1 **Osmium isotopes in Baffin Island and West Greenland picrites:**

2 **Implications for the ^{187}Os/^{188}Os composition of the convecting mantle**

3 **and the nature of high ^{3}He/^{4}He 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 and also has implications for the interpretation of Re-depletion ages in mantle peridotites. As partial melting preferentially samples components with the lowest melting temperatures, large degree melts such as picrites should most closely reflect the peridotitic components within the source. Thus, Re-Os analyses of thirty picrites from Baffin Island and West Greenland are thought to provide a good estimate of the bulk $^{187}Os/^{188}Os$ composition of their convecting mantle source, which is indistinguishable from DMM in terms of lithophile isotopes and trace elements. In addition, the high $^{3}He/^{4}He$ of these rocks allows us to comment on the possible origins 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%) from old recycled oceanic crust, including sediment. However, Baffin Island and the earliest West Greenland picrites are remarkably uniform in composition with $^{187}Os/^{188}Os$ between 0.1267 and 0.1280, and a mean and mode of 0.1272 ± 0.0007. Such Os isotope compositions are less radiogenic than estimates of primitive upper mantle 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 dominated by peridotite.

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 ($^{187}Os/^{188}Os$: <0.115), unless radiogenic Os, but not He, has been subsequently added. Conversely, a bulk outer core contribution would impart a supra-chondritic $^{187}Os/^{188}Os$ signature to the picrites, and thus Os isotopes preclude the core as a

37   source of high ^{3}He/^{4}He, unless core-mantle transfer of Os and He is decoupled. It is possible to

38   broadly account for the Os-He and Os-Nd isotope variations by mixing of depleted MORB mantle,

39   recycled oceanic crust and primitive mantle with high ^{3}He/^{4}He, but it is difficult to explain each

40   individual sample composition in this way. Alternatively, as the high ^{3}He/^{4}He signature is found in

41   samples with variable Os and Nd isotope compositions, it seems likely that He is decoupled from

42   other isotopic tracers and is dominated by minor addition of a He-rich, high ^{3}He/^{4}He component

43   probably of primordial nature, although the ultimate source is unclear from our data.

44   Keywords: osmium isotopes, helium isotopes, Baffin Island, West Greenland, picrite, convecting

45   mantle, depleted mantle.

46   **1. Introduction**

47   Mantle rocks and mantle-derived melts display a broad range of Os isotope compositions, both

48   depleted and enriched with respect to bulk Earth and primitive upper mantle (PUM) estimates. Due

49   to the differential compatibility of Re and Os during mantle melting, crustal rocks typically have

50   very high Re/Os ratios and, over time, evolve to radiogenic ^{187}Os/^{188}Os compared to the mantle.

51   Depleted mantle evolves complementary unradiogenic ^{187}Os/^{188}Os ratios, with the timing of Re

52   depletion indicated by the extent to which ^{187}Os/^{188}Os deviates below a chondritic evolution curve.

53   Therefore, the Re-Os system is a powerful tool with which to assess contributions from depleted

54   mantle and enriched recycled materials to the source of mantle-derived melts (e.g. Shirey and

55   Walker, 1998).

56   Significant Os isotope heterogeneity exists in the mantle at a variety of length-scales from mineral-

57   to vein- to slab-scale due to the recycling of enriched crustal materials and depletion of peridotite,

58   and due to the highly siderophile nature of both Re and Os. Variable degrees of melting of such a

59   heterogeneous mantle will lead to melts that vary in isotopic composition, where melts with the

60 most radiogenic signature are typically derived from the smallest degrees of melting, and large-

61 degree melts give the best estimate of the ^{187}Os/^{188}Os composition of the *bulk* mantle source. This

62 study of North Atlantic (NA) picrites from Baffin Island (BI) and West Greenland (WG) should

63 therefore provide a good estimate for the average Os isotope composition of the whole source

64 volume. Given the lithophile isotope similarity of this source to depleted mid-ocean ridge basalt

65 (MORB) mantle (DMM, e.g. Ellam and Stuart, 2004), this signature should also provide an estimate

66 for the ^{187}Os/^{188}Os of typical convecting mantle without significant contribution from enriched

67 components. Such an estimate is a valuable addition to current constraints on the Os isotope

68 evolution of the Earth and peridotite melting ages (e.g. Meisel et al., 2001; Walker et al., 2002b).

69 The Baffin Island and West Greenland picrites define the high ^{3}He/^{4}He end-member of the mantle

70 range (up to 50 Ra, Stuart et al., 2003; Starkey et al., 2008, in press), which includes the canonical

71 MORB range of 8 ± 1 R_a (Graham, 2003; where $R_a = {}^{3}$He/^{4}He$_{atmosphere} = 1.39$ x 10^{-6}). The picrites

72 were erupted at about 61 Ma, probably due to the onset of the proto-Iceland plume (Saunders et al.,

73 1997), and this melting region continues to produce mantle melts with high ^{3}He/^{4}He in the Iceland

74 region today (Macpherson et al., 2005). Due to the incompatibility of He, conventional wisdom

75 posits that high ^{3}He/^{4}He mantle reservoirs are less degassed than the convecting upper mantle, and

76 retain a component of the Earth's primordial volatile inventory (e.g. Kurz et al., 1982; Moreira et

77 al., 2001; Porcelli et al., 2002). However, the observation that U and Th may be less compatible

78 than He in an olivine-rich mantle assemblage has led to the suggestion that high ^{3}He/^{4}He may result

79 from the greater loss of U and Th than He during ancient melt depletion (Graham et al., 1990; Class

80 and Goldstein, 2005; Parman et al., 2005). The Re-Os isotope system has the ability to retain

81 information about mantle melting events, even in the convecting mantle (e.g. Brandon et al., 2000;

82 Meibom et al., 2002; Harvey et al., 2006; Pearson et al., 2007), and thus is the most suitable tracer

83 to test whether high ^{3}He/^{4}He signatures can be directly linked to ancient depletion events.

84 In this study, Os isotope data for North Atlantic picrites from Baffin Island and West Greenland

85 have been (i) used to gain an estimate of the average $^{187}Os/^{188}Os$ composition of the convecting

86 mantle, and (ii) combined with existing He and Nd isotope data to re-assess the nature of the highest

87 known $^3He/^4He$ mantle component.

88 **2. Samples: setting and chemistry**

89 The picrites in this study were collected from the eastern margin of Baffin Island (BI) at Cape

90 Searle, Padloping Island and Durban Island and from Disko Island (Qeqertarsuaq) and the

91 Nuussuaq peninsula in West Greenland (WG). The WG picrites were collected from the three

92 members of the Vaigat formation, from oldest to youngest, the Anaanaa, Naujánguit and

93 Ordlingassoq Members. Sampling focussed on olivine-rich samples, but is otherwise representative

94 of the units sampled. The BI picrites are undifferentiated stratigraphically, but can be grouped

95 chemically into the enriched (E-type) lavas (DUR-8, DI-23 and PAD-6) and normal (N-type) lavas

96 (all others) first identified by Francis (1985). Sample CS-7 is from a cross-cutting dyke, and not

97 from the picrite lava succession. Volcanism was largely contemporaneous on Baffin Island and in

98 West Greenland and commenced ~61 Ma (Storey et al., 1998). Picrites from BI and the Anaanaa

99 and lower Naujánguit Members of WG possess normal magnetisation whereas the subsequent WG

100 melts are reversely magnetised (Pedersen et al., 2002). All members, and ~80% of the entire

101 Paleocene volcanic sequence, were erupted within 1 million years (Storey et al., 1998).

102 Petrography, major and trace element chemistry and Sr, Nd and Pb isotopes are described in more

103 detail in previous studies (e.g. Francis, 1985; Holm et al., 1993; Graham et al., 1998; Larsen and

104 Pedersen, 2000; Kent et al., 2004).

105 All North Atlantic (NA) picrites have high MgO contents compared to most mantle melts (up to 27

106 wt. % in this study) which, although in part a result of olivine accumulation, reflects the Mg-rich

107 nature of parental melts. Estimates for the parental melts of WG picrites, based on Fo-rich olivine

108 phenocrysts, vary between 18 and 21 wt. % MgO (Pedersen, 1985; Larsen and Pedersen, 2000;

109 Herzberg and O'Hara, 2002). Such MgO contents indicate generation by a high degree of melting -

110 10-11% for a depleted source (Herzberg and O'Hara, 2002; twice as high as MORB melting) - of

111 anomalously hot mantle (1540-1600°C) at depths of 60-90 km (Pedersen, 1985; Gill et al., 1992;

112 Herzberg and O'Hara, 2002). The high degree of melting generates Os-rich melts which are less

113 susceptible to interactions with crust and lithospheric mantle – critical when looking at samples

114 erupted through ancient continental crust and lithosphere.

115 The three West Greenland picrite members have similar major element characteristics. However,

116 for a given MgO content, TiO_2 increases with time, possibly reflecting a changing contribution from

117 enriched material in the picrite source (cf. Prytulak and Elliott, 2007), with the older WG picrites

118 most closely resembling the Baffin picrites (Holm et al., 1993). Chondrite-normalised REE patterns

119 are flattest in the Anaanaa and Naujánguit picrites, while the Ordlingassoq samples have higher

120 LREE and incompatible element concentrations (Holm et al., 1993). Neodymium and Sr isotope

121 compositions of the Ordlingassoq picrites resemble the least depleted Iceland picrites (Holm et al.,

122 1993).

123 **3. Analytical techniques**

124 Approximately 1g of each whole rock powder was digested and equilibrated with a mixed ^{190}Os–

125 ^{185}Re-enriched spike, using inverse aqua regia (2.5 mL 12 mol L^{-1} HCl and 5 mL 16 mol L^{-1} HNO$_3$)

126 in a quartz high-pressure asher (HPA) vessel or borosilicate Carius tube. HPA vessels were placed

127 in an Anton-Paar HPA at Durham University at 300°C and >110 bars for at least 12 hours, and

128 Carius tubes were placed in an oven at 240°C for at least 36 hours. Osmium was extracted using

129 CCl$_4$, back-extracted using HBr, and then microdistilled (Cohen and Waters, 1996). The aqua regia

130 was dried, re-dissolved in 0.5 mol L^{-1} HCl, and Re was separated using AG1X-8 (100-200#) anion-

131 exchange resin (Pearson and Woodland, 2000).

6

132 Osmium was loaded onto Pt filaments and measured as OsO_3^- ions by negative-thermal ionisation

133 mass spectrometry (N-TIMS) using the ThermoFinnigan Triton at Durham University. All Os

134 isotope beams and mass 233, corresponding to $^{185}ReO_3^-$, were measured sequentially using an axial

135 secondary electron multiplier. Raw data were corrected offline for O isotope interference, mass

136 fractionation (using $^{192}Os/^{188}Os = 3.08271$) and spike unmixing. Interference from $^{187}ReO_3^-$ was

137 insignificant (<2 cps). Analyses of 170 pg aliquots of the University of Maryland Os standard

138 solution (UMd, or UMCP) gave mean $^{187}Os/^{188}Os$ of 0.11384±16 (2σ, n=19) and 0.11379±14 (2σ,

139 n=39) for the two periods of analysis, April - November 2002 and April - May 2008, respectively.

140 These values are in good agreement with a value of 0.113787±7 for 10-100 ng/g aliquots measured

141 on the same mass spectrometer in Faraday cup mode (Luguet et al., 2008a). Rhenium was analysed

142 by inductively-coupled plasma mass spectrometry (ICP-MS) on a ThermoFinnigan[®] Element 2.

143 Solutions were introduced using a micro-concentric nebuliser and a dual-cyclonic quartz spray

144 chamber. A standard Re solution (1 ng/g) was analysed at the start, middle and end of each session

145 to determine mass fractionation.

146 Carius tube and HPA digestions gave, respectively, mean total procedural blanks of 0.43 and 0.32

147 pg Os, 2.5 and 1.9 pg Re, and $^{187}Os/^{188}Os$ of 0.143 and 0.192. Blank corrections relate to the

148 appropriate reagent batch rather than a long-term mean, but were always less than 0.1% for

149 concentration and isotope composition.

150 *Reproducibility of samples.* Duplicate digestions of an Os-rich (~2 ng/g) and Os-poor sample (~0.5

151 ng/g) indicate $^{187}Os/^{188}Os$ reproducibility of 0.15% and 0.7% (2 RSD, n=3), respectively (Table 1).

152 Os concentrations are reproducible to ~5% and 11% (2 RSD) for the two samples, respectively, and

153 Re concentrations (n=2) vary by less than 1% (2 RSD) for both samples. The external accuracy of

154 analyses is more difficult to evaluate, but reproducibility using two different digestion techniques

155 for these samples and for reference materials (Dale et al., 2008; Dale et al., 2008, in press) suggests

156 that incomplete digestion and/or sample-spike equilibration is unlikely to be a significant

157 consideration.

158 **4. Results**

159 **4.1 Re and Os elemental data**

160 Osmium concentrations range from 1.50 to 4.02 ng/g in the West Greenland picrites and from 0.435

161 to 3.45 ng/g in the Baffin Island picrites (Table 1). The WG suite has a higher median of 2.49 ng/g

162 Os compared to 1.66 ng/g for the Baffin suite. All samples have much greater Os concentrations

163 than MORB, which range from <0.001 to 0.25 ng/g (Roy-Barman and Allegre, 1994; Gannoun et

164 al., 2007). The highest Os concentrations are greater than approximate averages for primitive or

165 depleted mantle (~3.1-3.7 ng/g, Morgan et al., 2001; Becker et al., 2006; Harvey et al., 2006).

166 Rhenium concentrations vary from 0.113 to 0.506 ng/g in the BI picrites, with a median of 0.32

167 ng/g, while the WG samples have a larger range (0.063 to 1.14 ng/g) but a similar median of 0.29

168 ng/g. Such Re abundances are typically lower than MORB (0.34 – 2.28 ng/g, median ~1 ng/g, Sun

169 et al., 2003; Gannoun et al., 2007).

170 Figure 1 here.

171 Osmium concentrations decrease with decreasing MgO (Figure 1) and Ni (not shown). For a given

172 MgO content, WG picrites tend to be more Os-rich than BI picrites, which are both richer in Os

173 than recent Iceland picrites (Brandon et al., 2007). Co-variation of MgO and Re is less systematic

174 than with Os, but a negative co-variation exists in the BI samples and is well-defined in two

175 regional sub-sets (see Figure 1). With the exception of several samples, Re/Os ratios increase with

176 decreasing MgO content in all suites (compare Figure 1a & b).

177 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 picrite has the lowest Os concentration (~0.45 ng/g, Figure 3), consistent with a previous conclusion of crustal contamination for this sample (Stuart et al., 2003); this sample has been excluded from further discussion. Thus, the range of initial $^{187}Os/^{188}Os$ is considerably more limited, from 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 0.1272, which is identical to the BI peak (Figure 2). 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 heterogeneity is a possible explanation for this difference but as the samples of Kent et al. (2004) have unusually high Os concentrations for their MgO content (Figure 1), and Os co-varies with $^{187}Os/^{188}Os$, we contend that these samples may have been modified during interaction with an Os-rich, low $^{187}Os/^{188}Os$ lithospheric mantle component (cf. Larsen et al., 2003).

The WG picrites extend to more radiogenic Os isotope compositions than the BI picrites, and $^{187}Os/^{188}Os$ clearly varies with stratigraphy. Picrites from the Anaanaa and Naujánguit Members of the Vaigat formation possess largely uniform and unradiogenic $^{187}Os/^{188}Os$ (Figure 3). In contrast, all samples from the slightly younger Ordlingassoq Member have more radiogenic $^{187}Os/^{188}Os_{initial}$ ratios of 0.1294 to 0.1322. Such $^{187}Os/^{188}Os$ ratios range to more radiogenic values than estimates of putative present-day primitive mantle (0.1296±0.0008, Meisel et al., 2001) but do not extend as high as previous picrite data for the WG Vaigat formation (up to 0.1371, Schaefer et al., 2000) or recent picrites from Iceland erupted within the last 1 Ma (up to 0.1378, Brandon et al., 2007).

202   Figure 2 here.

203   There is no co-variation of $^{187}Os/^{188}Os$ with Os concentration in either BI or WG picrites (Figure 3,

204   excluding CS-7, already discussed). Significant assimilation of crust or sub-continental lithospheric

205   mantle (SCLM) would likely result in samples from the same parental melt falling on mixing lines

206   towards either an Os-poor, ancient, extremely radiogenic crustal component or an Os-rich, ^{187}Os-

207   depleted SCLM; such trends are not observed. Equally, different parental melts would be variably

208   susceptible to contamination, according to their Os concentrations, and thus display a range of

209   isotope compositions. Previous WG data display increased $^{187}Os/^{188}Os$ in samples with <0.5 ng/g

210   Os (Schaefer et al., 2000), suggesting that Os-poor samples may be affected by crustal assimilation

211   and/or preferential sampling of radiogenic material during mantle melting; these samples have been

212   omitted from subsequent figures.

213   Figure 3 here.

214   **5. Discussion**

215   **5.1 Re-Os elemental behaviour**

216   Olivine accumulation in the NA picrites studied is indicated by sample MgO contents of up to 27.0

217   wt. %, compared to an estimated parental magma of 18-21 wt. % MgO (Pedersen, 1985; Larsen and

218   Pedersen, 2000; Herzberg and O'Hara, 2002). Osmium concentrations decrease with decreasing

219   MgO (Figure 1) and with Ni (not shown), reflecting the compatibility of Os within the olivine-rich

220   crystallising assemblage, which probably includes significant co-precipitated sulphide (as in global

221   MORB and OIB, e.g. Burton et al., 2002). Therefore, Os concentrations are likely controlled by

222   olivine accumulation and fractionation. On the basis of the Os content at MgO = 18 wt %, BI and

223   WG parental melts are estimated to contain ~1 and ~1.7 ng/g Os, respectively (Figure 1). As Os has

224   a high sulphide-silicate melt partition coefficient (at low pressures, e.g. ~10,000, Crocket et al.,

225   1997), such high parental melt Os concentrations probably require complete consumption of

226   sulphides in at least part of the melting column. The consumption of sulphide in a fertile upper

227   mantle source is thought to occur at melt fractions of >20-25%, based on the sulphide content of the

228   mantle and sulphur solubility in mafic melts (e.g. Mavrogenes and O'Neill, 1999; Lorand et al.,

229   2003), or at lower melt fractions for a previously depleted mantle source as proposed for the NA

230   picrites (e.g. Ellam and Stuart, 2004). These melt fractions are consistent with estimates based on

231   MgO content: ~20-28% for a fertile source or 10-11% for a depleted source (Herzberg and O'Hara,

232   2002). In addition to sulphide-silicate partitioning, physical entrainment of liquid sulphide within a

233   large degree silicate melt (Ballhaus et al., 2006) may, at least in part, account for the very high Os

234   concentrations of the NA picrite parental melts.

235   Rhenium concentrations in the picrites are lower than MORB, consistent with the moderate

236   incompatibility of Re during mantle melting and the higher melt fraction of picritic melts. Re/Yb

237   ratios are comparable to MORB (~0.00025), illustrating that Re maintains similar compatibility to

238   Yb at higher melt fractions than MORB. The preference of Re for olivine-poorer assemblages is

239   illustrated by increasing Re and Re/Os ratios with decreasing MgO content in almost all the picrite

240   units.

241   **5.2 Osmium-neodymium isotope systematics**

242   The majority of NA picrites analysed here have $^{143}Nd/^{144}Nd_{initial}$ of ~0.51307 (Graham et al., 1998;

243   Starkey et al., 2008, in press), consistent with a source dominated by a depleted mantle component,

244   as previously noted by Holm et al. (1993). Such Nd isotope compositions are indistinguishable

245   from the range of estimates for DMM corrected to 60 Ma (0.51304 - 0.51311, Salters and Stracke,

246   2004; Workman and Hart, 2005) and the proposed depleted end-member of the Iceland array (e.g.

247   Taylor et al., 1997). There is considerable $^{143}Nd/^{144}Nd$ variation in BI and Anaanaa (WG) picrites,

248   while $^{187}Os/^{188}Os$ is largely constant at around 0.127 (Figure 4). The $^{143}Nd/^{144}Nd$ range may be

249   partially due to crustal contamination (samples CS-7, 400444, Starkey et al., 2008, in press; and by

250   inference 408001.233) but still remains significant (0.512845 – 0.513072). In contrast, the younger

251   Ordlingassoq Member of the WG suite displays a negative co-variation of Os and Nd isotopes with

252    $^{143}Nd/^{144}Nd$ decreasing from 0.51308 to 0.51291 as $^{187}Os/^{188}Os$ increases from 0.1267 to 0.1322

253   (Figure 4). Literature data for the Naujánguit and Ordlingassoq Members (Schaefer et al., 2000),

254   possess similar systematics to the Ordlingassoq samples in this study, but display greater scatter,

255   probably due to careful screening during our sample selection to avoid potential crustal

256   contamination.

257   **5.2.1 Assessing the effects of post-melting crustal or lithospheric interaction**

258   Assessment of the potential effects of continental crust assimilation is of particular importance due

259   to the Proterozoic age of the crust through which the NA picrites were erupted. Careful screening

260   has removed samples which appear obviously contaminated by crust, but quantitative assessment of

261   the isotopic effects of minor contamination by continental crust is not a simple task due to its

262   marked heterogeneity, particularly in terms of Re-Os (e.g. Peucker-Ehrenbrink and Jahn, 2001).

263   The Os isotope composition of local crust has not been measured, and measurements, even if

264   numerous, are unlikely to provide a sufficiently well-constrained average due to the varied

265   basement geology. Baffin Island Proterozoic basement rocks also display a considerable range of

266    $^{143}Nd/^{144}Nd$ and Nd concentrations (Theriault et al., 2001), as do local Cretaceous West Greenland

267   sediments, derived from Proterozoic basement, which are the most likely contaminants (Goodrich

268   and Patchett, 1991, Larsen, unpubl.). Estimated Re and Os concentrations (Table 2) have been used

269   to calculate the $^{187}Os/^{188}Os$ composition of 2 Ga basement or basement-derived sediment, which has

270   then been combined with average measured Nd concentrations and isotope compositions (Table 2).

271 Mixing of average/best estimate 2 Ga basement or sedimentary cover with parental picritic melt

272 does not generate the uniform ^{187}Os/^{188}Os BI array, nor the negative co-variation of Nd and Os

273 isotopes observed in Ordlingassoq picrites. However, approximately 5% assimilation of two

274 different extreme crustal components may be able to account for both the BI and Ordlingassoq co-

275 variations (low Sm/Nd - low Re/Os and high Sm/Nd - high Re/Os crust, respectively, Figure 4), but

276 such very different and extreme contaminants are considered unlikely given the similar average

277 composition of the country rock in the two regions. Crust-melt mixing may also result in co-

278 variation of ^{187}Os/^{188}Os with Os concentration and this is not observed (Figure 3).

279 Combining Nd isotopes and Nb/Zr ratios provides a further line of evidence that crustal assimilation

280 is not a significant factor in controlling the isotope compositions of NA picrites (Starkey et al.,

281 2008, in press). The broad array of increasing Nb/Zr with decreasing ^{143}Nd/^{144}Nd cannot be

282 accounted for by assimilation of any low-Sm/Nd crustal material. Very small amounts of crustal

283 contamination can explain some of the spread in ^{143}Nd/^{144}Nd, but Nd isotopes (and by inference Os

284 isotopes) predominantly reflect the mantle source.

285 Figure 4 here.

286 **5.2.2 Generation of the osmium and neodymium isotope variations in the mantle source**

287 The upper part of the Vaigat formation (WG) records increasing ^{187}Os/^{188}Os combined with

288 decreasing ^{143}Nd/^{144}Nd. If this is unlikely to result from melt-crust interaction, such a shift in Os

289 and Nd isotope compositions may reflect an increased source input from an enriched component.

290 For instance, modelled mixing of DMM with 2 Ga recycled oceanic crust and associated sediment

291 can broadly produce the negative array in Os-Nd isotope space (Figure 4, e.g. ~5% recycled

292 component containing ~10% sediment). However, this model is non-unique and it is possible that

293 other enriched material such as metasomatised oceanic lithosphere (e.g. Niu and O'Hara, 2003)

294 could produce the negative array. Equally, mixtures of a hybrid putative primitive mantle –

295   recycled oceanic crust component with DMM can also explain the arrays of both the Ordlingassoq

296   and Iceland picrites, by varying the proportions of primitive mantle and recycled crust in the hybrid

297   component (Figure 4).

298   The variation of ^{143}Nd/^{144}Nd with no complementary change in ^{187}Os/^{188}Os seen in Baffin and

299   Anaanaa picrites is more difficult to explain because old recycled oceanic crust (plus sediment), and

300   old continental crust, are both likely to possess complementary high ^{187}Os/^{188}Os and low

301   ^{143}Nd/^{144}Nd. Mixing of primitive mantle and DMM may be able to account for such an array

302   (Figure 4). Alternatively, such a signature may reflect a mantle source containing a minor

303   (sulphide-poor?) pyroxenite component that, due to its high Nd content and low Os content,

304   significantly affected the Nd isotope composition of the melt without noticeably affecting Os.

305   Regardless of the ultimate source of Nd isotope heterogeneity, the uniform ^{187}Os/^{188}Os of BI and

306   early WG picrites indicates that any enriched component has had little influence on Os isotope

307   compositions.

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

309   Volcanism associated with the onset of the proto-Iceland plume was derived from high degree

310   melting and was characterised by approximately chondritic Os isotope compositions, the highest

311   known ^{3}He/^{4}He ratios and variable, but fairly radiogenic ^{143}Nd/^{144}Nd ratios. Osmium isotope

312   compositions are consistent with a source more depleted than estimates of PUM (e.g. Meisel et al.,

313   2001), with no significant contribution from any enriched component including, based on current

314   models, the outer core (e.g. Walker et al., 1995; Brandon et al., 1998) or metasomatic sulphide

315   (Luguet et al., 2008b). Subsequent melts (Ordlingassoq Member), erupted within 1 Ma of the

316   earliest picrites, have elevated ^{187}Os/^{188}Os indicating a contribution from enriched material, possibly

317   old recycled oceanic crust plus sediment, which was not tapped in the first phase of plume melting,

318   but was soon brought into the zone of melting presumably by continued upwelling. Crustal and

319 lithospheric interactions appear to mask the source composition of 58 Ma East Greenland samples

320 (e.g. Peate et al., 2003), but ^{3}He/^{4}He up to 21 R_a has been measured (Marty et al., 1998).

321 Elevated ^{187}Os/^{188}Os ratios and low ^{143}Nd/^{144}Nd isotope data, relative to DMM, in recent Iceland

322 picrites (Brandon et al., 2007) can be explained by mixing of a similar recycled crustal component,

323 including ~5% sediment, with a DMM component (Figure 4). The absence of ^{186}Os enrichment in

324 these picrites, relative to DMM (Brandon et al., 2007), combined with the NA picrite Os data,

325 suggests no bulk core contribution to the plume over its history. High ^{3}He/^{4}He ratios persist in the

326 Iceland plume today (>30 Ra, Macpherson et al., 2005) and, intriguingly, ^{3}He/^{4}He increases with

327 ^{187}Os/^{188}Os in recent picrites (Brandon et al., 2007; Figure 6b), despite the proposed involvement of

328 recycled oceanic crust which would possess very low ^{3}He/^{4}He (~4, Brandon et al., 2007).

329 5.4 The ^{187}Os/^{188}Os composition of shallow convecting mantle

330 Trace element and Nd-Sr isotope evidence (Holm et al., 1993; Stuart et al., 2003; Ellam and Stuart,

331 2004; Kent et al., 2004) indicates that the early NA picrite source is typically depleted with respect

332 to putative primitive mantle (e.g. Zindler and Hart, 1986), and is indistinguishable from the MORB

333 source mantle (DMM, e.g. Salters and Stracke, 2004; Workman and Hart, 2005). The ^{187}Os/^{188}Os

334 compositions of the earliest NA picrites (0.1267 to 0.1280) are consistent with a depleted mantle

335 source containing no significant contribution from recycled crust, pyroxenite, sediment, recycled

336 SCLM, metasomatised peridotite or outer core, nor any isolated ancient depleted domains.

337 Global ocean island basalts have variable reference TiO_2 concentrations (defined as the TiO_2

338 concentration on the olivine control line at the estimated parental MgO content). This has been

339 interpreted to reflect variable source contributions from enriched components, primarily recycled

340 oceanic crust (Prytulak and Elliott, 2007). The reference TiO_2 content for the early NA picrites is

341 ≤1% (at ~18 wt. % MgO), lower than all OIB suites compiled by Prytulak and Elliott (2007), and

342 comparable to Iceland. By this measure the NA picrites define the least enriched end-member for

343   within plate magmas, i.e. their sources are dominated by peridotite, with minimal 'enriched'

344   component. This conclusion is also supported by the low Ni/Mg ratios of Fo-rich picritic olivines

345   (0.74-0.94, Fo: 89-91, respectively; Larsen and Pedersen, 2000) which are comparable to MORB

346   and peridotite but lower than most within-plate magmas (Sobolev et al., 2007). Although Ni/Mg in

347   olivine may simply reflect the depth of melting, with which it appears positively correlated (Niu and

348   O'Hara, 2007), low Ni/Mg ratios combined with relatively deep melting definitely suggests an

349   insignificant pyroxenite source contribution.

350   Due to the large degree of melting during the generation of the NA picrites, their Os isotope

351   composition will closely reflect the composition of the bulk mantle source. This, coupled with the

352   chemical similarity to DMM and insignificance of enriched components, gives the potential for

353   providing an estimate of the bulk $^{187}Os/^{188}Os$ composition of the peridotitic convecting upper

354   mantle/DMM sampled by these magmas. The dominance of the DMM signature, in a proposed

355   plume-head, has probably arisen from extensive entrainment during the latter stages of plume

356   upwelling, and a disproportionate contribution due to melting in the shallower part of the melting

357   column. To facilitate comparison with other data such as those for MORB, initial $^{187}Os/^{188}Os$ for

358   picrites, platinum-group alloys and chromites have been recalculated to the present day by assuming

359   chondritic evolution of their sources since the time of mantle melting (Figure 5). Strictly speaking,

360   post-melting evolution of Os isotopes will be depressed relative to chondrite, due to Re depletion.

361   Thus, for a depleted NA picrite mantle source, with 0.12 ng/g Re (Sun et al., 2003), the present day

362    $^{187}Os/^{188}Os$ would be lower by 0.0002 than the values discussed below.

363   On the basis of the probability peak for the NA picrites, the $^{187}Os/^{188}Os$ composition of present day

364   convecting mantle underlying this region is estimated to be 0.1276 ± 0.0007 (2SD of data

365   contributing to peak, Figure 5). This value is indistinguishable from the least radiogenic recent

366   Iceland melts (<0.5 Ma, Skovgaard et al., 2001, Figure 3) and from the largest platinum-group alloy

367   peak (0.1276), derived mainly from Tibetan ophiolites (Pearson et al., 2007; Shi et al., 2007).

368   Phanerozoic ophiolite-derived chromites define a peak $^{187}Os/^{188}Os$ of 0.1283, while regression of

369   $^{187}Os/^{188}Os$ versus time gives a lower intercept of 0.1281 (Walker et al., 2002b). Subduction-related

370   enrichment of Re (see Becker, 2000; Dale et al., 2007) or radiogenic Os (e.g. Brandon et al., 1996)

371   has the potential to elevate this value, although this must also be considered for the ophiolite-

372   derived platinum-group alloys. The $^{187}Os/^{188}Os$ estimate defined by NA picrites and platinum-

373   group alloys is intermediate between averages for carbonaceous, enstatite and ordinary chondrites

374   (0.1262, 0.1281, 0.1283, respectively, Walker et al., 2002a). It is, however, significantly lower than

375   the proposed $^{187}Os/^{188}Os$ of 0.1296 ± 0.0008 for putative PUM, based on co-variation of Al_2O_3 and

376   $^{187}Os/^{188}Os$ in mantle xenoliths (Meisel et al., 2001). This difference is presumably due to the effect

377   of time-integrated mantle Re depletion through the generation of continental crust and/or generation

378   and isolation of subducted oceanic crust, although the potential effects of coupled metasomatic

379   increases in Al_2O_3 (e.g. Pearson et al., 2003) and $^{187}Os/^{188}Os$ (e.g. Alard et al., 2005) may also need

380   to be considered. The similarity between the modal NA picrite Os isotope composition and other

381   estimates of convecting mantle indicate that a $^{187}Os/^{188}Os$ value somewhere between 0.1274 and

382   0.1281 might be an appropriate estimate for ambient shallow convecting mantle.

383   Figure 5 here.

384   The least radiogenic MORB ($^{187}Os/^{188}Os$ of 0.1261-0.1272) are isotopically similar to the NA

385   picrite $^{187}Os/^{188}Os$ peak of 0.1276, and therefore support this proposal. Most MORB are

386   significantly more radiogenic with $^{187}Os/^{188}Os$ ratios up to 0.148 (Gannoun et al., 2007). This

387   probably reflects preferential sampling of enriched components and/or ^{187}Os-rich metasomatic

388   sulphides within the DMM, by MORB melts of smaller degree than the NA picrites (e.g. Alard et

389   al., 2005; Escrig et al., 2005). If such enriched components are present in the initial NA picrite

17

390   source (as suggested by the Nd isotope data), their Os isotope signature is greatly diluted compared

391   to the contribution from depleted peridotite.

392   **5.5 Implications for high ^{3}He/^{4}He in the mantle**

393   Based on the possibility of greater compatibility of He than U and Th during mantle melting

394   (Graham et al., 1990; Parman et al., 2005; Heber et al., 2007), it has been proposed that ancient melt

395   depletion will lead to the retention of high ^{3}He/^{4}He in the residue, through reduced ingrowth of ^{4}He

396   (Class and Goldstein, 2005; Parman et al., 2005). The timing of this depletion event, for ^{3}He/^{4}He as

397   high as 50 R_a, has been variously estimated at 3.7 and 3.1 Ga (Parman, 2007; Porcelli and Elliott,

398   2008). Preservation of high ^{3}He/^{4}He through depletion probably requires a large degree of melting

399   (Parman et al., 2005; Porcelli and Elliott, 2008) and this would likely result in near complete Re

400   depletion. Depletion of Re during such mantle melting will lead, over time, to ^{187}Os/^{188}Os ratios

401   which deviate well below the chondrite evolution curve. Thus, in contrast to the broadly chondritic

402   ^{187}Os/^{188}Os of the NA picrites, a source entirely depleted in Re at 3.7 or 3.1 Ga (T_{RD}), and

403   subsequently isolated, would have ^{187}Os/^{188}Os of ~0.102 or ~0.107, respectively. Even partial Re

404   depletion of the source at these ages would reduce ^{187}Os/^{188}Os evolution to between 0.113 and

405   0.115 (if Re was reduced to 0.12 ng/g, equal to an estimate of the DMM (Sun et al., 2003), Figure

406   6). The lowest initial ^{187}Os/^{188}Os for the NA picrites corresponds to a T_{RD} of only ~0.4 Ga, and

407   therefore Os isotopes are not consistent with ancient depletion as a mechanism for the preservation

408   of the highest known mantle ^{3}He/^{4}He ratios. Equally, mixing of high ^{3}He/^{4}He depleted mantle and

409   DMM entrained in the plume, cannot explain the range of ^{3}He/^{4}He at a near constant ^{187}Os/^{188}Os as

410   the former is unlikely to have a sufficiently high He content (Figure 6). Alternative models, such as

411   a sub-continental lithospheric mantle source for the high ^{3}He/^{4}He are also not supported by the Os

412   isotope data, because they too demand significantly sub-chondritic ^{187}Os/^{188}Os (see Larsen et al.,

413   2003 for WG lithosphere). Furthermore, high ^{3}He/^{4}He has not been found in sub-continental mantle

414   xenoliths or melts (Dunai and Baur, 1995; Day et al., 2005). For the NA picrites considered here,

415   only subsequent addition of radiogenic Os, without He, could reconcile the Os data with an ancient

416   depleted source for the high ^{3}He/^{4}He. Such a specific flux seems unlikely as fluid-mobile or

417   incompatible elements, including both U and Th, and probably He, would also presumably be

418   involved.

419   Figure 6 here.

420   Table 2 here.

421   The core has been proposed as a possible source of He enriched in ^{3}He (e.g. Macpherson et al.,

422   1998; Porcelli and Halliday, 2001). The proposed partitioning behaviour of Re and Os between the

423   inner and outer core has led to the hypothesis that the outer core possesses supra-chondritic Re/Os

424   ratios (Walker et al., 1995). Based on this model, the approximately chondritic ^{187}Os/^{188}Os of high

425   ^{3}He/^{4}He NA picrites precludes a bulk contribution from core material, as much less than 1% Fe-rich

426   core input would impart a radiogenic signature due to its much greater Os concentration. However,

427   decoupled transfer of He and Os across the core-mantle boundary, for instance by diffusion, could

428   produce high ^{3}He/^{4}He without correspondingly high ^{187}Os/^{188}Os.

429   In an attempt to account for the Os-He isotope data, mixing calculations for various possible mantle

430   components have been performed (Figure 6). However, constraints on He abundances are poor and

431   can be varied in order to better fit the data. We find such flexibility unsatisfactory in that it leads to

432   non-uniquely constrained models, thus we have cautious conclusions from our modelling.

433   Furthermore, such modelling implicitly assumes that source contributions of He and Os are

434   coupled, while the parameters are not sufficiently constrained to allow assessment of this

435   assumption.

436   Mixing between DMM and a subsidiary primitive mantle component (up to 25% in this model) can

437   broadly explain the Os-He array for the picrites with constant, approximately chondritic $^{187}Os/^{188}Os$

438   (Figure 6). A primitive high $^{3}He/^{4}He$ source of >50 with sufficient He, would permit a small

439   contribution derived from 2 Ga recycled oceanic crust, while retaining $^{3}He/^{4}He$ of >45 as observed

440   in some WG Ordlingassoq picrites (also proposed for Iceland, Brandon et al., 2007). However, the

441   difference in He concentration between a proposed primitive mantle-recycled oceanic crust end-

442   member and DMM gives rise to convex-up mixing curves which means that Ordlingassoq samples

443   require unique proportions of primitive mantle, recycled oceanic crust and DMM, rather than

444   variable contributions along a single mixing line. Convex-up mixing lines are also problematic for

445   the quasi-linear Icelandic array, although use of a higher He concentration for the DMM than that

446   used by Brandon et al. (2007) (Table 2), generates mixing lines that more closely approach linearity

447   (Figure 6). Mixing of DMM, PM and recycled oceanic crust plus sediment can also broadly explain

448   the NA and Iceland picrite Os-Nd variations (Figure 4), but such mixing requires somewhat

449   different component proportions in Os-He and Os-Nd isotope space and does not easily account for

450   each individual sample.

451   It has been proposed that a high $^{3}He/^{4}He$ component could be widespread in the upper mantle

452   (Meibom et al., 2003), and such a component with MORB-like $^{187}Os/^{188}Os$ of ~0.1276 cannot be

453   ruled out on the basis of these data, as long as it contains sufficient He to retain high $^{3}He/^{4}He$

454   despite mixing of a minor recycled oceanic crust component. However, while the mechanisms for

455   such a scenario are not well understood, it seems unlikely that this material would only be sampled

456   during episodes of large degree melting of hot mantle (as opposed to MORB melting) and even if

457   so, that it would not also impart an ancient depleted Os isotope signature, complementary to the

458   unradiogenic high $^{3}He/^{4}He$ signature.

459   Given that high ^{3}He/^{4}He ratios (>45) in NA picrites are not restricted to samples with a specific Os

460   isotope signature and mixing models do not satisfactorily explain isotope co-variations in detail, it

461   seems most likely that Os and He are decoupled (and Nd-He, Starkey et al., 2008, in press).

462   Diffusion or mixing of high ^{3}He/^{4}He from a He-rich primordial reservoir could impart the necessary

463   He signature without significantly changing the previously established Os and Nd isotope

464   heterogeneity. Constraining the ultimate source of this high ^{3}He/^{4}He component is difficult, but as

465   it was tapped most efficiently during melting of a plume-head and persists in Icelandic volcanism

466   today, it seems most plausible that it diffused across, or was entrained at, a deep boundary layer.

467   **6. Concluding remarks**

468   Osmium concentrations in NA picrites are high for mantle melts (1-1.7 ng/g parental Os content)

469   and suggest complete consumption of sulphide in at least part of the source due to a large degree of

470   melting. This is consistent with previous evaluations of the degree of melting based on MgO-rich

471   olivines: 10-11% for depleted mantle (Herzberg and O'Hara, 2002). Initial Os isotope compositions

472   in the earliest picrite melts from West Greenland (early Vaigat formation) and Baffin Island are

473   uniform and broadly chondritic (probability peak of ^{187}Os/^{188}Os = 0.1272). In terms of Os isotope

474   mass balance, this initial volcanism cannot contain any significant contribution from the outer core

475   or from old recycled crustal material, as both would impart a radiogenic ^{187}Os/^{188}Os signature. In

476   addition, the minimal presence of enriched pyroxenitic components in the source of NA picrites is

477   suggested by various parameters such as Ni content of olivine and bulk-rock TiO_2 content. The

478   absence of enriched components, coupled with a large degree of melting and Nd isotope

479   compositions which are indistinguishable from DMM, means that the average ^{187}Os/^{188}Os may

480   reflect the bulk ^{187}Os/^{188}Os of convecting mantle. This value, when corrected to the present day, is

481   similar to the least radiogenic MORB (e.g. Gannoun et al., 2007) and the most common ^{187}Os/^{188}Os

482   ratios found in platinum-group alloys from Tibetan ophiolites (Pearson et al., 2007; Shi et al.,

483   2007).   Subsequent melts sampled from the upper Vaigat formation, erupted within 1 Ma of the

484   earliest melts (Storey et al., 1998), possess supra-chondritic initial $^{187}Os/^{188}Os$ ratios of up to

485   0.1321.   Such ratios, also seen in recent Iceland plume melts (Brandon et al., 2007), can be

486   accounted for (though not uniquely) by a greater contribution from recycled oceanic crust.

487   Models seeking to explain high mantle $^3He/^4He$ ratios by ancient depletion (Class and Goldstein,

488   2005; Parman, 2007) are not supported by the uniform and largely chondritic $^{187}Os/^{188}Os$ ratios of

489   NA picrites.   Ancient melting and isolation would lead to significantly sub-chondritic $^{187}Os/^{188}Os$ in

490   the NA picrite source, which possesses the highest known mantle-derived $^3He/^4He$ ratios (up to 50

491   R_a, Stuart et al., 2003).   Outer core material would nominally impart radiogenic Os to the plume,

492   and so core material is also not supported as a source of high $^3He/^4He$, unless the mechanism of He

493   transfer to the plume (diffusion?) is decoupled from Os.   Therefore, three possible explanations for

494   the high $^3He/^4He$ signature are: (i) it is present in a typical upper mantle source, entrained in the

495   plume-head, but is only tapped during episodes of high-degree melting of hot mantle; (ii) it is

496   derived from a primitive mantle component which has been mixed with recycled oceanic crust and

497   DMM in order to broadly explain the Os, He and Nd isotope variations or, (iii) He is largely

498   decoupled from Os and Nd and is dominated by addition of a He-rich, high $^3He/^4He$ component,

499   probably primordial in nature, without complementary addition of other elements.   The difficulties

500   presented above for (i) and (ii) and the fact that high $^3He/^4He$ is not restricted to a source with a

501   particular Os (or Nd) isotope composition make the latter our favoured model.

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509   main text: 6380

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Figure captions

Figure 1. Co-variation of Os and Re with MgO in picrites from West Greenland (WG) and Baffin Island (BI). Other WG, BI and Iceland picrite data from Schaefer et al. (2000), Kent et al. (2004) and Brandon et al. (2007), respectively.

Figure 2. Probability density plot for initial $^{187}Os/^{188}Os$ in (a) North Atlantic (NA: Baffin Island & West Greenland) picrites from this study and (b) NA and Iceland picrites from the literature (at same scale). Other data: [1] Schaefer et al. (2000), [2] Brandon et al. (2007) and Skovgaard et al. (2001), [3] Kent et al. (2004). NA picrite samples with <0.5 ng/g Os have been omitted due to potential crustal contamination. For comparison, Iceland samples have been corrected to 61 Ma, based on chondritic evolution. 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 data sources: WG: Schaefer et al. (2000), BI: Kent et al. (2004), Iceland[1]: Brandon et al. (2007), Iceland[2]: Skovgaard et al. (2001).

Figure 4. $^{187}Os/^{188}Os$ vs. $^{143}Nd/^{144}Nd$ (corrected to 61 Ma) for NA picrites in this study (Nd data from Graham et al., 1998; Starkey et al., 2008, in press) and published NA and Iceland picrites

796 (Schaefer et al., 2000; Kent et al., 2004; Brandon et al., 2007). (a) Best estimates for the effects of

797 possible continental crust (CC) assimilation. (b) Mixing between possible mantle components:

798 DMM, PM (primitive mantle), 2 Ga ROC (recycled oceanic crust; gabbro:basalt - 50:50), 2 Ga

799 SROC (ROC:sediment - 95:5, except SROC*: 90:10), and mixing of PM/SROC hybrid component

800 with DMM (superscript numbers refer to the relative proportions in the hybrid). Tick marks

801 represent 10% proportion increments, except where stated. Displayed mantle mixing curves only

802 represent a part of the large possible range due to uncertain parameters and variable mixing

803 proportions. Parameters – see Table 2.

804 Figure 5. Probability density plot for $^{187}Os/^{188}Os$ of NA picrites and other direct and indirect

805 indicators of mantle composition. $^{187}Os/^{188}Os_{present\ day}$ denotes that data, where appropriate, has

806 been corrected for ingrowth since emplacement and then recalculated to a present day mantle

807 composition (assuming chondritic evolution). NA picrites with <0.5 ng/g Os have been omitted due

808 to potential crustal contamination. A 'bandwidth' uncertainty of 0.001 was applied to the

809 $^{187}Os/^{188}Os$ ratio of all samples (~0.8% relative uncertainty). Data sources: [1] This study, Schaefer et

810 al. (2000) and Kent et al. (2004), [2] Pearson et al. (2007) and Shi et al. (2007), [3] Walker et al.

811 (2002b; * mean value taken from the authors' regression using 187Os/188Os vs. time), [4] Roy-

812 Barman and Allegre (1994), Snow and Reisberg (1995), Brandon et al. (2000) and Harvey et al.

813 (2006), [5] Gannoun et al. (2007), Global OIB[6] data from samples with >30 pg/g Os from: Martin,

814 (1991), Hauri and Hart (1993), Reisberg et al. (1993), Martin et al. (1994), Marcantonio et al.

815 (1995), Bennett et al. (1996), Hauri et al. (1996), Widom and Shirey (1996), Widom et al. (1999).

816 EC, OC and CC are enstatite, ordinary and carbonaceous chondrites, respectively (Walker et al.,

817 2002a), PUM is primitive upper mantle (Meisel et al., 2001).

818 Figure 6. He isotope composition against $^{187}Os/^{188}Os$ at the time of eruption (61 Ma) for NA

819 picrites (this study, He data from Stuart et al., 2003; Starkey et al., 2008, in press) and recent

820   Icelandic picrites (Brandon et al., 2007). Symbols: see Figure 1. (a) Mixing of isolated depleted

821   3.1 Ga mantle and 2 Ga recycled oceanic crust plus sediment (SROC, ROC:sediment 90:10) and

822   mixing of this hybrid with DMM. Isolated depleted mantle Os and He isotope compositions are

823   based on the He evolution models of [1] Parman et al. (2007), [2] Porcelli and Elliott (2008) and Os

824   isotope evolution assuming a residual [Re] of 0.12 ng/g, similar to an estimate for the DMM (Sun et

825   al., 2003), and [Os] of 3.4 ng/g. As relatively high degree melting is required to generate

826   sufficiently high $^{3}He/^{4}He$ depleted domains (Parman et al., 2005; Porcelli and Elliott, 2008) such a

827   Re content, and therefore also the evolved $^{187}Os/^{188}Os$, is considered an upper limit. (b) Mixing of

828   primitive mantle, DMM, SROC (ROC:sediment 90:10), and PM/SROC hybrid (superscript

829   numbers refer to the relative proportions in the hybrid). Parameters: see Table 2. Note that there

830   are poor constraints on all He concentrations, and Re-Os abundances in ancient recycled sediment

831   are likely to be extremely heterogeneous. Grey dashed curve in (b) is DMM – $PM^{50}/SROC^{50}$

832   mixing modelled with a DMM [He] of 0.269 $\mu cm^{3}(STP)/g$ used in Brandon et al. (2007).

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
Click here to download Figure: Fig 1 - MgO vs Os-Re.eps
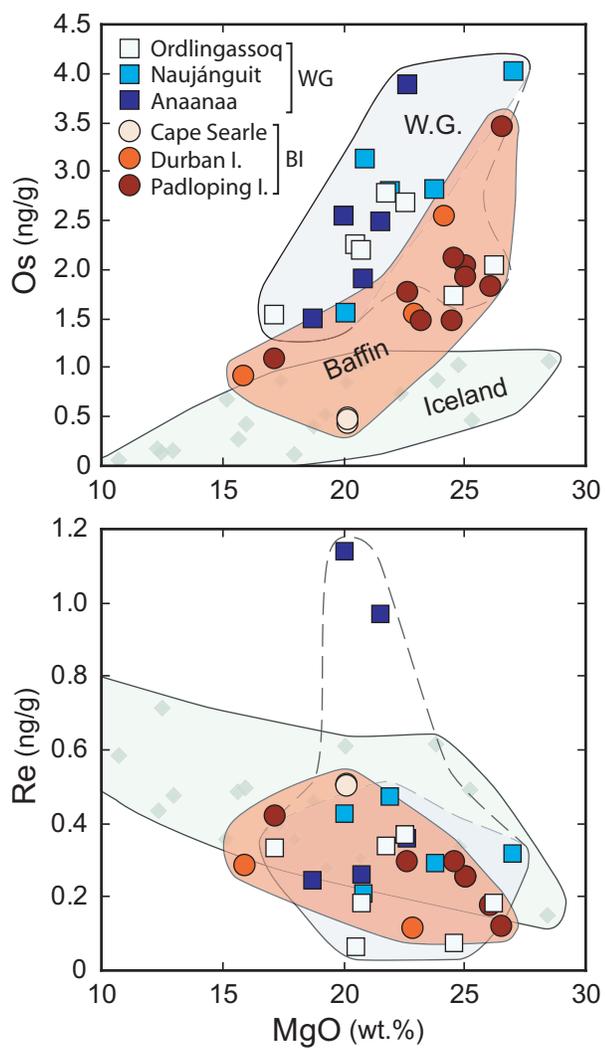
Figure 2
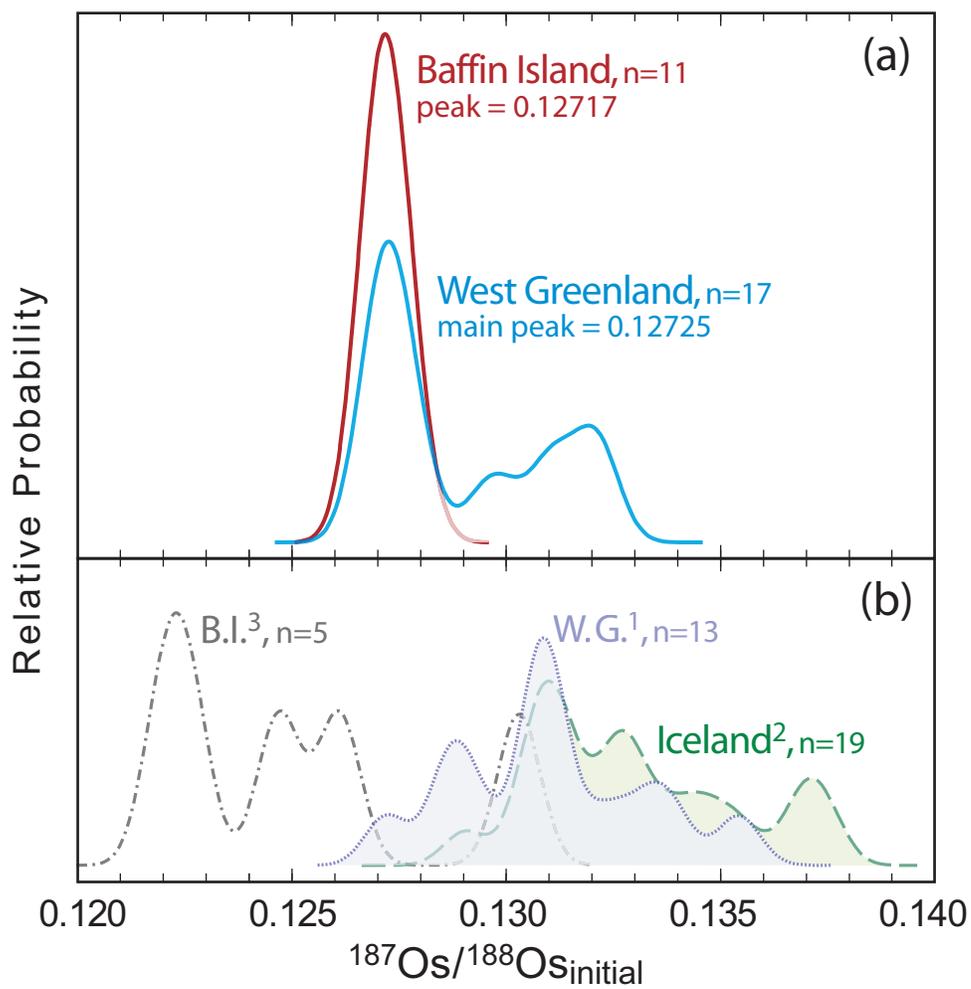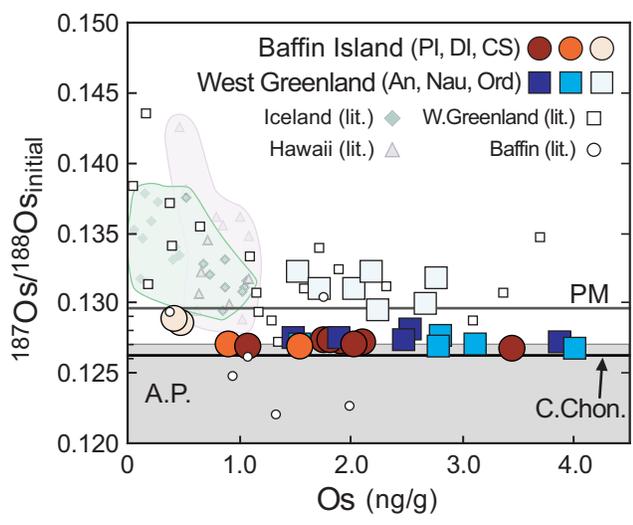
Figure 3

Figure 4
Click here to download Figure: Fig 4 - 187-188Os v Nd.eps

Figure 5