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Recognising early Archaean mantle: a reappraisal

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Abstract This paper examines 3.8 Ga peridotites from Greenland and Labrador to test claims that these samples are unmodified early Archaean mantle. Geochemical criteria were applied in which samples were compared to the mantle array in Mg/Si versus Al/Si (wt%) space, their REE patterns were compared to those of different mantle types and their chromite compositions were compared to mantle chromite compositions as expressed by their cr# and fe#. Geochemical data were used from the previously published works of Friend et al. (2002) and Bennett et al. (2002). Only two samples, from the region south of Isua satisfied all criteria, indicating that the area south of the Isua Greenstone Belt in west Greenland is a suitable place to search for early Archaean mantle. This study also confirms the observation by Friend et al. (2002) that early Archaean mantle from south of Isua is of a different character from Archaean mantle from the subcontinental lithosphere. Calculations presented here show that some mantle fragments from south of Isua experienced a lower degree of melt extraction and were probably more oxidising than early Archaean mantle preserved in the subcontinental lithosphere. Elemental concentrations of Os in early Archaean mantle are lower than the new estimate for the primitive upper mantle of Becker et al. (2006). Peridotites from the Isua greenstone belt are not mantle, but have an affinity with the layered intrusions found south of Isua.

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Introduction

Understanding the geochemical evolution of the Earth's mantle over time is a major quest of geochemistry. For our knowledge of mantle evolution is central to understanding the large-scale processes of Earth evolution, not least in understanding the formation of the Earth's core and the continents. As we go back in time our knowledge of the Earth's mantle becomes increasingly fragmentary, so that in the early Archaean and the Hadean—when interactions within the Earth system were extreme—we have very little material on which to base our models. In particular there are very few reliable outcrops of early Archaean mantle peridotite. This is because they are frequently metamorphosed and so can easily be confused with ultramafic rocks of a different origin—namely, peridotitic komatiites or peridotites of cumulate origin formed within layered intrusions. Furthermore, early Archaean peridotites are frequently enclosed by felsic gneisses and so are susceptible to chemical alteration.

And yet, these samples are of huge importance to models of the Earth. There is a growing body of evidence to indicate that the Earth's mantle differentiated very early in Earth's history. Short-lived radioactive isotopes such as ^{142}Nd suggest that the Earth's mantle experienced a major differentiation event within about 30 Ma of the formation of the solar system (Boyet and Carlson 2005). Further evidence of very early fractionation of the Earth comes from trace element partitioning experiments on the lower mantle phase perovskite and suggests that the mantle might have fractionated during the magma ocean stage of Earth accretion (Corgne et al. 2005). It has been suggested that this very early event may indicate that a primitive mafic to ultramafic crust was removed from the surface of the Earth to the D''-layer at the core-mantle boundary (see for example Tolstikhin et al. 2006).

In addition, other geochemical studies of early Archaean rocks suggest that the mantle also fractionated within a few hundred million years of planetary formation. Studies by Vervoort et al. (1994), Kamber et al. (2003) and Frei et al. (2004) on Archaean mafic and ultramafic rocks indicate that within the first few hundred million years of Earth history the mantle was differentiated into compositionally distinct domains, which persisted through the Archaean. These two different timescales of Hadean mantle evolution indicate that from almost the end of accretion the Earth's mantle has been a dynamic system. The record of these early processes may be preserved in samples of ancient mantle, which formed prior to the onset of 'modern' convective processes. Hence there is a need to identify those samples of the early Archaean mantle, which have remained chemically unmodified since they were first formed.

In this review paper, three geochemical criteria are developed for recognising early Archaean mantle peridotites. The focus here is on the earliest samples of possible Archaean mantle and so the emphasis is on peridotites found in rocks of about 3.8 Ga in age. The discriminants used are based upon the compositional relationships observed in geologically recent mantle samples, preserved either as mantle xenoliths or as tectonic slices in ophiolites and other orogenic settings. In this study, early Archaean mantle samples are positively identified on the basis of their

- major element chemistry as defined by the mantle array in a Mg/Si–Al/Si plot, from
- their rare earth element (REE) chemistry and
- on the basis of the composition of the chrome spinels present.

These criteria were selected because they are robust geochemical indicators of former mantle, and because they effectively discriminate between residual mantle and mantle melts. Isotopic criteria are not used here for two reasons. First, because the isotopic characterisation of early Archaean samples frequently produces ambiguous results. This arises from the uncertainty as to whether or not they have been metasomatised during emplacement or isotopically disturbed during later thermal events. Second, because if samples of early Archaean mantle can be identified with some certainty using other criteria, then isotopic results obtained on these samples can be accorded a higher degree of reliability.

On the basis of these criteria, claims made for early Archaean mantle are evaluated and possible mantle samples identified. The geochemical data used here are taken from a number of previously published studies.

Samples of early Archaean mantle

Possible samples of early Archaean (ca 3.8 Ga) mantle have been reported from Labrador (Collerson et al. 1991) and from the Itsaq gneisses in the area south of Isua, in west Greenland (Friend et al. 2002). In addition, this study also examines ultramafic rocks from the Isua greenstone belt, described by Dymek et al. (1988) and Frei and Jensen (2003).

Labrador

Collerson et al. (1991) and Wendt and Collerson (1999) reported ultramafic rocks from within early Archaean felsic gneisses in the Saglek–Hebron area of northern Labrador. They subdivided these into two suites. First, a group of spinel-bearing meta-peridotites and metapyroxenites, forming units 2–300-m thick and 1–2-km long, are thought to represent fragments of ancient residual mantle tectonically interleaved within the felsic gneisses. The interiors of these units are relatively massive and preserve what is thought to be an original compositional layering. A second suite of samples, comprising mostly pyroxenites, is associated with bands of metamorphosed mafic volcanic rocks up to 2 km long and 100 m wide. These are thought, on field and geochemical grounds, to be metamorphosed komatiites. Both suites are metamorphosed to amphibolite or granulite grade. Sm–Nd isochrons for the two suites yielded ages of 3.8 Ga for the residual mantle and 4.0 Ga for the komatiites.

South of Isua

Peridotites with ages >3.8 Ga have been described from the area south of the Isua Greenstone belt in the Itsaq gneisses of west Greenland (Friend et al. 2002; Nutman et al. 1996). These peridotites are located about 15 km south of the Isua Greenstone Belt where they form enclaves of ultramafic rocks up to 100 m long enclosed in the felsic gneisses of the Itsaq Gneiss complex. Typically the ultramafic rocks are spinel–dunite or harzburgite, although at their margins they are altered to amphibole-chlorite schists. Tonalitic veins cutting the ultramafic rocks contain zircons with ages of ca 3.8 Ga (Friend et al. 2002).

Friend et al. (2002) showed that there are two peridotite associations in this area. One peridotite suite forms part of a series of layered gabbro–harzburgite–dunite complexes and is closely associated with an amphibolite (metabasalt)-iron formation assemblage. The other, a dunite–harzburgite suite, is distinct from this assemblage and separated from it by a tectonic break. It is this latter suite that is thought to be residual mantle. These dunites and harzburgites are either massive or irregularly layered and cut through by olivine veins.

An excellent example of the layered gabbro–peridotite–dunite association is also found in this area, about 20 km SE of the Isua Greenstone Belt, and was recently described by Rollinson et al. (2002). This layered body is chromitiferous and shows well-defined chromite-rich layers in the dunites. Chromites from this body and from the residual mantle peridotites were used by Bennett et al. (2002) to constrain the Os-isotopic evolution of the mantle in the early Archaean. Lowry et al. (2003) studied the oxygen isotope composition of olivine in this layered peridotite complex ($\delta^{18}O = +4.49 - 4.89‰$) and noted a difference from that of olivine in a nearby dunite enclave ($\delta^{18}O = +5.25 - 5.27‰$). They pointed out that the latter value is close to the value of olivine in modern mantle peridotites apparently confirming the distinction between the two types of ultramafic association.

The Isua Greenstone Belt

Peridotites from the 3.7–3.8 Ga Isua Greenstone Belt were described by Dymek et al. (1988) and more recently by Frei and Jensen (2003). Although these authors made no claim that the Isua peridotites are fragments of Archaean mantle, their great antiquity and their uncertain origin make this a question worth asking and for this reason they are included in this study. The Isua peridotites contain primary olivine and orthopyroxene but are frequently altered either to tremolite-chlorite schists or serpentinite. One group of peridotites is associated with a thick basaltic pillow lava sequence, the 'garbenschiefer unit', and is probably cumulate in origin and so unlikely to be Archaean mantle. The remaining samples are examined here in order to see if they could be of mantle origin.

Characterising mantle peridotites

Defining the mantle array using major element chemistry

One of the criteria used here to recognise mantle samples is the major element chemistry expressed as the weight ratio of Mg/Si plotted against the weight ratio of Al/Si. This plot was first employed by Jagoutz et al. (1979) who showed that ultramafic mantle xenoliths define a trend of increasing Mg/Si with decreasing Al/Si. They showed that this 'mantle magmatic trend' contrasts with the trend for unfractionated meteorites, which define a 'cosmochemical trend' of increasing Mg/Si and increasing Al/Si. Jagoutz et al. (1979) suggested that the point of intersection of the two trends represents the composition of the bulk silicate Earth and hence the primitive mantle (Fig. 1a). This geochemical plot was also used by Collerson et al. (1991) and Friend et al. (2002) and effectively discriminates between rock compositions which represent fertile and depleted mantle and those rock compositions which represent former melts and cumulates from those melts. Melt compositions and cumulates plot to the right of and below the Jagoutz et al. (1979) mantle magmatic trend, whereas mantle compositions plot within the mantle array.

In this study, the mantle array is redefined and constrained with samples from two different sources. These include the following:

- *Mantle xenoliths.* Compositional data for almost 600 peridotite xenoliths, mostly from the subcontinental mantle and from Archaean to Recent in age (Pearson et al. 2003) define a scattered field, which broadly conforms to the trend of Jagoutz et al. (1979), (Fig. 1a). The main field of xenolith data points is well defined and the outlying analyses are thought to define the extremes of metasomatism. The main field has a curvilinear trend, shifting to high Mg/Si ratios at low Al/Si. Griffin et al. (2003) have shown that the average composition of peridotite xenoliths from the sub-continental lithospheric mantle varies with age and that these compositions define a trend of increasing Mg/Si and decreasing Al/Si with increasing age (Fig. 1a).
- *Mantle in ophiolite sequences and orogenic peridotites.* Harzburgites and dunites from the Oman ophiolite (Godard et al. 2000; Gerbert-Gaillard 2002) and orogenic lherzolites from the Alps (Lorand et al. 2000; Luguet et al. 2004) and lherzolites from the Horoman peridotite in Japan (Takazawa et al. 2000) lie within the field of mantle xenoliths (Fig. 1a). They too define a curvilinear trend such that the dunites increase in Mg/Si with little change in Al/Si. As a group the ophiolitic and orogenic peridotites show a more limited compositional range than mantle xenoliths, with fewer samples plotting away from the main compositional field. Within this subset of the data the orogenic peridotites have higher Al/Si ratios than the peridotites of the Oman ophiolite. A best-fit line for the ophiolitic and orogenic peridotite data is shown in Fig. 1a. This trend-line differs from that of Jagoutz trend in that it is curved, not straight and that at low Al/Si values (0.1–0.9) it plots at lower Mg/Si values.

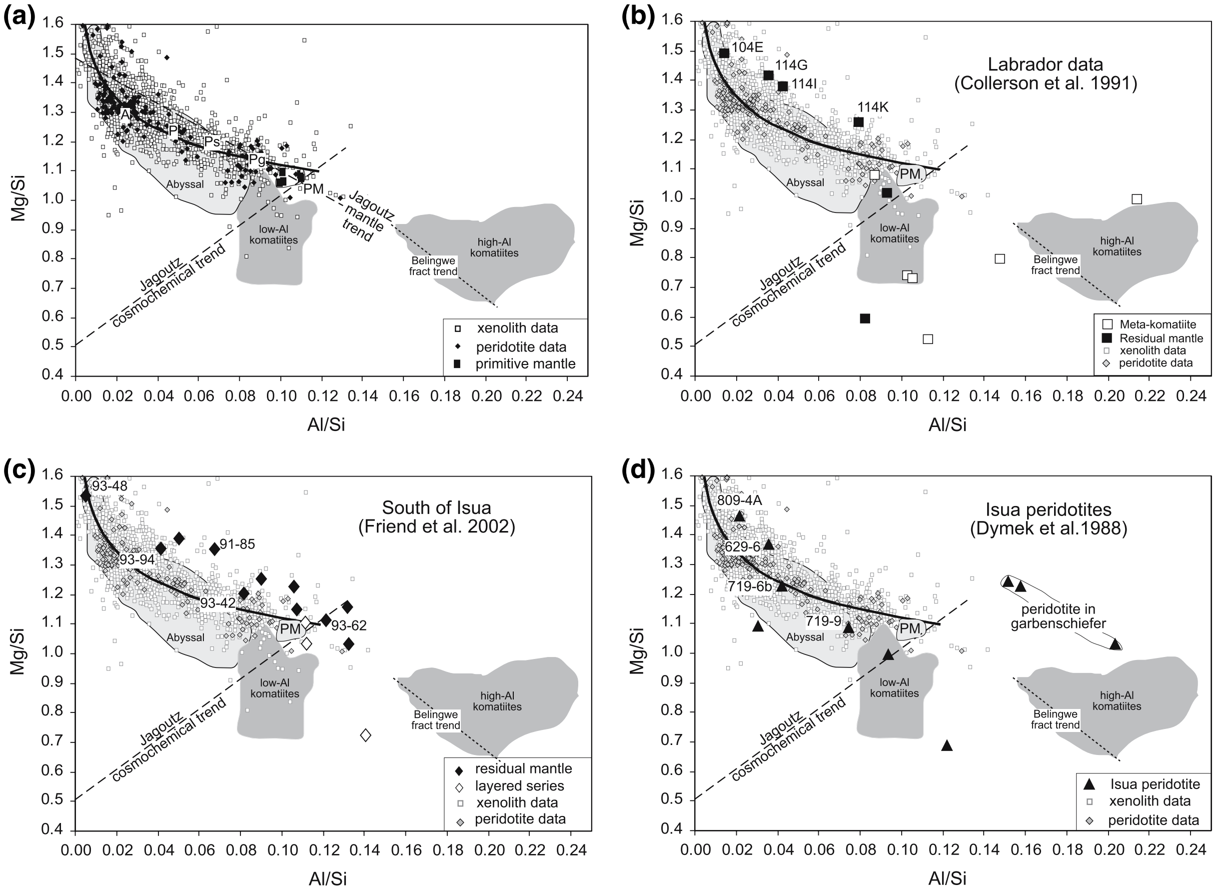

Fig. 1 Mg/Si versus Al/Si (wt%) plots for mantle rocks. **a** Data for orogenic and ophiolitic peridotites are from Lorand et al. (2000), Luguet et al. (2004), Takazawa et al. (2000), Godard et al. (2000) and Gerbert-Gaillard (2002), for abyssal peridotites from Niu (2004), Harvey et al. (2006) and for mantle xenoliths from Pearson et al. (2003). Also shown are the mantle and cosmochemical trends of Jagoutz et al. (1979), estimates of the composition of the primitive mantle [PM—from Jagoutz et al. 1979; Hart and Zindler 1986; Allegre et al. 1995; Ringwood 1991; McDonough and Sun 1995] and the fields for average subcontinental lithosphere of Archaean age (*A*), Proterozoic age (*P*), Phanerozoic age (spinel facies) (*Ps*), Phanerozoic age (garnet facies) (*Pg*) from Griffin et al. (2003). The high- and low-alumina komatiite data are from Parman et al. (2004) and Rollinson (1999) and the Belingwe komatiite trend from Nisbet et al. (1987). The *curved dark line* is a best-fit line to the orogenic and ophiolitic peridotite data points. **b** 3.8 Ga peridotites from Labrador (Collerson et al. 1991) plotted relative to the mantle and komatiite arrays shown in **a**. **c** 3.8 Ga peridotites from south of Isua (Friend et al. 2002) plotted relative to the mantle and komatiite arrays shown in **a**. Note this plot is different from that shown in Friend et al. (2002) because here the Mg/Si versus Al/Si values are calculated on a wt% basis. **d** 3.8 Ga peridotites from the Isua Greenstone Belt (Dymek et al. 1988) plotted relative to the mantle and komatiite arrays shown in **a**

Abyssal peridotites and the problem of serpentinisation

Also shown on Fig. 1a is the field for *abyssal peridotites.* These are mantle samples, which have been dredged or drilled from the ocean floor and are widely regarded as the melting residues of mid-ocean ridge basalts. Abyssal peridotites from the North Atlantic (Niu 2004; Harvey 2006) define a curvilinear field which is similar at low Al/Si ratios to that for mantle xenoliths and for ophiolitic dunites, but which at higher Al/Si ratios extend to lower Mg/Si ratios than are normally found in the xenolith field or in orogenic peridotites. Niu (2004) suggested that this is because the samples have lost MgO during alteration and for this reason abyssal peridotites are not used to define the mantle array.

The mechanism of MgO-loss in abyssal peridotite is not fully understood. It could be a product of sea-floor weathering following the serpentinisation of olivine (Niu 2004), whereby the peridotites lose MgO to seawater, giving them a low Mg/Si ratio. The rare earth element signature of abyssal peridotites also indicates that these rocks have experienced alteration for they show both positive and negative Ce and Eu anomalies. Given that these two elements have more than one oxidation state this variability may reasonably attributed to later alteration. However, the olivine breakdown hypothesis is inconsistent with the observation that in Mg/Si versus Al/Si space the greatest discrepancy between abyssal peridotites and the ‘unaltered mantle array’ is for the lower Mg/Si harzburgites and lherzolites—not the olivine-rich dunites. Furthermore, in the Labrador data set there is no correlation between the degree of serpentinisation and Mg/Si ratio, although this serpentinisation may be late and a retrograde phenomenon, following metamorphism.

Here, very few of the early Archaean peridotite samples examined plot in the low Mg/Si area of the abyssal peridotite field, rather they plot with high Al/Si ratios, above the mantle trend (see the ''Discussion''below). For this reason, it is argued that major element mobility through sea-floor alteration and serpentinisation is not a serious problem in these early Archaean peridotites.

Comparison between suboceanic and subcontinental mantle

Many years ago Boyd (1989) pointed out that there are geochemical differences between suboceanic and subcontinental mantle. In contrast, the observations discussed here show that in Mg/Si–Al/Si space, subcontinental mantle xenoliths, and samples of the suboceanic mantle represented by ophiolitic and orogenic peridotites have broadly the same major element compositions (although there is a greater compositional scatter in the xenolith data). Hence both suboceanic and subcontinental mantle samples can be used here to define the mantle array on an Mg/Si–Al/Si diagram and used in Figs. 1b–d to evaluate the mantle origin of the early Archaean samples.

The meaning of the mantle array

The spread of compositions within the mantle Mg/Si versus Al/Si array is thought to reflect the progressive extraction of melt from the primitive mantle, so that samples with low Al/Si are those, which are the most melt depleted. Average MORB plots along the extension of the mantle array, in the melt field at ca. Al/Si = 0.36, Mg/Si = 0.26. However, mantle xenoliths also record the process of metasomatism or melt enrichment, in which previously depleted mantle is refertilised with melt. This process is also seen in orogenic, ophiolitic and abyssal peridotites and has the effect that depleted bulk compositions move back towards the primitive mantle composition. In the Oman data set refertilised clinopyroxene harzburgites plot on the mantle trend but with higher Al/Si and lower Mg/Si ratios, whereas melt-impregnated dunites plot to the right of the mantle trend with high Al/Si ratios and slightly higher Mg/Si ratios (Godard et al. 2000; Gerbert-Gaillard 2002). The mantle array defined here does not conform exactly to the Jagoutz trend, but lies slightly below it. Further, as already noted, it is curvilinear in form, shifting to higher Mg/Si ratios at low Al/Si values. At pressures of 10 kb this process probably reflects the exhaustion of clinopyroxene and then orthopyroxene during peridotite melting. This view is supported by data from experimental studies of peridotite residues, progressively depleted during partial melting (eg. Wasylenki et al. 2003). In Mg/Si versus Al/Si space these residues plot very close to the best-fit line for the peridotite data shown in Fig. 1a. At lower pressures the curvature could be due to olivine crystallisation and orthopyroxene dissolution during melt-rock reaction, as described by Kelemen et al. (1995).

It was shown above that the modern mantle is compositionally variable because it records multiple magmatic events. Here it is argued that there is no a priori reason why early Archaean mantle should be any simpler. For this reason the mantle compositions in Fig. 1a are thought to be a realistic measure against which to assess putative early Archaean mantle compositions.

The komatiite field

It has already been noted that some early Archaean ultramafic rocks might be komatiitic in origin. For this reason the fields for Archaean komatiites are also plotted in Fig. 1. Low-alumina Barberton type komatiites (Parman et al. 2004) define a compositional field with a similar Al/Si ratio to that of the primitive mantle, but with a lower Mg/Si ratio. In contrast high-alumina, Munro-type komatiites from the Sula Mountains greenstone belt (Rollinson 1999) define a field with high Al/Si and low Mg/Si. Similarly the very fresh Belingwe komatiites (Nisbet et al. 1987) also fall in this field although with lower Al/Si ratios, indicating that the Sula Mountains samples experienced some element mobility (Rollinson 1999). The compositional differences between low and high alumina komatiites relative to the primitive mantle as seen on the Mg/Si–Al/Si diagram, imply very different mantle-melting processes.

Rare earth element geochemistry

REE data for abyssal and orogenic peridotites and for harzburgites and dunites from the Oman ophiolite, the Mariana forearc and the Horoman peridotite have been compiled from Bodinier and Godard (2003), Godard et al. (2000), Gerbert-Gaillard (2002), Niu (2004), Parkinson and Pearce (1998) and Takazawa et al. (2000). Chondrite-normalised Ce/Sm ratios are plotted against Yb for the different types of peridotite in Fig. 2 and individual REE patterns are shown for refractory orogenic peridotites (after Bodinier and Godard 2003) in Fig. 3. All samples have been normalised to the chondritic values of Sun and McDonough (1989).

The REE compositions of mantle peridotites may record more than one geological process. Hence in their review Bodinier and Godard (2003) subdivide mantle peridotites

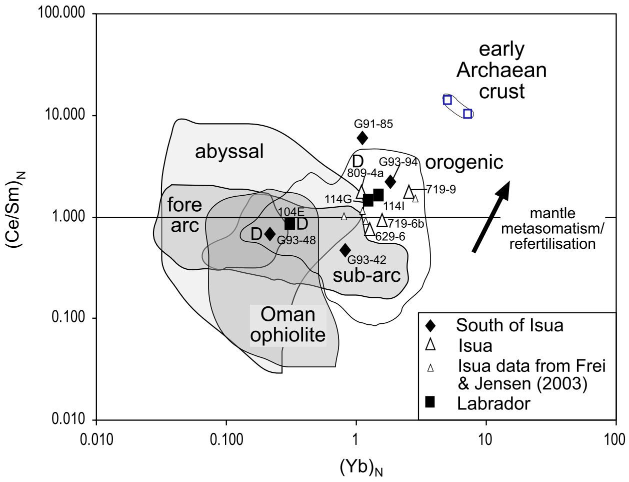

Fig. 2 (Ce/Sm)$_n$ versus Yb$_n$ for peridotites from South of Isua (Friend et al. 2002), Isua (Dymek et al. 1988; Frei and Jensen 2003) and Labrador (Collerson et al. 1991), plotted relative to the fields of mantle peridotites for the Oman ophiolite (Godard et al. 2000; Gerbert-Gaillard 2002), orogenic peridotites (Bodinier and Godard 2003), abyssal peridotites (Niu 2004), sub-arc peridotites from the Horoman peridotite (Takazawa et al. 2000) and forearc peridotites from the Mariana arc (Parkinson and Pearce 1998). Mantle metasomatism and refertilisation tends to produce an increase in (Ce/Sm)$_n$ and Yb$_n$. Metasomatism by the enclosing felsic gneisses has a similar effect and peridotites with a high Ce/Sm ratio may be formed by mixing between depleted mantle peridotite and early Archaean crust (data from Kamber et al. 2002; Condie 2005). *D* dunite

into those which are refractory, those which are fertile and those which have been refertilised or metasomatised. Each of these processes may be recorded in the different types of mantle peridotite. Refractory peridotites typically have low absolute REE concentrations and so will plot to low Yb$_{(n)}$ values. Mantle metasomatism and refertilisation have the effect of increasing the absolute REE concentrations and of increasing the Ce/Sm ratio (Fig. 2). However, some caution is needed here, for in early Archaean peridotite xenoliths enclosed in felsic gneisses, a similar pattern of enrichment may be produced by the post-emplacement metasomatism of the peridotite by fluids from the felsic gneisses. Figure 2 shows two estimates of average early Archaean felsic crust and illustrates how elevated Ce/Sm ratios may be obtained through the mixing of felsic crust and 'depleted' mantle peridotite.

Chromite chemistry

Chromite is refractory phase common in mantle peridotites. In metamorphosed ultramafic rocks chromite grains may preserve, in their cores, the composition of the original, primary magmatic chromite. Here chromite compositions are reported in terms of their Cr/(Cr + Al) ratio—the cr#, and their Fe(II)/(Fe(II) + Mg) ratio—the fe#. On a cr#–fe# diagram the composition of the modern mantle array has been defined by three suites of mantle peridotites (Fig. 4). These are chromites from 'fertile' lherzolitic mantle xenoliths (Chazot et al. 1997), chromites from depleted mantle harzburgite from the Oman ophiolite (Le Mee et al. 2004) and chromites from mantle chromitites, also from the Oman ophiolite (Rollinson 2005). These samples define an array from ca. cr# = 0.15, fe# = 0.2 to ca. cr# = 0.8, fe# = 0.4. Hellebrand et al. (2001) proposed an algorithm for mantle harzburgites which relates the cr# of chromites

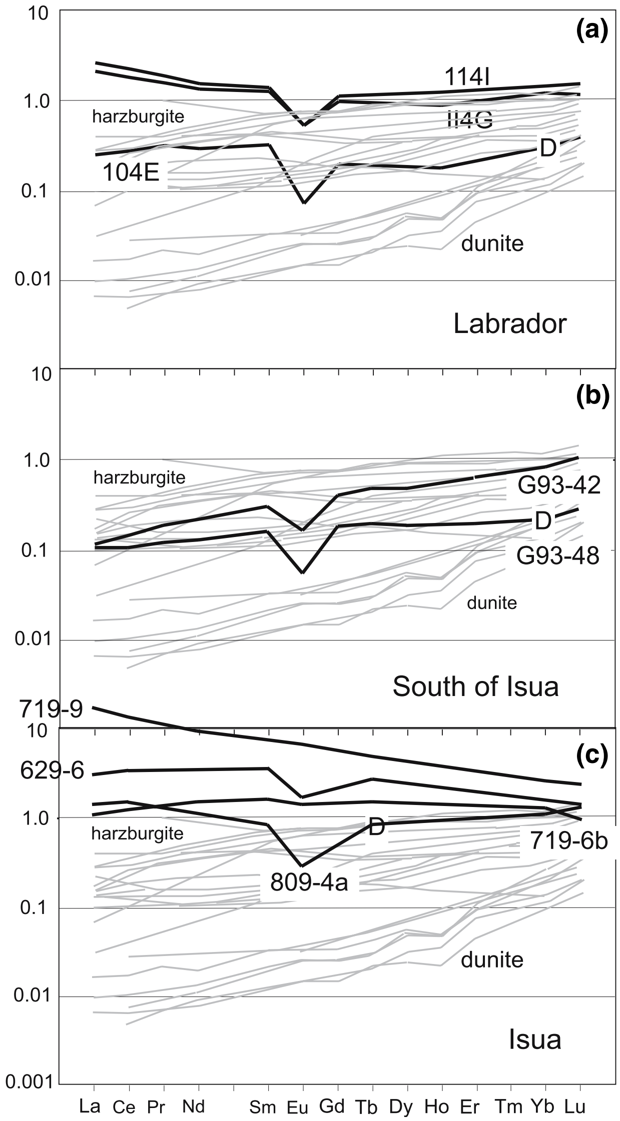

Fig. 3 REE patterns for those peridotites, which satisfy the major element mantle criteria plotted relative to refractory orogenic harzburgites and dunites (*grey lines*—after Bodinier and Godard 2003). The variability in composition within the orogenic peridotites (*grey lines*) is attributed to different degrees of melt extraction (low absolute values) and refertilisation (negative curvature in the light REE); **a** Labrador peridotites (Collerson et al. 1991); **b** peridotites from south of Isua (Friend et al. 2002); **c** Isua peridotites (Dymek et al. 1988). *D* dunite

to the % melt extraction from the harzburgite, assuming pure fractional melting. Chromite compositions in the Oman mantle harzburgites imply between 8% (cr# = 0.20) and 21% (cr# = 0.72) melt extraction (Rollinson 2005).

The difference in fe# between the lherzolite and harzburgite suites indicates that during melt extraction mantle chromite becomes more Fe-rich. The difference in composition between chromites from mantle chromitites and harzburgites shows that the chromitites are not simply residues from harzburgite melting but are produced by a process in which they become enriched in Mg relative to the harzburgitic chromites. This observation lends support to the hypothesis that mantle chromitites were produced by a melt migration—melt reaction process within the mantle (Rollinson 2005).

Recognising early Archaean mantle

In this section the criteria developed above are applied to 3.8 Ga peridotites from Labrador, south of Isua and the Isua greenstone belt. The geochemical data used here are taken from previously published studies on the samples discussed.

Labrador

Of the six residual mantle samples reported by Collerson et al. (1991) only four plot within or close to the mantle array. Of these three plot away from the peridotite field at the margin of the xenolith field (Fig. 1b). All plot on the high Al/Si side of the array. However, one of these four samples (114K) has 6 wt% Al_2O_3 and so is unlikely to be unmodified mantle. Of the meta-komatiite samples reported by Collerson et al. (1991), five plot outside the mantle array and one within it. Of the three peridotites which might be early Archaean mantle two (114I and 114G) are light-REE enriched and have $(Ce/Sm)_n > 1.0$ (Figs. 2, 3a) and plot outside the fields of ophiolitic, subarc and abyssal peridotites. They do however plot within the field of orogenic peridotites, although in an ambiguous region in which samples may be either the products of melt metasomatism or crustal contamination (Fig. 2). All have negative Eu-anomalies (Fig. 3a), a feature which is most commonly found in abyssal peridotites, indicating that these samples could have been abyssal peridotites. Although caution is required here for Eu is potentially mobile in during alteration and so no great significance can be attached to Eu anomalies. In any case two of the three samples plot outside the abyssal peridotite field (Fig. 2) and none of the samples has a negative Ce anomaly also typical of abyssal peridotites (Niu 2004). Sample 104E plots as a dunite in Fig. 1b and lies within the fields of orogenic, abyssal and arc peridotites in Fig. 2. However, it also has a high $(Ce/Sm)_n$ ratio, higher than that expected in a dunite (Fig. 3a). Given that these samples are known to have a disturbed Nd isotope signature (Wendt and Collerson 1999), it is possible that the light-REE enriched nature of these samples is due to element mobility after their emplacement. Hence it is unsafe to infer that the light-REE

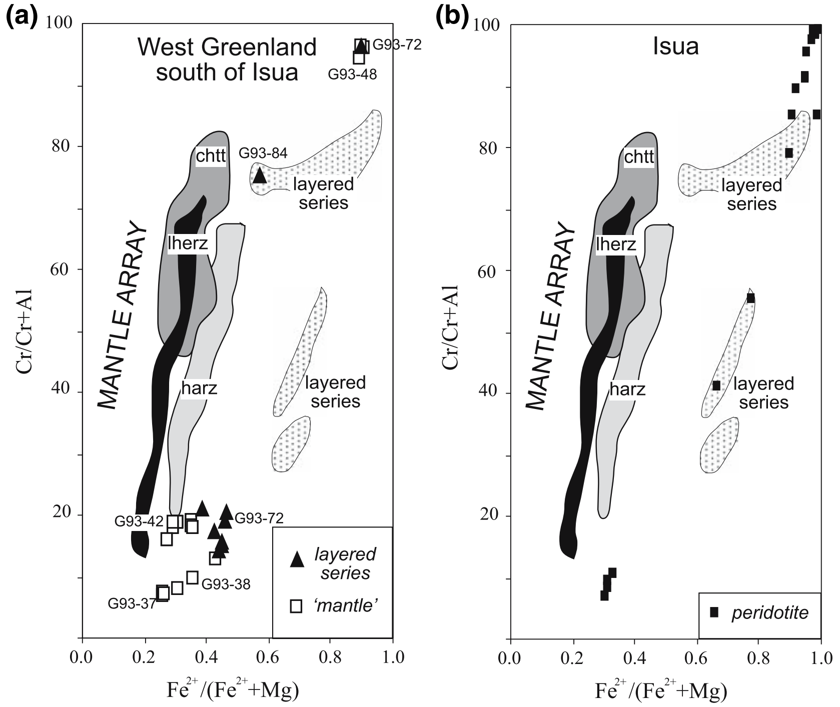

Fig. 4 cr#–fe# plot for chromites in early Archaean peridotites compared to mantle chromite compositions. The mantle array is defined by chromites from Oman harzburgites (harz—Le Mee et al. 2004), fertile mantle lherzolites (lherz—Chazot et al. 1997) and mantle chromitites from the Oman ophiolite (chtt—Rollinson 2005). **a** Samples from the area south of Isua from Nutman et al. (1996) and Friend (2002) compared to the mantle array and the composition of chromites in early Archaean layered gabbro-peridotites (Rollinson et al. 2002); **b** chromites from the Isua Greenstone Belt compared to the mantle array and the early Archaean layered series

enrichment observed in these peridotites is primary. There are no published chromite data for the Labrador samples and so this discriminant cannot be used.

South of Isua

Of the 11 samples ascribed to early Archaean mantle by Friend et al. (2002) all lie to the high Al/Si side of the mantle array. Three samples plot to the right of the primitive mantle. Of the remaining eight samples, four lie close to the peridotite array and four more at the margin of the xenolith field. Only sample 93-48 plots in the abyssal peridotite field. Two members of the layered series also plot close to the mantle trend and one does not, showing the importance of using field observations as a discriminant in addition to geochemistry. Of the eight samples, which appear, on the basis of their major element chemistry, to be mantle peridotites, there are REE data for four. Of these, two are strongly light-REE enriched, samples G91-85 and G93-94. Sample G91-85 plots outside the field of mantle peridotites (Fig. 2). The other, sample G93-94 contains 5% phlogopite, a mineral which can contain high levels of REEs and although phlogopite is known in mantle peridotites, in this terrane it is present as a late alteration mineral and unlikely to be primary (Rollinson et al. 2002). For this reason it is thought that peridotite G93-94 has been enriched in light-REE after its emplacement into the gneisses. In contrast, the two samples G93-42 and G93-48 have $(Ce/Sm)_n$ ratios less than 1.0 more typical of refractory orogenic or ophiolitic peridotites (Fig. 3).

A mantle origin for harzburgite G93-42 is supported by its spinel composition. This is the only sample of those reported by Nutman (1996) and Friend et al. (2002), which plots within the mantle chromite array (Fig. 4a). Figure 4a also shows that chromites from peridotites associated with a layered ultramafic intrusion from the same area plot in a different part of the diagram and are much more Fe-rich (Rollinson et al. 2002). This means that it is easy to discriminate on a cr# versus fe# diagram between chromites from peridotites and dunites in the layered series and those which are from mantle peridotites. In fact some of the other samples reported by Nutman (1996) and Friend et al. (2002) plot as the low cr# extension of this layered series and so are probably not mantle peridotite (Fig. 4a). Chromite in sample G93-48 is highly oxidised, probably as a result of secondary processes, and plots as a ferrit–chromite (Fig. 4a)

Isua

Four of the ten Isua peridotites described by Dymek et al. (1988) plot close to the mantle trend. The three samples, which are associated with the garbenschiefer (pillow lava) sequence plot in the melt field away from the mantle trend (Fig. 1d). Of the four peridotites plotting close to the mantle trend, all have relatively high REE abundances with Yb_n >1.0; two samples have $(Ce/Sm)_n$ ratios >1.0 (Figs. 2, 3c), suggesting that they have been metasomatised. Ultramafic rocks from the western part of the Isua belt with between 33 and 42 wt% MgO analysed by Frei and Jensen (2003) show a similar range of REE patterns (Fig. 2). However, these authors found that ε_{Nd} values for these rocks at 3.8 Ga varied between +3.9 and +10.6, implying that the Nd isotopic system and the REE systematics were disturbed by one or more metamorphic events which took place after the crystallisation of the Isua belt (Frei and Jensen 2003). Spinels from these rocks were analysed by Dymek et al. (1988) and plot on a cr#–fe# diagram in the same compositional range as that of the layered series defined from south of Isua (Fig. 4b). There is no evidence from these compositions that any of these chromite-spinels formed in mantle peridotites, as already inferred from the REE data. For this reason, it is not possible to infer an mantle origin for any of the ultramafic rocks in the Isua greenstone belt, rather the spinel data would suggest that they are analogous to the layered intrusions found south of Isua.

Summary

The application of geochemical discriminants to early Archaean peridotites shows that neither the peridotites from Labrador described by Collerson et al. (1991), nor the peridotites from the Isua greenstone belt can be unambiguously identified as early Archaean mantle.

A particular feature of the early Archaean samples discussed here is that most plot to the high Al/Si side of the mantle trend. There are a number of possible explanations. One is that the peridotites originally contained a small fraction of aluminous melt. However, as noted earlier the effect of melt metasomatism in harzburgites is simply to move the melt composition along the mantle trend, not away from the trend; only the metasomatism of dunites moves melts to compositions with higher Al/Si. This however is at high Mg/Si ratios and atypical of the Archaean samples studied here. A second possibility is that the peridotites are residues of deep mantle melting in which unmelted garnet was present in the residue. This hypothesis was tested by calculating the composition of melting residues containing garnet, formed between 5 and 7 GPa, from the experimental study of Walter (1998). These residues do resemble the aluminous sample compositions described in this study when plotted in Mg/Si–Al/Si space. However, they do not reproduce the most magnesian compositions found in the early Archaean peridotites and even moderately magnesian compositions reflect a very high degree of melt extraction (40–50 wt% melt) inconsistent with their trace element chemistry. For this reason the preferred explanation for the high Al/Si ratios in these samples is chemical alteration, probably during metamorphism. Samples from south of Isua contain traces of the aluminous phases chlorite, phlogopite and amphibole and some of the Labrador samples contain significant amounts of clinopyroxene (up to 32% in one sample), more than is typical in either primary lherzolite or refertilised harzburgite.

Of the harzburgites and dunites from the area south of Isua described by Friend et al. (2002) and Bennett (2002), only two samples, harzburgite G93-42 and dunite G93-48 have the geochemical characteristics of unmodified mantle peridotite, which has not undergone metasomatism. Both have the REE character either of refractory orogenic peridotite, abyssal peridotite or sub-arc peridotite and the chromite in harzburgite G93-42 has a mantle composition. This study therefore supports the findings of Friend et al. (2002), who argued that these peridotites were samples of the early Archaean mantle. It should be emphasised however, that the two samples identified here, as probable Archaean mantle are not primitive mantle, for both have experienced melt extraction. However, these rocks are probably not abyssal peridotites as originally claimed by Friend et al. (2002) for they have neither a major element chemistry similar to that of peridotites dredged from the ocean floor nor do they show the negative Ce-anomaly in their REE pattern typical of modern abyssal peridotites. Furthermore, some of the samples reported by Friend et al. (2002) as mantle appear to have been metasomatised by the enclosing felsic gneisses so that their original chemistry is disturbed. Others contain chromites, which suggest that they belong to related layered ultramafic intrusions.

Discussion

It might be argued that the search for samples of the Earth's early mantle is relatively unimportant since we can access the early mantle through the proxies of mantle-derived melts and their associated cumulates. Whilst this argument is true for isotope ratios, it does not hold good for studies, which seek to determine element concentrations in the early mantle. Determining element concentrations in the early mantle and in its mineral components is important for it allows estimates to be made of melt extraction and re-fertilisation. It also allows the calculation of the equilibration conditions of early mantle and in the case of the highly siderophile elements they may have a bearing on the separation of the core. Thus, in this study a small, but more robust sample set of early Archaean mantle has been identified from the work of previous authors and these samples are now used to evaluate the character of the Earth's mantle at 3.8 Ga.

Comparison with Archaean sub-continental lithospheric mantle

Friend et al. (2002) showed that samples of early Archaean mantle from south of Isua were different in composition from samples of Archaean sub-continental lithospheric mantle (SCLM) from beneath Archaean cratons. This conclusion was based upon the olivine-spinel mantle array (OSMA) chromite-cr# versus olivine-mg# diagram of Arai (1994). However, it was shown above that Cr–spinel compositions in the mantle peridotites from south of Isua are very variable and not all are of mantle origin, and so the use of the OSMA diagram is potentially misleading. For this reason the data for harzburgite G93-42 and dunite G93-48 are plotted on the 'percentage modal olivine versus mg# of olivine' diagram of Boyd (1989) (Fig. 5). It is clear that the Greenland samples plot with lower mg# and in a different field from samples from the Archaean SCLM, supporting the original claim of Friend et al. (2002). For sample G93-42 this difference is also seen on an OSMA plot (not shown), (Arai 1994) where the sample plots away from the SCLM array. However, both samples plot on or very close to the mantle peridotite Mg/Si–Al/Si trend as defined here, as does the average Archaean SCLM composition as calculated by Griffin et al. (2003). A possible explanation of these relationships is that the early Archaean mantle was more Fe-rich than the modern mantle—a fea-

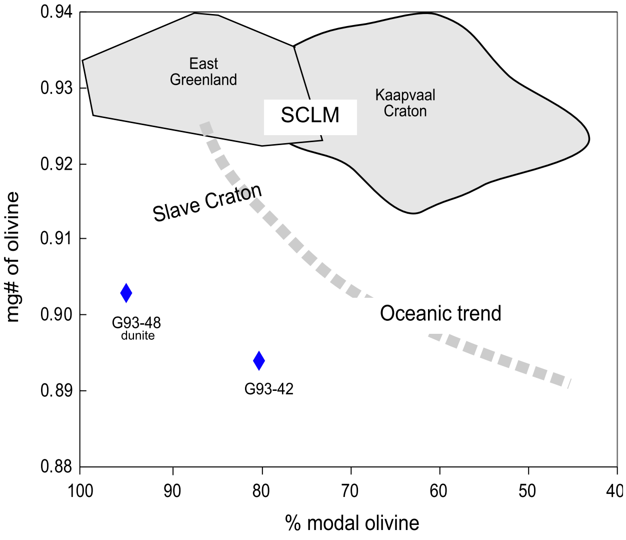

Fig. 5 Unmodified early Archaean mantle from south of Isua plotted on a 'Boyd' mantle plot showing their distinctive compositions relative to subcontinental lithosphere in the East Greenland, Kaapvaal and Slave cratons and the oceanic lithosphere [data from Berstein et al. (2006) and Boyd (1989)]

ture, which would be consistent with the more Fe-rich nature of average Archaean tholeiites as noted by Arndt et al. (1997).

Although the Greenland sample-set is small this result is important for it indicates that the mantle preserved south of Isua is of a different composition—principally lower in SiO_2 and higher in FeO—and maybe of a different type, from that preserved in the Archaean SCLM. Furthermore, if the algorithm of Hellebrand et al. (2001) is used linking the composition of chromite in mantle harzburgite to the degree of melt extraction experienced by the harzburgite, then chromites from harzburgite sample G93-42 from south of Isua have compositions which indicate that the host harzburgite experienced between 6 and 7% melt extraction. Chromite compositions in the harzburgites of the Oman ophiolite vary between cr# 21 and 67 and this ratio correlates inversely with Yb concentrations—a measure of melt depletion in the host harzburgite. Thus the low cr# in chromite in G93-42 is consistent with a relatively small amount of melt extraction from the harzburgite. A low degree of melting is also consistent with the relatively high Al_2O_3 content in the harzburgite (3.2 wt%) relative to 4.0–4.5 wt% in PUM (Becker et al. 2006) and is in contrast to the much higher levels of melt extraction normally proposed for Archaean sub-continental mantle (Boyd 1989). On the other hand the dunitic nature of sample G93-48, for which there is no good chromite data, would imply a very high degree of melt extraction.

The application of the Balhaus et al. (1991) olivine–orthopyroxene–chromite oxygen barometer to harzburgite G93-42 indicates that it equilibrated at 3 log units above the QFM buffer. This is higher than most estimates for the Archaean mantle, which tends to be QFM ±2 log units (see compilation in Rollinson 2007), and implies more oxidising conditions for these mantle samples.

Mantle Os concentrations

For many years our thinking about Os abundances in the Earth's mantle has been governed by what has been identified as the 'siderophile element excess' problem. Briefly stated, the highly siderophile elements, of which Os is one, should have partitioned into the Earth's core during core formation. However, when this effect is calculated there is a mismatch between actual mantle concentrations (which are too high) and calculated concentrations. Hence a siderophile element excess in the mantle. Solutions to the siderophile element excess problem include the notion that siderophile elements were added to the Earth, as a 'late veneer' and were mixed into the mantle after the formation of the core. Alternatively, it is possible that the partition coefficients used for the partitioning of the high siderophile elements between silicate and metal were wrong and that partition coefficients determined at very high pressures are more appropriate. Both hypotheses have their merits.

Bennett et al. (2002) measured Os concentrations in the early Archaean mantle peridotites from south of Isua and argued that Os concentrations in 3.8 Ga mantle are the same as those in modern spinel peridotites (about 2.6 ± 1.0 ppb). From this they inferred that any 'late veneer' of siderophile elements was added to the Earth before 3.8 Ga. Two factors have changed since the study of Bennett et al. (2002). The first is that the results of this study provide a more robust estimate of the Os concentration of 3.8 Ga peridotites from south of Isua than was used by Bennett et al. (2002). Here it is argued that only one sample from the studies of Bennett et al. (2002) and Friend et al. (2002) is a suitable candidate for undepleted mantle peridotite—sample G93-42 which contains 2.05 ± 0.09 ppb Os, (3.2 wt% Al_2O_3). The second factor is that there is now a revised estimate of Os in the Primitive Upper Mantle (PUM) =3.9 ± 0.5 ppb (4.0–4.5 wt% Al_2O_3), (Becker et al. 2006). There are also differences in the Re/Os ratio. In sample G93-42 the ratio is between 0.0043 and 0.0046 (Bennett et al. 2002), whereas in PUM the ratio is 0.090 ± 0.002 (Becker et al. 2006). It is possible that the differences between early Archaean mantle and PUM are analytical and are a function of the dissolution method used prior to Os analysis. Alternatively, there is a real difference between the composition of the early Archaean mantle and the modern mantle with respect to Os and its Re/Os ratio.

Conclusions

1. The principal finding of this study is that the area south of Isua in west Greenland contains fragments of early Archaean (3.8 Ga) mantle, which have not experienced mantle metasomatism. This mantle is not primitive mantle for it is compositionally distinct from estimates of the primitive mantle indicating that it has experience melt depletion. This finding supports the earlier claims of Bennett et al. (2002) and Friend et al. (2002), but the application of more robust criteria for recognising rocks of mantle origin means that only two samples, from their larger sample-set, can be reliably identified as early Archaean mantle.
2. Given the criteria for recognising mantle rocks established in this study, the area south of Isua is, at present, the only locality in the world from which early Archaean mantle is known. For this reason it should be investigated more thoroughly.
3. This study also confirms the observation by Friend et al. (2002) that early Archaean mantle from south of Isua appears to be of a different character from Ar-

chaean mantle from the subcontinental lithosphere. Calculations presented here show that at least one of the two mantle fragments from south of Isua experienced a lower degree of melt extraction and equilibrated under more oxidising conditions than early Archaean mantle preserved in the subcontinental lithosphere.

4. Elemental concentrations of Os in early Archaean mantle are lower than the new estimate for the primitive upper mantle of Becker et al. (2006). Further work needs to be carried out in this area to establish whether this is the product of different analytical techniques, or whether there is a geological explanation.

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