Evidence against an ancient non-chondritic mantle source for North Atlantic Igneous Province lavas

James M.D. Day

Geosciences Research Division, Scripps Institution of Oceanography, La Jolla, CA 92093-0244, USA (jmdday@ucsd.edu)

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Abstract

North Atlantic Igneous Province (NAIP) lavas host olivine with the highest $^3\text{He}/^4\text{He}$ ever measured for terrestrial igneous rocks (up to 50 R_A , or $^4\text{He}/^3\text{He} = \sim 15,300$). The relationship of high- $^3\text{He}/^4\text{He}$ with Pb isotope compositions close to the terrestrial geochron and $^{143}\text{Nd}/^{144}\text{Nd}$ plausibly consistent with supra-chondritic mantle Sm/Nd in Baffin Island and West Greenland lavas has been interpreted to reflect an ancient ‘non-chondritic’ mantle source signature. Alternatively, assimilation of continental crustal rocks with unradiogenic Pb isotope compositions and low $^{143}\text{Nd}/^{144}\text{Nd}$, into magmas with high- $^3\text{He}/^4\text{He}$, and derived from variably depleted mantle sources, could impart similar geochemical signatures. Radiogenic and stable isotope data for NAIP lavas are consistent with origins as melts from upper mantle sources that contain low- $^{18}\text{O}/^{16}\text{O}$ recycled lithosphere and/or hydrothermally altered crust, or that have experienced pervasive contamination by crustal gneisses. Olivines from NAIP lavas with $^3\text{He}/^4\text{He}$ spanning from 8 to 48 R_A have $\delta^{18}\text{O}$ ranging from 3.5 to 5.5‰. These compositions are consistent with sources of ambient mantle and low- $\delta^{18}\text{O}$ recycled lithosphere, or with concomitant crustal assimilation and He-loss during fractional crystallization. Limited assimilation ($\leq 1\%$) of incompatible element rich crustal gneisses with low $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ by melts from variably depleted mantle sources can explain Nd-Pb isotope compositions of Baffin Island and West Greenland picrites. Icelandic lavas provide supporting evidence that the ancestral mantle plume responsible for generating NAIP magmatism sampled variably enriched and depleted mantle, with no evidence for ancient non-chondritic mantle sources. Pervasive crustal contamination and partial melting of heterogeneous mantle sources, generated by plate tectonic processes, can account for the compositions of continental flood basalts (CFB), without the requirement of a non-chondritic terrestrial reservoir. Combined with increasing evidence that the $^{142}\text{Nd}/^{144}\text{Nd}$ composition of the bulk silicate Earth is due to nucleosynthetic *S*-process deficits in chondrite meteorites, these observations cast doubt that NAIP lavas sampled a non-chondritic mantle source with Sm/Nd higher than in chondrites. If short-lived radiogenic (e.g., ^{146}Sm - ^{142}Nd , ^{182}Hf - ^{182}W) isotope anomalies are found in CFB, they must either reflect assimilation of isotopically anomalous crustal materials, or partial melting of early-formed mantle heterogeneities produced by differentiation and late accretion.

1. Introduction

A long-standing paradigm for Earth is that it possesses a composition similar to some chondrite meteorite groups (e.g., Jacobsen & Wasserburg, 1980; Sun & McDonough, 1989; Javoy et al., 2010). This paradigm has recently been challenged by the observation that the accessible Earth has $^{142}\text{Nd}/^{144}\text{Nd}$ approximately 20 ppm greater than in ordinary or CV chondrites (Boyet & Carlson, 2005). Because ^{142}Nd forms from the decay of ^{146}Sm , with a half-life of 103 Ma (Meissner et al., 1987), this difference has been interpreted to require a supra-chondritic Sm/Nd ratio for the accessible Earth, presumably generated during differentiation in the first 30 Ma of Solar System formation (Boyet & Carlson, 2005). To account for a supra-chondritic accessible Earth reservoir, a complementary low Sm/Nd reservoir must have existed, now isolated in Earth, or that was lost by erosion during catastrophic impacts in the final stages of planetary accretion (e.g., Campbell & O'Neill, 2012). Jackson et al. (2010) and Jackson & Carlson (2011) recognized that, if a supra-chondritic Earth is valid, all known modern terrestrial mantle reservoirs should have evolved with Sm/Nd ratios approximately 5% higher than that of chondrites. They reasoned that combinations of Pb isotopic compositions on or close to the geochron, high- $^3\text{He}/^4\text{He}$ and unradiogenic $^{143}\text{Nd}/^{144}\text{Nd}$ would represent characteristics expected for an ancient non-chondritic 'primitive terrestrial reservoir'. This hypothetical reservoir is referred to as the ancient non-chondritic mantle source for the remainder of this article.

Helium isotopes in particular provide key geochemical information on the origin of lavas and their parental magmas. $^3\text{He}/^4\text{He}$ measured in olivines from LIP lavas can be higher than mid-ocean ridge basalt (MORB = $8 \pm 1 R_A$; Graham, 2002; where R_A is the present-day $^3\text{He}/^4\text{He}$ ratio in the atmosphere, 1.38×10^{-6}), continental intraplate alkaline volcanic rocks (CIAV = $5.9 \pm 1.2 R_A$; Day et al., 2005), or the continental lithospheric mantle ($6.1 \pm 2.1 R_A$; Gautheron & Moriera, 2002; Day et al., 2015). Continental flood basalts (CFB), such as the 65 Ma Deccan Traps (Basu et al., 1993), the ~250 Ma Siberian Traps (Basu et al., 1995), the <17.5 Ma Columbia River Flood Basalts (Dodson et al., 1997), and quaternary lavas from Ethiopia (Marty et al., 1996) all have lavas containing olivines with $^3\text{He}/^4\text{He}$ higher than canonical MORB or CIAV. These results suggest that less-degassed mantle sources with elevated abundances of primordial ^3He are sampled by many large igneous provinces. The highest $^3\text{He}/^4\text{He}$ ratios measured in any terrestrial

materials have come from olivine-rich basalts from the North Atlantic Igneous Province (NAIP) (**Figure 1**). Olivine separates from lavas at Baffin Island (up to 50 R_A; [Stuart et al., 2003](#); [Starkey et al., 2009](#)), West Greenland (up to 48 R_A; [Graham et al., 1998](#); [Starkey et al., 2009](#)), North East Greenland (up to 20 R_A; [Marty et al., 1998](#)), East Greenland (18 R_A; [Peate et al., 2003](#)), and Skye in the British Palaeogene Igneous Province (up to 22 R_A; [Stuart et al., 2000](#)), indicate derivation from a mantle source with similar He isotope characteristics to present-day Icelandic volcanism (up to 38 R_A; e.g., [Condomines et al., 1983](#); [Hilton et al., 1999](#); [Breddam et al., 2000](#); [Macpherson et al., 2005](#)). A long-lived, deep-seated mantle plume that has sustained volcanism since the Tertiary in the North Atlantic region has been posited to explain this relationship (e.g., [Saunders et al., 1997](#)).

It has recently been suggested that NAIP lavas with high-³He/⁴He, lead isotope compositions close to the 4.5 Ga terrestrial geochron, and unradiogenic ¹⁴³Nd/¹⁴⁴Nd sample an ancient non-chondritic mantle source ([Jackson et al., 2010](#)). Alternatively, it has been proposed that noble gases are strongly decoupled from other geochemical tracers at Baffin Island, West Greenland ([Stuart et al., 2003](#); [Starkey et al., 2012](#)) and elsewhere, due to the highest He concentration mantle reservoir dominating the ³He/⁴He composition during upper mantle partial melting processes ([Day & Hilton, 2011](#)). There is also strong evidence that trends in ¹⁴³Nd/¹⁴⁴Nd and Pb isotopes reflect contamination by continental crust (e.g., [Thirlwall & Jones, 1983](#); [Peate et al., 2008](#); [Larsen & Pedersen, 2009](#)). It is therefore equally possible that high-³He/⁴He melts from depleted mantle sources, which assimilate crustal rocks with low ²⁰⁶Pb/²⁰⁴Pb and enriched ¹⁴³Nd/¹⁴⁴Nd compositions, could explain the isotopic signatures preserved in NAIP lavas. Indeed, a persistent problem for evaluating potential mantle source contributions to large igneous provinces is that they transit through crust and lithosphere on their way to Earth's surface, so that there is always potential for crust-mantle interaction.

It is well-demonstrated that crustal contamination modifies Sr-Nd-Pb-Os isotope compositions in CFB (e.g., [Carter et al., 1978](#); [Cox, 1980](#); [Carlson et al., 1981](#); [Carlson, 1984](#); [Day et al., 2013](#)), and in NAIP lavas in particular ([Carter et al., 1978](#); [Dickin et al., 1981](#); [1984](#); [Thompson, 1982](#); [Thirlwall & Jones, 1983](#); [Gibson, 1990](#); [Lightfoot et al., 1997](#); [Geldmacher et](#)

al., 2002; Larsen & Pedersen, 2009). Fresh minerals from igneous rocks from Skye in the NAIP have anomalously low- $^{18}\text{O}/^{16}\text{O}$, due to assimilation of hydrothermally altered country rock (Taylor & Forester, 1971; Gilliam & Valley, 1998; Monani & Valley, 2001). This phenomenon also occurs in Iceland, where the assimilation of hydrothermally altered country rock by melts leads to variations in O isotopes and incompatible trace-element abundances, and leads to large range in $^3\text{He}/^4\text{He}$ due to degassing and assimilation processes (Condomines et al., 1983; Gautason & Muehlenbachs, 1998; Macpherson et al., 2005; Bindeman et al., 2005; Thirlwall et al., 2006; Pope et al., 2013). In this study, combined O-He-Nd-Pb isotope and trace element abundance data for picrites (>13.5 wt.% MgO), olivine-rich basalts and intrusive rocks from Skye (Scotland), East and West Greenland, Baffin Island and Iceland are used to evaluate whether ancient ‘non-chondritic’ mantle reservoirs can be unambiguously identified. NAIP picrites from Baffin Island and West Greenland represent near-primary highly-magnesian magma compositions, interpreted to reflect melting at mantle potential temperatures 100 to 300°C hotter than ambient asthenosphere (Francis, 1985; Gill et al., 1992; Bell & Williamson, 1994; Brown & Leshner, 2014). The lavas have both ‘enriched’ and ‘depleted’ Sr-Nd-Pb isotope and trace-element melt compositions, similar to modern Icelandic lavas (Robillard et al., 1992; Holm et al., 1993; Kent et al., 2004; Starkey et al., 2009; 2012). Combined, these petrogenetic features and identification of some NAIP lavas as potentially originating from an ancient non-chondritic mantle source (Jackson et al., 2010) make samples from this large igneous province important for further study.

2. Samples and Methods

Olivine and clinopyroxene major-element analyses were performed on some of the NAIP rocks using a *Jeol 8900* electron microprobe analyser at the University of Maryland, College Park (*Tables S1 & S2*). Mineral compositions were determined in wave-length dispersive spectral mode using an accelerating potential of 15 keV, a 10 nA beam current, with beam focus of 1 μm . Peak and background counting times of 20 s and standard ZAF correction procedures were used. Natural and synthetic standards were employed for calibration. Drift was within counting error through the analytical session. Detection limits (3σ above background) were <0.03 wt. % for all elements listed. Trace elements on Icelandic samples were determined on 100 mg of

homogenized whole rock powder using a *Perkin Elmer* ELAN 6000 inductively coupled plasma mass spectrometer at the University of Durham using standard analytical procedures (Ottley et al., 2003). Data were calibrated and monitored relative to accepted values of international standards (BHVO-1, AGV-1, BE-N, SRM 688) and internal standards (peridotite GP13, El Hierro lava EH01), with a reproducibility generally better than $\pm 3\%$ for basaltic materials (see Day et al., 2010; 2013).

Oxygen isotope analyses were performed on visibly ‘fresh’ olivine grains from crushed rocks. These grains were separated by hand from the matrix and ultrasonically cleaned in ultrapure water (18.2 M Ω MQ-H₂O) and then methanol. Samples were pre-fluorinated prior to analysis. Values of $\delta^{18}\text{O}$ ($\delta^{18}\text{O}_n$ is the per mil [‰] deviation of $^{18}\text{O}/^{16}\text{O}$ in n from the international standard [std] V-SMOW given by the relationship: $\delta^{18}\text{O}_n = 1000 \times ([^{18}\text{O}/^{16}\text{O}_n]/[^{18}\text{O}/^{16}\text{O}_{\text{std}}]-1)$) were measured at Royal Holloway, University of London on 1 to 2 mg splits using the laser fluorination protocols outlined in Matthey et al. (1994). Yields for all unknowns and standards in this study were $95 \pm 5\%$. Precision and accuracy of analyses was monitored with three internal standards; two different olivine populations from San Carlos, and UW garnet. The isotopic composition of these minerals relative to the international standard biotite NBS-30, and deviations of replicate analyses over the analytical period were: RHUL SCOL I $+4.84\text{‰}$ (± 0.16 , 2σ , $n = 27$), RHUL SCOL II $+5.22\text{‰}$ (± 0.16 , 2σ , $n = 19$) and UW GMG 2 garnet $+5.71\text{‰}$ (± 0.18 , 2σ , $n = 22$). The veracity of the laser fluorination O isotope method employed in this study has been outlined in Matthey et al. (1994), and latterly, Day et al. (2012).

In addition to the new data, previously published He-O-Sr-Nd-Pb isotope and trace-element abundance data are compiled for basalts from Iceland (Day, 2004; Thirlwall et al., 2004; Macpherson et al., 2005), Scotland (Stuart et al., 2000), Ireland (Ellam & Stuart, 2000), East Greenland (Brown et al., 1996; Peate et al., 2003), West Greenland (Larsen et al., 2009) and Baffin Island (Kent et al., 2004). These data are compiled in **Table 1** and in *Tables S3* and *S4*.

3. Results

Olivines from Icelandic basalts have forsterite contents ranging from 76 to 92, with the highest Ni contents (0.4 wt.%) occurring in olivine with the highest Mg/Fe ratios. Comparison of new and published olivine mineral chemistry data for Icelandic lavas versus igneous rocks from the British Palaeogene Igneous Province, East Greenland and Baffin Island indicate a comparable range of primary forsterite contents, from 86-91, indicating broadly similar, MgO-rich parental melt compositions, of between 13-15 wt.% MgO (e.g., [Francis, 1985](#); [Stracke et al., 2003](#)). New trace-element abundance data for Icelandic olivine-bearing basalts are presented in *Table S3* together with previously published trace-element abundance data for Baffin Island glasses from [Kent et al. \(2004\)](#), for which O and He isotope data on olivine grains from the same rocks are also available. Trace element abundances for SEL 97 and NAL 625 are identical within the uncertainties, and differ from trace element abundances measured for glasses from Baffin Island picrites with $^3\text{He}/^4\text{He}$ ratios up to 24 R_A ([Kent et al., 2004](#)), mainly in greater depletion of the heavy rare earth elements (HREE) in Icelandic samples (**Figure 3**). The close correspondence of trace element abundances between SEL 97, an ankaramite from the western Fjords, with high- $^3\text{He}/^4\text{He}$ (37.7 R_A ; [Hilton et al., 1999](#)) and NAL 625, an ankaramite with the highest $^3\text{He}/^4\text{He}$ from the central Icelandic rift zone (34.3 R_A ; [Macpherson et al., 2005](#)), suggests a similarly incompatible element enriched high- $^3\text{He}/^4\text{He}$ source for the parental melts of these two samples. When Sm/Yb ratios of the Icelandic lavas are plotted with the Group 1 Icelandic glasses identified by [Macpherson et al. \(2005\)](#) as the ‘least contaminated’ glasses from central Iceland, there is a broad positive correlation with $^3\text{He}/^4\text{He}$ and negative correlation with $\delta^{18}\text{O}$ (**Figure 4**). Baffin Island glass compositions measured by [Kent et al. \(2004\)](#) lie on the apparent correlations defined by Icelandic lavas, while Baffin Island and West Greenland lavas measured by [Starkey et al. \(2009\)](#) show no clear correlations of Sm/Yb against $^3\text{He}/^4\text{He}$.

Laser fluorination oxygen isotope analyses are presented for fresh, pre-fluorinated olivine separates from igneous rocks from Scotland, East Greenland and Iceland, along with previously reported olivine oxygen isotope data from Baffin Island in **Table 1**. The range of $\delta^{18}\text{O}$ values in olivine grains from lavas from Scotland (Skye Main Lava Series $\delta^{18}\text{O}_{\text{Ol}} = 4.95$ to 5.23‰), East Greenland ($\delta^{18}\text{O}_{\text{Ol}} = 4.83$ to 5.55‰) and Baffin Island ($\delta^{18}\text{O}_{\text{Ol}} = 4.84$ to 5.22‰ ; [Kent et al., 2004](#);

Starkey et al., 2016) are similar, and do not extend to the low $\delta^{18}\text{O}_{\text{OI}}$ values measured in plutonic rocks from the Skye Cuillin Complex (CC), Skye Dyke Swarm (SDS) or Little Minch Sill Complex (LMSC) (3.26 to 5.26‰). The range observed in the lavas extends across the mantle range of $\delta^{18}\text{O}_{\text{OI}}$ established from peridotites and basalts (Mattey et al., 1994; Eiler, 2001). Oxygen isotope ratios for the Icelandic lavas reported in **Table 1** also range to low $\delta^{18}\text{O}_{\text{OI}}$ (3.51 to 5.08‰; Day, 2004; Macpherson et al., 2005). Oxygen isotope ratios for the lavas are within the range of mantle peridotites (Mattey et al., 1994) and ocean island basalts from the Canary Islands (Day et al., 2010) and Hawai'i (Eiler et al., 1996), whereas the plutonic samples and Icelandic lavas trend to low $\delta^{18}\text{O}_{\text{OI}}$, consistent with previously reported values for Icelandic lavas (e.g., Thirlwall et al., 2006) (**Figure 2**).

4. Discussion

4.1 Evidence for pervasive crustal contamination in NAIP lavas

Crustal contamination processes that modify mantle-derived magmatic compositions have been acting throughout the eruptive history of NAIP and Icelandic magmatism. Evidence for crustal contamination of NAIP lavas has been extensively documented using trace-element abundances and Sr-Nd-Pb isotope systematics (e.g., Carter et al., 1978; Dickin et al., 1981; 1984; Thompson, 1982; Thirlwall & Jones, 1983; Gibson, 1990; Lightfoot et al., 1997; Geldmacher et al., 2002; Larsen & Pedersen, 2009). For example, Lightfoot et al. (1997) have identified West Greenland lavas with low Th/Nb, Zr/Nb and high La/Sm and $^{206}\text{Pb}/^{204}\text{Pb}$ that they interpreted to have experienced contamination from relatively mafic lower crustal rocks. Studies of both West and East Greenland lavas have indicated pervasive crustal contamination, with estimates of local crustal sediments or gneisses in the lavas as high as 5 to 30% (Fram & Leshner, 1997; Peate et al., 2008; Larsen & Pedersen, 2009). Despite these clear indications of crustal contamination, it has been noted that assimilation processes can be difficult to detect in other CFB lava suites (Larsen & Pedersen, 2009). This is partly due to uncertainties in initial melt composition and end-members for crustal contaminants, the nature and chemical distinctiveness of crustal contaminants (e.g., hydrothermally altered basalt versus gneiss), and partly due to the insensitivity of some trace elements to anything but significant (>5%) addition of crustal

contaminants. For example, [Huang et al. \(2010\)](#) reanalysed Baffin Island and West Greenland picrite data, previously considered to be crustally uncontaminated ([Jackson et al., 2010](#)), showing that Nb and Th concentrations, higher $^{87}\text{Sr}/^{86}\text{Sr}$ and lower $^{143}\text{Nd}/^{144}\text{Nd}$ occur with increasing $^{208}\text{Pb}^*/^{206}\text{Pb}^*$, consistent with crustal contamination ($^{208}\text{Pb}^*/^{206}\text{Pb}^*$ measures time-integrated Th/U, and is defined as: $[(^{208}\text{Pb}/^{204}\text{Pb})_{\text{sample}} - (^{208}\text{Pb}/^{204}\text{Pb})_{\text{CDT}}] / [(^{206}\text{Pb}/^{204}\text{Pb})_{\text{sample}} - (^{206}\text{Pb}/^{204}\text{Pb})_{\text{CDT}}]$, where CDT = Canñn Diablo Troilite, or primordial Pb).

Other potentially sensitive measures of crustal contamination are O and He isotopes, especially in early-formed olivine grains in basaltic rocks. Oxygen isotopes are particularly useful for tracking crustal contamination, especially if the contaminants do not differ significantly in major- or trace-element composition relative to melts, as is the case in assimilation of recently emplaced, hydrothermally altered basaltic crust into mafic magmas. Early studies of O isotopes showed that hydrothermal alteration during NAIP magmatism was pervasive (e.g., [Taylor & Forester, 1971](#); [Muehlenbachs et al., 1974](#); [Condomines et al., 1983](#); [Gilliam & Valley, 1998](#); [Monani & Valley, 2001](#); [Macpherson et al., 2005](#); [Burnard & Harrison, 2005](#); [Pope et al., 2013](#)). In a classic study, [Forester & Taylor \(1977\)](#) showed that concentric hydrothermal systems existed around the intrusive centres of Skye, Mull and Ardnamurchan, based on $\delta^{18}\text{O}$ variations from basalts and basaltic dykes. They found that the lowest $\delta^{18}\text{O}$ values occurred in the centre of the intrusive complexes, to values as low as -5‰. The new oxygen isotope data reported here for fresh olivine grains from the Skye Dyke Swarm and Cuillin Complex demonstrate that assimilation of low- $\delta^{18}\text{O}$ hydrothermally altered rocks was occurring prior to crystallization of some NAIP mafic magmas, with olivine $\delta^{18}\text{O}$ values as low as 3.3‰. These results are supported by observations of low- $\delta^{18}\text{O}$ in zircons from the Cuillin Complex, Skye ([Monani & Valley, 2001](#)). Similarly, there is strong evidence for assimilation of hydrothermally altered crustal components in Icelandic lavas (e.g., [Condomines et al., 1983](#); [Macpherson et al., 2005](#); [Bindemann et al., 2006](#)).

Ageing and ingrowth of ^4He due to alpha-decay of U and Th should theoretically not be a major factor in $^3\text{He}/^4\text{He}$ values measured by crushing olivine *in vacuo* in relatively young rocks (<100 Ma; for example, see calculations of [Day et al. \[2015\]](#)). Crushing of olivine releases

helium and other gases trapped in melt and fluid inclusions, rather than lattice-based He, which includes both helium retained from the primitive melt, as well as ingrown ^4He (e.g., Scarsi et al., 2000). Graham et al. (1998) performed both crushing and melting experiments on olivine from 61 Ma West Greenland olivine grains, showing that the crushed $^3\text{He}/^4\text{He}$ values were higher, and He concentrations lower, than corresponding melting experiments (**Figure 5b**). A combined dataset of NAIP He isotopes is inconsistent with ingrowth of ^4He from U and Th decay after olivine crystallization (**Figure 5**). Low He concentrations in degassed parental melts of lavas make them susceptible to minor amounts of crustal contamination *prior* to crystallization of the olivine and demonstrate that these samples have experienced crustal interaction. Degassing of He leads to lower concentrations of He in the melt, reducing the He/(U+Th) ratio. After emplacement, low He/(U+Th) reservoirs will evolve to low- $^3\text{He}/^4\text{He}$, through the ingrowth of ^4He .

Applying models of contamination by low- $\delta^{18}\text{O}$, low- $^3\text{He}/^4\text{He}$ crust with a mantle source end-member composition with high- $^3\text{He}/^4\text{He}$ indicates that variable k -values (where k is dominated by the concentration of He in the crust versus the mantle melt component) are required to explain the O-He isotope systematics of NAIP and Icelandic olivines and glasses (**Figure 5a**). The range in observed $^3\text{He}/^4\text{He}$ requires variable and extensive He-loss in melts prior to assimilation of hydrothermally altered crust and crystallization of the olivine (e.g., Condomines et al., 1983; Macpherson et al., 2005). Crustal contamination and outgassing of He occurs significantly prior to the eruption of lavas, explaining the highly variable $^3\text{He}/^4\text{He}$ values measured in olivines from locations such as Baffin Island (3 to 50 R_A ; Stuart et al., 2003; Starkey et al., 2009; Jackson et al., 2010), and mixing between crustal components and mantle-derived melts can therefore have a strong control on Nd-Pb isotope compositions in NAIP lavas.

Some Icelandic picrites have both low- $\delta^{18}\text{O}$ and high- $^3\text{He}/^4\text{He}$ (>8 to 38 R_A ; **Figure 5**), implying crustal-like oxygen isotopes preserved in olivine grains with primitive noble gas isotope systematics (c.f., SEL 97, SK-1, NAL 626, NAL 626, Pjor, TH-29; **Table 1**). Alternatively, some lavas may have high $^3\text{He}/^4\text{He}$ olivine, but a bulk-rock composition consistent with derivation from a crustally-contaminated melt. Sample LL1 from the Skye Main Lava Series (SMLS) is an example of this phenomena, with high- $^3\text{He}/^4\text{He}$ (22 R_A), low- $\delta^{18}\text{O}_{\text{olivine}}$ (4.95‰) and a highly

unradiogenic Pb isotope composition ($^{206}\text{Pb}/^{204}\text{Pb} = 16.3$). Consequently, it is unsafe to assume that high- $^3\text{He}/^4\text{He}$ measured in olivines from a volcanic rock means that the volcanic rock itself has not experienced crustal modification. Helium, O, Sr, Nd and Pb isotope data for NAIP lavas further reinforce that highly magnesian magmas are not impervious to crustal contamination (cf., Cox, 1980; Thirlwall & Jones, 1983).

4.2 *Crustal contamination, not a non-chondritic source for NAIP magmas*

NAIP picrites and high-magnesian basalts from Baffin Island, Greenland and Scotland have all traversed through continental crust and there is clear evidence from Sr-Nd-Hf-Os-Pb isotope systematics for crustal contamination of magmas (e.g., Dickin, 1981; Dickin et al., 1984; Lightfoot et al., 1997; Fram & Lesher, 1997; Geldmacher et al., 2002; Peate et al., 2003; 2008; O'Driscoll et al., 2009; Larsen & Pedersen, 2009). Indeed, the combined evidence from He-O-Nd-Pb isotope and trace-element abundance systematics strongly support previous arguments that minor to extensive crustal contamination is a probable cause for the isotopic compositions of NAIP lavas (e.g., Thirlwall & Jones, 1983; Dickin et al., 1984; Lightfoot et al., 1997; Peate et al., 2008; Larsen & Pedersen, 2009). Archaean to Proterozoic crustal protoliths from Greenland and Scotland are characterized by having much higher Pb and Nd concentrations than basaltic melts (**Figure 3**), and have unradiogenic Pb isotope compositions and low $^{143}\text{Nd}/^{144}\text{Nd}$. In $^{206}\text{Pb}/^{204}\text{Pb}$ - $^{207}\text{Pb}/^{204}\text{Pb}$ space the crustal gneisses generally plot to the left of the 4.568 Ga geochron (**Figure 6**) and, in a plot of $^{206}\text{Pb}/^{204}\text{Pb}$ versus $^{143}\text{Nd}/^{144}\text{Nd}$, the gneisses trend to low Pb and Nd isotope compositions (**Figure 7a**). Conversely, Icelandic lavas lie to the right of the geochron, have high $^{143}\text{Nd}/^{144}\text{Nd}$, and can be interpreted as originating from a depleted source, similar, although not identical, to the depleted MORB mantle (e.g., Chauvel & Hemond, 2000; Fitton et al., 2003; Stracke et al., 2003; Thirlwall et al., 2004).

Mixing models between an average Greenland gneiss composition and the most enriched and depleted Icelandic melt compositions measured by Thirlwall et al. (2004) illustrate that less than 1% crustal contribution can explain the Nd-Pb isotope systematics of West Greenland and Baffin Island lavas (**Figure 7a**). The robustness of these models is partly reliant on end-member parameters and, on levels of melt-depletion expected for a depleted mantle component; an even

more depleted mantle source end-member, as has been suggest for NAIP magmas (e.g., [Fitton et al., 2003](#)) would lead to greater degrees of crustal contamination. Furthermore, while some Icelandic lavas may have contributions from a low- $\delta^{18}\text{O}$ mantle source with relatively high $^{206}\text{Pb}/^{204}\text{Pb}$ (Model I2i) that is clearly distinct from Archaean crustal gneisses, much of the O-Pb isotope variations in NAIP lavas can also be explained by contributions from a recycled crustal protolith (**Figure 7b**). Some have argued that this protolith is recycled oceanic crust (e.g., [Macpherson et al., 2005](#)). To further emphasise that crustal contamination is a pervasive process in NAIP magmas, some East Greenland and British Palaeogene Igneous Province basaltic lavas plot to the left of the geochron (**Figure 6**). For example, LL1, with a $^3\text{He}/^4\text{He}$ ratio of 22 R_A has $^{206}\text{Pb}/^{204}\text{Pb}$ of 16.3. As mentioned previously, this means that lavas with significant crustal contributions can contain olivines with high- $^3\text{He}/^4\text{He}$ and $\delta^{18}\text{O}_{\text{olivine}}$ close to mantle compositions. Based on the overwhelming evidence for crust-mantle melt interactions during formation of the NAIP, it is probable that the Pb isotope compositions of Baffin Island and West Greenland picrites, which lie close to the 4.568 to 4.45 Ga geochrons, are coincidental and the result of variable contamination by high Pb and Nd concentration, low $^{206}\text{Pb}/^{204}\text{Pb}$, low $^{143}\text{Nd}/^{144}\text{Nd}$ continental crust.

The chemical identity of Baffin Island and West Greenland picrites represents variable mixtures between depleted mantle components and crustal contaminants. For example, the lower- $^3\text{He}/^4\text{He}$ (3 to 24 R_A) in the Baffin Island samples studied by [Jackson et al. \(2010\)](#) relative to other Baffin Island and West Greenland picrites (30 to 50 R_A ; [Graham et al., 1998](#); [Stuart et al., 2003](#); [Starkey et al., 2009](#)) most likely reflects concomitant degassing of He and crustal contamination. Trace-element abundance systematics for Baffin Island lavas have similar absolute and relative abundances to enriched, high- $^3\text{He}/^4\text{He}$ Icelandic picrites considered to come from non-primitive mantle sources and that have not traversed continental crust. Both Baffin Island and West Greenland picrites show evidence for enriched and depleted melt compositions ([Robillard et al., 1992](#); [Holm et al., 1993](#); [Kent et al., 2004](#); [Starkey et al., 2009](#); [2012](#)), and have significant variability in Nd-Pb isotopes ($^{206}\text{Pb}/^{204}\text{Pb} = 17.74\text{-}18.33$; $^{143}\text{Nd}/^{144}\text{Nd} = 0.51213\text{-}0.51311$), more consistent with assimilation of unradiogenic crustal protoliths than with a unique, non-chondritic mantle composition.

4.3 *Potential identification of early-formed mantle plume reservoirs*

Minor assimilation of crust in hot, highly magnesian magmas has been recognized as an important process for more than thirty-six years (Cox, 1980). In addition to NAIP picrites, a compilation of large igneous province (LIP) basaltic rocks (Kerguelen Plateau, Deccan Traps (India), Antarctic Karoo, Siberia and Ontong Java Plateau) was used by Jackson & Carlson (2011) to reinforce the concept of a non-chondritic primitive mantle with Pb isotope compositions that plot near to the geochron, as the source of these LIP. Of these LIP, only the Ontong Java Plateau did not erupt onto continental crust and, while high- $^3\text{He}/^4\text{He}$ ratios have been measured for some samples from LIP (e.g., Deccan and Siberia; Basu et al., 1993; 1995), to date, helium isotope data have not been reported for Ontong Java Plateau basalts due to difficulties in obtaining fresh olivine-phyric samples. A definitive link between high- $^3\text{He}/^4\text{He}$ in olivines from lavas and a hypothetical non-chondritic primitive mantle is therefore lacking. It is notable that high- $^3\text{He}/^4\text{He}$ has been measured in near-primary picrite flows from Baffin Island and West Greenland that span a range of incompatible element compositions (Starkey et al., 2009). This is consistent with noble gas systematics of intraplate lavas being controlled by the most gas-rich reservoir involved in mantle melting (Day & Hilton, 2011). It is likely that high- $^3\text{He}/^4\text{He}$ magmas predominantly reflect derivation from a less depleted mantle source with sub-equal to higher He contents than depleted mantle. Helium isotopes can therefore be inherited into melts generated from mantle sources with distinct O-Sr-Nd-Hf-Pb-Os isotope compositions, or that have been crustally contaminated.

Ancient continental crust lies to the left of the 4.568 Ga Pb isotope geochron and is highly variable in Sr-Nd-Pb isotope compositions, while almost all MORB and ocean island basalts lie to the right of the 4.55 Ga Pb isotope geochron. Inevitably, mixing between these end-members during crustal-mantle melt interactions will lead to variable Nd and Pb isotope compositions in resulting lavas. An expectation of crust-mantle melt interactions is that Nd-Pb isotope compositions should vary, even in magnesian basalts and picrites. Conversely, an ancient primitive mantle source composition (e.g., Jackson et al., 2010; Jackson & Carlson, 2011) should be reflected in a relatively homogeneous end-member composition, due to the large-scale

differentiation event involved during its formation (e.g., Boyet & Carlson, 2005). Jackson & Carlson (2011) identified primitive mantle-derived LIP samples based on their similarity to the range of present-day $\epsilon^{143}\text{Nd}$ ($\epsilon^{143}\text{Nd} = [({}^{143}\text{Nd}/{}^{144}\text{Nd})_{\text{sample}}/({}^{143}\text{Nd}/{}^{144}\text{Nd})_{\text{CHUR}} - 1] \times 10,000$) of 5.3 to 9.0 for ocean island basalts that they argued were consistent with a primitive mantle source with Sm/Nd ratios 4.2-7.3% higher than that of chondrites. Consequently, their select samples from the Kerguelen Plateau, Deccan Traps (India), Antarctic Karoo, Siberia and Ontong Java Plateau have initial $\epsilon^{143}\text{Nd}$ values spanning a range between 5.2 and 8.4. For this relatively restricted range of selected ${}^{143}\text{Nd}/{}^{144}\text{Nd}$ compositions, the ranges of ${}^{206}\text{Pb}/{}^{204}\text{Pb}$ (17.5-18.5) and ${}^{207}\text{Pb}/{}^{204}\text{Pb}$ (15.38-15.68) are remarkably large.

The range in Pb isotope compositions for North Atlantic gneisses and in global compilations of lower crustal rocks with low ${}^{143}\text{Nd}/{}^{144}\text{Nd}$ (${}^{206}\text{Pb}/{}^{204}\text{Pb} = 13.5-19.5$; ${}^{207}\text{Pb}/{}^{204}\text{Pb} = 14.6-15.8$; Murphy et al., 2002), make it equally plausible that the spread in compositions presented by Jackson & Carlson (2011) is an artefact of the choice of filter applied and reflect contamination by crustal rocks, rather than a preservation of a primitive mantle signature. To demonstrate this point, an unfiltered dataset of measured Nd and Pb isotopes for relatively young CFB (Ethiopian CFB, Columbia River Flood Basalt [CRFB], the Deccan Trap CFB, and the NAIP) was compiled from the GEOROC database, along with associated data from the Ontong Java Plateau (OJP) and a global mid-oceanic ridge basalt dataset from the PETDB database (Figure 8). The data presented in the figure are not corrected for Nd or Pb ingrowth since emplacement, but the relatively short time-span since emplacement means ageing has limited effect on the observed isotopic dispersion. A number of important observations come from such diagrams. First, many CFB lavas must be strongly contaminated by crust with unradiogenic Nd and Pb isotope compositions. Second, CFB lavas that plot at the appropriate $\epsilon^{143}\text{Nd}$ composition to be consistent with a non-chondritic Sm/Nd source span a remarkable range in ${}^{206}\text{Pb}/{}^{204}\text{Pb}$ isotope compositions (~16 to 19). Third, many of the NAIP lavas are more depleted than the estimated non-chondritic Earth composition, yet have ${}^{206}\text{Pb}/{}^{204}\text{Pb}$ close to the 4.568-4.45 Ga geochrons. Fourth, lavas with more radiogenic ${}^{206}\text{Pb}/{}^{204}\text{Pb}$ typically have ${}^{143}\text{Nd}/{}^{144}\text{Nd}$ that is less radiogenic than the non-chondritic Earth value and overlapping chondritic values. Collectively, these observations are not easily explained by an accessible Earth with supra-chondritic Sm/Nd.

Instead, they appear to be more consistent with the generation of depleted and enriched reservoirs in the upper mantle through long-term crust-mantle differentiation that are sampled by CFB that subsequently undergo crustal contamination. There is clearly some concern about filtering such complex datasets using He, Nd and Pb isotopes, especially in the absence of He isotope data for many samples (cf. Ontong-Java Plateau lavas).

Crustal contamination is a dominant process in generating isotopic and trace-element abundance variations in CFB. In contrast, [Jackson & Carlson \(2011\)](#) argued that crustal contamination could not be a significant issue for Ontong Java Plateau basalts. This is because Ontong Java Plateau melts were erupted through oceanic lithosphere. However, there is evidence for continental crustal contributions during formation of the Ontong Java Plateau. The presence of garnet pyroxenites from Malaita, Solomon Islands with relatively low $^{206}\text{Pb}/^{204}\text{Pb}$ (17.9-18.4) and $^{143}\text{Nd}/^{144}\text{Nd}$ (0.51217-0.51275) has been interpreted by [Ishikawa et al. \(2007\)](#) to reflect the presence of an ancient recycled crustal component in the mantle source of the Ontong Java Plateau. [Ishikawa et al. \(2007\)](#) argued that evaluation of any primitive mantle-like source would be required because of the potential contribution of recycled eclogite/pyroxenite. Ontong Java Plateau lavas plot to the lowest $^{143}\text{Nd}/^{144}\text{Nd}$ and highest $^{206}\text{Pb}/^{204}\text{Pb}$ and are not especially consistent with an ancient non-chondritic mantle source.

4.4 Chondritic versus non-chondritic accessible Earth?

Original geochemical models of terrestrial composition have assumed a ‘chondritic Earth’, meaning a planet with the bulk composition similar to chondrites and to the bulk photosphere of the Sun (e.g., [Jacobsen & Wasserburg, 1980](#); [Campbell & O’Neill, 2012](#)). Measurements of $^{142}\text{Nd}/^{144}\text{Nd}$ differences between ordinary and CV chondrites and Earth materials have challenged this view, implying a supra-chondritic Sm/Nd ratio for the accessible Earth ([Boyet & Carlson, 2005](#)). Recent work suggests that the difference in $^{142}\text{Nd}/^{144}\text{Nd}$ between the accessible Earth and chondrites may not be caused by radiogenic decay of ^{146}Sm and early terrestrial differentiation. Instead, a number of studies show that nucleosynthetic *S*-process deficits in modern-day chondrites may be responsible for the observed 10-20 ppm apparent offset in $^{142}\text{Nd}/^{144}\text{Nd}$ (e.g, [Gannoun et al., 2011](#); [Marks et al., 2014](#); [Burkhardt et al., 2016](#); [Fukai &](#)

Yokoyama, 2016). Correction of these nucleosynthetic anomalies reveals no resolvable difference in $^{142}\text{Nd}/^{144}\text{Nd}$ between the Earth and chondrites. These results have important implications for the building blocks of Earth, and obviate the need for a non-chondritic accessible Earth. Thus, the accessible Earth is likely ‘chondritic’ with respect to elemental abundance. The evidence shown here for variation in He-O-Nd-Pb isotopes of NAIP lavas being caused by variable crustal contamination of mantle-derived melts potentially leads the debate on a chondritic Earth composition full circle. Namely, crust-mantle differentiation, combined with plate tectonic processes, can account for the range of isotopic compositions in the majority of terrestrial lavas (e.g., O’Nions et al., 1979; Allegre, 1982; Zindler & Hart, 1986). Recent discoveries of short-lived isotopic heterogeneities, including ^{182}W isotopic heterogeneities in komatiites (Touboul et al., 2012), leads to the possibility that some CFB may also exhibit similar isotopic heterogeneities. In the absence of a non-chondritic accessible Earth, these anomalies can be interpreted to reflect either sampling of poorly-mixed, early-formed mantle reservoirs formed by a combination of large-scale magmatic differentiation and variable metal-silicate equilibrium, or from contamination by ancient crustal reservoirs with high W and $^{182}\text{W}/^{184}\text{W}$ values. Examples of such crustal rocks occur in the region of the NAIP and include the 3.96 Ga Acasta Gneiss Complex (NW Canada) (Willbold et al., 2015), or the 3.3-3.8 Ga Isua Crustal Belt mafic and ultramafic rocks, SW Greenland (Rizo et al., 2016).

5. Conclusions

Complexities of crustal contamination and the presence of recycled crust and lithosphere in the sources of LIP prevent the unambiguous identification of ancient non-chondritic mantle sources. This is the case for NAIP lavas, where combined He-O-Nd-Pb isotope compositions and trace-element abundances indicate that they derived from both enriched and depleted mantle sources and experienced variable concomitant crustal assimilation, as well as He-loss during crystallization of host olivine. NAIP lavas have experienced volumetrically high (>20%) to low (<1%) proportions of crustal contamination, even in samples with high- $^3\text{He}/^4\text{He}$. These crustal assimilants can have high Nd and Pb abundances and low $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{143}\text{Nd}/^{144}\text{Nd}$, indicating that the Pb isotope composition of Baffin Island and West Greenland lavas reflects mixing between enriched and depleted mantle sources and the continental crust. Similar enriched and

depleted sources are responsible for present-day magmatism in Iceland where high $^{143}\text{Nd}/^{144}\text{Nd}$ indicates highly depleted mantle sources. Combined with increasing evidence that the accessible Earth is in fact chondritic in bulk composition, these results reaffirm that CFB compositions primarily reflect mixing between enriched crust and variably depleted mantle reservoirs. It is likely that high- $^3\text{He}/^4\text{He}$ magmas predominantly reflect derivation from a less depleted mantle source with sub-equal to higher He contents than depleted mantle and that He isotopes can be strongly dissociated from radiogenic isotopes.

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Figures and figure captions

Figure 1 – Location map of olivine-rich igneous rocks from the North Atlantic Igneous Province discussed in this study (filled squares). Also shown is the inferred plume-track from 60 Ma to the present day (after [Saunders et al., 1997](#)). Age information and discussion of locations are provided in the text. Abbreviations: BPIP = British Palaeogene Igneous Province; BI = Baffin Island; WG = West Greenland; EG = East Greenland; KR = Kolbeinsey Ridge; RR = Reykjanes Ridge; MR = Mohns Ridge; JMFZ = Jan Mayen Fracture Zone; CGFZ = Charlie Gibbs Fracture Zone.

Figure 2 – Histogram plots of $\delta^{18}\text{O}_{\text{olivine}}$ for North Atlantic Igneous Province igneous rocks (filled) versus (a) Western Canary Island lavas (Day et al., 2010) and Hawai'ian lavas (Eiler et al., 1996) and (b) Icelandic lavas (Thirlwall et al., 2006 and references therein) and mantle olivines from peridotite nodules (Mattey et al., 1994). Average and two standard deviations are reported.

Figure 3 – Primitive-mantle normalised incompatible trace element patterns for Icelandic picrites and olivine-rich basalts compared with glass compositions from Baffin Island picrites (Kent et al., 2004; grey shading) and shown with corresponding ranges in $^3\text{He}/^4\text{He}$ for olivine grains included within the samples. Also shown are the representative ranges of Greenland crustal gneiss compositions, which have high abundances of incompatible trace elements (Rb, Ba, Th, U, Nb, Ta, K and La abundances plot off scale), including Pb (from Shimizu et al., 1988 and references therein). Primitive mantle normalisation from Sun & McDonough (1989).

Figure 4 – Samarium/ytterbium versus (a) $^3\text{He}/^4\text{He}$ and (b) $\delta^{18}\text{O}$ for olivines from Iceland and Baffin Island (Kent et al., 2004) compared with the most primitive Group-1 glasses from Central Iceland (Macpherson et al., 2005) and Baffin Island picrites (BI and WG; Starkey et al., 2009). Field of MORB helium isotope composition in (a) is from Graham (2002) and for mantle olivine $\delta^{18}\text{O}$ in (b) is from Matthey et al. (1994). Values of $\delta^{18}\text{O}$ for Icelandic Group 1 glasses have been converted to $\delta^{18}\text{O}_{\text{olivine}}$ using an empirically-derived $\Delta_{\text{glass-olivine}}$ fractionation factor of 0.5‰ (Eiler, 2001). Ratios of Sm/Yb in depleted MORB mantle (DMM = 0.65 to 0.67) and primitive mantle/CI-chondrites (PM/CI = 0.92) from Workman & Hart (2005) and Sun & McDonough (1989), respectively.

Figure 5 –Plots of helium isotope composition versus (a) $^{18}\text{O}/^{16}\text{O}$ (as $\delta^{18}\text{O}$) and (b) He concentration for NAIP igneous rocks. Mixing models in (a) are shown between Icelandic crust (C: $^3\text{He}/^4\text{He} = 0.05 R_A$; $[\text{He}] = 6.5 \times 10^{-8} \text{ cm}^3 \text{ STP/g}$ [Condomines et al., 1983]; $\delta^{18}\text{O} = 0\text{‰}$ [Eiler et al., 2000]) with low $\delta^{18}\text{O}$ and high $\delta^{18}\text{O}$ (iC: He same as for C; $\delta^{18}\text{O} = 10\text{‰}$ [Eiler, 2001]) with a high- $^3\text{He}/^4\text{He}$ mantle-derived melt (P: $^3\text{He}/^4\text{He} = 50 R_A$ [Stuart et al., 2003]; $\delta^{18}\text{O} = 5.2\text{‰}$ [Mattey et al., 1994; Eiler, 2001]) for different values of k [$=(\text{He}/\text{O})_C/(\text{He}/\text{O})_P$], where lower k -values correspond to higher $[\text{He}]$ in the mantle-derived melt. Black model lines in (b) are 10-1000 times degassing of He in basaltic melts with low- $^4\text{He}/^3\text{He}$ and addition of variable crustal $[\text{He}]$ with high- $^4\text{He}/^3\text{He}$ to simulate degassing and crustal assimilation prior to crystallization of olivine. Grey tie-lines are between crushing in vacuo measurements and melting measurements (grey circles, or off figure) of the same olivines from West Greenland (Graham et al., 1998) and Baffin Island (Jackson et al., 2010). Small black dots are Baffin Island data from Stuart et al. (2003) and Starkey et al. (2009). Note that the vectors for degassing and crustal contamination and in-growth of ^4He after eruption are different. Ice. Picrites = Iceland Picrites. Field of depleted MORB mantle (DMM), derived from He-O isotope systematics in MORB, shown for comparison.

Figure 6 – $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ diagram showing the Pb geochron at 4.568, 4.55, 4.5 and 4.45 Ga and the northern hemisphere reference line (NHRL; Hart, 1984). NAIP igneous rocks (Baffin Island, West Greenland, East Greenland and Skye) are plotted with published British Paleogene Igneous Province (BPIP) volcanic rocks (from Ellam & Stuart, 2000 and references therein) and versus crustal gneisses from the Lewisian complex of Scotland (Whitehouse & Robertson, 1995) and from Greenland (Moorbath et al., 1986), with Pb isotopic compositions corrected to 60 Ma. Field of Lewisian granulite and amphibolite (grey shaded region) from Geldmacher et al. (2002). While high- $^3\text{He}/^4\text{He}$ Iceland, West Greenland and Baffin Island samples plots on, or to the right of the 4.5 Ga geochron, Isle of Skye olivine basalt LL 1 ($22 R_A$) plots far to the left of the 4.568 Ga geochron.

Figure 7 – Plots of (a) $^{143}\text{Nd}/^{144}\text{Nd}$ and (b) $\delta^{18}\text{O}_{\text{olivine}}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ for NAIP igneous rocks. Mixing models are shown between end-member melt compositions for Icelandic parental melts (I1 = $^{206}\text{Pb}/^{204}\text{Pb}$ = 17.94-18.5; [Pb] = 0.7 ppm; $^{143}\text{Nd}/^{144}\text{Nd}$ = 0.5131; [Nd] 6.5 ppm; $\delta^{18}\text{O}_{\text{olivine}}$ = 5.2‰; I2 = $^{206}\text{Pb}/^{204}\text{Pb}$ = 19.2; [Pb] = 0.7 ppm; $^{143}\text{Nd}/^{144}\text{Nd}$ = 0.51296; [Nd] 6.5 ppm; $\delta^{18}\text{O}_{\text{olivine}}$ = 0 [I2ii] to 4 [I2i] ‰) versus average Greenland gneiss compositions (AC = $^{206}\text{Pb}/^{204}\text{Pb}$ = 14; [Pb] = 20 ppm; $^{143}\text{Nd}/^{144}\text{Nd}$ = 0.5105; [Nd] 25 ppm; $\delta^{18}\text{O}_{\text{olivine}}$ calculated from 0 [AC] to 10 [HAC] ‰), and intermediate crustal composition (IC = $^{206}\text{Pb}/^{204}\text{Pb}$ = 16.5; [Pb] = 20 ppm; $\delta^{18}\text{O}_{\text{olivine}}$ = 0‰). Icelandic parental melt parameters are from this study and Thirlwall et al. (2004; 2006). Crustal compositions are from Moorbath et al. (1986) and Bennett et al. (1993). I1, I2i and I2ii = Icelandic mantle compositions; AC = ‘Average Crust’; HAC = high- $\delta^{18}\text{O}$ ‘Average Crust’; IC = Intermediate Crust. Note that Icelandic compositions span a range from moderately enriched compositions with low $\delta^{18}\text{O}_{\text{olivine}}$ to strongly depleted, with mantle-like $\delta^{18}\text{O}_{\text{olivine}}$. The non-chondritic Earth and chondritic compositions are from Jackson et al. (2010). Data sources for Iceland are from Thirlwall et al. (2004; 2006), with other data presented in Table 1.

Figure 8 – Plots of (a) $^{143}\text{Nd}/^{144}\text{Nd}$ and (b) $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ for major continental flood basalts emplaced in the last ~65 Ma, versus data for Cretaceous Ontong Java Plateau volcanic rocks and global mid-ocean ridge basalts (Gray squares). Plots show the Pb geochron at 4.568, 4.55, 4.5 and 4.45 Ga and the northern hemisphere reference line (NHRL; Hart, 1984), the chondritic composition for Nd and the estimated composition of the ‘non-chondritic’ reservoir (from Jackson et al., 2010). Data for CFB were accessed from the GEOROC database (<http://georoc.mpch-mainz.gwdg.de/georoc/>) on the 7th April 2016 and data for MORB were accessed from the PETDB database (<http://www.earthchem.org/petdb/search>) on the 16th June 2016.

Table 1: Oxygen, helium, lead and forsterite content data for olivines in NAIP rocks

Sample	Location	$\delta^{18}\text{O}$		$^3\text{He}/^4\text{He}$		[4He]	$^{206}\text{Pb}/^{204}\text{Pb}$		Fo content		Data Sources
		(‰)	$\pm 2\sigma$	(R/R _A)	$\pm 2\sigma$	(nccSTP/g)		$\pm 2\sigma$		$\pm 2\sigma$	
Icelandic Lavas											
Sel 97	Westfjordur	4.60	-	37.7	2.0	0.9	-	-	86.8	0.5	[1], [2]
SK1	Westfjordur	4.14		37.7							[8]
Th-29	Northern Rift Zone	4.70	0.06	12.2	0.3	1.3	-	-	90.2	0.3	[1], [3]
NAL 239	Northern Rift Zone	4.01	-	10.6	0.3	7	-	-	-	-	[1], [3]
NAL 688 Gr	Northern Rift Zone	4.60	0.21	8.0	0.4	1.2	-	-	89.6	0.9	[1], [3]
NAL 688 Cc	Northern Rift Zone	5.08	0.27	5.9	0.3	1.3	-	-	91.1	1.1	[1], [3]
NAL 611	Northern Rift Zone	3.92	0.35	16.6	2	1	-	-	85.5	0.7	[1], [3]
NAL 625	Northern Rift Zone	4.16	0.23	34.3	4	0.35	-	-	86.2	1.1	[1], [3]
NAL 626	Northern Rift Zone	3.51	0.09	17.3	0.3	17.4	-	-	76.3	-	[1], [3]
Tro-53	Central Iceland Rift Zone	5.00	-	8.3	5	0.3	-	-	-	-	[1], [3]
Pjor	Southern Iceland Rift Zone	4.68	-	18.3	0.6	3.2	-	-	86.9	0.2	[1], [3]
British Palaeogene Igneous Province											
SK 266	SMLS, Isle of Skye	5.14	0.05	3.5	0.7	0.62	17.226	0.010	-	-	[1], [4]
BB1	SMLS, Isle of Skye	5.23	-	18.4	2.1	0.64	18.043	0.007	-	-	[1], [4]
LL1	SMLS, Isle of Skye	4.95	0.06	22.1	3.6	0.93	16.286	0.008	-	-	[1], [4]
SK291	SMLS, Isle of Skye	5.14	-	1.2	0.6	1.1	16.839	0.010	-	-	[1], [4]
INV1	LMSC, Isle of Skye	5.17	-	2.3	0.4	0.59	16.787	0.007	-	-	[1], [4]
BH1	LMSC, Isle of Skye	5.26	-	4.1	0.8	1.32	17.558	0.013	-	-	[1], [4]
SR81	LMSC, Isle of Skye	5.26	-	2.5	1.3	0.88	17.678	0.005	-	-	[1], [4]
SK056	SDS, Isle of Skye	4.79	-	2.9	0.1	11.6	17.534	0.015	-	-	[1], [4]
SK057	SDS, Isle of Skye	4.79	-	8.0	0.4	1.92	17.378	0.005	-	-	[1], [4]
SK204	SDS, Isle of Skye	3.26	0.54	1.1	0.04	9.57	16.806	0.006	85.0	1.8	[1], [4]
SK208	Cuillin Complex, Isle of Skye	4.74	-	6.6	0.25	0.66	17.377	0.006	-	-	[1], [4]
East Greenland											
EG2240	Prinsen af Wales, E. Greenland	5.55	0.08	9.7	0.8	-	17.551	0.008	89.5	1.5	[1], [5]
EG2244	Prinsen af Wales, E. Greenland	4.84	-	1.8	0.8	-	14.733	0.004	84.1	1.6	[1], [5]
EG3359	Prinsen af Wales, E. Greenland	4.83	-	10.5	1	-	18.328	0.006	-	-	[1], [5]
West Greenland											
400230		4.99		47.6							[8]
400485		4.99		40.3							[8]
Baffin Island											
BI/P1/27	Padopling Island, Baffin	5.12		43.10							[8]
BI/CS/7	Padopling Island, Baffin	5.07		43.90							[8]
10004	Padopling Island, Baffin	5.04	0.01	-	-	-	-	-	-	-	[6], [7]
10013	Padopling Island, Baffin	5.22	0.01	14.37	0.54	0.32	-	-	87.9	-	[6], [7]
10014	Padopling Island, Baffin	5.05	0.05	-	-	-	18.129	-	85.7	-	[6], [7]
10019	Padopling Island, Baffin	-	-	4.65	0.66	0.28	18.030	-	88.6	-	[6], [7]
10021	Padopling Island, Baffin	5.13	0.03	8.95	0.6	0.4	17.880	-	87.4	-	[6], [7]
10024	Padopling Island, Baffin	5.06	0.02	23.79	0.71	0.62	17.957	-	86.6	-	[6], [7]
10029	Padopling Island, Baffin	5.16	0.01	6.34	0.42	0.31	17.949	-	88.2	-	[6], [7]
10064	Padopling Island, Baffin	4.84	-	3.01	0.49	0.61	18.326	-	85.7	-	[6], [7]

Presented uncertainties for oxygen isotopes are two standard deviations of replicate measurements, otherwise uncertainties are $\pm 0.16\text{‰}$ (2 σ). Data sources: [1] = this study; [2] Hilton et al., 1999; [3] Macpherson et al., 2005; [4] Stuart et al., 2000; [5] Ellam and Stuart, 2000; [6] Kent et al., 2004; [7] Jackson et al., 2010; [8] Starkey et al., 2016.

Table S1: Olivine compositions (wt.%) for some NAIP rocks									
Sample	MgO	FeO	Al ₂ O ₃	CaO	SiO ₂	MnO	NiO	Total	Forsterite Content
NAL688 CC	49.1	10.18	0.10	0.36	40.5	0.17	0.40	100.8	89.6
	48.9	10.31	0.10	0.35	40.5	0.15	0.36	100.6	89.4
	49.0	10.86	0.08	0.37	40.3	0.16	0.32	101.0	88.9
	49.9	9.76	0.06	0.35	40.6	0.14	0.33	101.1	90.1
	49.6	9.68	0.08	0.33	41.0	0.13	0.33	101.1	90.1
NAL688 GR	50.9	8.14	0.13	0.31	40.9	0.12	0.40	101.0	91.8
	50.2	8.96	0.14	0.32	40.8	0.13	0.42	101.0	90.9
	49.6	9.34	0.06	0.33	40.7	0.14	0.31	100.5	90.4
NAL 626	40.1	22.19	0.04	0.08	38.5	0.38	0.06	101.3	76.3
NAL 625	47.5	12.76	0.05	0.30	40.7	0.17	0.31	101.8	86.9
	46.8	13.93	0.06	0.30	39.6	0.17	0.32	101.1	85.7
	47.5	12.99	0.09	0.26	40.2	0.18	0.35	101.5	86.7
	46.6	13.87	0.08	0.28	39.8	0.23	0.21	101.1	85.7
PJOR	47.6	12.81	0.08	0.28	40.0	0.19	0.31	101.3	86.9
	47.5	12.77	0.08	0.32	40.1	0.18	0.34	101.3	86.9
	47.0	12.72	0.08	0.30	39.9	0.17	0.32	100.5	86.8
	47.6	12.64	0.11	0.30	40.5	0.20	0.36	101.8	87.0
	46.3	14.00	0.07	0.27	40.2	0.17	0.26	101.3	85.5
NAL 611	47.0	13.72	0.08	0.29	39.8	0.18	0.25	101.3	85.9
	46.4	13.98	0.07	0.29	39.8	0.19	0.29	101.0	85.5
	46.4	14.59	0.06	0.27	39.9	0.24	0.24	101.7	85.0
	45.6	15.24	0.04	0.32	39.5	0.21	0.19	101.1	84.2
SK204	46.9	13.65	0.05	0.37	40.1	0.23	0.15	101.4	86.0
	46.8	13.71	0.07	0.33	39.9	0.21	0.17	101.2	85.9
	45.6	15.38	0.03	0.31	39.8	0.22	0.21	101.5	84.1
	46.1	14.98	0.08	0.22	39.9	0.24	0.36	102.0	84.6
EG2244	44.3	16.43	0.06	0.22	39.5	0.21	0.30	101.0	82.8
	45.4	14.71	0.09	0.25	39.9	0.22	0.26	100.8	84.6
	45.6	14.77	0.08	0.22	40.2	0.21	0.31	101.4	84.6
EG 2240 Rim ^a	41.8	11.75	0.00	0.19	46.7	0.00	0.30	100.7	88.0
EG 2240 Core ^b	41.7	8.25	0.00	0.25	48.9	0.00	0.38	99.5	91.0
^a From Brown et al., 1996.									

Table S2: Pyroxene compositions (wt.%) for NAL 626									
Sample	MgO	FeO	Al ₂ O ₃	CaO	SiO ₂	MnO	NiO	Total	Mg number
NAL626_02	17.2	5.52	2.68	20.9	51.8	0.15	0.028	98.3	84.7
NAL626_03	17.5	5.83	2.80	20.7	52.8	0.11	0.014	99.7	84.2

Table S3: Trace element abundance data (in ppm) for Icelandic and Baffin Island whole-rocks

Sample	SEL 97	NAL-625	NAL-688	NAL-688 gl	Pior	Trö-53	NAL-611*	Th-29 ^b	NAL 626 Pl	10013 ^c	10014 ^c	10019 ^c	10021 ^c	10024 ^c	10029 ^c	10064 ^c
Type	Whole-Rock	Whole-Rock	Whole-Rock	Whole-Rock	Whole-Rock	Whole-Rock	Whole-Rock	Whole-Rock	Whole-Rock	Matrix Glass	Matrix Glass	Matrix Glass	Matrix Glass	Matrix Glass	Matrix Glass	Matrix Glass
Location	Vestfirðir	Vadadla	Egget	Egget	Pjorsardalur	Trolladyngja	Kisufell	Thiðstreykkir	Fjalalsendi	Baffin Island	Baffin Island	Baffin Island	Baffin Island	Baffin Island	Baffin Island	Baffin Island
Lithology	Ankaramite	Picrite	Picro-basalt	Picro-basalt	Picro-basalt	Picro-basalt	Picro-basalt	Picrite	Picro-basalt	Picrite	Picrite	Picrite	Picrite	Picrite	Picrite	Picrite
MgO (wt.%)	18.74	-	-	9.73	-	11.13	10.45	15.73	-	22.11	11.97	21.94	17.91	14.97	21.82	14.4
ppm																
Sc	34.72	35.98	33.12	44.45	21.74	33.10	37.40	4.00	1.41	51.1	53	48.7	51.9	48.5	44.3	45.1
V	212.3	218.9	174.2	238.3	150.5	190.0	257.0	207.0	6.42	307	337	303	324	357	336	309
Cr	1608.9	1685.0	2297.7	993.8	2089.1	390.0	384.0	2176.0	9.80	365	354	361	359	364	392	339
Co	87.6	87.0	90.6	63.2	109.9	48.0	49.6	63.0	3.68	45.9	47.4	44	43.4	49.4	48.5	46.1
Ni	893.9	872.5	1174.7	459.3	1663.9	211.0	223.0	559.0	7.75	-	-	-	-	-	-	-
Cu	13.27	15.83	19.17	109.55	17.40	116.00	111.00	95.00	3.40	-	-	-	-	-	-	-
Ga	12.33	12.76	8.49	11.87	8.48	-	-	50.00	16.85	-	-	-	-	-	-	-
Rb	0.64	0.72	0.45	0.60	1.16	-	1.37	0.24	0.07	1.02	3.42	1.09	3.98	3.96	1.43	2.5
Sr	191.86	197.15	35.98	49.17	76.56	159.00	239.00	51.00	212.9	130	175	130	149	157	142	150
Y	15.29	15.72	10.49	15.18	11.40	15.00	18.10	14.10	0.37	24.9	28.6	23.7	25.8	23.7	20.7	22.3
Zr	67.55	69.65	14.03	20.15	46.10	46.00	47.80	19.10	0.40	62.9	91.7	60.5	72.3	65.6	58.5	65.5
Nb	8.07	8.35	0.64	0.83	4.41	-	3.00	0.47	0.16	2.07	7.59	2.09	6.38	6.7	2.51	4.51
Cs	0.01	0.01	0.00	0.01	0.04	-	0.05	-	0.00	-	-	-	-	-	-	-
Ba	21.31	21.32	4.96	6.56	10.22	29.00	50.00	3.67	4.82	15.1	66.8	14.5	62.1	79.3	17.7	41.8
La	6.79	6.97	0.56	0.73	3.27	-	2.69	0.54	0.23	2.74	6.69	2.53	5.36	5.2	2.74	4.15
Ce	15.58	15.98	1.63	2.28	8.29	-	7.26	1.94	0.47	7.83	15.5	7.28	12.3	13	8.59	10.8
Pr	2.22	2.28	0.28	0.39	1.30	-	1.08	0.36	0.07	1.46	2.46	1.32	1.87	1.86	1.43	1.68
Nd	10.34	10.66	1.82	2.60	6.32	-	5.82	2.21	0.31	7.38	12.1	7.16	9.2	9.11	7.33	8.42
Sm	2.69	2.74	0.75	1.07	1.76	-	2.01	1.00	0.08	2.53	3.52	2.57	2.93	3	2.48	2.72
Eu	0.97	0.99	0.31	0.46	0.61	-	0.81	0.45	0.16	1	1.29	1	1.12	1.17	1	1.06
Gd	3.11	3.18	1.23	1.81	2.08	-	2.60	1.72	0.07	3.42	4.33	3.4	3.65	3.34	2.87	3.3
Tb	0.49	0.51	0.24	0.35	0.34	-	0.45	0.32	0.01	-	-	-	-	-	-	-
Dy	2.82	2.86	1.61	2.36	2.00	-	3.06	2.20	0.06	4.27	4.98	4.17	4.59	4.32	3.6	3.99
Ho	0.56	0.56	0.37	0.54	0.41	-	0.71	0.48	0.01	-	-	-	-	-	-	-
Er	1.39	1.42	1.05	1.54	1.07	-	1.75	1.41	0.03	2.6	3.08	2.58	2.81	2.5	2.15	2.35
Tm	0.21	0.22	0.18	0.27	0.17	-	0.29	0.22	0.01	-	-	-	-	-	-	-
Yb	1.23	1.25	1.11	1.61	1.00	-	1.86	1.45	0.03	2.36	2.77	2.41	2.45	2.4	2.15	2.26
Lu	0.19	0.19	0.18	0.27	0.16	-	0.32	0.23	0.004	-	-	-	-	-	-	-
Hf	1.85	1.94	0.68	0.65	1.39	-	-	0.62	0.01	1.61	2.32	1.6	1.82	1.67	1.5	1.72
Ta	0.54	0.55	0.10	0.06	0.33	-	0.22	0.04	0.03	0.15	0.47	0.13	0.38	0.37	0.15	0.27
Pb	0.43	1.12	0.75	0.16	0.90	-	-	-	<0.01	0.38	0.81	0.45	0.63	1.08	0.58	0.65
Th	0.51	0.52	0.06	0.07	0.30	-	0.13	0.02	0.004	0.19	0.59	0.16	0.67	0.46	0.14	0.35
U	0.09	0.09	0.02	0.02	0.11	-	0.02	0.01	0.001	0.04	0.14	0.03	0.13	0.11	0.04	0.09

Data for: a - NAL 611 from Breddam et al. (2000); b - Th-29 from MacLennan et al. (2001); c - Baffin Island samples from Kent et al. (2004)

Table S4: Whole-rock Sr-Nd-Pb isotope data for NAIP rocks									
Sample	Location	$^{87}\text{Sr}/^{86}\text{Sr}$	$^{143}\text{Nd}/^{144}\text{Nd}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$			
		$\pm 2\sigma$	$\pm 2\sigma$	$\pm 2\sigma$	$\pm 2\sigma$	$\pm 2\sigma$			
Icelandic Lavas									
NAL 625	Central Iceland Rift Zone	0.703463	0.512986						
British Palaeogene Igneous Province									
SK 266	SMLS, Isle of Skye			17.226	0.010	15.321	0.012	37.14	0.02
BB1	SMLS, Isle of Skye	0.704481	0.000017	0.512877	0.000006	18.043	0.007	15.476	0.005
LL1	SMLS, Isle of Skye			16.286	0.008	15.079	0.008	36.05	0.01
SK291	SMLS, Isle of Skye			16.839	0.010	15.200	0.010	36.85	0.03
INV1	LMSC, Isle of Skye	0.704563	0.000018	0.512564	0.000006	16.787	0.007	15.209	0.006
BH1	LMSC, Isle of Skye	0.704844	0.000018	0.512874	0.000006	17.558	0.013	15.399	0.014
SR81	LMSC, Isle of Skye	0.704093	0.000021	0.51289	0.000014	17.678	0.005	15.397	0.004
SK056	SDS, Isle of Skye			17.534	0.015	15.361	0.019	37.04	0.03
SK057	SDS, Isle of Skye	0.703487	0.000017	0.512777	0.000009	17.378	0.005	15.355	0.005
SK204	SDS, Isle of Skye			16.806	0.006	15.244	0.006	37.27	0.02
SK208	Cuillin Complex, Isle of Skye	0.703599	0.000018	17.377	0.006	15.452	0.005	37.53	0.01
East Greenland									
EG2240	Prins af Wales, E. Greenland	0.70403	0.51280	17.551	0.008	15.149	0.007	37.31	0.02
EG2244	Prins af Wales, E. Greenland			14.733	0.004	14.620	0.005	35.74	0.01
EG3359	Prins af Wales, E. Greenland			18.328	0.006	15.352	0.005	38.41	0.02
West Greenland									
332828	Ordlingassooq, W. Greenland	0.702975	0.513113	17.938		15.404		37.62	
332788	Ordlingassooq, W. Greenland	0.703478	0.512974	18.105		15.482		38.03	
362149	Nuajanguit, W. Greenland	0.703027	0.513114	17.972		15.410		37.76	
362275	Nuajanguit, W. Greenland	0.703147		17.740		15.399		37.57	
400457	Anaanaa, W. Greenland	0.703077	0.512125	17.813		15.393		37.51	
Baffin Island									
10013	Padopling Island, Baffin	0.703005	0.512999						
10014	Padopling Island, Baffin	0.703306	0.51289	18.129		15.451		38.09	
10019	Padopling Island, Baffin	0.70306	0.512994	18.030		15.465		37.87	
10021	Padopling Island, Baffin	0.703813	0.512825	17.880		15.395		37.89	
10024	Padopling Island, Baffin	0.70322	0.512905	17.957		15.379		37.88	
10029	Padopling Island, Baffin	0.70299	0.512989	17.949		15.417		37.74	
10064	Padopling Island, Baffin	0.703604	0.512906	18.326		15.468		38.22	
Data from: BPIP = Stuart et al., 2000; Ellam & Stuart, 2000; East Greenland = Brown et al, 1996; Ellam & Stuart, 2000; West Greenland = Graham et al. 1998; Baffin Island = Kent et al., 2004; Jackson et al., 2010									

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