
0.2
0.4
ε<sub>Nd</sub>
ΔNb

Figure 13