



The neodymium stable isotope composition of the silicate Earth and chondrites



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ABSTRACT

The non-chondritic neodymium (Nd) $^{142}\text{Nd}/^{144}\text{Nd}$ ratio of the silicate Earth potentially provides a key constraint on the accretion and early evolution of the Earth. Yet, it is debated whether this offset is due to the Earth being formed from material enriched in *s*-process Nd isotopes or results from an early differentiation process such as the segregation of a late sulfide matte during core formation, collisional erosion or a some combination of these processes. Neodymium stable isotopes are potentially sensitive to early sulfide segregation into Earth's core, a process that cannot be resolved using their radiogenic counterparts. This study presents the first comprehensive Nd stable isotope data for chondritic meteorites and terrestrial rocks. Stable Nd measurements were made using a double spike technique coupled with thermal ionisation mass spectrometry. All three of the major classes of chondritic meteorites, carbonaceous, enstatite and ordinary chondrites have broadly similar isotopic compositions allowing calculation of a chondritic mean of $\delta^{146/144}\text{Nd} = -0.025 \pm 0.025\%$ (± 2 s.d.; $n = 39$). Enstatite chondrites yield the most uniform stable isotope composition ($\Delta^{146/144}\text{Nd} = 26$ ppm), with considerably more variability observed within ordinary ($\Delta^{146/144}\text{Nd} = 72$ ppm) and carbonaceous meteorites ($\Delta^{146/144}\text{Nd} = 143$ ppm). Terrestrial weathering, nucleosynthetic variations and parent body thermal metamorphism appear to have little measurable effect on $\delta^{146/144}\text{Nd}$ in chondrites. The small variations observed between ordinary chondrite groups most likely reflect inherited compositional differences between parent bodies, with the larger variations observed in carbonaceous chondrites being linked to varying modal proportions of calcium–aluminium rich inclusions. The terrestrial samples analysed here include rocks ranging from basaltic to rhyolitic in composition, MORB glasses and residual mantle lithologies. All of these terrestrial rocks possess a broadly similar Nd isotope composition giving an average composition for the bulk silicate Earth of $\delta^{146/144}\text{Nd} = -0.022 \pm 0.034\%$ ($n = 30$). In the samples here magmatic differentiation appears to only have an effect on stable Nd in highly evolved magmas with heavier $\delta^{146/144}\text{Nd}$ values observed in samples with >70 wt% SiO_2 . The average stable Nd isotope composition of chondrites and the bulk silicate Earth are indistinguishable at the 95% confidence level. However, mantle samples do possess variable stable Nd isotope compositions ($\Delta^{146/144}\text{Nd} = 75$ ppm) with an average $\delta^{146/144}\text{Nd}$ value of -0.008% . If these heavier values represent the true composition of pristine mantle then it is not possible to completely rule out some role for core formation in accounting for some of the offset between the mantle and chondrites. Overall, these results indicate that the mismatch of ^{142}Nd between the Earth and chondrites is best explained by a higher proportion of *s*-process Nd in the Earth, rather than partitioning into sulfide or S-rich metal in the core.

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1. Introduction

The planets of our Solar System are thought to have originated from the accumulation of dust and gas in the young Sun's protoplanetary disk, in which case the initial composition of the ter-

restrial planets should be similar to that of primitive 'chondritic' meteorites, fragments of material that escaped planetary differentiation. Although it has long been known that Earth's composition cannot be readily ascribed to any particular group of chondrites (e.g. Drake and Righter, 2002), the notion that the Earth has 'chondritic' refractory lithophile element ratios has persisted. The challenge to this assumption came from pioneering studies demonstrating that the $^{142}\text{Nd}/^{144}\text{Nd}$ isotope composition of chondritic meteorites is 18 ± 5 ppm lower than Earth's mantle

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(Boyett and Carlson, 2005; Carlson et al., 2007). The measured difference in $^{142}\text{Nd}/^{144}\text{Nd}$ requires the Sm/Nd ratio of the silicate Earth to be $\sim 6\%$ above the average chondritic value (known to within $\pm 0.3\%$; Bouvier et al., 2008) providing definitive evidence that Earth's silicate mantle at the present day is non-chondritic. One potential explanation is that Earth's mantle may have experienced an early depletion event resulting in the formation of an incompatible element enriched reservoir with a low Sm/Nd ratio balancing that seen in the mantle at the present-day (Andreasen et al., 2008; Boyett and Carlson, 2005; Carlson et al., 2007). One possibility, is that this reservoir resides at the base of the mantle, in the seismically anomalous D'' layer overlying the core. Thus far, however, there is little chemical or thermal evidence for such a reservoir (Campbell and O'Neill, 2012). Moreover, because this reservoir must have been formed within the first ~ 30 Myr following accretion, while ^{146}Sm was extant, it has been argued that it would have been destroyed by the Giant-Impact that formed the Moon (Caro and Bourdon, 2010). Alternatively, it has been suggested that the Earth was initially assembled from chondritic material and was subsequently modified during planetary construction through the removal of its early silicate crust through, so-called, collisional erosion (O'Neill and Palme, 2008). Nevertheless, collisional erosion implies that the heat-producing elements are depleted up to 50% of their chondritic values, implying unlikely cooling rates for Earth over geological history (e.g. Campbell and O'Neill, 2012).

An alternative deep reservoir within the Earth is the metallic core, however, experimental data suggests that neither Sm nor Nd are partitioned into Fe–Ni metal at the conditions of core formation, nor is Nd preferentially incorporated over Sm (Bouhifd et al., 2015). Nevertheless, it has long been known that Earth's liquid outer core must be alloyed with ~ 10 wt.% 'light' elements (e.g. Birch, 1952) which may have been added late to the core during accretion. An oft cited candidate for that light element is sulfur (e.g. Boujibar et al., 2014; Labidi et al., 2013) and experimental data suggest that Sm and Nd, and other incompatible elements, may be substantially incorporated into sulfide or sulfide-rich metal (Wohlert and Wood, 2015). In particular, Wohlert and Wood (2015) have shown that the partition coefficients of U, Nd and Sm are strong functions of the FeO content of silicate melts, increasing dramatically (with D becoming >1) as the FeO content decreases below 1 wt.%. Furthermore, D_{Nd} is always significantly greater than D_{Sm} with $D_{\text{Nd}}/D_{\text{Sm}}$ approaching 1.5 in some cases. In this case, in a growing planet the segregation of a sulfide (or S-rich metal) from reduced FeO-poor silicate (conditions analogous to core formation) will lead to enrichment of the metallic phase in U and in Nd relative to Sm relative to the residual silicate mantle, providing a significant heat source to the core and a mantle with a superchondritic Sm/Nd ratio. If such a body represented Earth early in its history then the mantle would have developed a positive ^{142}Nd anomaly relative to chondrites (as observed) and much of the energy deficit identified for core convection (Labrosse et al., 2001) would be supplied by the additional U (and Th), while maintaining a chondritic complement of heat-producing elements for long-term heating of the Earth. This raises the possibility that Earth's missing refractory elements are simply located in a sulfide or sulfur-rich metal phase in the core, rather than being lost through collisional erosion or hidden in the deep mantle. However, this result has been questioned by more recent experimental work which indicates that there is little elemental fractionation between Sm and Nd at higher temperatures, closer to those that pertain to core formation (up to 2100 °C; Wohlert and Wood, 2017).

In parallel, recent high precision Nd and Sm isotope data indicate that, compared to chondrites, the Earth is enriched in Nd produced by the slow neutron capture process (*s*-process) of nucleosynthesis (Bouvier and Boyett, 2016; Burkhardt et al., 2016). This *s*-process excess leads to higher $^{142}\text{Nd}/^{144}\text{Nd}$ ratios, relative

to chondrites. Therefore, the ^{142}Nd offset between the Earth and chondrites most likely reflects a higher proportion of *s*-process Nd in the Earth, rather than ^{146}Sm decay from a super-chondritic terrestrial reservoir (Bouvier and Boyett, 2016; Burkhardt et al., 2016; Saji et al., 2016). Nevertheless, the possibility remains that some part of the ^{142}Nd excess seen in the silicate Earth may be due to the presence of a hidden reservoir (up to 5 ± 2 ppm; Burkhardt et al., 2016). These different processes are not mutually exclusive and it is conceivable that the suprachondritic terrestrial $^{142}\text{Nd}/^{144}\text{Nd}$ is a consequence of some combination of nucleosynthetic processes, collisional erosion, and partitioning of Nd into the core.

If there is indeed a sulfide-rich reservoir in the deep Earth that has contributed to the ^{142}Nd discrepancy between chondrites and the terrestrial mantle, then Nd stable isotopes have the potential to trace this sulfide segregation event. Theory predicts that equilibrium stable isotope fractionation between silicate material (such as the mantle) and metal or sulfide (such as the core, depending on its S content) is driven by contrasts in bonding environment and oxidation state (e.g. Polyakov, 2009; Rustad and Yin, 2009). There is a significant contrast in bonding environment between sulfide and silicate, therefore heavy isotopes should be preferentially incorporated into high force-constant bonds involving rare earth element (REE) 3+ ions in silicate minerals. Preliminary measurements by Andreasen and Lapen (2015) indicate that mantle rocks may indeed possess heavier Nd isotope compositions than chondritic meteorites, consistent with the removal of light Nd into sulfide in the core, driving the residual mantle to heavier values. This study presents high-precision double spike stable Nd isotopic data for chondritic meteorites and terrestrial samples to assess the extent of fractionation between the silicate Earth and chondrites.

2. Samples and methods

2.1. Sample preparation and Nd separation

The samples investigated during this study include eleven carbonaceous chondrites (CI, CM, CO, CK, CV), seven enstatite chondrites (EH, EL) and fifteen ordinary chondrites (H, L, LL). For comparison a range of terrestrial materials were analysed and these include: twelve international rock standards that range from ultramafic to rhyolitic in composition; four optically pristine mid-ocean ridge basalt (MORB) glasses and four abyssal peridotites from the Garrett Fracture Zone, on the East Pacific Rise (Niu and Hékinian, 1997; Wendt et al., 1999); and two spinel lherzolite xenoliths from Kilbourne Hole, New Mexico (Burton et al., 1999; Jagoutz et al., 1980).

The majority of samples analysed herein were obtained as powders. When new powders were required samples were coarse crushed and unaltered chips, without visible fusion crust, were selected and then ultrasonically cleaned in distilled water and subsequently powdered using an agate mortar and pestle. Between 0.1 and 1 g of sample was taken to obtain between 100 and 200 ng of natural Nd and then spiked with our ^{145}Nd – ^{150}Nd double spike, comprising 29% ^{145}Nd and 66% ^{150}Nd , with an ideal sample to spike ratio of 60:40 based on the calculations given in Rudge et al. (2009; Fig. A1). Two digestion methods were employed during this study: 1) samples were placed in 15 mL Savillex beakers and dissolved using a concentrated HF–HNO₃ (3:1) mixture on a hot-plate at ≥ 130 °C for 72 h; or 2) samples were placed in Carius tubes with a concentrated HCl–HNO₃ (5:4) mixture and sealed and heated at 220 °C for four days, following cooling all of the sample material was extracted and subsequently underwent a standard HF–HNO₃ dissolution to dissolve the silicate fraction. Carius tube digestions were implemented to ensure the complete digestion of samples containing refractory phases, such as the Cr-spinel present in some mantle samples (see Table ES1). Following dissolution the

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