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# Stable isotope genealogy of meteorites

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One of the oldest problems in meteoritics is that of taxonomically grouping samples. In recent years the use of isotopes, particularly oxygen isotopes has proved very successful in this respect. Other light-element systematics potentially can perform the same function. For example, nitrogen in iron meteorites, and nitrogen and carbon in ureilites and SNC meteorites. These measurements will serve to extend and augment existing classification schemes and provide clues to the nature of meteorite parent bodies. They can also aid in the recognition of the isotopic signatures relating to inaccessible regions of the Earth.

## Introduction

Light-element and noble-gas isotopes provide a wealth of information relevant to the formation and evolution of the Solar System. Isotopic anomalies (these are usually exotic isotopic signatures that cannot be explained by Solar System processes) in meteorites demonstrate that dust of interstellar origin has survived the events involved in the collapse of the solar nebular and can be used to recognize the nucleosynthetic environments in which the grains condensed (Swart *et al.* 1983*a*; Anders 1988). Clearly, however, the vast majority of the pre-existing material was reasonably well mixed during condensation in the solar nebula but fortunately not so well mixed that patterns in isotopic composition leftover, or subsequently introduced by fractionation, cannot be used to recognize genetic links between meteorites and allow predictions concerning the parent bodies of meteorite families. This subject, which I propose to call stable isotope genealogy, has foundations based on oxygen-isotopic determinations (see Clayton, this symposium, for a review and Pillinger (1984)). In this paper I explore the potential of stable isotope genealogy using examples involving nitrogen and carbon and to a lesser extent, noble-gas isotope measurements on iron meteorites, urelites and SNC meteorites; the problem of the carbon isotopic composition of the Earth's mantle is also discussed.

## Nitrogen isotope studies of iron meteorites

A simple view of iron meteorites is that these objects could be the cores of differentiated, but now disintegrated, small parent bodies (Scott 1972). However, not all irons can have had a magmatic origin. Trace-element (Ga, Ge, etc.) data which have been most successfully used in classifying iron meteorites into at least sixteen groups (for a review see Dodd 1981) suggest that four, possibly five, of these were formed non-magmatically, possibly by a process that mobilized the metal phase in a chondritic megaregolith (Wasson *et al.* 1980). The existence of isotopically anomalous nitrogen in the metal of the unusual polymict breccia Bencubbin (Prombo & Clayton 1985; Franchi *et al.* 1986) suggests that at least some iron in the Solar System is either a primordial condensate (seeded by interstellar grains?) or is produced by impact melting in a planetary regolith and may therefore contain remnants of the projectile which was the source of the exotic nitrogen.

Including Bencubbin, the martian atmosphere (Nier & McElroy 1977; Owen *et al.* 1977), Cδ (Ash *et al.* 1987) and solar-wind implanted species (L. P. Carr *et al.* 1985), the range of nitrogen isotope compositions in Solar System materials gives rise to $\delta^{15}$N values from −375 to +1035‰. This tremendous scale implies that there is considerable scope for matching compositions and recognizing genetic relations. Bulk $\delta^{15}$N measurements on iron meteorites alone reveal a spread of *ca.* 250‰ from −96 to +153‰ (Prombo & Clayton 1983; Franchi *et al.* 1988), which is greater than for any other group. Irons must therefore play host to several different kinds of nitrogen.

To evaluate the use of nitrogen abundance and isotopic composition in the isotope genealogy of iron meteorites, Franchi *et al.* (1988) have used a data set of 46 samples, with measurements made in my laboratory together with those acquired by Prombo & Clayton (1983). There is a major difference in technique between the two groups of analysts in that Franchi *et al.* (1988) use much smaller samples but for meteorites investigated in both laboratories, there is relatively good agreement. Analyses of 17 specimens from the groups IC, IIAB, IIIAB and IIIE (defined by trace elements) display a range of $\delta^{15}$N values from −96 to −76‰ with concentrations of 5–37 p.p.m. N (figure 1). For 13 members of groups IAB and IIICD, the two main non-magmatic groups (Wasson *et al.* 1980), $\delta^{15}$N values are much more tightly constrained between −66 and −60‰ but concentrations (from 8 to 85 p.p.m.) are marginally more variable. The remaining trace element defined groups, excepting IIC, but including the non-magmatic candidate IIE, have $\delta^{15}$N values of between −16 and +47‰ for relatively low N abundance, 0.4–28 p.p.m. Group IIC has a distinct composition, three analyses having $\delta^{15}$N values from

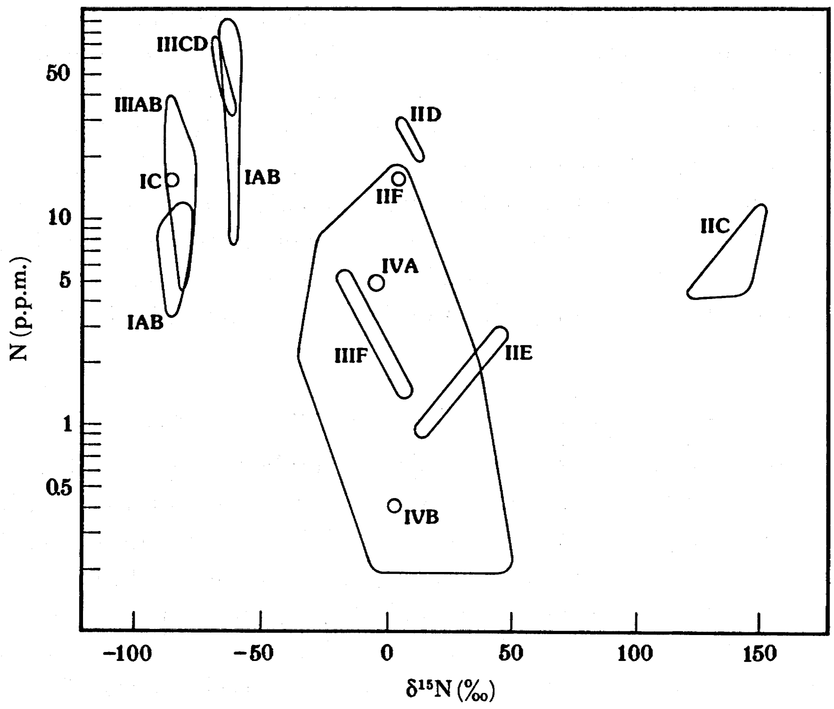

FIGURE 1. Nitrogen abundance against $\delta^{15}$N for several groups of iron meteorites.

+125 to +153‰ for between 4 and 10 p.p.m.; this is particularly interesting because IIC is not well resolved from other groups on the basis of trace elements. Similarly, it is noteworthy that limited $\delta^{15}$N data on pallasites (not in figure) seem to fall outside the range measured for IIIAB to which it is linked by Ge, Ga measurements.

The initial impression is that, from nitrogen measurements, iron meteorites fall into four distinct families, however, an expanded dataset, especially for the less common groups, may change this picture substantially. Additionally, the high-sensitivity methods used by Franchi *et al.* (1988) will permit the in depth study of the nitrogen carriers. Groups not distinguished by bulk measurements could be recognized by the distribution and identity of host phases. Within a particular group, nitrogen, being a volatile, may exhibit fractionation trends which provide information about evolution of the parent body, for example, impact and melting processes implicated in the formation of non-magmatic metal. It will be interesting to see whether nitrogen isotope genealogy can be successful in categorizing any of the 15% of iron meteorites which are unclassified by other trace-element methods or in providing links to chondrites or other stony meteorites through the analysis of metal fragments contained therein.

## UREILITES: MIXED OR MORE THAN ONE PARENT BODY?

The ureilites are a most unusual group of achondrite meteorites composed principally of olivine and clinopyroxene but distinguished by diamond-bearing carbonaceous veins, rich in metal, siderophile elements and primordial noble gases. The number of samples available for study has been doubled in recent years by about ten finds made on the Antarctic ice sheet; most specimens so far collected from this new resource seem to represent individual falls rather than part of a shower (Takeda 1987). Two schools of thought have developed concerning the origin of the ureilites: one (e.g. Binz *et al.* 1975) would have them as a melt residue with the veins injected following an impacted event; the other (e.g. Berkley & Jones 1982; Janssens *et al.* 1988) sees them as an ultramafic cumulate formed from a carbon-rich iron melt, so that the veins are indigenous.

Because of the abundance of carbon in ureilites almost all known specimens have been the subject of carbon abundance and isotope measurements. These are being augmented slowly by nitrogen isotope determinations. Because the vast majority of the samples involved are finds, analytical techniques to distinguish terrestrial contamination from the indigenous species are used (Swart *et al.* 1983*b*). Although within any given ureilite, graphite and diamond, where it occurs, has variable combustion properties, which are not yet understood, the $\delta^{13}$C values obtained suggest a common composition, seemingly confirming hypotheses that the diamond formed from graphite by an impact event which did not fractionate carbon isotopes. The carbon isotopic composition of graphite or diamond intergrowths among ureilites is, however, by no means constant, neither does it display a continuous variation. Instead the ureilites fall into distinct groups, which are best illustrated by plotting bulk $\delta^{13}$C against the composition of cores olivine grains (figure 2).

The $\delta^{13}$C values for five samples are tightly grouped between −9 and −11‰; the olivine for the same meteorites is highly magnesian, more than $FO_{84}$. The second group has a greater number of cohorts with $\delta^{13}$C values clustered around −2‰ and olivine compositions primarily close to $FO_{78}$ (Grady *et al.* 1985). It would be tempting to invoke some kind of reduction process to account for the relation between $\delta^{13}$C and olivine composition, particularly as a suitable

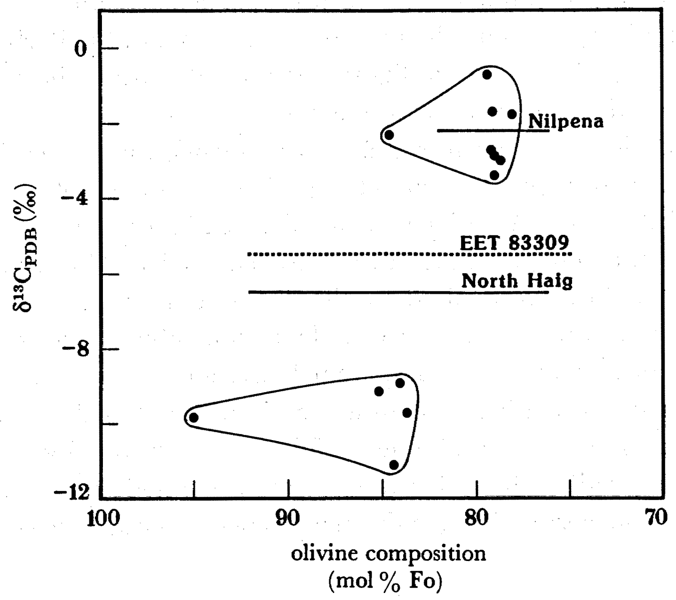

FIGURE 2. $\delta^{13}C$ against olivine composition for ureilites.

candidate is readily available, (iron silicates in ureilites are reduced to metal at grain boundaries). However, the absence of a continuous trend and the lack of any relation between carbon abundance and $\delta^{13}C$ mitigate against such a hypothesis. Two related alternative explanations of the discrete populations seen in figure 2 are that they reflect (i) the existence of more than one ureilite parent body or (ii) a parent body that is heterogeneous on a massive scale.

The heterogeneous parent body hypothesis is supported by recognition of a distinct subset of polymict ureilites (Prinz *et al.* 1987). These are characterized by their brecciated nature and the presence of lithic clasts and mineral fragments derived from different meteorite types. In addition, their olivines display a wide range in Fe/(Fe + Mg) ratios and two of the three samples now known (the exception being Nilpena) have $\delta^{13}C$ values which plot intermediate between the main groups in figure 2.

The consanguinity of the polymict ureilites is adequately recognized by nitrogen isotope measurements (Grady & Pillinger 1988). Much of the nitrogen released from these samples has very elevated $\delta^{15}N$ values up to +527‰ in contrast with five main group ureilites studies for nitrogen which have no $\delta^{15}N > +20‰$. The heavy nitrogen is variable in abundance in the three specimens but appears concentrated in EET 83309, which has been used in experiments to try and identify the carrier. Over 75% of the $^{15}N$ enriched appears to be trapped possibly as an interstitial gas or present in a labile form in the original sample because it can be released by acid dissolution. The acid residue still contains substantial quantities of $^{15}N$ and its release pattern during stepped combustion shows a remarkable similarity to that obtained by analysis of a carbonaceous clast hand-picked from another polymict ureilite Nilpena. A working hypothesis to place polymict ureilites in relation with the main group contemporaries would appear to be that the heavy nitrogen was introduced during a brecciation event caused by a carbonaceous chondrite-like projectile which impacted in a region of contact between different lithologies in a heterogeneous parent body. None of the nitrogen or carbon isotope measurements to date provide any evidence that the ureilite parent body derived from CV3 type material (Janssens *et al.* 1987), but the high $\delta^{15}N$ and its release pattern from acid residues could suggest a connection with the unusual breccia Bencubbin.

## SNC METEORITES

The Shergottite, Nakhlite and Chassigny (SNC) meteorites are conspicuous because of their relatively recent crystallization ages. Active volcanism 1.3 Ga BP requires a major planetary body and by default Mars is an obvious candidate (Wood & Ashwal 1981). Fortunately, we are in a reasonable position to examine this hypothesis because the *Viking* missions of the mid-1970s gathered geochemical data on the martian surface and made abundance and isotopic measurements of species in the atmosphere of the planet. This entry science has proved critical in demonstrating that SNC meteorites are not just samples of minor bodies in the Solar System.

The martian atmosphere has a very distinctive nitrogen isotopic composition with a $\delta^{15}N$ value of about +620‰ (Nier & McElroy 1977; Owen *et al.* 1977). Assuming that the martian atmosphere would be derived from outgasing of the planet, Fallick *et al.* (1983) looked for evidence of a heavy nitrogen component in the type specimens of the SNC meteorites but none was detected. Bogard & Johnson (1983) analysed shock-produced glass fractions (lithology C) of an Antarctic shergottite EETA 79001 for the complete spectrum of noble gases from He to Xe and reported an excellent agreement between the relative abundance pattern and that measured for the atmosphere by *Viking*. Clearly, this sample was the one most appropriate for the definitive nitrogen isotopic composition experiment. In due course high-temperature fractions during stepped analysis of EETA 79001 were shown to afford $\delta^{15}N$ values as high as +201‰ (Becker & Pepin 1984) or even +310‰ (Wiens *et al.* 1984) which, when corrected for the presence of a magmatic component can match the martian atmosphere. More important, the abundance of nitrogen is in martian atmosphere proportions relative to the noble gases. The concordance between nitrogen isotope measurements and martian atmospheric composition is shown in a plot of $\delta^{15}N$ against $^{14}N/^{36}Ar$ which can be interpreted in favour of the nitrogen released from the samples as a simple two component mixture.

The noble gases and nitrogen found in EETA 79001 lithology C are believed to have been emplaced in glass by shock and laboratory experiments have shown this hypothesis to be feasible (Wiens & Pepin 1986; Bogard *et al.* 1986). Heavy nitrogen has subsequently been found in the type specimen, Shergotty, suggesting that shocked produced glass may be heterogeneously distributed in this meteorite (Wright *et al.* 1986).

As SNC meteorites demonstrably trap components of the martian atmosphere, they ought to show evidence of interaction with carbon dioxide, which is by far the most abundant gas on Mars. The carbon budget of SNC meteorites is extremely variable and in some cases surprisingly high (R. H. Carr *et al.* 1985; Wright *et al.* 1986) by comparison with other achondrite meteorites (Grady & Pillinger 1985). It is possibly complicated by a considerable proportion of terrestrial contamination but it is presumptuous to explain everything away by this simplistic view. Figure 3*a* which is a plot of bulk $\delta^{13}C$ against bulk carbon abundance shows that EETA 79001 lithology C has the lowest bulk carbon yet measured for an SNC sample, and thus must be least affected by contamination as might be expected for an interior chip of a large meteorite preserved in the harsh Antarctic environment. If carbon released

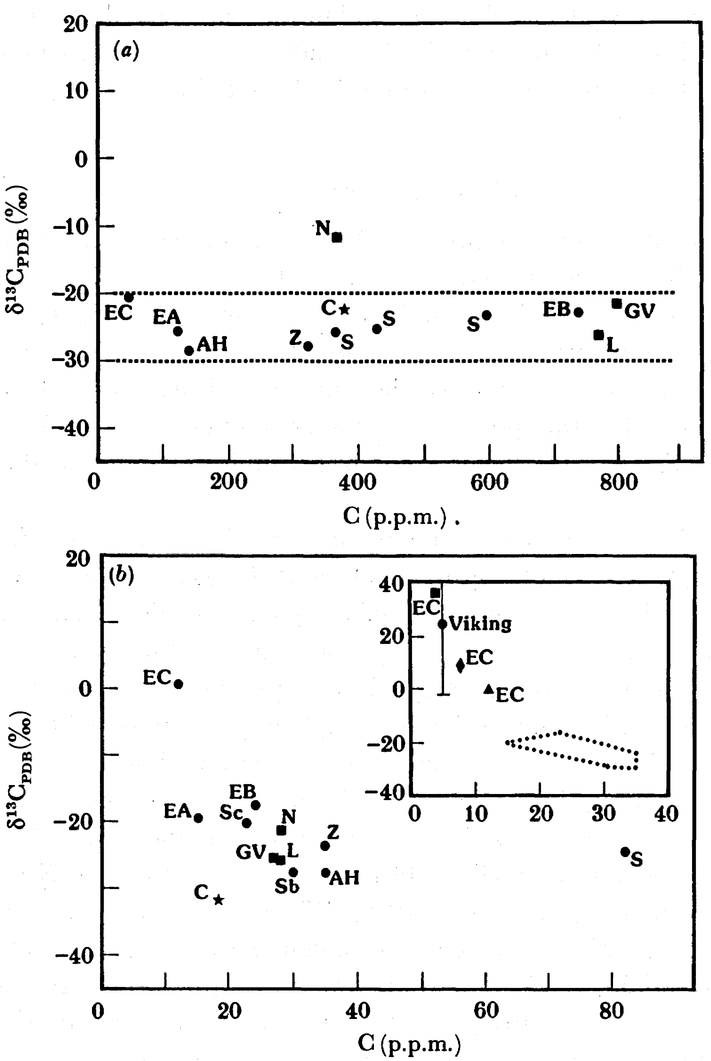

FIGURE 3. (*a*) Bulk carbon contents for SNC meteorites against $\delta^{13}C$. (*b*) Bulk carbon released above 600 °C against $\delta^{13}C$. Inset, SNC data for highest temperature steps compared with *Viking* atmospheric carbon dioxide measurements. The initials on the plot identify individual meteorites. EC and EA, lithologies C and A of EETA 79001; N, Nakhla, are the only samples whose names are germane to the discussion in the text.

above 600 °C is considered as contamination-free, 79001 lithology C clearly detaches itself from the main SNC group by moving towards higher $\delta^{13}C$ values (figure 3*b*: inset, ▲; main figure, EC). The detachment becomes more obvious, i.e. the $\delta^{13}C$ value even higher, if a 900 °C cutoff is used to resolve a possible contribution from magmatic carbon (inset figure 3*b*, ♦). The limiting value of $\delta^{13}C$ for a possible trapped atmospheric component in glass from EETA 79001 (■) is arrived at by subtracting a putative magmatic component estimated from lithology A from the lithology C high-temperature data: it can be compared with *Viking* measurements in figure 3*b* inset. The quantity of carbon dioxide trapped in lithology C is more than might be expected from its relative abundance in the martian atmosphere but laboratory experiments demonstrate that $CO_2$ is preferentially retained in shock events (Wiens *et al.* 1985).

An estimate for the isotopic composition of carbon dioxide in the martian atmosphere provides an explanation (R. H. Carr *et al.* 1985) for the anomalous bulk $\delta^{13}C$ found for Nakhla (figure 3*a*). For this meteorite, the carbon which gives rise to the unusually heavy $\delta^{13}C$ value is labile at relatively low temperature, i.e. incompatible with residence within a mineral which needs to be melted before gas release occurs. The properties of the carbonaceous substance giving low temperature heavy carbon from Nakhla are in keeping with it being a carbonate (release is equally facile for pyrolysis or combustion and the component yields carbon dioxide on treatment with phosphoric acid). Although carbonate remains to be petrologically recognized in Nakhla, a search for minor lithological units in EETA 79001 have revealed a new material termed 'white druse' as calcium carbonate (Gooding *et al.* 1988), its response to various extraction techniques is identical to Nakhla and $\delta^{13}C$ and $\delta^{18}O$ performed for both meteorites are remarkably similar (Wright *et al.* 1988*a*). Moreover, they fall in a region on a plot of $\delta^{13}C$ against $\delta^{18}O$ which distinguishes them from both terrestrial and other meteorite carbonates. It is therefore proposed that carbonates in SNC meteorites are weathering products synthesized in the martian environment. The similarity of Nakhla and EETA 79001 in this respect clearly strengthens arguments for a common provenance.

On the basis of measurements made of the oxidation state of sulphur (Burgess *et al.* 1988) in SNC meteorites there is a strong indication that alteration is a progressive process which has affected Nakhla to the greatest extent. Such arguments could indicate that a portion of the extensive carbon content of Nakhlite samples is indigenous to the samples and not just contamination; this idea is under consideration (Wright *et al.* 1988*b*).

## THE ISOTOPIC COMPOSITION OF PRIMORDIAL TERRESTRIAL MANTLE CARBON

Ever since the first measurements of carbon isotopes in samples of biological, geological and extraterrestrial origin, it has been topical to speculate concerning the role of meteorite determinations in deciding the primordial isotopic composition of terrestrial carbon (Pillinger 1984). The range of $\delta^{13}C$ values recognized for diamonds over 37‰ from +2 to −35‰, questions the concept of a mantle characterized by a single primitive carbon isotope ratio. The mode of all diamond carbon isotope analyses lies between −5 and −6‰ and is usually taken as the mantle signature. However, this also might be ill advised because of biased sampling; there is a preponderance of measurements from diamonds extracted from a relatively small number of South African mines in the dataset. Nevertheless, several authors have equated the −5 to −6‰ peak with meteorite compositions (Deines *et al.* 1987): it is close to a value for graphite in iron meteorites or matches the bulk $\delta^{13}C$ composition of type I and M carbonaceous chondrites (Hoefs 1972). In a more sophisticated approach based on $\delta^{13}C$, $\delta^{18}O$, $\delta D$ and $\delta^{15}N$, Javoy & Pineau (1983) have modelled the Earth from an enstatite chondrite-like progenitor. A requirement of the model is that $\delta^{15}N$ values measured for a suite of diamonds from Mbuji Mayi (Zaire) project a deep-seated nitrogen composition which tends towards −20‰ or less (Javoy *et al.* 1985).

Boyd *et al.* (1987) have now made more detailed studies of the distribution of nitrogen and its isotopic composition within diamonds (using a laser dissection technique to determine a growth profile) and coupled these with Fourier-transform-infrared (to ascertain the aggregation state of nitrogen) and carbon-isotope determinations. Primarily the investigation has been directed towards coated stones (diamonds having a morphology that is symptomatic of at least two stage growth), which characteristically are a large percentage of the Mbuji Mayi mine production. The study has revealed that diamonds of cubic habit including coat material were found to be indistinguishable with $\delta^{13}C$ in the range $-6.5$ to $-7.5$‰ and nitrogen, not extensively aggregated, with $\delta^{15}N$ between $-4$ and $-8$‰. Diamond of octahedral habit including the cores of the coated diamonds was much more variable: $\delta^{13}C$ $-5.0$ to $-9.8$‰ and complex forms of nitrogen with $\delta^{15}N$ from $-3.4$ to $+13.4$‰. The data for the octahedral diamonds indicate a number of epochs of diamond genesis separated possibly both in time and space and obviously predating the coats. Once formed they remained in the mantle for a period sufficient for thermal maturation. Later some of these octahedral diamonds were involved in a common event which could have been the kimberlite eruption. More recent analysis (S. R. Boyd, unpublished results) of coated stones from other diamond pipes, and conjoint carbon and nitrogen studies on less complex stones, tends to indicate that the reservoir sampled by the coats is widespread, possibly even worldwide, and located within the asthenosphere or the upper mantle. There are strong indications that the late-stage (coat) carbon may be the provenance of the diamonds giving the mode value in the $\delta^{13}C$ distribution and as such it may not be appropriate for providing the primordial composition of the Earth.

The range of isotopic compositions encountered for core samples and more mature diamonds could be indicative of an incompletely mixed mantle, which retains inhomogeneities introduced during accretion. However, other scenarios including large-scale fractionation processes in magmas or recycling of crustal material (which was fractionated in low temperature events) to a poorly mixed mantle, have to be fully evaluated before unambiguous solutions can be proposed for a perennial question.

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