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Rare evidence for the existence of a Hadean enriched mantle reservoir

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Abstract

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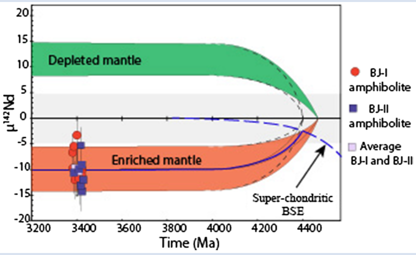

Short lived isotopic systems can help unravel the complex early differentiation history of the Earth's mantle. Excesses in neodymium-142 (^{142}Nd) measured in several occurrences of Archean mantle-derived rocks, compared to the modern upper mantle, imply the formation of an early depleted mantle in the Hadean. However, the existence of a complementary enriched reservoir, which should have also stemmed from such early differentiation event, remains equivocal. New data on 3.4 billion year old amphibolites from the São José do Campestre Massif, NE Brazil, show well resolved ^{142}Nd deficits compared to the modern upper mantle, down to -14.1 ppm. This provides the first clear evidence for an early enriched mantle source, which may represent the missing concomitant reservoir complementary to Earth's early depleted mantle.

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Introduction

The formation of the Earth was associated with considerable thermal energy, leading to large scale differentiation shortly after its formation. It has been suggested that the Earth formed, at least in part, from already differentiated bodies (*e.g.*, Kruijer *et al.*, 2014; Frossard *et al.*, 2022). Giant impacts, such as the Moon-forming event, with ensuing magma ocean formation and crystallisation, also contributed to extensive differentiation of the early mantle. These differentiation processes were consequential on the composition of Earth's various geochemical reservoirs (Carlson *et al.*, 2015), but have been greatly obscured by more than four billion years of geological activity. High precision measurements of short lived isotopic systems can however provide insights into the early differentiation of the silicate Earth and growing evidence from systems such as ^{146}Sm-^{142}Nd, ^{182}Hf-^{182}W and ^{129}I-^{129}Xe, supports a complex differentiation history of the mantle during the first few tens to hundreds of million years of Earth's evolution (Bennett *et al.*, 2007; Willbold *et al.*, 2011; Tucker and Mukhopadhyay, 2014).

The short lived ^{146}Sm-^{142}Nd isotopic system is a powerful tool to understand the early differentiation of the silicate Earth. Variations in $^{142}Nd/^{144}Nd$ ratios imply Sm-Nd fractionation occurring in the Hadean (>4.0 billion year old [Ga]), while ^{146}Sm ($t_{½} = 103$ Myr) was decaying. Positive $\mu^{142}Nd$ values, where $\mu^{142}Nd = \left[\left(\frac{^{142}Nd}{^{144}Nd_{sample}} / \frac{^{142}Nd}{^{144}Nd_{standard}}\right) - 1\right] \times 10^6$, have been measured in Eoarchean (4.0 to 3.6 Ga) mantle-derived rocks (Caro *et al.*, 2006; Bennett *et al.*, 2007; Rizo *et al.*, 2011; Li *et al.*, 2017; Morino *et al.*, 2017), supporting the formation of a depleted mantle reservoir *circa* 4.4 Ga (Caro *et al.*, 2006; Rizo *et al.*, 2011; Morino *et al.*, 2017; Hasenstab-Dübeler *et al.*, 2022). Such a differentiation event should have produced a complementary enriched reservoir, but evidence for its existence is more tenuous. Negative $\mu^{142}Nd$ values have been measured in ancient rocks from NE and NW Canada, central China, and Antarctica (O'Neil *et al.*, 2008; Caro *et al.*, 2017; O'Neil and Carlson, 2017; Reimink *et al.*, 2018; Guitreau *et al.*, 2019; Wang *et al.*, 2023), but are believed to be associated with early crust formation or crustal reworking, rather than tracing an enriched mantle. Only sparse Palaeoarchean (3.6 to 3.2 Ga) mantle-derived rocks hint at an early enriched mantle source, with few resolved negative $\mu^{142}Nd$ anomalies (Rizo *et al.*, 2012; Puchtel *et al.*, 2016; Schneider *et al.*, 2018; Boyet *et al.*, 2021), whose existence remains unclear.

Here we present ^{142}Nd data obtained on ≥3.4 Ga mafic amphibolite xenoliths from the São José do Campestre Massif, NE Brazil, which are the first results reporting ^{142}Nd anomalies in rocks from South America. More importantly, our results provide clear evidence for the existence of a Hadean enriched mantle, potentially complementary to the early depleted reservoir recorded by the Eoarchean mantle-derived rocks from the North Atlantic and North China cratons (Caro *et al.*, 2006; Bennett *et al.*, 2007; Rizo *et al.*, 2011; Li *et al.*, 2017; Morino *et al.*, 2017).

Results

The São José do Campestre Massif is an Archean basement inlier occurring within the Neoproterozoic Borborema Province of

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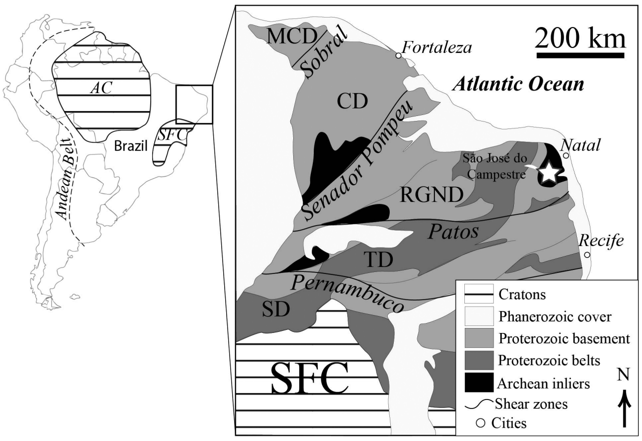

Figure 1 Simplified geological map of the Borborema Province. Tectonic domains; MCD – Médio coreau domain; CD – Ceará domain; RGND – Rio grande do norte domain; TD- transversal domain; SD – Southern domain. Archean cratons; AC- Amazon craton; SFC – São Francisco craton. See Figure S-1 for detailed geological map.

NE Brazil (Dantas *et al.*, 2013) (Fig. 1). It includes gneisses from the tonalite-trondhjemite-granodiorite (TTG) series and supracrustal sequences, with ages from 3.4 to 2.7 Ga (Dantas *et al.*, 2013). The oldest rocks are from the Bom Jesus unit, occurring in two distinct locations (BJ-I and BJ-II; Fig. S-1). The felsic rocks from BJ-I are finely banded migmatites, whereas BJ-II consists of tonalitic gneisses, both including metre scale mafic amphibolite xenoliths (Fig. S-1). The Bom Jesus tonalite has been dated at 3412 ± 8 Ma (Dantas *et al.*, 2013), providing a minimum age for the amphibolite xenoliths. Besides a few migmatite hosted samples, the amphibolites are mafic in composition (7.6–12.7 wt. % MgO; 46.5–54.0 wt. % SiO_2,) with elevated Fe_2O_3 (≤17.5 wt. %). They generally exhibit high incompatible trace element concentrations and variable degrees of LREE enrichment with relatively flat HREE (Supplementary Information for details). Their composition is therefore consistent with derivation from a mantle source, rather than a crustal progenitor. To investigate the source of these Palaeoarchean mafic rocks, 15 amphibolite xenoliths (8 samples from BJ-I and 7 samples from BJ-II) have been analysed for their ^{146}Sm-^{142}Nd compositions, as well as 2 tonalitic host samples. Except for 1 sample, the amphibolite xenoliths exhibit ^{142}Nd/^{144}Nd ratios lower than the terrestrial Nd standard (including 11 samples showing well resolved negative anomalies), with μ^{142}Nd as low as −14.1 and an average of −10.2 ± 5.0 ppm (2 sd, n = 15) (Fig. 2, Table S-2). The tonalitic samples yield μ^{142}Nd (−6.7 ± 3.6 and −3.3 ± 3.8) higher than most amphibolite samples (Fig. 2, Table S-2). Besides the most enriched mafic rocks from the Nuvvuagittuq greenstone belt in Canada (O'Neil *et al.*, 2012), the Bom Jesus amphibolite μ^{142}Nd values are the lowest measured in mantle-derived rocks.

Discussion

The amphibolite xenoliths are included in felsic rocks and have been metamorphosed to upper amphibolite facies, which could raise concerns about the inherency of their light REE content and Nd isotopic composition. The amphibolites present variable Nb/La ratios with some showing evidence of Th mobility (Figs. S-3, S-4). Post-magmatic disturbance is also apparent from the long lived ^{147}Sm-^{143}Nd isotopic system (Table S-3), yielding a ^{147}Sm-^{144}Nd *vs.* ^{143}Nd/^{144}Nd best fit line with an age of 4049.9 ± 832.8 Ma, MSWD = 290 (n = 13), holding no geochronological meaning and suggesting some extent of open system behaviour. The short lived ^{146}Sm-^{142}Nd system is however much less susceptible to post-crystallisation disturbance because processes fractionating the light REE after 4 Ga have no incidence on the ^{142}Nd/^{144}Nd ratios, since ^{146}Sm is extinct. Nevertheless, the possible effects of secondary alteration must be examined. The xenoliths high concentration in Ba, a large ion lithophile element (LILE), may indicate some secondary fluid alteration. Yet, they are not correlated with the μ^{142}Nd values (Fig. S4h) suggesting that the ^{142}Nd/^{144}Nd ratios were not disturbed despite possible element mobility. The felsic host exhibits higher concentrations in SiO_2, Al_2O_3, Na_2O when compared to the amphibolites, but the lack of correlations with μ^{142}Nd and their lower Nd and Nb content argues against crustal contamination affecting the amphibolites' ^{142}Nd/^{144}Nd ratios (Supplementary Information). Consequently, the measured μ^{142}Nd are interpreted as the original isotopic composition. Furthermore, the migmatite-hosted mafic xenoliths (BJ-I) show lower Sm/Nd ratios compared to the tonalite-hosted samples (BJ-II) (Fig. S-3, Table S-3). Still, all amphibolites exhibit uniform μ^{142}Nd values (Fig. 2) with average values of −9.6 ± 4.8 (2 sd, n = 7; BJ-I) and −10.6 ± 5.3 (2 sd, n = 8; BJ-II), suggesting that the ^{142}Nd/^{144}Nd ratios of the amphibolites were not significantly affected by the crustal reworking processes recorded by their host, and are thus interpreted as representative of their source. Although most amphibolite samples display lower μ^{142}Nd compared to the TTG samples, the average compositions for both rock types overlap within error. This could suggest an indistinguishable ^{142}Nd composition between the xenoliths and the TTG. If so, the most likely scenario would be that the TTG were produced by melting the mafic amphibolite, from which they inherited their ^{142}Nd composition.

Since variability in ^{142}Nd/^{144}Nd ratios requires Sm-Nd chemical fractionation during the Hadean, the isotopic composition measured in the 3.4 Ga Bom Jesus amphibolites, can only be explained by a few scenarios: 1) the amphibolites themselves are xenoliths of Hadean mafic rocks, 2) a recycled Hadean crustal

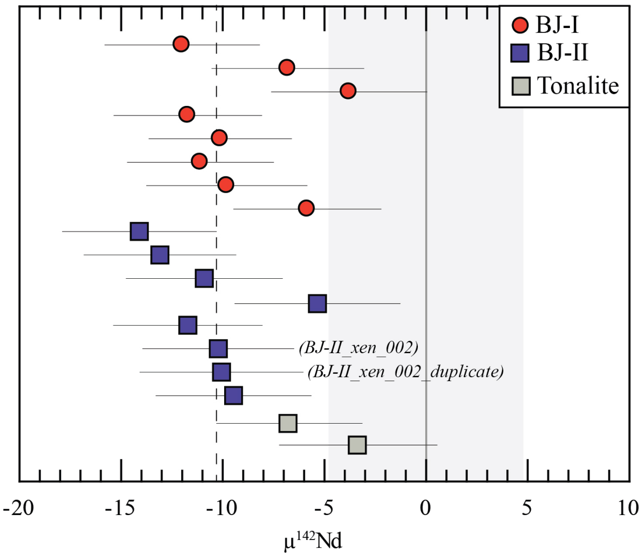

Figure 2 μ^{142}Nd for the Bom Jesus samples. Grey band shows external error on the standard. Dashed line shows the average μ^{142}Nd = −10.2 for the amphibolite samples exhibiting μ^{142}Nd outside of the JNdi-1 error. Errors on data points are 2σ. Duplicate samples are labelled.

component in their mantle source, and 3) derivation from a Hadean light REE-enriched mantle.

If older than 4 Ga, rocks with sub-chondritic Sm/Nd ratios would exhibit lower ^{142}Nd/^{144}Nd ratios compared to the terrestrial Nd standard. For example, a mafic crust with a ^{147}Sm/^{144}Nd ratio of 0.16 and formed at 4.32 Ga from a reservoir with chondritic Sm/Nd and present day ^{142}Nd/^{144}Nd corresponding to modern terrestrial mantle, would evolve to a μ^{142}Nd of ~ −10 (Figs. 3, S-5), similar to the average obtained for the Bom Jesus amphibolites. Their low ^{142}Nd/^{144}Nd ratios could therefore be consistent with preserved fragments of Hadean mafic crust. However, a Hadean suite of rocks with variable Sm/Nd ratios such as measured in the Bom Jesus amphibolites, would exhibit a range in μ^{142}Nd, correlated with their Sm/Nd ratios. Such correlation is not observed for amphibolites (Fig. S-5). Post-magmatic processes have likely affected, to some extent, the original Sm/Nd ratios of the amphibolites, which may have obscured a potential relationship with μ^{142}Nd, yet the relatively homogeneous μ^{142}Nd values of ~−10 ppm for most samples is more in accordance with post-4 Ga derivation from an enriched Hadean source.

Rather than being direct remnants of Hadean crust, the low ^{142}Nd/^{144}Nd ratios of the Bom Jesus amphibolites could be the result of a recycled Hadean crust in their mantle source (Hasenstab-Dübeler *et al.*, 2022; Tusch *et al.*, 2022). A similar model involving a contaminated source has been proposed to explain negative μ^{142}Nd values measured in mafic rocks from the Nuvvuagittuq belt in NE Canada (Caro *et al.*, 2017), but the correlation between their ^{142}Nd/^{144}Nd and Sm/Nd ratios has also been interpreted as reflective of their Hadean age (O'Neil *et al.*, 2008). Regardless, the distinct geochemical compositions of the Bom Jesus and Nuvvuagittuq mafic rocks, as well as the contrasting extent of variations in μ^{142}Nd values that they exhibit (Fig. 4), suggest that different processes were involved. If subduction-like processes occurred at the time of the Bom Jesus amphibolite formation, one could propose that a Hadean mafic crust carrying a low ^{142}Nd/^{144}Nd ratio, subducting during the Palaeoarchean, may have imprinted its ^{142}Nd isotopic composition in the mantle source of the amphibolites. This could produce mantle-derived rocks with a range in μ^{142}Nd, but expected to be correlated with light REE or other elements mobilised by such a process (Caro *et al.*, 2017). The homogenous μ^{142}Nd of the Bom Jesus amphibolites and lack of correlation with common subduction setting geochemical indicators (Figs. S-3, S-4), however, argues against inheritance of the ^{142}Nd composition from a Hadean subducting slab. This rather suggests that the low ^{142}Nd/^{144}Nd composition was characteristic of their mantle source. Alternative models without subduction may be

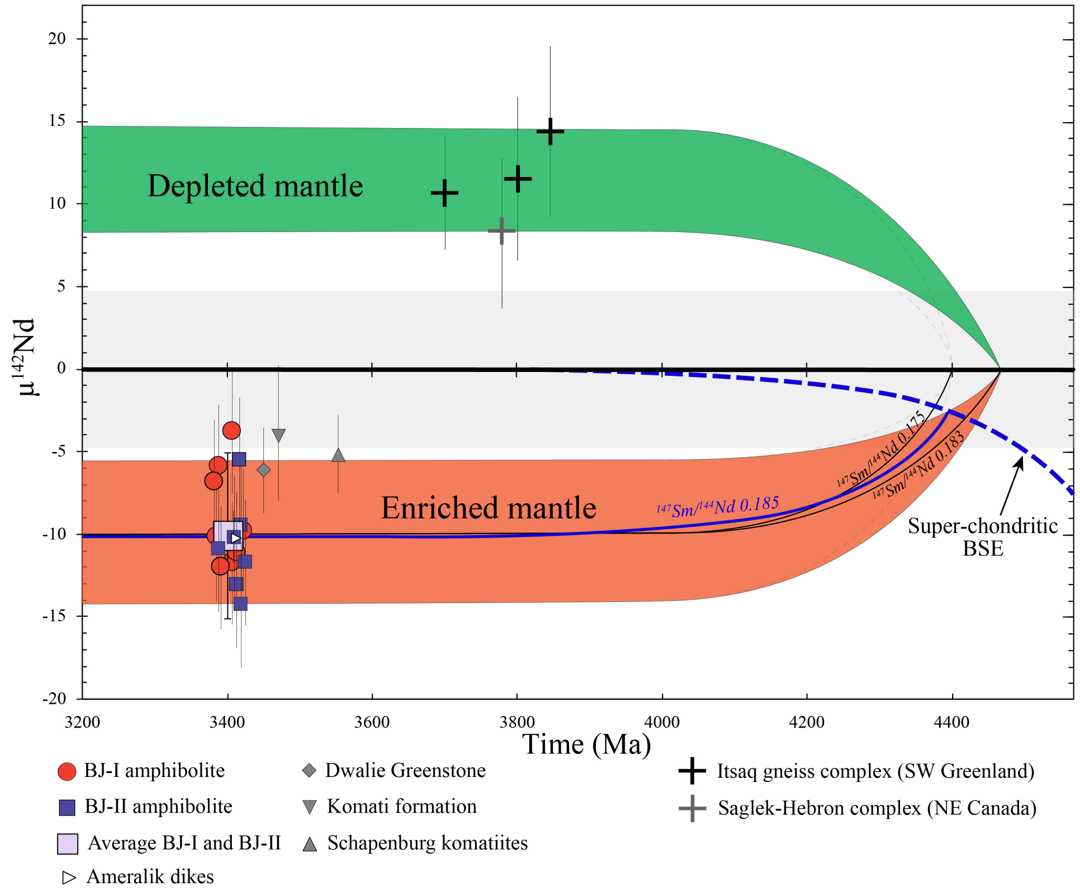

Figure 3 **μ^{142}Nd evolution of the source of the Bom Jesus amphibolites.** Horizontal line at μ^{142}Nd = 0 represents a reservoir with chondritic Sm/Nd and present day μ^{142}Nd corresponding to modern mantle. Blue dashed line represents a super-chondritic BSE (Frossard *et al.*, 2022; Johnston *et al.*, 2022). Grey band as in Figure 2. Red and green envelopes represent modelled enriched and depleted reservoirs formed at 4470 Ma. Black dashed lines show these reservoirs formed at 4400 Ma and evolving to the same present day μ^{142}Nd. Thin solid black lines correspond to the ^{147}Sm/^{144}Nd required to evolve to μ^{142}Nd = −10 (average amphibolite value). Blue line shows a 4400 Ma reservoir derived from a super-chondritic BSE. The Bom Jesus samples are displaced around their interpreted age (3.4 Ga) for clarity. SW Greenland and NE Canada data are average μ^{142}Nd values for distinct mantle-derived lithologies. Schapenburg komatiite, Dwalie Greenstone and Komati formation data show average values (Puchtel *et al.*, 2016; Schneider *et al.*, 2018; Boyet *et al.*, 2021). Ameralik dike data shows the lowest μ^{142}Nd value measured (Rizo *et al.*, 2012).

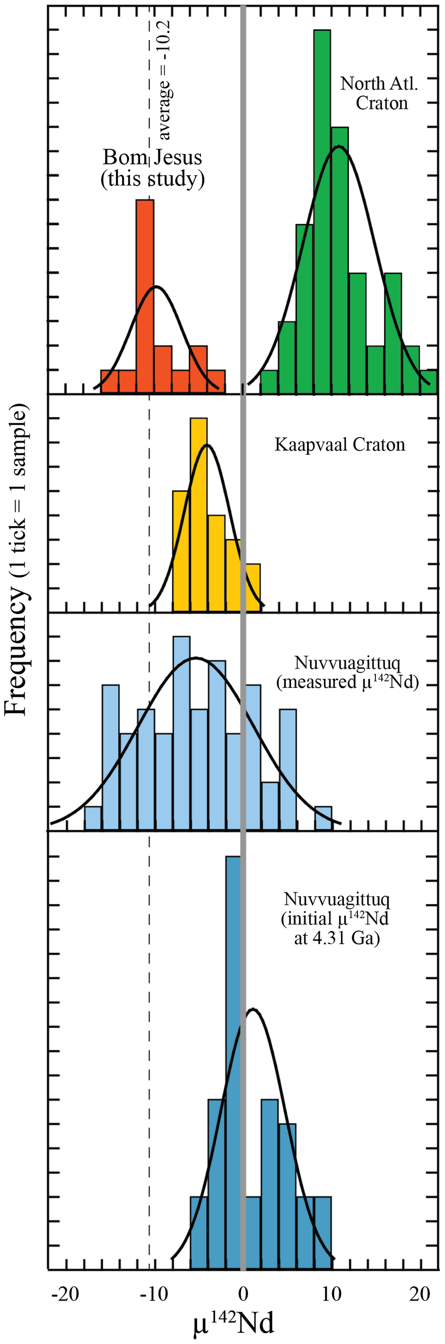

Figure 4 **μ^{142}Nd histograms for $\leq$3.4 Ga mantle-derived rocks.** Data from this study; Bennett *et al.* (2007); O'Neil *et al.* (2008, 2012); Rizo *et al.* (2013, 2016); Morino *et al.* (2017); Schneider *et al.* (2018); Boyet *et al.* (2021). Nuvvuagittuq data in top panel are measured ^{142}Nd compositions, while bottom panel shows the initial μ^{142}Nd at 4.31 Ga if the rocks are considered Hadean (O'Neil *et al.*, 2012).

able to produce crustal material with variable ^{142}Nd isotopic compositions interacting with the mantle. For example, a recent model proposed the differentiation of $\sim$4.5 Ga proto-crust, producing a restitic material that would then mix with an Archean mantle to produce a hybrid source (Tusch *et al.*, 2022). With the right amount of mixing, timing and extent of differentiations, such an intricate model could produce a mantle source with a ^{142}Nd isotopic composition consistent with that of the Bom Jesus amphibolites, but no evidence supports a similar complex multi-stage process. This hybrid source is also inconsistent with the high concentrations in incompatible trace elements of the Bom Jesus amphibolites (up to $\sim$55 ppm Nd) and would require an unrealistically low degree of partial melting ($<$2 %) to produce similarly enriched mafic magmas.

Silicate differentiation events occurring while ^{146}Sm was still extant would produce complementary incompatible trace element depleted and enriched reservoirs, respectively evolving to high and low ^{142}Nd/^{144}Nd ratios. The existence of an early enriched reservoir complementary to Earth's modern mantle has been proposed to account for the higher ^{142}Nd/^{144}Nd ratios of terrestrial rocks compared to chondrites (Boyet and Carlson, 2005), but recent studies proposed alternative scenarios that do not require early differentiation of the Earth's mantle to explain this difference in ^{142}Nd isotopic compositions (Bouvier and Boyet, 2016; Burkhardt *et al.*, 2016). However, the positive μ^{142}Nd values measured in a number Eoarchean mantle-derived rocks imply the formation of an early depleted mantle, perhaps formed through crystallisation of a magma ocean (Rizo *et al.*, 2011; Li *et al.*, 2017; Morino *et al.*, 2017), and would still entail the concomitant formation of an early enriched reservoir (*i.e.* characterised by negative μ^{142}Nd values). Not only the negative μ^{142}Nd values of the Bom Jesus mafic xenolith suggest derivation from such low Sm/Nd early sources, but their unusually high concentrations in most incompatible trace elements, compared to predominant Archean basaltic rocks (Fig. S-3c), is also consistent with an enriched mantle source.

Estimations of the chemical composition of an early formed enriched reservoir depend on its Sm/Nd ratio, size and μ^{142}Nd of Bulk Silicate Earth (BSE). Nevertheless, it would most likely exhibit higher Nd contents compared to BSE. For instance, Boyet and Carlson (2005) estimated that the Nd concentration of a small size, enriched reservoir (4 % of the mass of BSE), could be up to 7 times higher than that of the BSE. Although they considered the enriched reservoir as complementary to a depleted MORB mantle with an excess in ^{142}Nd compared to BSE, which recent work showed is not required, it nevertheless supports the fact that an early enriched reservoir would exhibit relatively high Nd content. It is however difficult to constrain the exact nature of an early enriched source. Hofmann *et al.* (2022) proposed an early enriched reservoir that originated as a mafic crust, while the early enriched reservoir modelled by Boyet and Carlson (2005) is ultramafic in composition. Regardless of the exact nature of the early enriched source of the Bom Jesus amphibolites, it needs to be capable of producing mafic magmas with high concentrations of incompatible trace elements.

The extent of the ^{142}Nd deficit characterising this early formed reservoir depends on the timing of its differentiation, its Sm/Nd ratio and the ^{142}Nd composition of the source reservoir. If we consider an early enriched mantle derived from a BSE with chondritic Sm/Nd and present day $\mu^{142}\text{Nd} = 0$, and formed coevally to the SW Greenland early depleted mantle at 4.47 Ga (Rizo *et al.*, 2011), it would require a $^{147}\text{Sm}/^{144}\text{Nd} = 0.183$ to evolve to a μ^{142}Nd $\sim\!-10$, consistent with the source of the Bom Jesus amphibolites (Fig. 3). A later differentiation at 4.40 Ga would require a lower ^{147}Sm/^{144}Nd ratio of 0.175 to evolve to $\mu^{142}\text{Nd} = -10$. As illustrated on Figure 3, derivation at 4.40 Ga from a slightly super-chondritic BSE with initial $\mu^{142}\text{Nd} = -7.6$ (average value proposed by Frossard *et al.*, 2022 and Johnston *et al.*, 2022) would increase the required ^{147}Sm/^{144}Nd ratio to 0.185 in order to produce the same μ^{142}Nd of -10.

Together, the geochemistry and ^{142}Nd isotopic composition of the Bom Jesus amphibolites provide evidence for a Hadean enriched mantle reservoir. A number of mantle-derived rocks from the Kaapvaal craton alluded to an early formed enriched source (Fig. 4), but its existence is better evidenced by the unequivocal and well resolved negative ^{142}Nd anomalies of the Bom Jesus amphibolites. With a μ^{142}Nd value of ~ -10, the range of plausible compositions and ages for this enriched mantle indicates that it could be complementary to an early depleted source previously recorded by several Eoarchean mafic and ultramafic rocks from the North Atlantic craton (Fig. 4). The confirmation of a Hadean enriched mantle reservoir has major implications on our understanding of the complex differentiation processes occurring on the Earth shortly after its formation and for the Archean geodynamics. Considering that the age of the mafic xenoliths is similar to their felsic hosts, the Bom Jesus amphibolites and other rare mantle-derived rocks hinting at the existence of an early enriched reservoir characterised by negative μ^{142}Nd values are interestingly of similar ages (Fig. 3), dated between 3.41 and 3.55 Ga, but located over distinct Archean cratons (Rizo *et al.*, 2012; Dantas *et al.*, 2013; Puchtel *et al.*, 2016; Schneider *et al.*, 2018; Boyet *et al.*, 2021). Several lines of evidence are suggesting a shift in global geodynamic setting in the mid to early Archean (*e.g.*, Næraa *et al.*, 2012; Bauer *et al.*, 2017; Reimink *et al.*, 2018; Hawkesworth *et al.*, 2019; Drabon *et al.*, 2022), which coincides with the emplacement of most rocks consistent with derivation from this Hadean enriched source, as well as the apparent disappearance, or at least attenuation, of rocks derived from the early depleted mantle.

Acknowledgments

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2336.

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