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Ancient, highly heterogeneous mantle beneath Gakkel ridge, Arctic Ocean

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The Earth's mantle beneath ocean ridges is widely thought to be depleted by previous melt extraction, but well homogenized by convective stirring. This inference of homogeneity has been complicated by the occurrence of portions enriched in incompatible elements. Here we show that some refractory abyssal peridotites from the ultraslow-spreading Gakkel ridge (Arctic Ocean) have very depleted $^{187}Os/^{188}Os$ ratios with model ages up to 2 billion years, implying the long-term preservation of refractory domains in the asthenospheric mantle rather than their erasure by mantle convection. The refractory domains would not be sampled by mid-ocean-ridge basalts because they contribute little to the genesis of magmas. We thus suggest that the upwelling mantle beneath mid-ocean ridges is highly heterogeneous, which makes it difficult to constrain its composition by mid-ocean-ridge basalts alone. Furthermore, the existence of ancient domains in oceanic mantle suggests that using osmium model ages to constrain the evolution of continental lithosphere should be approached with caution.

The Earth's mantle is known to be chemically heterogeneous on spatial scales ranging from the size of ocean basins down to kilo-metres or possibly metres, with incompatible-element-enriched portions surviving up to 3 Gyr (billion years)[1-5]. However, on the basis of the relatively uniform radiogenic isotope and trace element composition of normal mid-ocean-ridge basalts (N-MORB), the depleted MORB mantle (DMM) source has often been invoked as a large-scale homogeneous and degassed reservoir[3,6-8]. This homogeneity has been complicated by the mixing of the primary depleted basalts (N-MORB) with enriched basalts (E-MORB)[1-4], and also by the recognition of ancient depletion inherited in some MORB[9,10] and abyssal peridotites[11,12]. Geochemical studies of MORB largely revolve around elements that preferentially enter the melt phase during partial melting (that is, incompatible elements). Once clinopyroxene is exhausted during partial melting, these refractory peridotites cannot be re-melted under normal anhydrous conditions and thus a portion of the mantle is rendered invisible in magmas derived from any subsequent melting event. The refractory mantle domains are more likely to be preserved and detected at mid-ocean ridges with low spreading rates, because they are not obscured by melting processes as at other faster spreading ridges. Gakkel ridge is an ultraslow-spreading ridge and magmatic activity is relatively weak there[13,14]. Fresh abyssal peridotites recovered from Gakkel ridge[15] thus provide us an opportunity to study ancient depletion signatures inherited in the refractory mantle.

Ancient depletion signals in refractory mantle domains are well recorded by the Re–Os isotopic system, because of its unique geo-chemical properties. Unlike other long-lived isotopic systems (for example, Sm–Nd and Rb–Sr) in which both parent and daughter elements are lithophile, osmium is chalcophile and behaves as a compatible element in mantle peridotites during mid-ocean-ridge partial melting. Rhenium, on the other hand, behaves as a moderately incompatible element with a bulk partition coefficient similar to that of aluminium; even different phases probably control the partitioning of these two elements[16]. Furthermore, higher Os concentrations in peridotites relative to basaltic melts render the Os isotopes in peridotites more robust to late-stage metasomatism and contamination processes than lithophile isotope systems. Previous Re–Os studies on abyssal peridotites have indicated that some mantle domains retain evidence for ancient partial melting events[11,12,17].

Geological setting of samples

The 1,800-km-long Gakkel ridge is the slowest-spreading mid-ocean ridge in the world. It forms the North America/Eurasia plate boundary in the Arctic Ocean (Fig. 1). To the west it passes via the Lena trough and the Molloy fracture zone into the Knipovich ridge, and to the east it runs into the Siberian continent as a broad region of continental rifting on the Laptev shelf[14]. The spreading of Gakkel ridge propagates eastwards with a rate varying from 14.6 mm yr^{-1} on its western end to 6.3 mm yr^{-1} at its Siberian eastern end[14,18]. The ridge axis is continuous, with no transform offsets, and there are three main tectonic zones, from west to east, the Western Volcanic Zone (7° W to 3° E), the Sparsely Magmatic Zone (3° E to 29° E) and the Eastern Volcanic Zone (29° E to 85° E)[14]. Samples in the present study are selected from two dredge hauls, HLY0102-D70 (AMORE 2001)[14] and PS66-238 (ARK XX-2, 2004)[15] (Fig. 1). Dredge haul PS66-238 is located at the transition from the Western Volcanic to the Sparsely Magmatic Zone (84° 29.23′ N, 4° 12.64′ E), around which no basalts, only peridotites, have been recovered (3° E to 8° E), and it is thought to indicate an amagmatic segment. Dredge haul HLY0102-D70 represents the easternmost occurrence of peridotite found on Gakkel ridge so far and is located near a volcanic centre in the Eastern Volcanic Zone. The two dredge hauls straddle an important geochemical boundary, indicated by the radiogenic compositions of MORB samples globally between western Gakkel DUPAL-like mantle and eastern Gakkel mantle, which is similar in nature to that of the north Atlantic[19].

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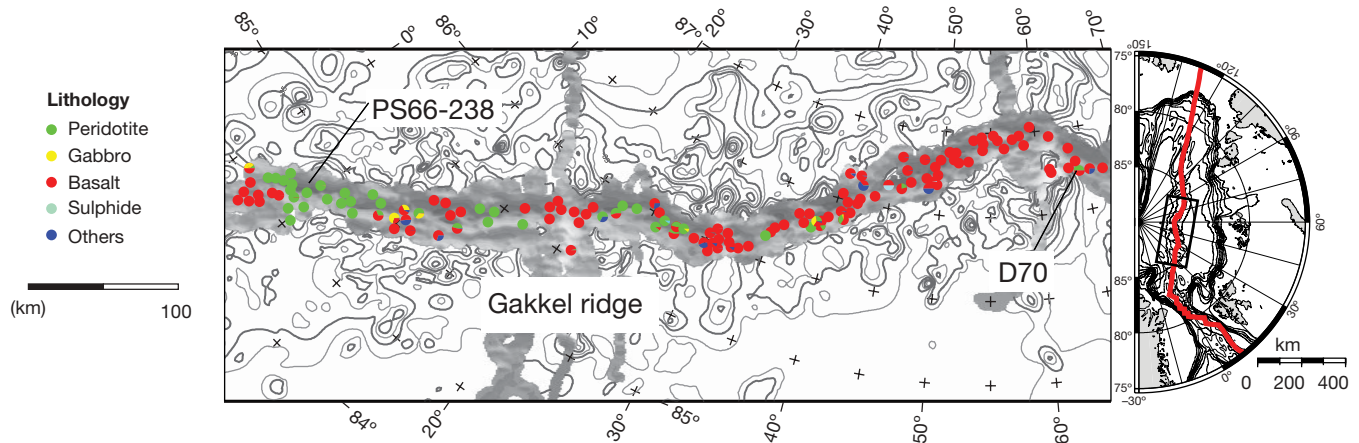
Figure 1 | Sketch map of dredge hauls on Gakkel ridge. Dredge haul PS66-238 is located at the transition from the Western Volcanic Zone to the Sparsely Magmatic Zone, in which only peridotites and no basalt have been recovered. Dredge haul D70 is recovered in the Eastern Volcanic Zone, which is the easternmost occurrence of peridotite found on Gakkel ridge as yet.

The samples from dredge PS66-238 comprise nine spinel lherzolites, one plagioclase lherzolite and one harzburgite. All of the samples from dredge PS66-238 are extremely fresh but with centimetre-thick brown-yellow weathering rims, which were separated from their fresh interiors. Most interior portions have a loss on ignition (LOI, an indicator of volatile components in the rock due to olivine serpentinization) close to 0% and only two of them are around 1%. Their weathered rims have slightly higher LOI (up to 3%) relative to their corresponding interiors, but these are still substantially lower than those in typical serpentinites ($\sim$14%). Samples from dredge D70, consisting of six lherzolites and one harzburgite, are typical serpentinized peridotites with large LOI (9–14%). Most of the peridotite samples from both dredges are relatively fertile with similar ranges of bulk Al_2O_3, 1.3–3.2% in D70 and 1.5–3.1% in PS66-238 (see Supplementary Table 1). Both D70 and PS66-238 samples have a similar range of chromium number, $Cr\# = $ molar $Cr/(Cr+Al)$, in their residual spinels ($\sim$0.12–0.28 and $\sim$0.15–0.28, respectively), which indicates that they were subjected to low degrees of partial melting[20] (Fig. 2a). The same conclusion can also be derived from the content of ytterbium (Yb) in the clinopyroxene (cpx). Clinopyroxenes have higher light rare earth elements, indicated by high Ce/Yb ratios, than predicted by simple partial melting models (Fig. 2b), implying late-stage enrichments after low-degree partial melting[21].

Re–Os isotopes of Gakkel abyssal peridotites

Except for one sample, all PS66-238 samples have higher Os concentrations than the D70 samples (Fig. 3a). This cannot be accounted for by any serpentinization effect because the more serpentinized sample rims are not systematically higher than the less serpentinized interiors; thus it may either be a source characteristic or reflect the different magmatic activity in the mantle beneath these two dredge hauls. Rhenium in the peridotites is potentially affected by secondary processes: that is, serpentinization and melt refertilization. No large gap in Re content, however, exists between the two dredge hauls, all of which are generally lower than the estimated value of the primitive mantle (0.34 parts per 10^9, ref. 22).

Replicate analyses indicate that Os contents are variable (Supplementary Table 1), because of a 'nugget effect' from different amounts of small sulphide phases in the sample aliquots, but that Os isotopes are highly reproducible. Previous studies have suggested that sulphides in mantle peridotites can be very heterogeneous in Os isotopic composition on the individual grain scale, from relatively depleted in the silicate-included sulphides to highly radiogenic in the interstitial sulphides[23]. The highly reproducible Os isotopes but variable Os concentrations in replicate analyses indicate that sulphides in Gakkel abyssal peridotites (both serpentinized and fresh) have similar Os isotopic compositions. Notably, $^{187}Os/^{188}Os$ ratios of samples from both dredge hauls have similar ranges, varying from depleted to relatively radiogenic values, despite their substantial differences in Os contents (Fig. 3a).

Neither the D70 nor the PS66-238 samples show any correlation between their Re/Os and Os isotopic ratios, whereas a good correlation exists between Al_2O_3 and $^{187}Os/^{188}Os$ in the D70 samples (Fig. 3b). Rhenium is more likely to be disturbed by late processes than aluminium, and this is the reason that bulk Al_2O_3 content is often used as a proxy for Re/Os to calculate the ages of partial melting events in mantle peridotites[24]. Although the PS66-238 samples have similarities with D70 samples in both bulk Al_2O_3 contents and $^{187}Os/^{188}Os$ ratios, an Os–Al_2O_3 correlation does not exist in PS66-238 samples (Fig. 3b). However, the most refractory sample does have the lowest $^{187}Os/^{188}Os$.

Ancient and recent partial melting

The correlation between $^{187}Os/^{188}Os$ and Al_2O_3 was first observed in orogenic peridotites[24], and has subsequently also been reported in the continental mantle from other tectonic settings, for example mantle xenoliths[25–27]. Until now, no such relationship has been found in the young asthenospheric mantle, that is, abyssal peridotites. This could be attributed to the convective stirring in the oceanic mantle, the juxtaposition of pieces of mantle with different melting histories, and subsequent partial melting and reactive melt migration beneath the ridge. The inherited depletions in Gakkel ridge abyssal peridotites record ancient melting events, which extracted Re and caused their unradiogenic $^{187}Os/^{188}Os$ ratios. Inherited ancient partial melting has also been previously invoked as the cause of unradiogenic $^{187}Os/^{188}Os$ ratios in abyssal peridotites from the Mid-Atlantic Ridge[11,12] and in forearc mantle peridotites from the Izu–Bonin–Mariana subduction zone[17]. Ancient depletions have also been inferred to exist in the mantle sources of some MORB[9,10].

Another possibility is that the D70 samples with a correlation between $^{187}Os/^{188}Os$ and Al_2O_3 represent a residual subcontinental lithospheric mantle, which is delaminated and incorporated into asthenosphere[28,29]. This is unlikely, however, because studies on MORB from the eastern Gakkel ridge suggest that they come from normal depleted 'Atlantic-like' oceanic mantle, and no signature of any continental mantle in their mantle source has been found in these MORB[19].

Harzburgites from both dredge hauls (D70-62 and PS66-238-22) give the oldest Re depletion ages yet found in global abyssal peridotites, around 2.2 Gyr (relative to primitive upper mantle[30]). These ancient depletion events are far older than, and therefore unrelated

to, any recent decompression partial melting beneath Gakkel ridge. This is particularly well illustrated by the fact that the correlation between Os isotopes and bulk Al_2O_3 content would not exist if they had been affected by substantial partial melting beneath Gakkel ridge.

Effects of seawater contamination and melt refertilization

Two processes that might affect Os isotopes of abyssal peridotites are interactions with metasomatic fluids or melts and seawater contamination. The potential for seawater contamination to increase the radiogenic Os isotopes of abyssal peridotites has been debated for some time[23,31,32]. Ratios of $^{187}Os/^{188}Os$ from abyssal peridotites with clear signs of seafloor weathering have been suspected of seawater contamination and typically discounted from representing the depleted MORB mantle in previous studies[31]. The current sample set provides an excellent test of this hypothesis, as we have perfectly fresh, weathered and serpentinized samples (although the latter are from a different area). The $^{187}Os/^{188}Os$ ratios in the five weathered rims from dredge PS66-238 are slightly higher than in their corresponding interiors, with a maximum Os isotope ratio that is 2.3% higher in the harzburgite. Such a difference is significant compared

with the external reproducibility of the analytical method (<0.4%), but small compared with the overall range in the data. The interiors of these samples are essentially very fresh, and thus are unlikely to show any seawater influence at all. Despite the very large difference in their alteration, both PS66-238 and D70 samples have similar ranges of $^{187}Os/^{188}Os$ ratios. Thus, seawater contamination has had a negligible effect on the Os isotopic compositions of these abyssal peridotites.

Melt refertilization is very common in abyssal peridotites from slow-spreading ridges[29]. Trace elements in clinopyroxenes indicate that Gakkel abyssal peridotites were refertilized by late-enriched melts after low-degree partial melting. Basaltic melts generally have more radiogenic $^{187}Os/^{188}Os$ ratios (0.125–0.23 compared with 0.12–0.13) but much lower Os contents (1–50 parts per 10^{12} (p.p.t.) compared with 1–4 parts per 10^9 (p.p.b.)) relative to abyssal peridotites[33]. Even the addition of 25% of an extreme hypothetical basaltic melt ($^{187}Os/^{188}Os = 0.23$ and [Os] = 50 p.p.t.) to an abyssal peridotite ($^{187}Os/^{188}Os = 0.127$ and [Os] = 3.4 p.p.b.) would result in only 0.4% change in the Os isotopic composition of a typical mantle peridotite. Thus, simple binary mixing with basaltic melt cannot significantly affect Os isotopic compositions in mantle rocks. On
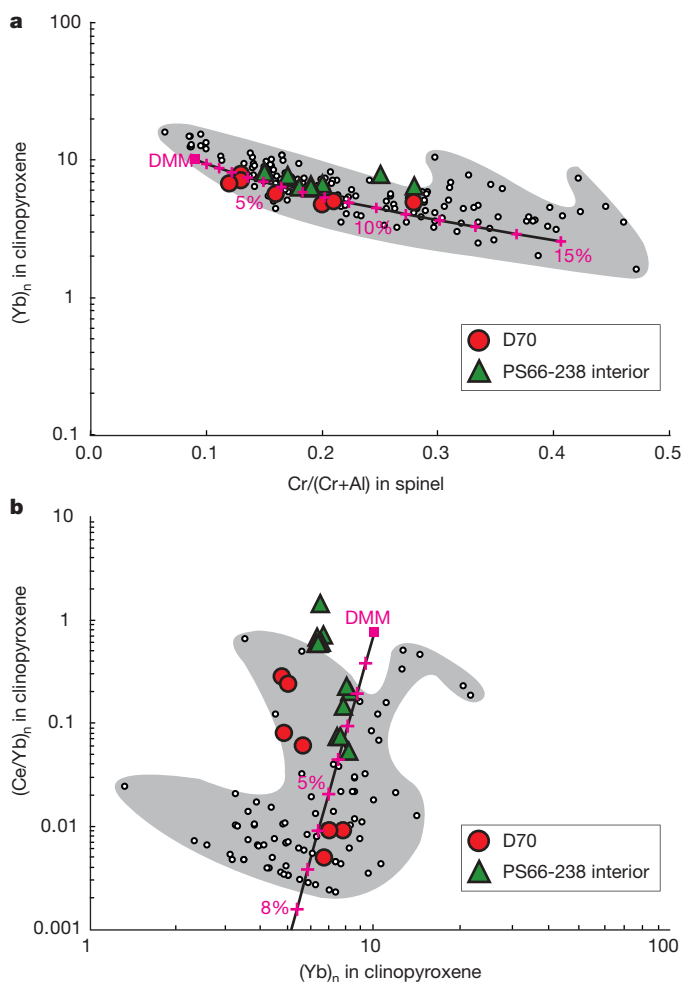

Figure 2 | Sample composition indicating partial melting and melt refertilization histories. a, Spinel Cr# versus $(Yb)_n$ in cpx (n, chondrite normalized). Low to moderate (5–12%) degrees of partial melting in Gakkel abyssal peridotites are estimated by Cr# (= Cr/(Cr+Al)) of the residual spinels, using a method described previously[21]. **b,** $(Ce/Yb)_n$ versus $(Yb)_n$ in cpx. Ce/Yb ratios in cpx from both D70 and PS66-238 deviate from the fractional partial melting trend, indicating the occurrence of melt refertilization. Chondrite normalized values are from ref. 47. The equation and parameters used in fractional partial melting modelling are from ref. 21 and DMM compositions are from ref. 8. The white small circles are abyssal peridotite data from the literature[21,48,49]

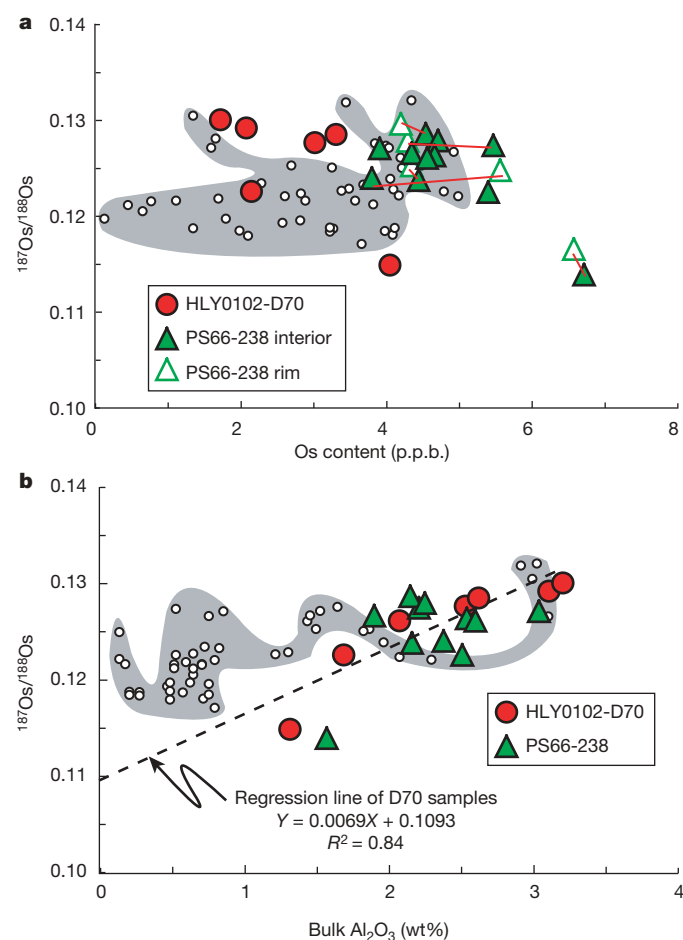**Figure 3 | Diagram of $^{187}Os/^{188}Os$ versus Os content and bulk Al_2O_3. a,** Osmium contents of D70 samples are systematically lower than those of PS66-238 samples. Fresh interiors and their corresponding altered rims from the same sample are connected by the red lines. The altered rims generally have slightly higher $^{187}Os/^{188}Os$ ratios than their corresponding interiors, whereas there is no systematic variation of Os contents between rims and interiors. **b,** Both PS66-238 and D70 samples have similar ranges in both $^{187}Os/^{188}Os$ ratios and bulk Al_2O_3 contents. Bulk Al_2O_3 contents of D70 samples show good correlation with their $^{187}Os/^{188}Os$ ratios, which, however, is absent in PS66-238 samples. The white small circles are abyssal peridotite data from the literature[11,12,17,31,50].

the other hand, percolation of sulphide melts rather than basaltic melts would more effectively increase the $^{187}Os/^{188}Os$ ratios of the mantle peridotites[34]. For example, *in situ* study of sulphides by the laser ablation method has detected radiogenic $^{187}Os/^{188}Os$ in interstitial sulphides, which were explained as secondary sulphides and addition from melts[23]. However, the Al_2O_3–$^{187}Os/^{188}Os$ correlation in the D70 peridotites suggests that their Os isotopes have not been significantly affected by addition of either sulphides or basaltic melts. Otherwise, such a correlation would have been erased. The correlation between Re/Os and bulk Al_2O_3 content in the PS66-238 peridotites (Fig. 4) also indicates that the addition of pure sulphide melt did not occur in these samples.

Os isotopic heterogeneity in the oceanic mantle

Currently, there is no consensus on the Os isotopic composition of the DMM[31,35]. On the basis of the $^{187}Os/^{188}Os$ ratios of abyssal peridotites, it has been suggested that DMM has subchondritic $^{187}Os/^{188}Os$ ratios around 0.125, which implies a long-term Re depletion in DMM relative to primitive upper mantle (PUM)[31]. The continental crust does not balance the amount of Re depleted from the upper mantle, and therefore subducted oceanic crust, possibly stored in the lower mantle, has been proposed as a potential reservoir for such 'missing Re'[36]. Other studies, however, inferred DMM to have PUM-like $^{187}Os/^{188}Os$ ratios ≈ 0.129, indicating that little Re has been removed from the DMM as a whole[35]. This would imply that the refractory peridotites with unradiogenic $^{187}Os/^{188}Os$ ratios generally do not contribute to the MORB source region during melting in magmatically active or inactive ridge segments. Gakkel ridge is an extreme case, because the unradiogenic peridotites are refractory harzburgites and the overall degree of melting at the ridge is very low. They may thus preserve ancient signatures from the asthenosphere unaffected by a few per cent of partial melting at Gakkel ridge. On other ridges, however, pervasive wetting by melt results in compositional changes that largely obscure the relationship between the depleted and fertile portions of the Gakkel ridge mantle that we have sampled.

The wide range of $^{187}Os/^{188}Os$ ratios in Gakkel ridge abyssal peridotites reflects the coexistence (on the kilometre scale sampled by dredging) of fertile mantle domains with relatively radiogenic near-PUM $^{187}Os/^{188}Os$ ratios and refractory domains with unradiogenic $^{187}Os/^{188}Os$ ratios. The distribution of refractory and fertile upper mantle domains with different Os isotopic characteristics in the upwelling asthenosphere is, perhaps, also similar to a 'plum pudding' mantle[37]. Thus, a single Os isotopic composition of DMM cannot be
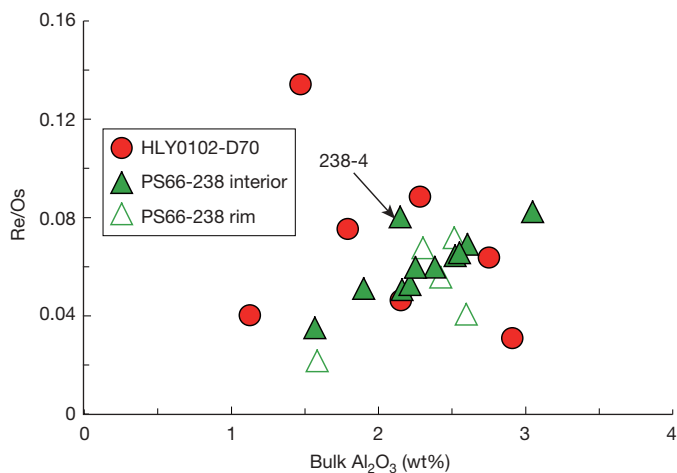

Figure 4 | Correlation between Re/Os ratios and bulk Al_2O_3 contents. All fresh interiors but one (238-4) show good positive correlation. The absence of such a relationship in both D70 samples and the altered rims of PS66-238 samples indicate the disturbance of Re during seawater alteration.

established. The domains with unradiogenic $^{187}Os/^{188}Os$ ratios, represented by the harzburgites, have been subjected to ancient partial melting. The widely reported occurrences of unradiogenic $^{187}Os/^{188}Os$ ratios in both abyssal peridotites[11,12,17] and ophiolites[38–40] suggest that ancient partial melting events are ubiquitously recorded in the global asthenosphere[41]. On the other hand, PUM-like $^{187}Os/^{188}Os$ ratios in many lherzolites from ocean ridges suggest that either no significant Re has been removed in some fertile domains or they have been affected by components enriched in $^{187}Os/^{188}Os$.

Long-time survival of ancient Os isotopic signals in asthenospheric mantle could also have substantial implications for intra-oceanic or continental mantle keel sources. A correlation between $^{187}Os/^{188}Os$ and Al_2O_3 in asthenospheric mantle, like in the D70 samples, could be produced by mixing of peridotites with different fertilities and ages, which have been preserved in the convecting asthenospheric mantle for a long time and delivered to mid-ocean ridges by mantle convection. Such a correlation, therefore, cannot be used to discriminate between subcontinental lithospheric mantle and asthenospheric mantle as has been previously proposed[25]. If mixtures of refractory and MORB-source (that is, relatively fertile) mantle characterize the asthenospheric mantle, young asthenospheric mantle could bear refractory domains with ancient Os isotopic signatures. The observation of relatively unradiogenic Os isotopic signatures in ophiolites (and mantle rocks generally) could simply represent the ambient heterogeneity of depleted harzburgitic domains in the oceanic mantle rather than definite evidence of delaminated subcontinental lithospheric mantle. In the meantime, the use of Os model ages of mantle xenoliths to constrain the ages of subcontinental lithospheric mantle should be approached with caution.

Is DMM the source of MORB?

Traditionally, the terms 'DMM' and 'MORB source' have been considered synonymous with each other, and with upwelling of a depleted asthenospheric mantle that may also contain enriched 'plums' or 'blobs'[3,7,8]. For incompatible elements, this assumption is valid. But it fails to take into account regions of mantle too refractory to take part in partial melting such as sections of Gakkel ridge. These refractory mantle domains have formed by ancient partial melting and been carried passively in the mantle until their emplacement on the sea floor. If observations from Gakkel ridge can be generalized to mantle beneath mid-ocean ridges as a whole, the source of MORB preferentially samples the more fertile portion of the total upwelling mantle, and not the coexisting domains of refractory material that do not melt and thus contribute little to MORB formation. Domains with different melting histories have been mechanically juxtaposed but not homogenized by mantle convection. The fertile mantle domains may or may not have a genetic relationship with neighbouring refractory domains[42].

If refractory domains are heterogeneously distributed in the asthenospheric mantle (Fig. 5), there are several interesting consequences for models of mantle dynamics. First, MORB does not sample the entire upwelling asthenosphere beneath the ocean ridges, but only the non-refractory domains, and thus MORB compositions do not necessarily constrain the nature or composition of the overall asthenosphere. This complicates the interpretation of geochemical data in much the same way as the hypothesis of enriched mantle veins[43,44], which our data do not address. Second, without knowing *a priori* which mantle domains participate in partial melting and to what extent, it is impossible to calculate a DMM (that is, asthenospheric mantle) Os isotopic composition relevant to the formation of MORB from observations in asthenospheric mantle peridotites. The same might also be true for other isotopic systems (for example, Rb–Sr, Sm–Nd, U–Pb and Lu–Hf). Third, observations of asthenospheric mantle composition based on observations on MORB that do not take refractory components into account will tend to overestimate the fertility of the upper mantle, particularly on ultraslow-spreading ridges.
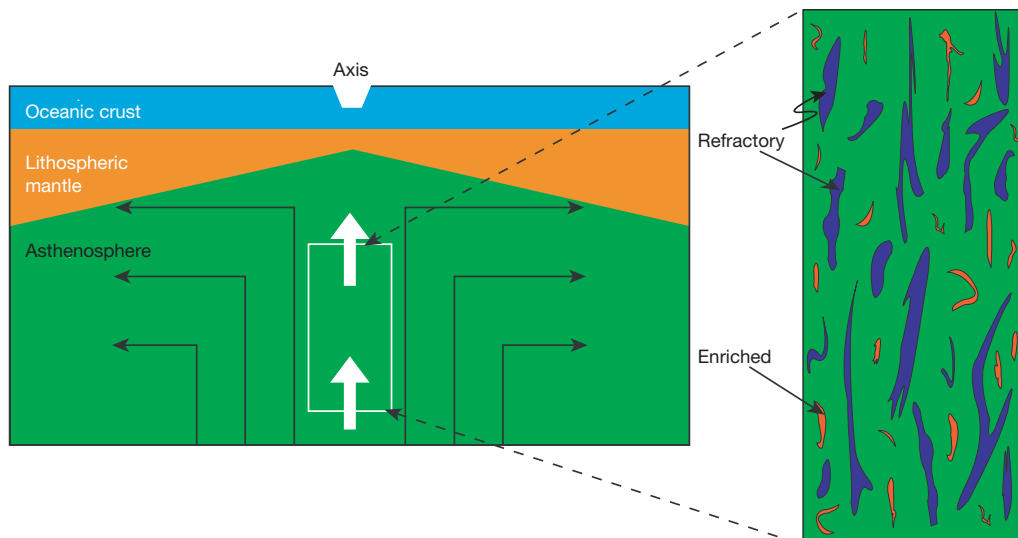
Figure 5 | Schematic diagram of the upwelling asthenospheric mantle beneath mid-ocean ridges. Both enriched and refractory mantle are distributed among the 'matrix' of normally depleted mantle. The enriched mantle and the normally depleted mantle can be sampled by E-MORB and N-MORB, respectively. However, it is hard to sample the refractory mantle with inherited ancient depletion signals because it contributes little to the genesis of MORB. Therefore, they are not a part of the 'MORB source' *sensu stricto*, although the latter has been generally taken to be synonymous with asthenospheric mantle.

METHODS SUMMARY

Major and trace elements. Major elements were analysed using X-ray fluorescence at the University of Mainz. Major elements of spinels were measured by JEOL JXA 8900RL electron microprobe at the University of Mainz, using an acceleration potential of 20 kV, a beam current of 12 nA and a spot size of 2 μm. Clinopyroxene trace-element data of D70 serpentinites were measured by Cameca ims-3f at the Max-Planck Institute of Chemistry (MPI), Mainz, using a method described previously[45]. We measured the trace elements of clinopyroxenes in the fresh PS66-238 samples by laser ablation/inductively coupled plasma mass spectrometry at MPI. Ablation was achieved with a New Wave UP-213 Nd:YAG laser system, using a pulse repetition rate of 10 Hz and crater diameter of 80 μm. Analyses were performed on a single collector sector-field Thermo Finnigan Element-2 mass spectrometer in pulse counting mode. We used ^{43}Ca as internal standard element. We used two internal standards (NIST-612 and KL2-G) and one external standard (GOR132-G), and their values are from GEOREM (http://georem.mpch-mainz.gwdg.de).

Re–Os isotopes. We determined Re–Os isotopes of the Gakkel abyssal peridotites at the Max-Planck Institute for Chemistry. The method has been described previously[46]. Samples of powder (2 g) were digested in capped quartz tubes together with a mixed Re–Os isotope tracer, 3 ml of $12\,\mathrm{mol}\,\mathrm{l}^{-1}$ HCl and 7 ml $16\,\mathrm{mol}\,\mathrm{l}^{-1}$ HNO_3 for 16 h in a high-pressure asher (HPA-S) at 100 bar and 300 °C. We separated Os from the solution by solvent extraction with bromine and purified it by micro-distillation. Afterwards, we extracted Re from the solution in anion exchange columns. We made Os isotope measurements by negative thermal ionization mass spectrometer (N-TIMS) on a Finnigan MAT 262 instrument. Total reagent blanks were <6 pg for Os and <20 pg for Re. The blank ^{187}Os/^{188}Os ratios are less than 0.3176. Repeat measurements of ^{187}Os/^{188}Os in a standard containing 35 pg Os yielded external precision of 0.1% (2δ). Reproducibility based on duplicate analyses of three samples was <0.2% for ^{187}Os/^{188}Os and <25% for Os concentration. The quality of the Os data from the Mainz laboratory was also confirmed in a comparative analytical study of serpentinite standard UB-N. Rhenium was measured on a 'Nu Plasma' multi-collector inductively coupled plasma mass spectrometer. The external precision (2δ) of standard solution ($10–25\,\mathrm{ng}\,\mathrm{g}^{-1}$) measurements was 0.1–0.2% for Re.

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Author Contributions C.-Z.L. and G.B. did the Os-PGE measurements on samples collected by two Arctic expeditions proposed and carried out by J.E.S., together with E.H., A.v.d.H. and A.B., who also provided preliminary and supporting data. A.W.H. provided intellectual and material support. All authors contributed to interpretation of the results and writing of the manuscript.

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