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The Sub-lithospheric Source of North Atlantic Basalts: Evidence for, and Significance of, a Common End-member

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Palaeogene basalts from the margins of the North Atlantic often show geochemical variations that are consistent with their parental magmas having interacted with the lithosphere en route to the Earth's surface. These geochemical trends vary depending on the nature of the local lithospheric contaminants. Using examples from the British Tertiary Igneous Province and SE Greenland, we construct coherent contamination trends, which converge on a restricted Pb isotope composition, apparently indicating a common uncontaminated asthenospheric mantle component. Significantly, this composition is also suitable as one end-member of the Pb isotope arrays recorded in Recent Icelandic basalts. We conclude that this composition has been a persistent component of the Iceland plume over 60 my, dominating the mantle contribution to the Palaeocene phase of flood basalt magmatism but constituting only one end-member on Iceland. The Pb isotope composition of this 'North Atlantic end-member' is consistent with, but not necessarily demanding of, a primordial source. Recent evidence suggesting a lower-mantle origin for mantle plumes encourages investigation of whether the geochemical evidence supports that hypothesis. Helium isotope data from Palaeogene North Atlantic basalts support a lower-mantle contribution. However, mixing models suggest that it is unlikely that the lower-mantle contribution is large enough to dominate the Sr-Nd-Pb isotope compositions and lithophile trace element signatures of any plumederived basalts.

KEY WORDS: North Atlantic; Iceland; lower mantle; mantle plumes; flood basalts; isotopes

INTRODUCTION

Continental flood basalts (CFB) have ⁸⁷Sr/⁸⁶Sr, ¹⁴³Nd/ ¹⁴⁴Nd and Pb isotope ratios that are considerably more variable than those of mid-ocean ridge basalts (MORB) and ocean island basalts (OIB). Although the convecting mantle beneath continents is likely to be as heterogeneous as that beneath oceans, there is no obvious reason why it would be any more variable. The obvious corollary is that the additional isotopic heterogeneity is derived from the continent itself; either from the continental crust or the lithospheric mantle keel.

Characterization of sub-lithospheric mantle has therefore tended to concentrate on oceanic basalts, thereby minimizing the possibility of additional complication continental lithospheric contamination although the possibility remains that some of the OIB Sr-Nd-Pb isotope variation reflects a contribution from remnants of continental lithosphere in the upper mantle (e.g. Hawkesworth et al., 1986; Kamber & Collerson, 1999). However, there may be insights into the deep mantle that are available only by studying the voluminous continental flood basalt provinces. To exploit such information requires us to understand, quantify and ultimately strip away the effects of high-level differentiation and lithospheric interaction leaving a 'window' into sublithospheric sources and processes.

As more CFB provinces have been subjected to petrological and geochemical studies, distinguishing crustal contamination from 'remobilization' of enriched lithospheric mantle material has become a major preoccupation. Much effort has been expended in using the increasingly readily available isotope and trace element tracers to characterize the high 87Sr/86Sr, low 143Nd/ ¹⁴⁴Nd 'enriched' end-members and assess the relative likelihood that they are derived from crustal or enriched sub-continental mantle sources. Consensus has been slow to develop and the crustal contamination vs enriched mantle debate was often polemic. However, persuasive examples of both crustal and lithospheric contaminants are now documented and there can be little doubt that both crustal contamination and entrainment of enriched lithospheric mantle are valid processes in continental basalt petrogenesis (e.g. Carlson et al., 1984; Hawkesworth et al., 1984; Menzies et al., 1984; Devey & Cox, 1987; Ellam & Cox, 1991; Ellam et al., 1992).

Less tractable perhaps has been the sub-lithospheric contribution to CFB. Very few CFB have radiogenic isotope signatures that overlap the fields for MORB and OIB. This may merely indicate the rarity of CFB that have not interacted with continental lithosphere (e.g. Thompson et al., 1983). Alternatively, it may suggest that asthenospheric sources have not contributed to these CFB, with the implication that wholesale melting of lithospheric mantle has produced many CFB (e.g. Turner et al., 1996). One notable exception is the North Atlantic Igneous Province (NAIP) in which Palaeogene rift-related continental magmatism around the North Atlantic margin has given way to oceanic hotspot activity at Iceland today. Comparison of the Sr, Nd and Pb isotope compositions of the Palaeogene CFB and the Iceland OIB indicate that many of the former interacted strongly with the rifting continental lithosphere (Moorbath & Thompson, 1980; Dickin, 1981; Gariépy et al., 1983; Thirlwall & Jones, 1983; Thirlwall et al., 1994; Wallace et al., 1994; Kerr et al., 1995; Fram & Lesher, 1997; Lightfoot et al., 1997; Fitton et al., 1998; Hansen & Nielsen, 1999). Nevertheless, several of the Tertiary lavas have relatively low ⁸⁷Sr/⁸⁶Sr and high ¹⁴³Nd/¹⁴⁴Nd, which approach Icelandic compositions and suggest that largely uncontaminated basalts are present, which may allow us to identify a sub-lithospheric contribution and comment on its origins.

In this contribution we present new Pb isotope data for NAIP basalts from Northern Ireland, which we refer to as the Antrim basalts (Fig. 1) following common local terminology, although minor outcrops of Palaeogene basalts are found outside County Antrim. The petrography, major and trace element compositions and Sr-Nd isotope geochemistry of these samples have been described previously (Wallace et al., 1994; Wallace, 1995). A similar dataset sampling many of the same outcrops and sections has also been presented by Barrat & Nesbitt (1996). In addition, we present new Sr and Nd isotope data for lavas and intrusives from the Isle of Skye, NW Scotland (Fig. 1), which were previously analysed for elemental concentrations and He and Pb isotope ratios (Stuart et al., 2000). The Skye samples were chosen for their high proportion of fresh olivine phenocrysts to facilitate He isotope measurements. Sr and Nd isotope measurements were made on whole-rock powders prepared before olivine separation but, in view of the high

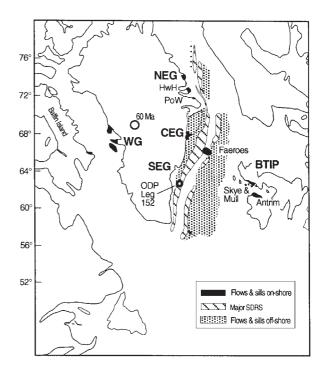


Fig. 1. Locality map with palaeogeography reconstructed at c. 55 Ma (after Saunders et al., 1997). Bold text labels define the various subprovinces identified within the North Atlantic Igneous Province: NEG, North East Greenland; CEG, Central East Greenland; SEG, South East Greenland; WG, West Greenland; BTIP, British Tertiary Igneous Province. Plain text labels identify specific localities within the subprovinces: HwH, Hold with Hope; PoW, Prince of Wales Mountains. Circle in central Greenland is the proposed position of the Iceland plume at 60 Ma (Lawver & Müller, 1994). SDRS, seaward-dipping reflector sequence.

olivine contents, it is possible that the whole rocks do not represent liquid compositions. New He and Pb isotope determinations are presented for picrites from the Prince of Wales Mountains in NE Greenland (Fig. 1). Petrography, major and trace element compositions and Sr-Nd isotope geochemistry of the Prince of Wales Formation were reported by Brown et al. (1996) but, again, the samples selected for He isotope measurements were chosen on the basis of their olivine content rather than the availability of other geochemical data.

The new data are combined with the literature database, to investigate the extent to which we can indeed quantify and strip away relatively high-level processes and thereby reveal the sub-lithospheric involvement in Tertiary North Atlantic magmatism, and compare it with the intra-oceanic magmatism at Iceland today. More specifically, we exploit differences in the intra-lithospheric evolution of various basalts from the NAIP to 'see through' to uncontaminated isotopic compositions. The results suggest remarkably similar primary magmas that appear to implicate a common sub-lithospheric component in the petrogenesis of North Atlantic basalts from 60 Ma to the present day. We discuss the nature and likely origin of this common end-member. Above all, in the North Atlantic at least, we demonstrate that the pre-break-up CFB carry important information about their deep mantle origins, which are complementary to those available from Iceland OIB and critical to unravelling the full temporal and spatial history of plume-related magmatism.

NORTH ATLANTIC IGNEOUS PROVINCE: ISOTOPE GEOCHEMISTRY

A thorough and up-to-date review of North Atlantic magmatism has been presented by Saunders *et al.* (1997). We follow their precedent by dividing the region into a series of sub-provinces (Fig. 1) for the purposes of data presentation and comparison. Our first-order observations are drawn from the British Tertiary Igneous Province (BTIP) and SE Greenland sub-provinces (Fig. 1).

Antrim basalts

The Antrim basalts are the most areally extensive remnant of the BTIP. The lava pile has been subdivided into Upper and Lower Basalt Formations and the Interbasaltic Formation, which represents a period of relative volcanic quiescence during which *in situ* laterites were formed (Old, 1975). The relatively minor Interbasaltic Formation magmatism includes eruption of the compositionally distinct Causeway Tholeiite Member. The magmatism is mostly tholeiitic, although occasional nepheline-normative flows occur (Lyle, 1980), and mostly mafic, but with minor differentiated tholeiitic andesites and occasional rhyolites (Wallace *et al.*, 1994).

Isotope data for the Tertiary lavas of Northern Ireland are a compilation of the data of Wallace et al. (1994), Wallace (1995) and new Pb isotope determinations (Table 1). As demonstrated previously (Wallace et al., 1994; Barrat & Nesbitt, 1996) there are correlations between isotopic composition and major and trace element differentiation indices in the Antrim basalts. For example (Fig. 2a), the two Antrim lava series that show substantial MgO variation in our database (Lower Basalt Formation and Causeway Tholeiite Member) appear to evolve increasing initial Sr isotope composition (87Sr/86Sr_i) with decreasing MgO; although the trends are not collinear and that for the Causeway tholeiites is most striking. Significantly, it is the more differentiated samples of each series that show elevated ⁸⁷Sr/⁸⁶Sr_i isotope composition. If we make the (reasonable) assumption that high 87Sr/ ⁸⁶Sr_i is a reflection of the incorporation of lithospheric material (either crust or mantle), then that incorporation seems to have progressed as the magmas differentiated and is thereby analogous to the assimilation—fractional crystallization (AFC) contamination mechanism. In the case of Antrim we do not claim to have necessarily discriminated crust from lithospheric mantle contaminants with any certainty. However, our purpose here is to 'see through' both crustal and lithospheric mantle contamination to sub-lithospheric mantle source compositions. Thus, when we are confident that contamination effects occurred in the continental crust we will refer to them as 'crustal contamination'. Elsewhere we will refer to a 'lithospheric' contribution without any implication that the contaminant was either crust or mantle lithosphere.

Pb–Sr isotope co-variations (Fig. 2b) are fairly coherent in the Antrim samples, with both the Upper and Lower Basalt Formations defining a positive ⁸⁷Sr/⁸⁶Sr-²⁰⁶Pb/²⁰⁴Pb trend, which implies that the contaminant inferred from the radiogenic Sr had elevated ²⁰⁶Pb/²⁰⁴Pb. The four Causeway Tholeiite Member samples lie off the Upper–Lower basalt trend, but also show radiogenic Pb associated with high ⁸⁷Sr/⁸⁶Sr. Thus, in Pb–Pb isotope space (Fig. 3a and e), where the Antrim lavas define linear arrays, it is the radiogenic Pb that is associated with high ⁸⁷Sr/⁸⁶Sr_i (and, indeed, low ¹⁴³Nd/¹⁴⁴Nd_i) and apparently most affected by lithospheric contamination. By contrast, the less contaminated samples are characterized by relatively unradiogenic Pb.

Comparisons with other NAIP lavas

The Antrim Pb isotope data are in marked contrast to Pb isotope data for other BTIP occurrences. For example, Pb isotope data from Skye (Dickin et al., 1981) and Mull (Kerr et al., 1995) define spectacular Pb-Pb arrays (Fig. 3a and e), which have been attributed to contamination of mantle-derived magmas by Archaean granulite facies (Lewisian) continental crust; the extremely unradiogenic Pb of the contaminant reflecting ancient U depletion thought to occur during granulite facies metamorphism. Similar data arrays (Fig. 3a and e), albeit slightly less coherent, were observed for basalts from the Ocean Drilling Program (ODP) Leg 152 cores (Fig. 1) drilled into the basalt flows that constitute the seaward-dipping seismic reflectors of the SE Greenland margin at 63°N (Fitton et al., 1998). Again, the trends towards unradiogenic Pb were attributed to contamination with ancient U-depleted granulite facies crust.

Thus, in the Skye, Mull and ODP Leg 152 cases, crustal contamination is thought to result in unradiogenic Pb and the least contaminated samples are those with highest ²⁰⁶Pb/²⁰⁴Pb. By contrast, in Antrim, the lithospheric contaminant (identified on the basis of elevated

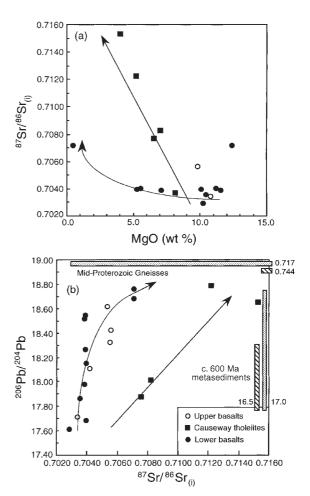
Table 1: Pb isotope compositions of lavas from Antrim, Northern Ireland

Sample	²⁰⁶ Pb/ ²⁰⁴ Pb ₍₀₎	²⁰⁷ Pb/ ²⁰⁴ Pb ₍₀₎	²⁰⁸ Pb/ ²⁰⁴ Pb ₍₀₎				
Lower Basalt Formation							
PL43	17.607 ± 10	15.441 ± 11	37.522 ± 21				
JP117	18·149 <u>+</u> 8	15.531 ± 9	$38 \cdot 165 \pm 35$				
PL55	18.676 ± 7	15.622 ± 7	$38 {\cdot} 744 \pm 18$				
GBY-2	17.971 ± 10	15.450 ± 8	$37{\cdot}690\pm21$				
GBY-4	$18{\cdot}260\pm7$	$15{\cdot}589\pm 6$	$38{\cdot}153\pm18$				
GBY-5	$18{\cdot}511\pm10$	$15{\cdot}589\pm10$	$38 \cdot 208 \pm 24$				
DNL-1	17.858 ± 7	15.618 ± 8	37.818 ± 22				
JP-2	17.677 ± 7	15.428 ± 5	$37\!\cdot\!472\pm13$				
GB-1	18⋅538 ± 9	15.584 ± 7	38.093 ± 21				
GBY-1	$18 {\cdot} 755 \pm 7$	$15{\cdot}575\pm 6$	$38{\cdot}448\pm17$				
Upper Basalt Formation							
PL42	$\textbf{18.421} \pm \textbf{11}$	$\textbf{15.612} \pm \textbf{11}$	38.518 ± 30				
BVAO	$17{\cdot}709\pm7$	$\textbf{15.511} \pm \textbf{6}$	$37{\cdot}590\pm18$				
AGH-4	$\textbf{18.613} \pm \textbf{13}$	$\textbf{15.576} \pm \textbf{14}$	$38{\cdot}477\pm28$				
PB-1	18.318 ± 7	$15{\cdot}554\pm7$	$36\text{-}898 \pm 19$				
BVB-1	$18{\cdot}106 \pm 10$	$15{\cdot}475\pm9$	$37{\cdot}914\pm23$				
Causeway Tholeiite Member							
PL1	$\textbf{18.649} \pm \textbf{10}$	$15{\cdot}603\pm10$	$38 \cdot 444 \pm 25$				
JP49	$\textbf{18.012} \pm \textbf{4}$	15.494 ± 4	$37{\cdot}981\pm11$				
CHQ-5	18.786 ± 6	$15{\cdot}607\pm8$	38.667 ± 18				
CHQ-1	17.870 ± 5	$15{\cdot}411\pm5$	$37{\cdot}639\pm17$				

Pb was separated from whole-rock powders by anion exchange using HBr and HCl elutions. Total procedure blanks averaged 350 pg and were always below 600 pg. Isotope ratios were measured on a VG54E single-collector mass spectrometer. Instrumental mass fractionation of 1.5% per a.m.u. was corrected by repeated analyses of the NBS981 standard. Errors are 2 SEM but precision is limited by the reproducibility of NBS981 of 0.2%.

⁸⁷Sr/⁸⁶Sr_i>0·707 for Lower basalts, >0·715 for Causeway basalts) appears to have had relatively high ²⁰⁶Pb/²⁰⁴Pb and it is the less radiogenic samples that approach most closely uncontaminated compositions. This difference is not unexpected, as it corresponds to major changes in crustal structure. Archaean (Lewisian) crust is a potential contaminant beneath Skye and Mull, and the occurrence of similar ancient granulite is likely beneath East Greenland. By contrast, there is unlikely to be Archaean crust beneath Northern Ireland.

The most likely crustal contaminants beneath Antrim are Neo-Proterozoic—Lower Palaeozoic (Dalradian) metasediments or mid-Proterozoic gneisses. Both are lithologically diverse and certainly heterogeneous in Sr and Pb isotope compositions (Fig. 2b; Dickin *et al.*, 1981;



 ${\bf Fig.~2.~(a)}$ $^{87}{\rm Sr}/^{86}{\rm Sr-MgO}$ relationships for the Palaeogene lavas of Northern Ireland. Evolved (low-MgO) lavas within the different lava series (Wallace et al., 1994) tend to have the highest 87Sr/86Sr, suggesting that they have incorporated radiogenic Sr and Pb during magmatic fractionation, which probably occurred within the continental crust, or perhaps, lithospheric mantle. (b) 206Pb/204Pb-87Sr/86Sr variations in the Antrim lavas, which indicate that elevated 87Sr/86Sr is associated with relatively radiogenic Pb (i.e. high 206Pb/204Pb). Uncontaminated asthenospheric mantle compositions are therefore likely to lie on extrapolations of the inferred contamination trends towards low 87Sr/ ⁶Sr and low ²⁰⁶Pb/²⁰⁴Pb. Likely potential crustal contaminants beneath Antrim are mid-Proterozoic gneisses (stippled boxes) and Neo-Proterozoic-Lower Palaeozoic (Dalradian) metasediments (cross-hatched boxes), which have variable Sr-Pb isotope compositions. Figures indicate the extreme isotope compositions for these potential contaminants when they fall outside the range of the plot axes.

Marcantonio *et al.*, 1988). However, the extremely low ²⁰⁶Pb/²⁰⁴Pb Archaean granulite signature is not anticipated beneath Antrim and it is entirely reasonable that a suitable mixture of lithologies exists beneath NE Ireland whose Pb isotope characteristics would result in contamination trends towards a relatively high ²⁰⁶Pb/ ²⁰⁴Pb lithospheric end-member.

Significantly, the Skye, Mull and ODP Leg $152\,\mathrm{samples}$ with most radiogenic Pb have very similar Pb isotope

compositions to the lowest ²⁰⁶Pb/²⁰⁴Pb Antrim samples. Thus, as we back-track along trends for these three datasets (Fig. 3a and e), which are thought to be mixing arrays between mantle-derived magmas and lithospheric contaminants, we arrive from diametrically opposed directions at relatively uncontaminated compositions that occupy a restricted region of Pb isotope space with ²⁰⁶Pb/²⁰⁴Pb ~ 17·5, ²⁰⁷Pb/²⁰⁴Pb ~ 15·4 and ²⁰⁸Pb/²⁰⁴Pb ~ 37·4

Thus, in the BTIP and SE Greenland sub-provinces where the basalts appear to be dominated by contamination of mantle-derived magmas by lithospheric materials it is possible to use the well-defined mixing trends to extrapolate towards uncontaminated compositions. Because the Skye–Mull–ODP Leg 152 and Antrim trends are towards very distinctive crustal contaminants, their intersection can be used to define a restricted range of uncontaminated composition, which appears to demand a common mantle end-member. For purely descriptive purposes we will refer to this putative end-member as NAEM (North Atlantic End-Member) to avoid any confusion with other postulated isotopic components (e.g. Zindler & Hart, 1986).

Elsewhere, the situation is less clear cut. Palaeogene basalts from West Greenland (Lightfoot et al., 1997; Graham et al., 1998) lack clear Pb–Pb isotope correlations (Fig. 3b and f) but the Pb isotope variation observed is not inconsistent with a source contribution from the NAEM. Lavas from NE Greenland (Thirlwall et al., 1994; Ellam et al., 1998), Central East Greenland (Hansen & Nielsen, 1999) and the Faeroes (Gariépy et al., 1983) do not demand the involvement of a NAEM (Fig. 3c and g) but either lie on linear trends that pass through the proposed NAEM composition or define more diffuse fields, which clearly demand more complex petrogenetic schemes than simple binary source mixtures (e.g. Thirlwall et al., 1994; Hansen & Nielsen, 1999).

Perhaps the most compelling evidence that the NAEM is indeed representative of a single component available throughout the NAIP comes from comparison of the Palaeogene flood basalts with Recent basalts from Iceland (Fig. 3). Iceland basalts define coherent Pb-Pb arrays (Fig. 3d and h), which, extrapolated to unradiogenic Pb isotope compositions, closely approach the NAEM composition and certainly intersect the area of overlap in Pb isotope space between the Skye, Mull, ODP Leg 152 and Antrim datasets. Thus, it seems plausible that the Iceland Pb-Pb arrays represent mixing between the NAEM composition and a more radiogenic Pb isotope composition with ${}^{206}\text{Pb}/{}^{204}\text{Pb} \ge 19.4, {}^{207}\text{Pb}/{}^{204}\text{Pb} \ge$ 15.56 and $^{208}\text{Pb}/^{204}\text{Pb} \ge 39.0$. If so, we are led to conclude that NAEM has made a pervasive contribution to NAIP magmatism over the past 60 my.

Interestingly, the Pb isotope compositions of Palaeogene BTIP that have not undergone lithospheric contamination (i.e. those that define the NAEM) are much more restricted than the present-day range for Icelandic basalts (Fig. 3). If the Iceland Pb—Pb arrays (Fig. 3d and h) document mixing between NAEM and some other end-member with relatively radiogenic Pb isotope ratios, then the latter appears not to have been as influential in generation of the Palaeogene CFB as it is in Icelandic petrogenesis today.

Pb–Sr and Pb–Nd co-variations (Fig. 4a and b) are not as coherent as those in Pb–Pb space, presumably indicating a substantial degree of Sr and Nd isotope heterogeneity in the various lithospheric contaminants that is not recorded by Pb isotopes. None the less, both diagrams show a fairly restricted area in which the Iceland, Skye, Mull, Antrim and ODP Leg 152 data overlap. This overlap provides a very rough constraint on the NAEM, suggesting $^{87}{\rm Sr}/^{86}{\rm Sr}\sim 0.703$, $\epsilon_{\rm Nd}\sim +9$ [it should be noted that the Skye field in Fig. 4c is much reduced because Nd isotope data are not available for the samples studied for Sr and Pb isotopes by Moorbath & Thompson (1980) and Dickin *et al.* (1981) and the field therefore contains only the few $^{143}{\rm Nd}/^{144}{\rm Nd}$ presented in Table 2 of this paper].

CHARACTERISTICS OF THE NAEM

The identification of a single Pb isotope end-member throughout the NAIP is in itself significant but there are possibly additional implications in the particular Pb isotope composition identified for the NAEM. Most strikingly, the proposed end-member composition (i.e. $^{206}\text{Pb}/^{204}\text{Pb} \sim 17.5, \,^{207}\text{Pb}/^{204}\text{Pb} \sim 15.4 \text{ and } ^{208}\text{Pb}/^{204}\text{Pb}$ ~ 37.4) in $^{207}\text{Pb}/^{204}\text{Pb}-^{206}\text{Pb}/^{204}\text{Pb}$ space (Fig. 3) falls close to the intersection of the 4.55 Ga geochron with the northern hemisphere reference line (Hart, 1984). The geochron is the locus of ²⁰⁷Pb/²⁰⁴Pb-²⁰⁶Pb/²⁰⁴Pb compositions that are generated for different ²³⁸U/²⁰⁴Pb (μ) over 4.55 Gy through single-stage (i.e. constant μ) Pb isotope evolution from an initial Pb isotope composition identical to that recorded by troilite (FeS) from the Canyon Diablo iron meteorite. Assuming that Canyon Diablo troilite represents initial Bulk Earth Pb, then the present-day Bulk Earth should lie on the geochron. However, most mantle and crustal rocks fall to the right of the geochron in ²⁰⁷Pb/²⁰⁴Pb-²⁰⁶Pb/²⁰⁴Pb space and the failure to identify a means by which such compositions could average to a position on the geochron is at the heart of the so-called Pb paradox (e.g. Zindler & Hart, 1986).

Identification of an NAEM Pb-isotope composition that lies on the 4.55 Ga geochron is therefore intriguing because such a composition could represent a primordial source that has remained intact since formation of the Earth. Speculation intensifies when the exact location of

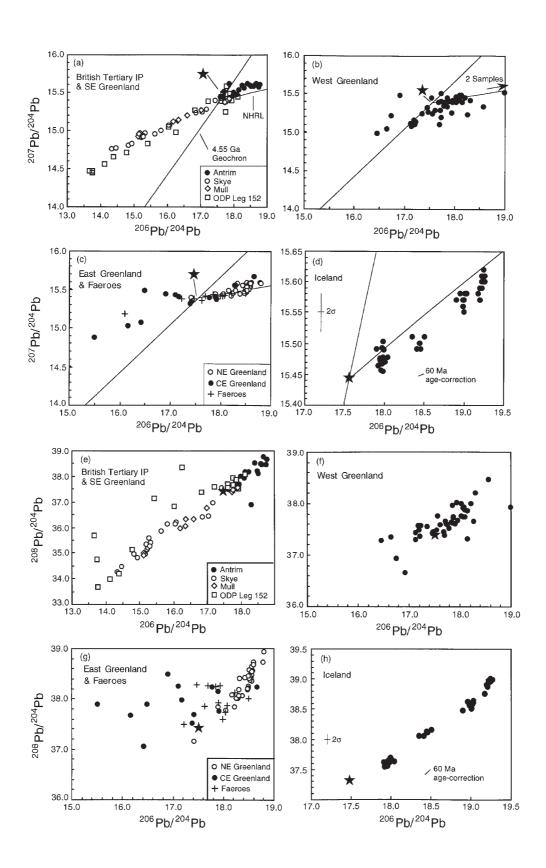


Table 2: Sr and Nd isotope compositions of lavas from Skye

Sample	⁸⁷ Sr/ ⁸⁶ Sr ₍₀₎	¹⁴³ Nd/ ¹⁴⁴ Nd ₍₀₎
SK208	0·703 599 <u>±</u> 18	_
BB1	0.704481 ± 17	0.512877 ± 6
BH1	0.704844 ± 18	0.512874 ± 6
INV1	0.704563 ± 18	0.512564 ± 6
SR81	0.704093 ± 21	0.512890 ± 14
SK057	$0.703\ 487\pm17$	0.512777 ± 9

Sr and Nd were separated using conventional ion exchange techniques with average total procedure blanks of 350 pg and 300 pg, respectively. Analyses were made using a VG Sector 54-30 multiple-collector mass spectrometer in dynamic mode. Ratios were normalized to $^{86}\text{Sr}/^{68}\text{Sr}=0.1194$ and $^{146}\text{Nd}/^{144}\text{Nd}=0.7219.$ NBS987 gave $^{87}\text{Sr}/^{68}\text{Sr}=0.710$ 243 \pm 20 (2 SD) and the laboratory Nd 'standard' (JM) gave 0.511 500 \pm 10 (2 SD). Errors for individual samples are 2 SE.

the proposed end-member on the geochron is considered because values of $^{206}\text{Pb}/^{204}\text{Pb}=17\cdot50$ and $^{207}\text{Pb}/^{204}\text{Pb}=15\cdot36$ correspond to a single-stage evolution of Pb with $\mu=7\cdot99,$ to all intents identical to the Bulk Earth μ value of $8\cdot0$ (White, 1993). Furthermore, fixing $\mu=7\cdot99$ yields a $4\cdot55$ Ga single-stage $^{208}\text{Pb}/^{204}\text{Pb}=37\cdot4$ for a $^{232}\text{Th}/^{238}\text{U}$ (κ) = $3\cdot93,$ which falls tantalizingly within the range of proposed Bulk Earth κ values of $3\cdot8-4\cdot2$ (Galer & O'Nions, 1985; Allègre *et al.*, 1986).

Thus, the Pb isotope composition of the proposed NAEM is consistent with a primordial component although we would caution from the outset that such a component is by no means demanded by this observation. None the less, given the recent seismic evidence (Helmberger *et al.*, 1998; Shen *et al.*, 1998; Bijwaard & Spakman, 1999) that the Iceland plume originates within the lower mantle, coupled with the implications of two-layer mantle convection models (i.e. that the lower mantle could preserve a primordial composition), it is worth examining further the extent to which the proposed NAEM might represent a lower-mantle contribution.

The first evidence against a lower-mantle origin for the NAEM must be that the potentially primordial Pb isotope signature is not accompanied by primordial Sr or Nd. As noted above, the NAEM appears to be characterized by relatively low $^{87}Sr/^{86}Sr$ and high $\epsilon_{Nd}.$ More specifically, the least contaminated basalts from Antrim have ⁸⁷Sr/⁸⁶Sr_i <0.703 (Wallace et al., 1994) and ¹⁴³Nd/¹⁴⁴Nd_i >0.5130 (Wallace, 1995), which correspond more closely to MORB values, thought to represent depleted upper mantle, than to the primordial (or Bulk Earth) values $(^{87}Sr/^{86}Sr = 0.7045-0.7055, ^{143}Nd/$ 144 Nd = 0.512638) expected for lower mantle. Similarly, relatively uncontaminated basalts from Skye (Thirlwall & Jones, 1983; Table 2), Mull (Kerr et al., 1995) and ODP Leg 152 (Fitton et al., 1998) all have high 143Nd/ ¹⁴⁴Nd and low ⁸⁷Sr/⁸⁶Sr compared with Bulk Earth values. Furthermore, the present-day Iceland hotspot magmatism is also characterized by high 143Nd/144Nd and low 87Sr/86Sr compared with Bulk Earth values. Thus, in all these examples, the apparent tendency towards primordial Pb is not accompanied by a well-developed Sr-Nd isotopic signature that might be considered primordial.

HELIUM ISOTOPE EVIDENCE

Perhaps the least equivocal evidence for a primordial contribution to hotspot magmatism is the presence of high ³He/⁴He ratios in some OIB. Some hot-spots are characterized by He isotope compositions that are much higher than the uniform value of 8 \pm 1 R_a (where R_a represents the ³He/⁴He of a sample normalized to the atmospheric ³He/⁴He, 1·39 × 10⁻⁶) that characterizes MORB. Loihi seamount, thought to mark the present axis of the Hawaii plume, and Iceland are the best examples of high- 3 He hot-spots, having 3 He/ 4 He >20 $R_{\rm a}$ (Allègre et al., 1983, 1987; Kurz et al., 1983; Rison & Craig, 1983; Valbracht et al., 1997; Hilton et al., 1999). High ³He/⁴He values are indicative of a mantle source that through time has maintained a high ³He/(Th + U) and is therefore relatively undegassed compared with the MORB source, which is widely accepted as the relatively depleted and degassed upper mantle. A popular

Fig. 3. ²⁰⁷Pb/²⁰⁴Pb=²⁰⁶Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb=²⁰⁶Pb/²⁰⁴Pb plots for lavas from (a, e) British Tertiary and SE Greenland; (b, f) West Greenland; (c, g) East Greenland and Faeroe Islands; (d, h) Iceland. ²⁰⁷Pb/²⁰⁴Pb=²⁰⁶Pb/²⁰⁴Pb plots (a–d) also illustrate a 4·55 Ga geochron and the northern hemisphere reference line (NHRL) of Hart (1984). Star symbol indicates the position of the proposed North Atlantic end-member (NAEM). Error bars (d, h) are 2σ based on reproducibility of the NBS981 standard at SUERC. All Pb isotope data are present day; no attempt has been made to age-correct Palaeogene lavas to initial (eruption age) values because (1) U and Pb concentrations are not always available, (2) U/Pb ratios may be unreliable in altered samples, and (3) present-day data facilitate comparison between Tertiary lavas and Recent Iceland basalts assuming no significant (U,Th)–Pb fractionation in the petrogenesis of the former. However, an approximate 60 my age-correction based on ²³⁸U/²⁰⁴Pb = 8 and ²³²Th/²³⁸U = 4 is illustrated and shown to be small in comparison with the observed Pb isotope variations (d, h). Data sources are given in main text.

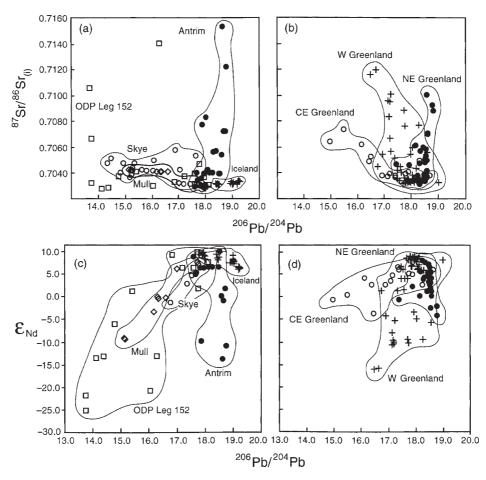


Fig. 4. (a, b) $^{206}\text{Pb}/^{204}\text{Pb}=^{87}\text{Sr}/^{86}\text{Sr}_i$ and (b) $^{206}\text{Pb}/^{204}\text{Pb}=\epsilon_{Nd}$ plots for North Atlantic Igneous Province lavas. Trends for the separate sub-provinces converge to define a relatively restricted area of overlap from which the approximate Sr and Nd isotope composition of the proposed North Atlantic end-member can be inferred.

paradigm equates this relatively undegassed reservoir with lower mantle beneath the 670 km seismic discontinuity (e.g. Kellogg & Wasserburg, 1990)—although there are alternative paradigms (Anderson, 1993; Phipps Morgan & Morgan, 1999). The high ³He/⁴He of Icelandic basalts (Condomines et al., 1983; Kurz et al., 1985; Poreda et al., 1986; Hilton et al., 1999) is at least consistent with seismic evidence suggesting that the Iceland plume originates at the core—mantle boundary (Helmberger et al., 1998; Shen et al., 1998; Bijwaard & Spakman, 1999) and for the purposes of the following discussion we will assume that the lower mantle is indeed the source of high ³He/⁴He.

The high diffusivity of He in most minerals at Earth's surface temperatures and the rapid accumulation in rocks of ⁴He through radioactive decay of U, Th and their daughter nuclides restricts the preservation of magmatic ³He/⁴He in ancient basalts. However, for basalts that include abundant He-retentive and U, Th-poor minerals, principally olivine, the helium isotope composition of

ancient plume basalts can be measured and the involvement of a high ³He/⁴He lower-mantle signature quantified (Basu *et al.*, 1993, 1995; Marty *et al.*, 1996, 1998; Graham *et al.*, 1998).

Helium isotope data are now available from olivinerich basalts that cover much of the subaerially preserved NAIP. High ${}^{3}\text{He}/{}^{4}\text{He} > 8$ R_{a} (Fig. 5) have now been recorded at Hold with Hope, NE Greenland (Marty et al., 1998), West Greenland (Graham et al., 1998) and Skye (Stuart et al., 2000). Here (Table 3) we present new data that extend He isotope coverage to the Prince of Wales Mountains (c. 69°N, 32°30'W, Fig. 1), in NE Greenland (Brown et al., 1996). Two of the Prince of Wales Mountains samples have ${}^{3}\text{He}/{}^{4}\text{He} > 8 R_{a}$, although one of these is within error of the MORB range (8 \pm 1 R_a). A third sample, whose unradiogenic Pb isotope compositions (Table 3) suggest substantial contamination with continental crust, has considerably lower ³He/⁴He of c. $2 R_a$. The new data are therefore consistent with the above-mentioned recent studies, which have all found

Table 3: Pb and He isotope results for olivine basalts from the Prince of Wales Mountains, East Greenland

Sample	²⁰⁶ Pb/ ²⁰⁴ Pb ₍₀₎	²⁰⁷ Pb/ ²⁰⁴ Pb ₍₀₎	²⁰⁸ Pb/ ²⁰⁴ Pb ₍₀₎	³ He/ ⁴ He (<i>R</i> / <i>R</i> _a)
EG2240	17·551±8	15·149±7	37.309 ± 18	9.7 ± 0.8 1.8 ± 0.8 10.5 ± 1.0
EG2244	14·733±4	14·620±5	35.741 ± 11	
EG3359	18·328±6	15·352±5	38.409 ± 15	

Pb techniques as for Table 1. He isotope ratios are normalized to the atmospheric ratio (1.39×10^{-6} ; $R_{\rm s}$) and blank corrected on the basis of measured Ne abundances. Helium was extracted by *in vacuo* crushing using techniques described by Stuart *et al.* (1995). All errors are 2 SE. Details of sample localities and additional geochemical data have been given by Brown *et al.* (1996).

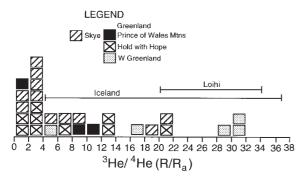


Fig. 5. Histogram showing recent helium isotope measurements on olivines from North Atlantic Igneous Province Palaeogene lavas and a comparison with the ranges from Iceland and Loihi Seamount, Hawaii. Data sources: Allègre et al. (1983, 1987); Kurz et al. (1983); Rison & Craig (1983); Valbracht et al. (1997); Graham et al. (1998); Marty et al. (1998); Hilton et al. (1999, table 3); Stuart et al. (2000).

³He/⁴He >8 *R*_a (Fig. 5) but also tended to discover substantial variability in ³He/⁴He. A range of ³He/⁴He is not unexpected because the samples studied all have complex petrogenesis—they are, after all, continental basalts. However, there is good reason to believe that the highest ³He/⁴He values measured from each province are minima because the processes that could have affected ³He/⁴He (i.e. crustal contamination and post-eruptive radiogenic in-growth) act to lower ³He/⁴He. Only accumulation of cosmogenic ³He would raise ³He/⁴He and all the recent studies have employed *in vacuo* crush release of magmatic helium, which minimizes the contribution from crystal-lattice-hosted helium that might be partly cosmogenic.

Thus, the observed high ³He/⁴He ratios appear indicative of a lower-mantle contribution to basalt petrogenesis throughout the North Atlantic province from West Greenland to the BTIP. However, ³He/⁴He ratios

that approach those of the highest values from Loihi seamount and Iceland (i.e. >30 $R_{\rm a}$) have been encountered only in the West Greenland picrites (Graham et al., 1998), which are thought to have been erupted within 200 km of the plume axis. We cannot rule out the possibility that towards the periphery of the plume similarly elevated magmatic ${}^{3}\text{He}/{}^{4}\text{He}$ ratios have been reduced by crustal contamination, but it is tempting to infer that West Greenland ${}^{3}\text{He}/{}^{4}\text{He}$ values are highest because they are closest to the plume axis and that the lower-mantle signature becomes diluted towards the periphery of the plume so that the NE Greenland and BTIP basalts preserve He isotope ratios (${}^{3}\text{He}/{}^{4}\text{He} \leq 21$ $R_{\rm a}$) intermediate between Iceland plume (${}^{3}\text{He}/{}^{4}\text{He} \geq 37$ $R_{\rm a}$) and MORB (${}^{3}\text{He}/{}^{4}\text{He} = 8 \pm 1$ $R_{\rm a}$).

He-Sr-Nd-Pb CO-VARIATIONS

Taking ${}^{3}\text{He}/{}^{4}\text{He} > 8$ R_{a} as indicative of a lower-mantle contribution, we might expect this signature to be most pronounced in samples with Pb isotope compositions close to the proposed NAEM. However, it is necessary to consider the possibility that distinctly non-linear mixing relationships might arise between upper- and lowermantle end-members in He-Pb-Sr-Nd space. Best estimates (e.g. Kellogg & Wasserburg, 1990; O'Nions & Tolstikhin, 1994) of upper- and lower-mantle He concentrations suggest that the relatively undegassed lower mantle has of the order of 100 times higher He concentration than the degassed upper mantle. This is rather more contrast than expected for the moderately incompatible trace elements Pb, Sr and Nd, for which a 10 times concentration contrast between primitive lower mantle and depleted upper mantle seems a realistic maximum (e.g. McKenzie & O'Nions, 1995). Modelling mixing between relatively depleted upper mantle and a relatively enriched lower mantle whose helium characteristics are similar to the 'primitive helium enriched mantle' (PHEM) of Farley et al. (1992) results in distinctly non-linear mixtures (Fig. 6a and b). Imposing a lowermantle ${}^{3}\text{He}/{}^{4}\text{He}$ of 35–50 $R_{\rm a}$ allows a mixture with 20 $R_{\rm a}$ to be achieved with as little as 0.5% PHEM contribution, which results in a Pb isotope shift that is little larger than analytical error (Fig. 6a).

For Sr and Nd, the lower-mantle-upper-mantle contrast is likely to be less than for Pb for two principal reasons. First, Sr and Nd are slightly more compatible than Pb during mantle melting. The depleted mantle is therefore expected to be less depleted in Sr and Nd than in Pb. Hence, the contrast between lower and upper mantle will be smaller for Sr and Nd concentrations than for Pb abundance. Second, Pb is significantly more enriched in average continental crust (e.g. Taylor & McLennan, 1985; Rudnick, 1995) than are Sr and Nd.

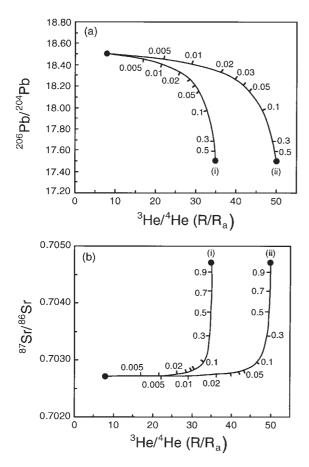


Fig. 6. Pb—He and Sr—He mixing relationships for mixtures of upper and lower mantle assuming the following characteristics. Lower mantle: He 1 × 10⁻⁹ mol/g, 3 He/ 4 He = (i) 35 $R_{\rm a}$; (ii) 50 $R_{\rm a}$; Pb 0·16 ppm, 206 Pb/ 204 Pb = 17·5, 87 Sr/ 86 Sr = 0·7047. Upper mantle: He 1 × 10⁻¹¹ mol/g, 3 He/ 4 He = 8 $R_{\rm a}$, Pb 0·016 ppm, 206 Pb/ 204 Pb = 18·5, 87 Sr/ 86 Sr = 0·7027. Numbered tick marks indicate proportions of lower mantle in mixture. Even small (<1%) amounts of lower mantle are able to raise the He isotope compositions of mixtures to above 20 $R_{\rm a}$ whereas Pb, and even more so Sr, remain dominated by the uppermantle end-member.

That enrichment is likely to be 'balanced' by relative deficiency of Pb in the depleted upper mantle if, as widely held, the upper mantle is residual to continental crust extraction. He–Sr (Fig. 6b) and He–Nd (not illustrated) mixing relationships are therefore likely to be even more non-linear than that of He–Pb. Thus, although He isotopes may be a very sensitive indicator of lower-mantle involvement in hotspot magmatism, it is unlikely that Sr, Nd and Pb isotope signatures of the lower mantle can be inferred from the compositions of basalts that display high ³He/⁴He.

One caveat must be that this inference depends greatly on our model assumption that even the highest ³He/⁴He values measured in basalts are lower than the lowermantle value. If, on the other hand, the basalts with highest ³He/⁴He faithfully record lower-mantle ³He/⁴He,

then clearly this is a lower-mantle signature for all isotope systems. None the less, it will remain possible to approach closely lower-mantle $^3{\rm He}/^4{\rm He}$ without approaching lower-mantle Sr, Nd or Pb isotope compositions. To date, values of $^3{\rm He}/^4{\rm He} > 30~R_{\rm a}$ are known only from Loihi seamount, Iceland and West Greenland basalts; these show similar, but not identical, Pb isotopes and, perhaps significantly, they do not approach the NAEM composition.

One additional matter that requires clarification is a potential misunderstanding that our observations above might be in conflict with the coherent and linear He-Pb isotope mixing relationships observed within the various Hawaiian volcanoes (Eiler et al., 1998). That observation demonstrates persuasively that a high ³He/⁴He Loihi component with distinctive Pb isotope characteristics contributes to the source regions of Hawaiian basalts and does so in similar measure for He and Pb. We do not dispute that interpretation but point out that the Loihi component itself is likely to be a mixture of source materials. The mixing process responsible for the Loihi component is therefore subject to exactly the same nonlinearity as illustrated in Fig. 6 and it is possible to produce the Loihi component with dominantly lowermantle He but upper-mantle Pb. Once constituted, the Loihi component can participate in further source mixing beneath Hawaii and effect similar leverage on such mixtures for both Pb and He. However, this in itself does not necessarily demonstrate a significant role for lowermantle Sr, Nd and Pb.

Only if the Loihi component could be demonstrated to represent unadulterated lower mantle would it be appropriate to infer that its Sr, Nd and Pb isotopic compositions are similarly representative of lower mantle. Plausible lower-mantle ${}^{3}\text{He}/{}^{4}\text{He}$ is no more constrained than >37 R_{a} (e.g. Hilton *et al.*, 1999) Thus, an *a priori* assumption that the highest ${}^{3}\text{He}/{}^{4}\text{He}$ measured at Loihi represents lower-mantle ${}^{3}\text{He}/{}^{4}\text{He}$ seems imprudent. There remains sufficient latitude in the lower-mantle He isotope composition that the observed range in basalt ${}^{3}\text{He}/{}^{4}\text{He}$ is consistent with a situation in which the lower-mantle contribution to even the most ${}^{3}\text{He}$ -rich OIB remains sufficiently small that Sr, Nd and Pb are predominantly derived from the upper mantle.

Similarly, other elements that are more evenly distributed between upper and lower mantle than is He will be subject to the same non-linear mixing relationships. Most probably, this applies to all the commonly analysed incompatible lithophile elements. In particular, we would not expect the Nb/Y and Zr/Y diagrams used to distinguish Iceland basalts from MORB (Fitton *et al.*, 1997) to be significantly more sensitive to small contributions of lower-mantle material than we propose are Sr, Nd and Pb isotopes. It is notable that the recent NAIP He isotope data include samples with high ³He/⁴He values

that have both positive (West Greenland, NE Greenland) and negative (BTIP) ΔNb (Fitton *et al.*, 1997). On the basis of their ΔNb value, Fitton *et al.* (1997) favoured a source for the Iceland plume predominantly within the upper mantle and an origin at the 670 km transition zone. Our data support that interpretation and also demand a minor contribution from high ³He/⁴He, Herich lower mantle.

The above analysis leads us to conclude that it is unlikely that any basalts sample lower mantle to sufficient degrees that their Sr, Nd and Pb isotope compositions approach lower-mantle values, and this conclusion is consistent with the simple observation that the high ³He/

⁴He OIB do not tend towards one particularly well-defined Sr, Nd and Pb isotope composition, and certainly do not approach Bulk Earth Sr or Nd. Similarly, the proposed NAEM is unlikely to represent lower-mantle Sr, Nd and Pb, and again this is consistent with its low ⁸⁷Sr/⁸⁶Sr and high ¹⁴³Nd/¹⁴⁴Nd. We conclude that the observation that the NAEM falls close to the 4·55 Ga geochron is not likely to be indicative of the involvement of primordial lower mantle and may be purely coincidental.

None the less, the implication remains that upper mantle with a Pb isotope composition that falls close to the geochron appears to be widely implicated in North Atlantic magmatism. Perhaps the most revealing aspect of the NAEM comes from comparison of the Palaeogene flood basalts and present-day Icelandic basalts. For Recent Icelandic magmatism, the NAEM is only one source component and the Iceland Pb–Pb arrays (Fig. 2d and h) trend away from the NAEM towards high ²⁰⁶Pb/²⁰⁴Pb, implicating at least one other distinct mantle source in the petrogenesis of Icelandic basalts, which we will term the 'Iceland radiogenic Pb end-member'.

In the examples we have used to identify the NAEM (i.e. Antrim, Skye, Mull and ODP Leg 152), the uncontaminated basalts always approach the NAEM itself, not an 'average Icelandic' composition lying between NAEM and the Iceland radiogenic Pb end-member. By contrast, relatively uncontaminated NE Greenland samples have Pb isotope compositions that fall towards the centre of the Iceland Pb-Pb arrays, and this has been interpreted (Thirlwall et al., 1994) to indicate the availability of both relatively low 206Pb/204Pb and relatively high 206Pb/204Pb Icelandic end-members beneath NE Greenland 60 my ago. Given the scatter in the various Pb-Pb isotope arrays it is probably imprudent to attempt to distinguish these two viewpoints but it is worth noting that both have significant implications for the structure of the proto-Iceland plume.

On the basis of the BTIP and SE Greenland data, we might speculate that the Iceland radiogenic Pb endmember was not available during CFB production. This could indicate that the mantle plume beneath Iceland has become progressively more heterogeneous as the radiogenic Pb end-member was incorporated into the plume. Alternatively, it may suggest that the source of radiogenic Pb is distinct from the plume and was simply not available to melt during the Tertiary; perhaps because it represents a small-scale upper-mantle heterogeneity currently convecting through the melting regime beneath Iceland. On the basis of the NE Greenland data, the radiogenic Pb Iceland end-member has been a pervasive component of the plume but only beneath Iceland today have conditions been such that it is sampled as a discrete entity. Elsewhere it was either efficiently mixed into the plume (NE Greenland) or absent (BTIP-SE Greenland). We are therefore inclined to speculate that the NAEM represents relatively deep-seated, but still predominantly upper-mantle, material whereas the Iceland radiogenic Pb end-member is derived from more transient shallow heterogeneities, which were sampled, at most only very sporadically, by the Palaeogene magmatism, but one of which is now present and fusible beneath Iceland.

CONCLUSIONS

(1) Pb isotope variations, which are best interpreted as mixing trends between asthenospheric magmas and lithospheric contaminants, are well developed in basalts from the BTIP but indicate very different contaminants, allowing extrapolation to uncontaminated compositions along vectors that are diametrically opposed and that converge upon a restricted field taken to indicate a single sub-lithospheric end-member. This North Atlantic endmember (NAEM) also falls close to an extension of the Iceland ²⁰⁶Pb/²⁰⁴Pb-²⁰⁷Pb/²⁰⁴Pb and ²⁰⁶Pb/²⁰⁴Pb-²⁰⁸Pb/ ²⁰⁴Pb arrays towards low ²⁰⁶Pb/²⁰⁴Pb, and therefore appears to have been present throughout the development of the 60 Ma to present North Atlantic-Iceland magmatic province. Data from other North Atlantic Tertiary centres are generally not inconsistent with the existence of the NAEM.

(2) The proposed NAEM has Pb isotope compositions (206Pb/204Pb ~ 17·5, 207Pb/204Pb ~ 15·4 and 208Pb/204Pb ~ 37·4) that lie close to the 4·55 Ga geochron. That observation carries the possible implication that the NAEM is primordial, and given the obvious link with the Iceland plume and the common view that such plumes originate at great depth in the mantle, it is tempting to equate NAEM with a lower-mantle source component. However, consideration of the likely mixing relationships between lower and upper mantle leads to the conclusion that NAEM is unlikely to represent unadulterated lower mantle and that the similarity with a credible lower-mantle Pb isotope composition is more likely to be coincidental.

(3) Simple mixing models also suggest that the lowermantle 'leverage' on mixtures of upper and lower mantle will be much greater for He than for Sr, Nd, Pb or, indeed, other lithophile trace elements. Unless we can be sure that the highest ³He/⁴He OIB sampled lower mantle unadulterated by upper mantle, we cannot assume that any OIB carries more than a small (<1%) lowermantle contribution and that is not sufficient for mixtures to approach lower-mantle Sr, Nd and Pb isotope compositions. Individual volcanic systems may well yield coherent He-Pb-Sr-Nd mixing relationships but this does not necessarily indicate that the high ³He/⁴He component in such mixtures carries significant lowermantle Sr, Nd and Pb. The high ³He/⁴He end-member is itself likely to be a mixture of lower- and upper-mantle sources in which He is dominated by lower mantle but Sr, Nd and Pb are overwhelmingly derived from the upper mantle.

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