Osmium isotopes in North Atlantic picrites with extreme $^{3}He/^{4}He$ ratios: implications for the nature of high $^{3}He/^{4}He$ mantle and the Os isotope composition of the convecting mantle

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Abstract

Identifying the Os isotope composition of the prevalent peridotitic convecting mantle places important constraints on the Earth's accretion, differentiation and evolution. Furthermore, the accurate interpretation of Re-depletion ages in mantle peridotites is dependent on this composition. Small degree mantle melts preferentially sample components with the lowest melting temperatures and so are poorly suited to identifying the Os isotope composition of peridotite components. In contrast, Os isotope studies of large degree melts such as picrites are much more likely to reflect the peridotitic components within the source. Thirty proto-Iceland plume picrites from Baffin Island and West Greenland, produced by large degree mantle melting, have been analysed for Re-Os isotopes in order to provide an estimate for the bulk $^{187}Os/^{188}Os$ composition of convecting mantle and to investigate the nature of high $^{3}He/^{4}He$ mantle. Ingrowth-corrected $^{187}Os/^{188}Os$ of the picrites ranges from 0.1267 to 0.1322. The higher $^{187}Os/^{188}Os$ samples have correspondingly lower $^{143}Nd/^{144}Nd$ which may reflect a contribution (~5-10%) from old recycled crust including sediment. However, samples from Baffin Island and the earliest West Greenland members are remarkably uniform in composition with $^{187}Os/^{188}Os$ between 0.1267 and 0.1280, and a mode of 0.1272. These Os isotope compositions are less radiogenic than estimates of PUM but are similar to the least radiogenic mid-ocean ridge basalts (MORB) and the most common composition of ophiolite-derived platinum-group alloys and chromites. These compositions appear to represent a source that contains no significant enriched component such as pyroxenite/eclogite.

The picrites studied record the highest known $^{3}He/^{4}He$ in the silicate Earth (up to 50 R_a). For this signature to reflect isolated domains of ancient melt depletion would require significantly less radiogenic Os isotope compositions than observed, if mantle Os and He remain coupled. Conversely, an outer core contribution would impart a supra-chondritic $^{187}Os/^{188}Os$ signature to the picrites, thus the near-chondritic Os isotope compositions also preclude the core as a source of high

$^{3}He/^{4}He$, unless the mechanism for transfer of He into the plume is decoupled from that of Os. However, as the high $^{3}He/^{4}He$ signature is found in samples with variable Os and Nd isotope compositions, it is likely that He is decoupled from other isotopic tracers and is dominated by minor addition of a He-rich component with high $^{3}He/^{4}He$, probably of primordial nature, although the ultimate source is unclear. Alternatively, a high $^{3}He/^{4}He$ mantle reservoir with $^{187}Os/^{188}Os$ of ~0.1275 cannot be ruled out, but the absence of any elevated $^{3}He/^{4}He$ in MORB, which has a similar Os isotope composition, suggests that such a component could only be sampled during episodes of high degree melting of hot mantle.

Keywords: osmium isotopes, helium isotopes, Baffin, convecting mantle, depleted mantle.

1. Introduction

Mantle rocks and mantle-derived melts display a broad range of Os isotope compositions, both depleted and enriched with respect to bulk Earth and primitive upper mantle (PUM) estimates. Osmium is a compatible element during mantle melting while Re, which is the parent to ^{187}Os through beta decay of ^{187}Re, is moderately incompatible. Thus, crustal rocks typically have very high Re/Os ratios and, over time, evolve to radiogenic $^{187}Os/^{188}Os$ compared to the mantle. Depleted mantle, which has preferentially lost Re over Os, evolves complementary unradiogenic $^{187}Os/^{188}Os$ ratios, with the timing of Re depletion indicated by the extent to which $^{187}Os/^{188}Os$ deviates from a chondritic evolution curve. Therefore, the Re-Os system is a potentially powerful tool with which to assess the contributions from depleted mantle and enriched recycled materials to the source of mantle-derived melts.

Significant Os isotope heterogeneity exists in the mantle at a variety of length-scales from mineral to vein to slab scale due to the recycling of enriched crustal materials and depletion of peridotite, and the highly siderophile nature of both Re and Os. Different degrees of melting of such a heterogeneous mantle will lead to melts that vary in isotopic composition, with the most radiogenic melts typically derived from the smallest degrees of melting, and large-degree melts giving the best estimate of the $^{187}Os/^{188}Os$ composition of the *bulk* mantle source. This study of picrites from Baffin Island (BI) and West Greenland (WG) should therefore provide a good estimate for the average Os isotope composition of the source and, given the similarity to depleted MORB mantle in terms of lithophile isotopes (e.g. Ellam and Stuart, 2004), provide an estimate for the $^{187}Os/^{188}Os$ of convecting mantle free from enriched components. Such an estimate is a valuable parameter for constraining the Os isotope evolution of the Earth and peridotite melting ages (e.g. Meisel et al., 1996; Meisel et al., 2001; Walker et al., 2002b).

Mantle $^{3}He/^{4}He$ ranges from ~ 5 to 50 R_a (where $R_a = {}^{3}He/^{4}He_{atmosphere} = 1.39 \times 10^{-6}$) and includes the canonical mid-ocean ridge basalt (MORB) range of 8 ± 1 R_a (Graham, 2003). The high $^{3}He/^{4}He$ end-member is defined by picrites from Baffin Island (BI) (~50 R_a, Stuart et al., 2003) and West Greenland (Graham et al., 1998; Starkey et al., 2007, Starkey et al. in review) which erupted at about 61 Ma as a result of elevated mantle temperatures and regional rifting due to the arrival of the Iceland plume (Saunders et al., 1997). Due to the incompatibility of He, conventional wisdom posits that high $^{3}He/^{4}He$ mantle reservoirs are less degassed than the convecting upper mantle, and retain a component of the Earth's primordial volatile inventory (e.g. Kurz et al., 1982; Moreira et al., 2001; Porcelli et al., 2002). However, the observation that U and Th may be less compatible than He in an olivine-rich mantle assemblage has led to the suggestion that high $^{3}He/^{4}He$ may result from the greater loss of U and Th than He during ancient melt depletion (Graham et al., 1990; Class and Goldstein, 2005; Parman et al., 2005; Parman, 2007). The Re-Os isotope system has the ability to retain information about mantle melting events, even in the convecting mantle (e.g. Brandon et

al., 2000; Meibom et al., 2002; Harvey et al., 2006; Pearson et al., 2007), and thus is the most suitable tracer to test whether high $^{3}He/^{4}He$ signatures can be directly linked to ancient depletion events.

The North Atlantic picrites (NAP) from Baffin Island and West Greenland represent some of the earliest known volcanic rocks of the proto-Iceland plume, which continues to produce mantle melts with high $^{3}He/^{4}He$ in the Iceland region today (e.g. Macpherson et al., 2005). In this study, Os isotope data have been (i) combined with existing He and Nd isotope data to re-assess the nature of the highest known $^{3}He/^{4}He$ mantle component, and (ii) used to gain an estimate of the average Os isotope composition of convecting mantle. The high MgO content of the samples (>15 wt. %) and correspondingly high Os concentrations mean that they are less susceptible to interactions with crust and lithospheric mantle – critical when looking at samples which have been erupted through ancient continental crust and lithosphere.

2. Samples: setting and chemistry

The high-Mg basalts and picrites in this study were collected from the eastern margin of Baffin Island (BI) at Cape Searle, Padloping Island and Durban Island and from Disko Island (Qeqertarsuaq) and the Nuussuaq peninsula in West Greenland (WG). The WG picrites belong to the Vaigat formation which is divided into three members, from oldest to youngest, the Anaanaa, Naujánguit and Ordlingassoq Members. The Baffin picrites are undifferentiated stratigraphically but can be grouped chemically into the enriched (E-type) lavas (DUR-8, DI-23 and PAD-6) and normal (N-type) lavas (all others) first identified by Francis (1985). Sample CS-7 is from a cross-cutting dyke, rather than the picrite succession. Volcanism was largely contemporaneous on Baffin Island and in WG and commenced ~61 Ma (Storey et al., 1998). Both the WG Anaanaa Member

and the BI picrites possess normal magnetisation whereas the subsequent WG melts are reversely magnetised (Pedersen et al., 2002). The whole sequence, and ~80% of the total Paleocene volcanic rocks, were erupted within 1 million years (Storey et al., 1998). Petrography, major and trace element chemistry and Sr, Nd and Pb isotopes are described in more detail in previous studies (e.g. Francis, 1985; Robillard et al., 1992; Holm et al., 1993; Lightfoot et al., 1997; Graham et al., 1998; Larsen and Pedersen, 2000; Stuart et al., 2003; Kent et al., 2004).

MgO contents of the NAP are very high (up to 27 wt. % in this study) which, although in part a result of olivine accumulation, reflects the Mg-rich nature of parental melts. Estimates for the parental melts of WG picrites, based on Fo-rich olivine phenocrysts, vary between 15 and 21 wt. % MgO (Pedersen, 1985; Lightfoot et al., 1997; Graham et al., 1998; Larsen and Pedersen, 2000). Such high MgO contents in an intra-continental plate setting indicate generation by a high degree of melting (10-11% depleted mantle, ~20-28% fertile mantle) of anomalously hot mantle (1540-1600°C) at moderate depths (60-90 km) (Pedersen, 1985; Gill et al., 1992; Herzberg and O'Hara, 2002).

The three West Greenland picrite members are largely similar in terms of major elements. However, for a given MgO content, there is an increase in TiO_2 with time, with the older WG picrites most closely resembling the Baffin picrites (Holm et al., 1993). Chondrite-normalised REE patterns are flattest in the Anaanaa and Naujánguit Members (not shown), while the Ordlingassoq samples have higher LREE and incompatible element concentrations (Holm et al., 1993; Lightfoot et al., 1997). Neodymium and Sr isotope compositions of the Ordlingassoq picrites appear to resemble the least depleted Icelandic picrites (Holm et al., 1993).

3. Analytical techniques and samples

Whole rock powders (~1g) were digested and equilibrated with a mixed ^{190}Os–^{185}Re-enriched spike, using inverse aqua regia (2.5 ml 12 mol l^{-1} HCl and 5 ml 16 mol l^{-1} HNO_3) in quartz high-pressure asher (HPA) vessels or borosilicate carius tubes. The HPA vessels were placed in the Durham University Anton-Paar HPA at 300°C and >110 bars for at least 12 hours, and the Carius tubes were placed in an oven at 240°C for at least 36 hours. Osmium was extracted using CCl_4, followed by back-extraction using HBr, and then microdistilled (Cohen and Waters, 1996). The aqua regia was dried and prepared for purification of Re using AG1X-8 (100-200#) anion-exchange resin (Pearson and Woodland, 2000).

Osmium was loaded onto Pt filaments, ionised as OsO_3^- and analysed by negative-thermal ionisation mass spectrometry (N-TIMS) using the ThermoFinnigan Triton at Durham University. All Os isotope beams and mass 233, corresponding to $^{185}ReO_3^-$, were measured sequentially using an axial secondary electron multiplier. All Os isotope raw data were corrected offline for O isotope interference, mass fractionation (using a $^{192}Os/^{188}Os$ ratio of 3.08271) and spike unmixing. Subtraction of a possible $^{187}ReO_3^-$ interference was not necessary due to insignificant counts on mass 233 (<2 cps). Repeated analyses of 170 pg aliquots of the University of Maryland Os standard solution (UMd, also referred to as UMCP) at Durham University gave $^{187}Os/^{188}Os$ mean values of 0.11384±16 (2σ, n=19) and 0.11379±14 (2σ, n=39) for the two time periods of analysis, April 2000 - November 2000 and April 2006 - May 2008, respectively. This is in good agreement with a value of 0.113787±7 for much larger aliquots (10-100 ng/g) measured on the same mass spectrometer in Faraday cup mode (Luguet et al., 2008). Rhenium was analysed by inductively-coupled plasma mass spectrometry (ICP-MS) on a ThermoFinnigan® Element 2. Solutions were introduced using a MicroMist micro-concentric nebuliser and ESI stable sample introduction system (dual-cyclonic quartz spray chamber). Standard solutions (1 ng/g Re) were analysed at the start, middle and end of each session to determine mass fractionation.

The Carius tube digestions (April 2000 – May 2006) and HPA digestions (November 2007 – May 2008) gave, respectively, mean total procedural blanks of 0.43 and 0.32 pg for Os, 2.5 and 1.9 pg for Re and $^{187}Os/^{188}Os$ ratios of 0.143 and 0.192. The blank corrections relate to the appropriate reagent batch rather than a long-term mean, but were always less than 0.1% for concentration and isotope composition.

Reproducibility of samples. Duplicate digestions of the same sample powder (n=3) indicate that reproducibility of $^{187}Os/^{188}Os$ is 0.15% and 0.7% (2 RSD) for an Os-rich (PI-26, ~2 ng/g Os) and Os-poorer sample (CS-7, 0.46 ng/g), respectively. Os concentrations are reproducible to ~5% (2 RSD) and 11% for PI-26 and CS7, respectively, and Re concentrations (n=2) vary by less than 1% (2 RSD) for both samples (Re: 0.25 and 0.50 ng/g, respectively). The accuracy of measurements is more difficult to evaluate, but reproducibility using two different digestion techniques for these samples and for reference materials (Dale et al., 2008) suggests that incomplete digestion and/or sample-spike equilibration is unlikely to be a significant consideration.

4. Results

4.1 Re and Os elemental data

Osmium concentrations range from 1.50 to 4.02 ng/g in the West Greenland picrites and from 0.435 to 3.45 ng/g in the Baffin Island picrites (Table 1). The median of 2.49 ng/g for the WG suite is higher than for the Baffin suite (1.66 ng/g). All samples have much greater Os concentrations than MORB, which have a range of <0.001 to 0.25 ng/g (Roy-Barman and Allegre, 1994; Gannoun et al., 2007), and the highest Os concentrations are greater than approximate averages for primitive or depleted mantle (~3.1-3.7 ng/g, Morgan et al., 2001; Becker et al., 2006; Harvey et al., 2006). Rhenium concentrations range from 0.113 to 0.506 ng/g in the BI picrites, with a median of 0.32 ng/g, while the WG samples have a larger range (0.063 to 1.14 ng/g, including two samples with

anomalously high Re concentrations from the Anaanaa Member) but a similar median of 0.29 ng/g. Such Re abundances are lower than MORB (0.34 – 2.28 ng/g, median ~1 ng/g, Schiano et al., 1997; Sun et al., 2003; Gannoun et al., 2007). There is little overall covariation of Os and Re in the picrites (not shown), although certain sub-sets, such as samples from Padloping Island, tend to have increasing Re concentrations with decreasing Os and this is broadly true of the BI picrites as a whole.

Figure 1 here.

Osmium concentrations decrease with decreasing MgO (Figure 1) and Ni (not shown). For a given MgO content, picrites from WG have higher Os abundances than those from Baffin Island, while Icelandic picrites are lower in Os than all the early plume North Atlantic picrites (NAP) (Brandon et al., 2007). Rhenium concentrations vary less systematically with MgO content, although a weak negative co-variation can be seen, particularly in the Baffin samples. With the exception of sample CS-7, and two Anaanaa (WG) samples with anomalously high Re content, Re/Os ratios tend to increase with decreasing MgO content in all of the suites (not shown). Re concentrations of Icelandic picrites are broadly comparable to the NAP, but the lower Os concentrations result in higher Re/Os ratios in the Iceland suite (Brandon et al., 2007).

Table 1 here.

4.2 Os isotope data

Baffin Island picrites have a range of $^{187}Os/^{188}Os$ from 0.1269 to 0.1344, which, when corrected for ingrowth of ^{187}Os since the time of emplacement (61 Ma), gives a narrow range of initial $^{187}Os/^{188}Os$ ratios from 0.1267 to 0.1287. The most radiogenic value corresponds to the picrite with the lowest Os concentration (~0.45 ng/g, Figure 3) and therefore crustal contamination is a likely mechanism for increasing $^{187}Os/^{188}Os$. If this value is omitted the range of $^{187}Os/^{188}Os$ is

considerably more limited (0.1267 to 0.1278). The WG picrites have a larger range of $^{187}Os/^{188}Os$ and $^{187}Os/^{188}Os_{initial}$ of 0.1271 – 0.1332 and 0.1267 - 0.1322, respectively. However, the least radiogenic WG samples form a peak on an initial $^{187}Os/^{188}Os$ probability density plot at 0.12725 (Figure 2), which is indistinguishable from the BI peak (0.12717). The sub-chondritic to supra-chondritic range of Os isotope values observed in a different suite of BI picrites (Kent et al., 2004) was not found in this study. Source variability or interaction with lithospheric mantle are both possible origins for the difference between the BI suites, although Kent et al. (2004) concluded that the latter was not consistent with other isotope data.

The WG picrites analysed here, and by Schaefer et al. (2000), extend to more radiogenic Os isotope compositions than the BI picrites. $^{187}Os/^{188}Os$ varies with stratigraphy in the Vaigat formation. The picrites from the Anaanaa and Naujánguit Members possess only the least radiogenic signature (Figure 3). In contrast, $^{187}Os/^{188}Os$ ratios matching this signature were not found in the slightly younger Ordlingassoq Member, which instead have initial $^{187}Os/^{188}Os$ of 0.1294 or greater. Such $^{187}Os/^{188}Os$ ratios are more radiogenic than estimates of putative present-day primitive mantle (0.1296, Meisel et al., 2001) but do not extend as high as WG picrite data from the literature (up to 0.1371, Schaefer et al., 2000). The much younger picrites from Iceland are characterised by more radiogenic $^{187}Os/^{188}Os$ (up to 0.1378, Brandon et al., 2007) which, in part, may correlate with their generally lower MgO contents. The main NAP $^{187}Os/^{188}Os$ peak (Figure 2 and Figure 5) is also significantly less radiogenic than almost all OIB (Figure 5).

Figure 2 here.

With the possible exception of the sample with the lowest Os concentration, there is no covariation of $^{187}Os/^{188}Os$ with Os concentration in either Baffin or West Greenland picrites (Figure 3). Significant assimilation of crust or sub-continental lithospheric mantle (SCLM) would likely result in samples falling on mixing lines towards either an Os-poor, ancient, extremely radiogenic crustal

component or an Os-rich, ^{187}Os-depleted SCLM; such trends are not observed. Equally, if Os-poor samples are derived from parental melts with lower Os content, then these melts would be more susceptible to crustal contamination, and thus are more likely to be radiogenic, and this is not observed. Previous WG data appear to display increased $^{187}Os/^{188}Os$ with decreasing Os concentrations (Schaefer et al., 2000). This may indicate crustal assimilation, or possibly preferential sampling of Os-poorer radiogenic material during mantle melting, for the least Os-rich samples. However, this is only true for samples with <0.5 ng/g Os; such samples have been omitted from subsequent figures.

Figure 3 here.

5. Discussion

5.1 Re-Os elemental behaviour

Olivine accumulation in all the NAP studied is indicated by sample MgO contents of up to 27.0 wt. %, compared to an estimated parental magma of 18.5-21 wt. % MgO (e.g. Pedersen, 1985; Graham et al., 1998; Larsen and Pedersen, 2000). Osmium concentrations decrease with decreasing MgO (Figure 1) and with Ni (not shown), reflecting the compatibility of Os within the olivine- and sulphide-rich crystallising assemblage, which has previously been recognised in global MORB and OIB (e.g. Burton et al., 2002). Therefore, Os concentrations are likely controlled by olivine fractionation and/or accumulation. Rhenium concentrations in the picrites are lower than MORB which is consistent with the moderate incompatibility of Re during mantle melting and the higher melt fraction of picritic melts. Re/Yb ratios are comparable to MORB (~0.00025). The relative incompatibility of Re in olivine and associated crystallising phases (e.g. sulphide +/- spinel), compared to Os, is also illustrated by increasing Re/Os ratios with decreasing MgO content in almost all the picrite units.

As osmium has a very high sulphide-silicate melt partition coefficient (e.g. ~10,000, Crocket et al., 1997), the estimated Os concentrations for the parental melts of NAP (1-2 ng/g) suggest complete consumption of sulphides in at least part of the melting column. The consumption of sulphide in a fertile upper mantle source is thought to occur at melt fractions of >20-25%, based on the sulphide content of the mantle and sulphur solubility in mafic melts (e.g. Mavrogenes and O'Neill, 1999; Lorand et al., 2003), or may occur at lower melt fractions given a previously depleted mantle source. These melt fractions are consistent with estimates based on MgO content: ~20-28% for a fertile source or 10-11% for a depleted source (Herzberg and O'Hara, 2002). In addition to sulphide-silicate partitioning, physical entrainment of liquid sulphide within a high-degree silicate melt (Ballhaus et al., 2006) may, at least in part, account for the very high Os concentration of the NAP parental melts.

West Greenland picrites have higher Os for a given MgO content compared to BI picrites, which could reflect a greater degree of melting. This has been suggested independently on the basis of lower MgO content in the Baffin picrites parental melt (Francis, 1985, based on interpretation of Fo-rich olivines as xenocrysts), although other studies have not concurred with this interpretation (e.g. Kent et al., 2004). Alternatively, assuming the same degree of melting, a sulphide-poorer source may result in complete sulphide consumption in a greater proportion of the melting region and therefore lead to higher Os abundances in melts with similar MgO.

5.2 Osmium-neodymium isotope systematics

The majority of NAP analysed here have Nd isotope compositions similar to Icelandic lavas and consistent with a source dominated by a depleted mantle component ($^{143}Nd/^{144}Nd$ ~0.51307), as previously noted by Holm et al. (1993). However, none of the picrites are as depleted as estimates of DMM (Salters and Stracke, 2004; Workman and Hart, 2005), or the depleted end-member of the Iceland array (e.g. Taylor et al., 1997; Thirlwall et al., 2004). There is considerable $^{143}Nd/^{144}Nd$

variation in BI and Anaanaa (WG) picrites, ranging from 0.51273 to 0.51305 (Figure 4; Graham et al., 1998; Starkey et al., 2007), while $^{187}Os/^{188}Os$ is largely constant at around 0.127. The lower Nd isotope limit is defined by sample CS7 which appears, on the basis of Os isotopes and a previous study (Stuart et al., 2003), to contain a crustal component, but even when omitting this sample there is significant $^{143}Nd/^{144}Nd$ variation. In contrast, the younger Ordlingassoq Member of the WG suite displays a negative co-variation of Os and Nd isotopes with $^{143}Nd/^{144}Nd$ decreasing from 0.51308 to 0.51291 as $^{187}Os/^{188}Os$ increases from 0.1267 to 0.1322 (Figure 4). The data of Schaefer et al. (2000), derived from the Naujánguit and Ordlingassoq Members, possess similar systematics to the Ordlingassoq samples in this study (but not the Naujánguit Member), although the data are more scattered.

5.2.1 Assessing the effects of post-melting crustal or lithospheric interaction

The NAP were erupted through old continental crust, therefore it is important to assess the potential effects of assimilation of such material. The difficulty in assessing the isotopic effects of such contamination is that continental crust is markedly heterogeneous in terms of Os (e.g. Peucker-Ehrenbrink and Jahn, 2001). The Os isotope composition of local crust has not been measured, and measurements, even if numerous, are unlikely to provide a sufficiently well-constrained average due to the varied basement geology. Baffin Island basement rocks display a considerable range of $^{143}Nd/^{144}Nd$ and Nd concentrations (Theriault et al., 2001), as do local West Greenland sediments, which are the most likely contaminants (Larsen, unpubl.). Using assumed Re and Os concentrations (e.g. Esser and Turekian, 1993; Peucker-Ehrenbrink and Jahn, 2001) and knowledge of Re-Os behaviour, the Os isotope composition of a possible contaminant has been estimated for 2.5 Ga continental crust, and assimilation of this material has been modelled. Osmium and Re concentrations of 5, 10, 30 pg/g and 500, 300, 200 pg/g (high Re/Os, best-estimate and low Re/Os, respectively) have been combined with average Nd concentrations and $^{143}Nd/^{144}Nd$ of 24 µg/g and 0.5109, and 39 and 0.5111 for BI basement and WG metasediments, respectively (Theriault et al.,

2001, Larsen, unpubl.). The area defined by possible mixing lines for the entire range of Nd and Os concentrations and varying Sm/Nd and Re/Os ratios is shown by the grey field in Figure 4.

Mixing of parental picritic melt with average/best estimate metasedimentary basement does not produce either of the data arrays observed (Figure 4). Assimilation cannot produce the observed variation in $^{143}Nd/^{144}Nd$ without a noticeable change in $^{187}Os/^{188}Os$. However, mixing of relatively high Sm/Nd crust could produce the array with Os and Nd isotope co-variation. Mixing curves in Os-Sr isotope space are also consistent with this conclusion (not shown).

On a qualitative level, the observation that Anaanaa and Ordlingassoq samples display different Nd-Os isotope trends, despite being emplaced in the same region, makes crustal contamination an unlikely possibility for both trends. However, such a difference could result from changes in magma plumbing and such stratigraphically variable contamination has been recorded in other flood-basalt suites (e.g. Thompson et al., 2001). The similarity of major element (TiO_2) and Os-Nd isotope compositions of Baffin Island and Anaanaa picrites also cannot easily be accounted for by assimilation. Crust-melt mixing may also result in co-variation of $^{187}Os/^{188}Os$ with Os concentration (depending on concentrations of contaminant and melt), and this is not observed (Figure 3).

A further and more conclusive line of evidence to suggest that crustal assimilation is not a significant factor in controlling the isotope compositions of the NAP is presented by Starkey et al. (2007) and Starkey et al. (in review). The overall trend of increasing Nb/Sm with decreasing $^{143}Nd/^{144}Nd$ in their dataset cannot be accounted for by assimilation of any low-Sm/Nd crustal material. Very small amounts of crustal contamination can explain the spread in $^{143}Nd/^{144}Nd$, but cannot also account for the relative enrichment in incompatible elements. Therefore, Nd isotope variations (and by inference Os isotopes) appear to be controlled by source heterogeneity of the mantle.

Figure 4 here.

5.2.2 Generation of the osmium and neodymium isotope variations in the mantle source

The upper part of the Vaigat formation (WG) records increasing $^{187}Os/^{188}Os$ combined with decreasing $^{143}Nd/^{144}Nd$. If this cannot be adequately explained by melt-crust interaction, such a shift to slightly more enriched Os and Nd isotope compositions may reflect an increased input from an enriched component. For instance, modelled mixing of depleted MORB mantle (DMM) with 2 Ga recycled oceanic crust and associated sediment can produce the negative array in Os-Nd isotope space (Figure 4, e.g. 5-10% recycled component containing 5-10% sediment). While this model indicates that recycled oceanic crust and sediments are a plausible contributor, such material is not necessarily required and it is possible that other enriched material, such as metasomatised oceanic lithosphere (e.g. Niu and O'Hara, 2003) could produce the negative array.

The variation of $^{143}Nd/^{144}Nd$ with no complementary variation of $^{187}Os/^{188}Os$ is more difficult to explain because old recycled oceanic crust (plus sediment), and old continental crust, both possess complementary high $^{187}Os/^{188}Os$ and low $^{143}Nd/^{144}Nd$. However, we suggest that the mantle source of the Baffin and Anaanaa picrites contained Nd isotope heterogeneity as a result of a minor (sulphide-poor?) pyroxenite component that, due to its high Nd content and low Os content, significantly affected the Nd isotope composition of the melt without noticeably affecting Os.

5.3 Evolution of the Iceland plume: Os, Nd and He evidence

Initial volcanism, derived from high degree melting, was characterised by approximately chondritic Os isotope compositions, the highest known $^{3}He/^{4}He$ ratios and variable, but fairly radiogenic

342 $^{143}Nd/^{144}Nd$ ratios. Osmium isotope compositions are consistent with a depleted, but not ancient
343 source, with no significant contribution from any enriched component, including the outer core.
344 Subsequent melts erupted within 1 Myr of the earliest picrites (Ordlingassoq Member) have
345 elevated $^{187}Os/^{188}Os$ indicating a contribution from enriched material, possibly old recycled crust
346 plus sediment, which was not tapped in the first phase of plume melting.

347 Osmium and Nd isotope data for Iceland picrites (Brandon et al., 2007) are most easily explained by
348 a similar recycled crustal component including a small percentage of sediment (~5%), mixed with a
349 DMM component with higher $^{143}Nd/^{144}Nd$ than the depleted end-member of early NAIP basalts
350 (Figure 4). High $^{3}He/^{4}He$ ratios persist in the Iceland plume (up to 38 Ra in late Tertiary volcanics,
351 Hilton et al., 1999). Brandon et al. (2007) report elevated ^{187}Os, but not ^{186}Os, indicating the
352 continued lack of core contribution to the Iceland plume source. Intriguingly, Brandon et al. (2007)
353 found that $^{3}He/^{4}He$ increases with $^{187}Os/^{188}Os$ in young Icelandic picrites (Figure 6b), despite the
354 probable requirement of recycled oceanic crust which would possess very low $^{3}He/^{4}He$. Brandon
355 and co-authors explained this by two-stage mixing (firstly combining high $^{3}He/^{4}He$ mantle and
356 recycled crust, then mixing with DMM), although the generation of a largely linear array in He-Os
357 isotope space is difficult to reconcile with the vastly different He concentrations in DMM and a
358 hybrid recycled crust-high $^{3}He/^{4}He$ source, regardless of the ultimate source of the latter.

359

360 **5.4 The $^{187}Os/^{188}Os$ composition of convecting mantle**

361 Trace element and Nd-Sr isotope evidence (Stuart et al., 2003; Ellam and Stuart, 2004; Kent et al.,
362 2004) indicates that the NAP source is typically depleted with respect to putative primitive mantle
363 (e.g. Zindler and Hart, 1986), and largely indistinguishable from the MORB source mantle (DMM,
364 e.g. Salters and Stracke, 2004; Workman and Hart, 2005). The $^{187}Os/^{188}Os$ compositions of the
365 earliest NAP (0.1267 to 0.1280) are consistent with a depleted source containing no significant

366 contribution from recycled crust, pyroxenite, sediment, recycled SCLM, metasomatised peridotite
367 or outer core, nor any isolated ancient depleted domains. The absence of enriched components in
368 the source is also supported by the Ni content of picritic olivines. Ni/Mg ratios in the most Mg-rich
369 olivines (Fo >89) from the Vaigat formation, WG (Larsen and Pedersen, 2000) are lower than those
370 of other within-plate magmas defined by Sobolev et al. (2007), and also in fact largely lower than
371 the peridotite range. Although this indicator of pyroxenite contribution has been questioned (see
372 Niu and O'Hara, 2007), such low Ni/Mg ratios suggest little or no pyroxenite contribution to the
373 melt. Prytulak and Elliott (2007) proposed that the reference TiO_2 concentration of a OIB suite
374 (estimated parental melt content, defined as the TiO_2 content on the liquid line of descent typically
375 at 12.5% MgO), increases with increasing contribution from recycled oceanic crust. The estimated
376 reference TiO_2 content for the early NAP is ≤1%, lower than all OIB suites compiled by Prytulak
377 and Elliott (2007) except Iceland, and thus the NAP define the low enriched component end-
378 member for within plate magmas, i.e. their sources are dominated by peridotite. The reference TiO_2
379 content for Iceland of ~1% (Prytulak and Elliott, 2007) seems low compared to many Icelandic
380 lavas and thus perhaps does not accurately represent the enriched components which have
381 contributed to recent Icelandic magmas.

382 Due to the large degree of melting during the generation of the NAP, their Os isotope composition
383 will closely reflect the composition of the bulk mantle source. This, coupled with the chemical
384 similarity to DMM and absence of enriched components, gives the potential for providing an
385 estimate of the bulk $^{187}Os/^{188}Os$ composition of the convecting upper mantle/DMM. To facilitate
386 comparison with other data such as those for MORB, initial $^{187}Os/^{188}Os$ for picrites, platinum-group
387 alloys (PGA) and chromites have been recalculated to the present day by assuming chondritic
388 evolution of their sources since the time of mantle melting. Strictly speaking the evolution of Os
389 isotopes since melting may be depressed in relation to chondrite (due to Re depletion) and the true
390 comparative value will be between the initial and re-corrected values. However, the errors

associated with such a correction, given the limited time periods, will be negligible (60 Ma for NAP, 177 Ma for Tibetan PGA).

On the basis of the probability peak for the NAP, the present day $^{187}Os/^{188}Os$ composition of convecting mantle is estimated to be 0.1276 (Figure 5). This is indistinguishable from the main PGA peak (0.1276), derived mainly from Tibetan ophiolites (Pearson et al., 2007; Shi et al., 2007). Recent to Phanerozoic ophiolite-derived chromites define a peak at $^{187}Os/^{188}Os = 0.1283$ and regression of $^{187}Os/^{188}Os$ versus time gives a mean of 0.1281, which is closer to the NAP value (Walker et al., 2002b). It is possible that the chromite value could be slightly elevated by subduction-related enrichment of Re (time-integrated) or radiogenic Os. The $^{187}Os/^{188}Os$ estimate defined by NAP and PGA is intermediate between averages for carbonaceous, enstatite and ordinary chondrites (0.1262, 0.1281, 0.1283, respectively, Walker et al., 2002a). It is, however, significantly lower than the proposed $^{187}Os/^{188}Os$ of 0.1296 for putative primitive upper mantle (PUM) (Meisel et al., 1996). In addition to the effect of time-integrated depletion of Re through the generation of continental crust, this difference may arise because many of the peridotites used for regression by Meisel et al. (2001) have been refertilised and radiogenic Os has been added. The similarity between the modal NAP Os isotope composition and other estimates of ambient convecting mantle indicate that a $^{187}Os/^{188}Os$ value somewhere between 0.1275 and 0.1281 might be an appropriate estimate for ambient convecting mantle from which to calculate Re-Os model ages for peridotites.

Figure 5 here.

The least radiogenic MORB ($^{187}Os/^{188}Os$ of 0.1261-0.1272) are isotopically similar to the NAP $^{187}Os/^{188}Os$ peak of 0.1276, but most MORB are significantly more radiogenic with $^{187}Os/^{188}Os$ ratios up to 0.148 (Gannoun et al., 2007). This probably reflects the presence of enriched components within the DMM and the preferential sampling of these components by the smaller

415 degree melts that constitute MORB, relative to the NAP (e.g. Escrig et al., 2005). If such enriched
416 components are present in the initial NAP source (which the Nd isotope data suggest), their
417 signature is insignificant compared to the contribution from depleted peridotite.

418 Walker et al. (2002b) estimated the amount of isolated mafic slab material required in the mantle to
419 account for the difference (1.2%) between their estimate of the $^{187}Os/^{188}Os$ composition of
420 convecting upper mantle (0.1281, defined by chromites) and the estimate for PUM (0.1296, Meisel.
421 Their calculations suggested 1.8% of the entire mantle mass must be isolated slabs for a 1.8 Ga
422 isolation age and 3.4% for 1 Ga isolation. The difference between convecting mantle and PUM
423 $^{187}Os/^{188}Os$ based on the NAP and Tibetan PGA grains is 1.5% and therefore >2% and ~4% isolated
424 slab material is required for 1.8 and 1 Ga isolation ages, respectively. However, the probable
425 extensive Re loss from basaltic oceanic crust during subduction, and the lower Re content of
426 gabbroic crust (Dale et al., 2007) would lead to greater required inputs, unless the lost Re was
427 incorporated and isolated in arc/continental crust. These calculations are based on depletion of only
428 half the total mantle mass, if whole-mantle convection is predominant, estimates could be at least
429 twice as high. However, as noted above, the likely metasomatic sulphide input to some of the more
430 radiogenic peridotites used to constrain PUM (Meisel et al., 2001) means that 'primitive' mantle
431 may not be accurately estimated.

432 **5.5 Implications for high $^{3}He/^{4}He$ in the mantle**

433 Based on the greater compatibility of He than U and Th during mantle melting (Graham et al.,
434 1990; Parman et al., 2005), it has been proposed that ancient melt depletion will result in reduced
435 ingrowth of ^{4}He and therefore high $^{3}He/^{4}He$ in the residue (Class and Goldstein, 2005; Parman,
436 2007). According to a model matching common $^{3}He/^{4}He$ values in oceanic basalts with continental
437 crust age peaks (Parman, 2007), the source of $^{3}He/^{4}He$ up to 50 R_a must have been depleted by
438 melting and isolated since ~3.7 Ga, or alternatively at least 1 Ga in another model by Class and

Goldstein (2005). Depletion of moderately incompatible Re during mantle melting leads, over time, to $^{187}Os/^{188}Os$ ratios which deviate below the primitive mantle evolution curve. A source depleted at 3.7 Ga or 1 Ga, and subsequently isolated, should have $^{187}Os/^{188}Os$ of ~0.102 or ~0.122, respectively. The Os isotope data for NAP are more radiogenic, and are therefore not consistent with ancient depletion as a source for high $^{3}He/^{4}He$. Even the lowest initial $^{187}Os/^{188}Os$ corresponds to a Re-depletion model age (T_{RD}) of only ~0.4 Ga. T_{RD} ages assume that Re is entirely depleted during melting, and therefore ingrowth of ^{187}Os ceases after depletion, therefore T_{RD} are minimum ages, but even minor Re depletion as a result of small degree melting would depress $^{187}Os/^{188}Os$ evolution further than observed. For depletion to be the mechanism by which high $^{3}He/^{4}He$ are generated, radiogenic Os, but not He, must have been added subsequently. Sulphide addition could provide such a mechanism but would likely be accompanied by a flux of other elements.

Figure 6 here.

The core has also been proposed as a possible source of He enriched in ^{3}He (e.g. Macpherson et al., 1998; Porcelli and Halliday, 2001). The proposed partitioning behaviour of Re and Os (and Pt) between the inner and outer core has led to the hypothesis that the outer core will possess supra-chondritic Re/Os ratios (and Pt/Os, e.g. Brandon et al., 1998). Therefore, the approximately chondritic $^{187}Os/^{188}Os$ of extremely high $^{3}He/^{4}He$ picrites studied here precludes a bulk contribution from core material. However, decoupling of core-mantle transfer of Os and He could result in high $^{3}He/^{4}He$ without correspondingly high $^{187}Os/^{188}Os$.

The high $^{3}He/^{4}He$ component cannot be linked to mantle with a specific Os isotope signature, because $^{3}He/^{4}He$ ratios up to 48 are found in picrites with $^{187}Os/^{188}Os$ of ~0.1267 to 0.132. However, if the elevated $^{187}Os/^{188}Os$ can be explained by a minor recycled crust component (small enough not to overwrite the high $^{3}He/^{4}He$ signature, c.f. Macpherson et al. (2005) for Iceland), or metasomatic sulphide transfer from such sources, then the end-member high $^{3}He/^{4}He$ component

would have a $^{187}Os/^{188}Os$ of ~0.1276 – defined by the NAP, indistinguishable from the main PGA peak and similar to the least radiogenic MORB. It is not possible to conclude, from these data, the ultimate source of the high $^{3}He/^{4}He$ signature that was tapped most efficiently during melting of the plume-head and persists in Icelandic volcanism today. However, it is possible that He is decoupled from Os and Nd and could reflect a 'primitive' component which is volumetrically small, hence not changing Os or Nd isotopes significantly, but rich enough in He to strongly affect the $^{3}He/^{4}He$ ratio. Alternatively, although the mechanisms are not well understood, it has been proposed that a high $^{3}He/^{4}He$ component could be present in some upper mantle material (e.g. Meibom et al., 2003), although, if so, such material must only be sampled during episodes of large degree melting of hot mantle.

6. Concluding remarks

Osmium concentrations in NAP are very high (up to 4.02 ng/g, mean: 2.15 ng/g) and suggest complete consumption of sulphide in at least part of the source due to large degree melting (≥20% for a fertile source). This is consistent with previous evaluations of the degree of melting based on MgO-rich olivines: 10-11% for depleted mantle, >20-28% for fertile mantle (Herzberg and O'Hara, 2002). Initial Os isotope compositions in the earliest picritic melts from West Greenland (Anaanaa and Naujánguit Members of the Vaigat formation) and Baffin Island are uniform and broadly chondritic (probability peak of $^{187}Os/^{188}Os = 0.1272$). This initial volcanism cannot contain any significant contribution from the outer core or from old recycled crustal material, as both would impart a radiogenic $^{187}Os/^{188}Os$ signature. In addition, minimal presence of enriched pyroxenitic components in the source of NAP is suggested by various parameters such as Ni content of olivine and TiO_2 content. The absence of enriched components, coupled with a large degree of melting, means that the average $^{187}Os/^{188}Os$ (corrected to 0.1276 for the present day) may reflect the bulk $^{187}Os/^{188}Os$ of convecting mantle. This value is similar to the least radiogenic MORB (e.g.

Gannoun et al., 2007) and the most common $^{187}Os/^{188}Os$ ratios found in PGA grains from Tibetan ophiolites (Pearson et al., 2007; Shi et al., 2007). Subsequent melts sampled from the Ordlingassoq Member (upper Vaigat formation), erupted within 1 Myr of the earliest melts (Storey et al., 1998), possess supra-chondritic initial $^{187}Os/^{188}Os$ ratios of up to 0.1321, which can be accounted for (though not exclusively) by a greater contribution from recycled crust. These more enriched signatures are seen in more recent melts from the Iceland plume (Brandon et al., 2007).

Models seeking to explain high $^{3}He/^{4}He$ ratios by ancient depletion (Class and Goldstein, 2005; Parman, 2007) are not supported by the uniform and largely chondritic $^{187}Os/^{188}Os$ ratios of NAP which possess the highest known mantle-derived $^{3}He/^{4}He$ ratios (up to 50 Ra, Stuart et al., 2003). If ancient melting and isolation had occurred, the depletion of Re during melting would lead to significantly sub-chondritic $^{187}Os/^{188}Os$ in the depleted source (~0.122 and ~0.102 for depletion ages of 1 Ga and 3.7 Ga, Class and Goldstein (2005), and Parman (2007) models respectively). Conversely, outer core material would impart radiogenic Os to the plume and so core material is also not supported as a source of high $^{3}He/^{4}He$ unless the mechanism of He transfer to the plume is decoupled from Os. High $^{3}He/^{4}He$ mantle is not otherwise isotopically distinct from a typical upper mantle source, in terms of Os or other radiogenic isotopes. Therefore two possible explanations for the high $^{3}He/^{4}He$ signature are: (i) it is present in a typical upper mantle source which has been entrained in the plume-head, but is only tapped during episodes of high-degree melting of hot mantle; (ii) the He isotope signature is entirely decoupled from other elements and is dominated by addition of a He-rich, high $^{3}He/^{4}He$ component, probably primordial in nature, without complementary addition of other elements. The latter is our favoured model.

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main text: ~6330

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Figure captions

Figure 1. Co-variation of Os and Re with MgO in West Greenland, Baffin Island and Iceland picrites. Icelandic picrite data from Brandon et al. (2007).

Figure 2. Probability density plot for initial $^{187}Os/^{188}Os$ in (a) Baffin Island & West Greenland picrites (NAP) from this study and (b) NAP and Iceland picrites from the literature. Other data: [1] Schaefer et al. (2000), [2] Brandon et al. (2007), [3] Kent et al. (2004). NAP samples with <0.5 ng/g Os have been omitted due to potential crustal contamination. A 'bandwidth' uncertainty of 0.001 was applied to the $^{187}Os/^{188}Os$ ratio of all samples (~0.8% relative uncertainty).

Figure 3. Initial $^{187}Os/^{188}Os$ (at 61 Ma) plotted against Os concentration. PI, DI and CS are Padloping Island, Durban Island and Cape Searle. An, Nau and Ord are, oldest to youngest, Anaanaa, Naujánguit and Ordlingassoq Members of the Vaigat formation. A lack of co-variation suggests insignificant assimilation of radiogenic ancient crust and/or unradiogenic SCLM. Only the duplicated low Os CS sample has slightly more radiogenic Os than others from Baffin Island, consistent with a minor crustal contribution. Primitive mantle estimate (without uncertainty) from Meisel et al. (2001), abyssal peridotites (AP) from Snow and Reisberg (1995) and Harvey et al. (2006), carbonaceous chondrite (without uncertainty) from Walker et al. (2002a). Other WG data from Schaefer et al. (2000), other BI data from Kent et al. (2004), Hawaii and Iceland data from Brandon et al. (1999; 2007, respectively).

766 Figure 4. (a) $^{187}Os/^{188}Os$ vs. $^{143}Nd/^{144}Nd$ (both age corrected to 61 Ma), for NAP in this study and
767 NAP and Iceland picrites from the literature (Schaefer et al., 2000; Kent et al., 2004; Brandon et al.,
768 2007, respectively). (b) Assessment of mixing enriched materials with mantle. Crustal
769 contamination: black lines – best estimate/average, (Sm-Nd data from Larsen (unpubl.) - thick, and
770 Theriault et al. (2001) - thin), grey area – whole possible range. Enriched plume components in the
771 source: thick green line – 2 Ga oceanic crust with 5-10% sediment, thin green line - 2Ga oceanic
772 crust. Literature data sources - see Figure 3, except DMM Nd estimate from Workman and Hart
773 (2005) and Salters and Stracke (2004). Mixing lines calculated using 2 Ga recycled crust Nd
774 isotope composition estimates from Stracke et al. (2003) and Os compositions calculated from
775 Schiano (1997), Gannoun et al. (2007) and Dale et al. (2007). Crustal Os compositions based on
776 average continental crust (Peucker-Ehrenbrink and Jahn, 2001) and Os data for granites (Dale,
777 unpubl.). Lighter depleted mantle ornament denotes likely end-member for Iceland array.

778

779 Figure 5. Probability density plot for $^{187}Os/^{188}Os$ of NAP (this study and literature) and other direct
780 and indirect indicators of mantle composition. $^{187}Os/^{188}Os$ of the picrites has been corrected for
781 ingrowth since emplacement, but then recalculated to a present day composition (by means of
782 deviation from chondrite evolution curve). Data for chromites[3] (Walker et al., 2002b) are also
783 corrected to present day (* mean value for chromites taken from the authors' regression using
784 $^{187}Os/^{188}Os$ vs. time, not the peak on this plot). Other data sources: [1] Schaefer et al. (2000) and
785 Kent et al. (2004), [2] Pearson et al. (2007) and Shi et al. (2007), [4] Roy-Barman and Allegre (1994),
786 Snow and Reisberg (1995), Brandon et al. (2000) and Harvey et al. (2006), [5] Gannoun et al. (2007),
787 Global OIB data from samples with >30 pg/g Os from: Martin, (1991), Hauri and Hart (1993),
788 Reisberg et al. (1993), Martin et al. (1994), Marcantonio et al. (1995), Bennett et al. (1996), Hauri
789 et al. (1996), Widom and Shirey (1996), Widom et al. (1999). EC, OC and CC are enstatite,
790 ordinary and carbonaceous chondrites, respectively (Walker et al., 2002a), PUM is primitive upper

mantle (Meisel et al., 2001). NAP with <0.5 ng/g Os have been omitted due to potential crustal contamination. A 'bandwidth' uncertainty of 0.001 was applied to the $^{187}Os/^{188}Os$ ratio of all samples (~0.8% relative uncertainty).

Figure 6. (a) He isotope composition against $^{187}Os/^{188}Os$ at the time of eruption (61 Ma) for picrites in this study, and (b) probability density plot for $^{187}Os/^{188}Os$ ratios (not corrected to present day) in picrites from this study, Icelandic picrites (Brandon et al., 2007), global ocean island basalts and picrites, and platinum-group alloys (PGA) from ophiolites in Tasmania, Tibet and the Urals (Pearson et al., 2007). (b) Symbols: see Figure 1. Mantle ages refer to the time of melt depletion. He compositions for these ages are based on: [1] Class and Goldstein (2005), [2] Parman et al. (2007). Helium isotope data from Stuart et al. (2003), Starkey et al. (2007) and Starkey et al. (in review). Also shown are picrites from Hawaii (Brandon et al., 1999).

Table 1

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Table 1. Re-Os isotope and elemental data for North Atlantic picrites (NAP, Baffin Island and Vaigat formation, West Greenland).

	Dig. method	Os ng/g	Re ng/g	Re/Os	$^{187}Os/^{188}Os$	$^{187}Os/^{188}Os_i$	$^{187}Re/^{188}Os$	γOs	MgO wt. %	Ni µg/g	Yb µg/g
Baffin Island											
Padloping Island											
PI-23	CT	1.477	-	-	0.12849	-	-	-	24.51	1127	1.28
PI-24	HPA	1.827	0.173	0.095	0.12775	0.12729	0.457	0.10	26.10	1228	1.27
PI-25	HPA	2.555	0.101	0.040	0.12780	0.12761	0.191	0.35	27.69	1336	1.01
PI-26	HPA	2.028	0.251	0.124	0.12758	0.12698	0.596	-0.15	25.09	1190	1.24
dupl.	HPA	1.927	0.253	0.131	0.12775	0.12711	0.631	-0.04	25.09	1190	1.24
dupl.	CT	1.974	-		0.12760	-	-	-	25.09	1190	1.24
PI-27	HPA	1.369	0.336	0.245	0.12896	0.12775	1.168	0.46	23.20	1058	1.30
dupl.	CT	1.483	-		0.12849	-	-	-	23.20	1058	1.30
PI-31	HPA	1.762	0.294	0.167	0.12816	0.12734	0.803	0.14	22.64	1027	1.48
PI-37	HPA	3.445	0.120	0.035	0.12692	0.12675	0.168	-0.33	26.57	1206	1.07
PI-43	HPA	2.106	0.295	0.140	0.12777	0.12708	0.674	-0.06	24.58	1081	1.37
PAD-6	HPA	1.084	0.419	0.387	0.12878	0.12689	1.86	-1.06	17.20	647	1.70
Durban Island											
DI-23	CT	2.549	0.097	0.038	0.12751	0.12732	0.182	0.12	24.14	1035	1.32
DI-26	HPA	0.909	0.286	0.315	0.12855	0.12701	1.52	-0.11	15.92	546	1.70
DUR-8	HPA	1.548	0.113	0.073	0.12714	0.12679	0.350	-1.16	22.89	856	1.28
Cape Searle											
CS-7	HPA	0.484	0.506	1.044	0.13356	0.12844	5.03	1.06	20.18	831	1.47
dupl.	HPA	0.435	0.504	1.158	0.13442	0.12874	5.58	1.31	20.18	831	1.47
dupl.	CT	0.471	-	-	0.13369	-	-	-	20.18	831	1.47
W.Greenland											
Anaanaa Member											
400444	CT	1.907	0.259	0.136	0.12812	0.12746	0.655	0.23	20.81	769	1.53
400452	HPA	2.489	0.966	0.388	0.12922	0.12732	1.87	-0.72	21.55	913	1.49
400457	CT	3.879	0.359	0.093	0.12765	0.12720	0.446	0.02	22.65	848	1.40
400492	CT	2.533	1.140	0.450	0.13024	0.12804	2.17	0.70	20.05	801	1.57
408001.233	CT	1.503	0.243	0.162	0.12824	0.12746	0.779	0.22	17.99	751	1.45
Naujanguit Member											
113210	CT	3.119	0.206	0.066	0.12725	*0.12693*	0.318	-0.19	20.88	920	
264217	CT	2.786	0.472	0.169	0.12772	0.12689	0.816	-0.21	21.97	949	1.50
332771	CT	1.553	0.425	0.274	0.12837	0.12704	1.32	-0.10	20.09	824	1.57
362149	CT	2.813	0.291	0.103	0.12814	0.12759	0.550	0.33	23.45	1329	1.33
400485	HPA	4.024	0.316	0.079	0.12713	0.12674	0.378	-1.19	27.02	1184	1.17
Ordlingassoq Member											
113333	CT	2.256	0.063	0.028	0.12955	0.12942	0.134	1.77	20.53	935	
138228	CT	1.524	0.330	0.216	0.13321	0.13216	1.04	3.92	17.17	850	
332788	CT	2.034	0.180	0.089	0.13120	0.13094	0.427	2.97	25.62	1306	1.13
332828	CT	2.680	0.367	0.137	0.13060	0.12994	0.660	2.17	22.23	1098	1.35
332901	HPA	1.727	0.071	0.041	0.13123	0.13103	0.198	2.15	24.60	1098	1.22
354754	HPA	2.198	0.179	0.082	0.13254	0.13214	0.393	3.02	20.74	810	1.45
400230	HPA	2.768	0.335	0.121	0.13235	0.13175	0.584	2.72	21.77	935	1.43

Notes:
Reference materials analysed during the period of analysis (using HPA digestion) have been published previously in Dale et al. (2008) and Dale et al. (in review)
$^{187}Os/^{188}Os_i$ – Os isotope composition corrected for ingrowth of ^{187}Os since the time of emplacement (61 Ma).
γOs – deviation of $^{187}Os/^{188}Os_i$ from the chondrite evolution curve: $((^{187}Os/^{188}Os_i/^{187}Os/^{188}Os_{chondrite})-1)*100$.
Digestion method: HPA – high-pressure asher, CT – Carius tube, both digestions in inverse aqua regia.
Sample mass digested approx. 1g in all cases.

Figure 1

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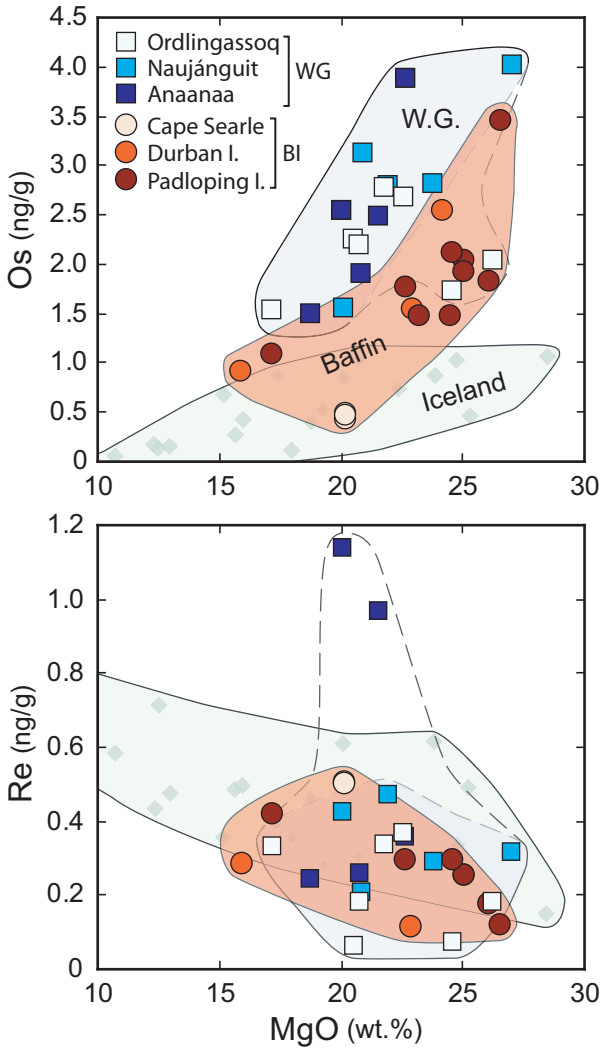

Figure 2

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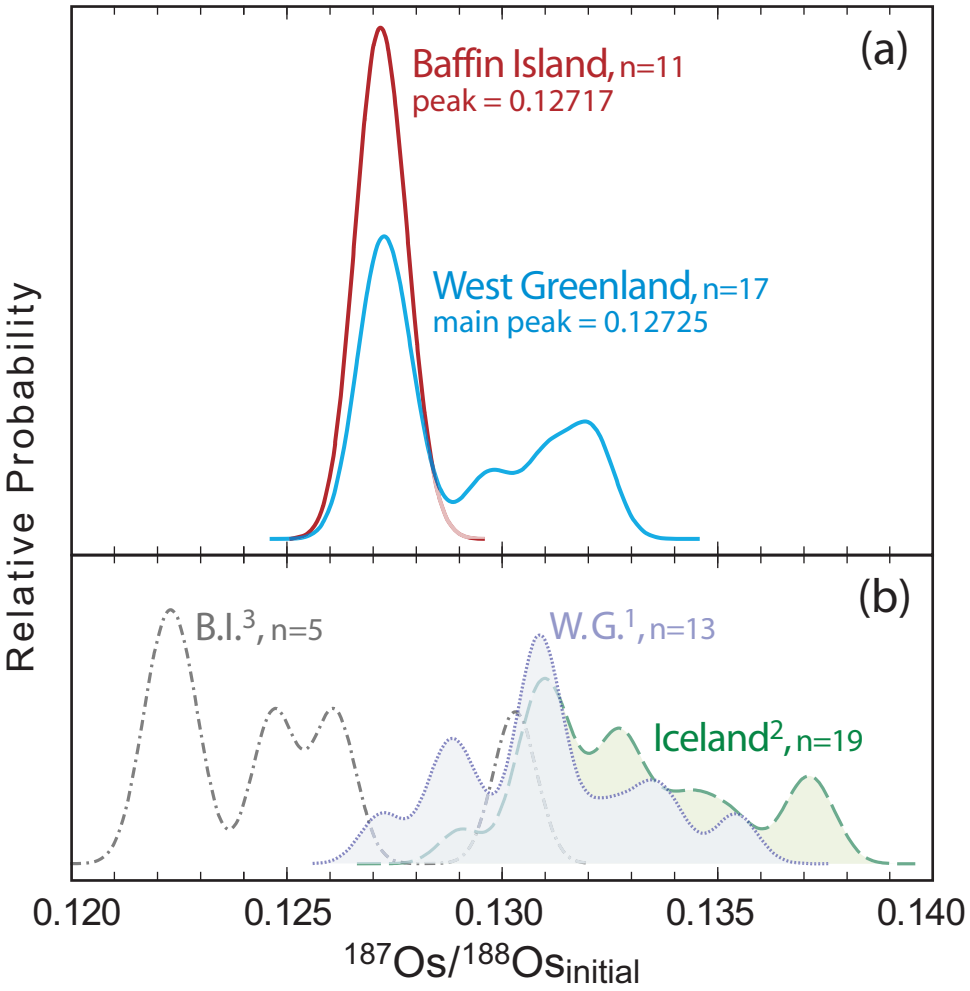

Figure 3

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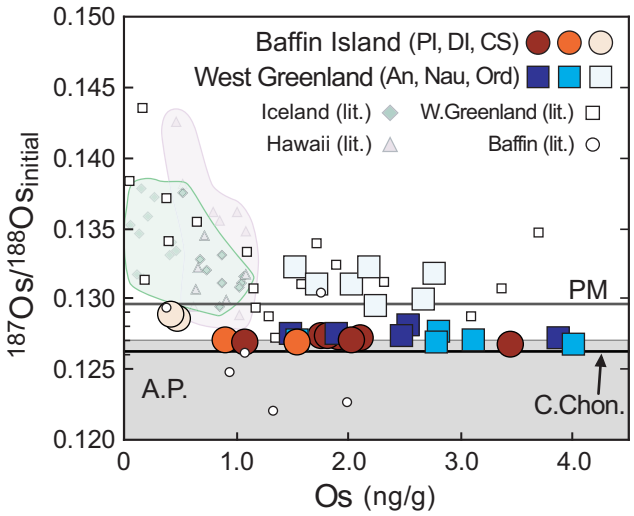

Figure 4

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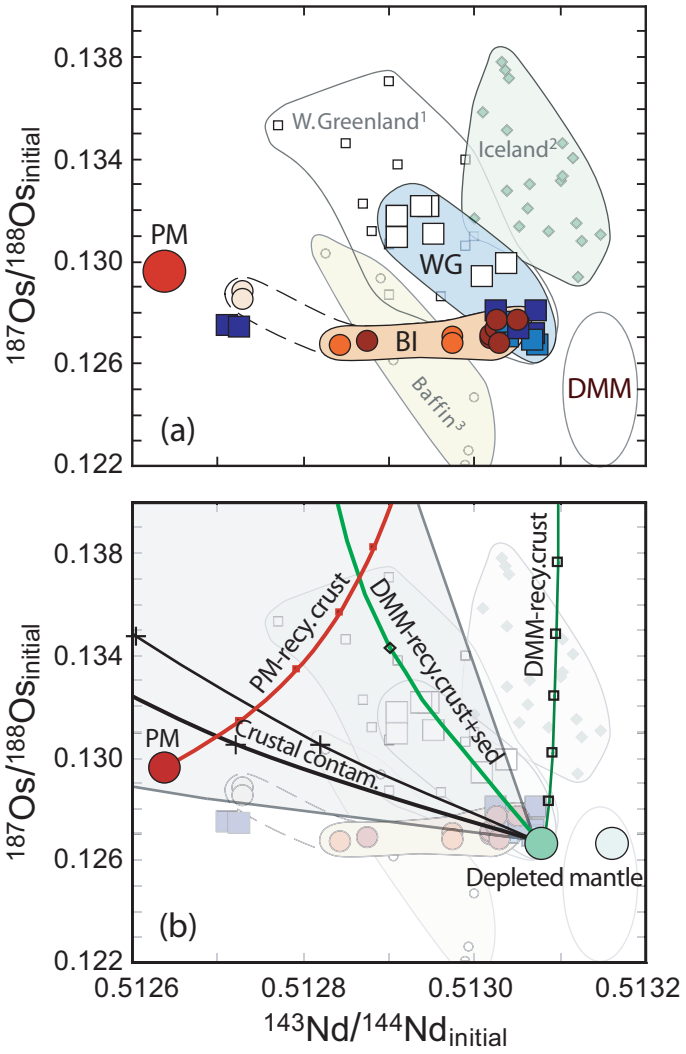

Figure 5

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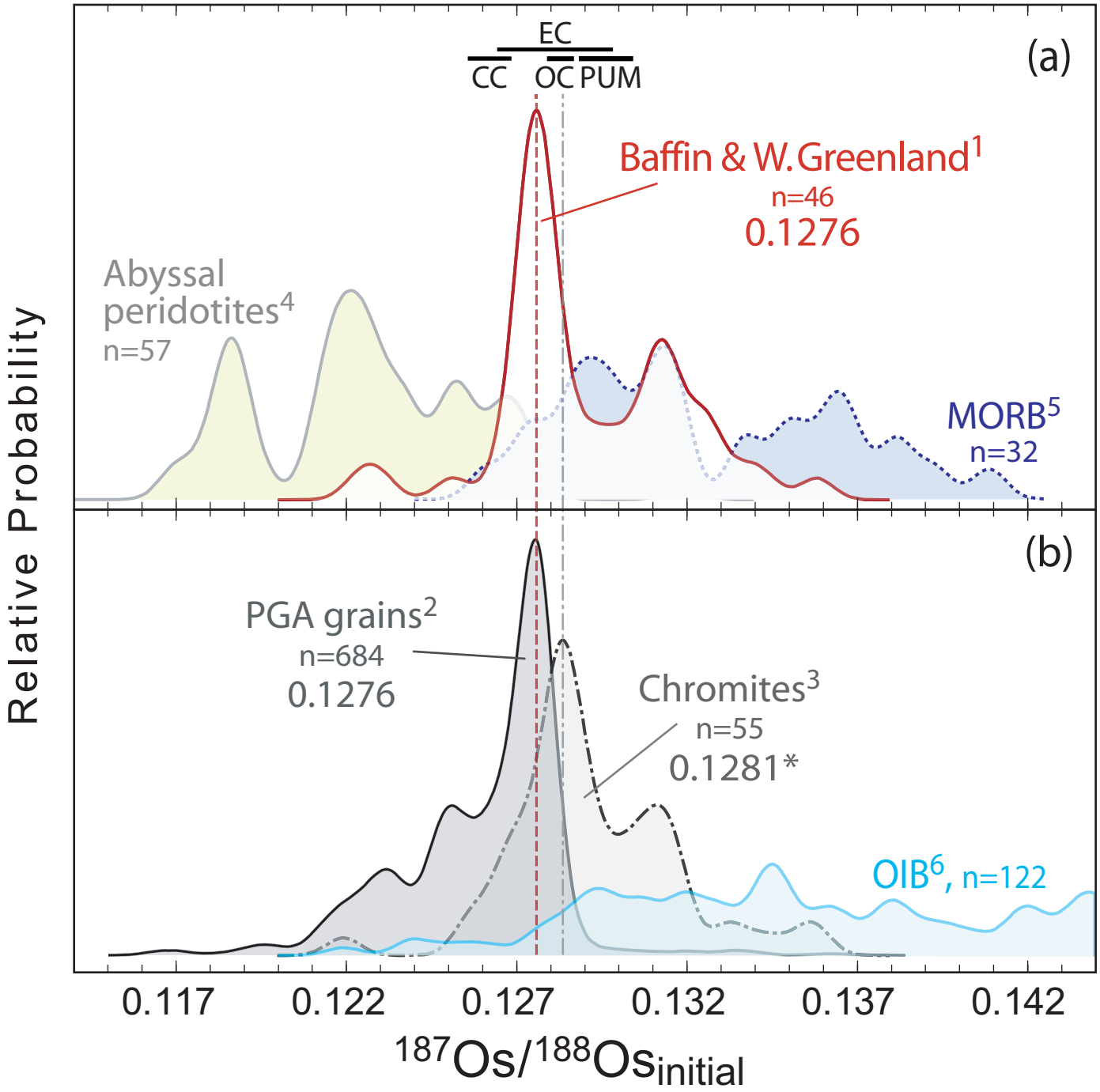

Figure 6

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