

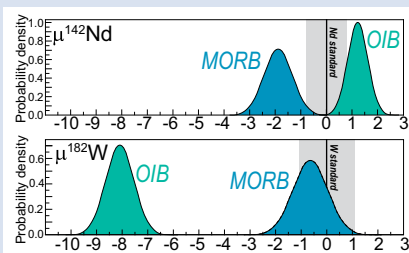
Comparative ^{142}Nd and ^{182}W study of MORBs and the 4.5 Gyr evolution of the upper mantle

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<https://doi.org/10.7185/geochemlet.2412>

Abstract



New high precision Nd and W isotopic compositions were obtained on the same basalt samples from the Pacific-Antarctic Ridge. These provide the best estimate so far for the $\mu^{142}\text{Nd}$ and $\mu^{182}\text{W}$ values of the depleted mantle source of mid-ocean ridge basalts known as DMM. The PAR basalts yield a mean $\mu^{142}\text{Nd} = -1.6 \pm 5.0$ (2 s.d.) and $\mu^{182}\text{W} = -1.9 \pm 3.5$ (2 s.d.), which together with the literature data allow the isotope composition of the DMM to be constrained. The present-day DMM $\mu^{182}\text{W}$ is 10–20 ppm lower than that of the Archean mantle. This decrease could be related to the broad incorporation of mantle plume material into the upper mantle, starting between 2.4 and 3 billion years ago, due to the onset of deep cold slab subduction, and its attendant return mantle flow.

Received 4 July 2023 | Accepted 28 February 2024 | Published 11 April 2024

Introduction

The Earth's mantle underwent significant chemical evolution during its early history. The short lived isotope systems ^{182}Hf - ^{182}W ($t_{1/2} = 8.9$ Myr) and ^{146}Sm - ^{142}Nd ($t_{1/2} = 103$ Myr) applied to the study of terrestrial rocks have provided valuable insights into understanding the nature of these changes, because they are especially capable of tracing the most ancient chemical fractionation processes. Notably, ancient mantle-derived rocks from various Archean cratons exhibit variations in ^{142}Nd abundances (e.g., Carlson *et al.*, 2019), implying silicate differentiation within the first ~500 million years of Earth's history. The recent detection of ^{182}W abundance variations in mantle-derived rocks of different ages have initiated debates regarding the main processes controlling ^{182}W variability, including early silicate differentiation as seen by the ^{146}Sm - ^{142}Nd system (e.g., Touboul *et al.*, 2012), core-mantle chemical interactions (e.g., Rizo *et al.*, 2019), differences in the mass of late accreted extra-terrestrial material into the mantle (e.g., Willbold *et al.*, 2011) and the recycling of ancient crust or sediments into the mantle (e.g., Tusch *et al.*, 2021).

Variations in ^{142}Nd and ^{182}W abundances, denoted as $\mu^{142}\text{Nd}$ and $\mu^{182}\text{W}$, reflect part-per-million deviations from laboratory standards set at $\mu = 0$. These standards are assumed to represent the compositions of the modern mantle, or the depleted mid-ocean ridge mantle (DMM) reservoir. However, few mantle peridotites ($n = 4$), or melts directly derived from the depleted upper mantle such as MORB ($n = 9$), have been analysed for $\mu^{142}\text{Nd}$ (e.g., Boyet and Carlson, 2006; Caro *et al.*, 2006; Cipriani *et al.*, 2011; Jackson and Carlson 2012; Hyung

and Jacobsen, 2020). The W isotopic composition of the DMM has never been properly determined, given that only two MORB samples have been studied to date (Rizo *et al.*, 2016; Mundl *et al.*, 2017). Therefore, $\mu^{142}\text{Nd}$ and $\mu^{182}\text{W}$ of the upper mantle are presently inadequately constrained, yet are crucial for understanding the composition and chemical evolution of the silicate Earth.

This study presents new $\mu^{142}\text{Nd}$ data for fourteen MORB samples from the Pacific-Antarctic Ridge (PAR), therefore, tripling the current MORB dataset. We also report W concentrations for 18 samples, and $\mu^{182}\text{W}$ measurements for seven of these PAR samples. The studied MORB samples were collected between 53°S and 42°S (Fig. S-1), a fast-spreading ridge section with no nearby hotspots. All fresh MORB samples were collected on-axis, and have previously been thoroughly characterised for their petrology, geochemistry and isotopic compositions (e.g., Sr, Nd, Pb, Hf, He, D, S, and Mo), showing the lack of plume influence (Hamelin *et al.*, 2011; see Supplementary Information for sample descriptions). The MORB $\mu^{142}\text{Nd}$ and $\mu^{182}\text{W}$ are used to provide the first robust estimate for the DMM supplying the Pacific-Antarctic Ridge.

The $\mu^{182}\text{W}$ and $\mu^{142}\text{Nd}$ of the Upper Mantle Sources Supplying the PAR Basalts

As mantle-derived melts devoid from continental crustal contamination, MORBs allow to constrain the $\mu^{142}\text{Nd}$ and $\mu^{182}\text{W}$ of the upper mantle. The low W abundance of MORB samples,

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however, makes them susceptible to disturbances related to element mobility or secondary hydrothermal alteration overprinting. This concern does not apply to the less mobile Nd, for which concentration in basalts is higher. Therefore, evaluating whether the W hosted in these rocks is of igneous origin is imperative.

Tungsten concentrations in the studied PAR MORBs ($n = 18$) vary from 6.8 ng g^{-1} to 220 ng g^{-1} (Data Table S-1) and are strongly coupled to other immobile and highly incompatible trace elements, such as Th and Nb (Fig. S-2a, b). Narrow ranges for $W/Th = 0.1\text{--}0.15$ (Fig. S-2d), $W/Ba = 0.0019\text{--}0.0027$, $W/Ta = 0.08\text{--}0.16$, and $W/U = 0.23\text{--}0.43$ (not shown) indicate that the W contained in the MORB samples is derived from their mantle sources and free from secondary hydrothermal W overprinting (e.g., König *et al.*, 2011). Therefore, the W isotopic analyses of these samples accurately establish the $\mu^{182}\text{W}$ of their mantle sources.

The $\mu^{182}\text{W}$ of the PAR MORBs range from -3.9 ± 4.1 (2 s.e.) to 1.0 ± 2.9 (2 s.e.) with a mean of -1.9 ± 3.5 (2 s.d.; $n = 7$) (Table 1 and Fig. 1a; Method and detailed isotope data in the Supplementary Information). These results are undistinguishable within errors from the $\mu^{182}\text{W}$ values of -0.8 ± 4.5 (2 s.e.) and 3.5 ± 4.0 (2 s.e.) in MORBs, respectively, from the East Pacific Rise (Rizo *et al.*, 2016) and the Central Indian Ridge (Mundl *et al.*, 2017). The narrow W isotopic variability observed across three mid-ocean ridge segments from a sparse worldwide coverage suggests that there is little $\mu^{182}\text{W}$ variation outside the analytical uncertainties within the DMM, on a regional and global scale.

The PAR MORB $\mu^{142}\text{Nd}$ range from -5.7 ± 2.3 (2 s.e.) to 2.8 ± 2.4 (2 s.e.), with a mean of -1.6 ± 5.0 (2 s.d.; $n = 14$) (Fig. 1b). Eleven out of 14 samples show $\mu^{142}\text{Nd}$ within the uncertainty of the JNdi-1, and three samples exhibit lower $\mu^{142}\text{Nd}$ between -5.7 ± 2.3 (2 s.e.) and -5.0 ± 2.4 (2 s.e.). Most $\mu^{142}\text{Nd}$ of PAR MORB are within errors of basalts collected from five other ridges (Fig. 1b), showing little $\mu^{142}\text{Nd}$ variability outside the analytical precision of measurements, regionally or globally, consistent with the $\mu^{182}\text{W}$.

Hadean Differentiation Between DMM and OIB Sources?

MORB and abyssal peridotite samples are the best proxies for the $\mu^{142}\text{Nd}$ of the DMM. Although the $\mu^{142}\text{Nd} \leq -5$ values of some MORB and mantle peridotite samples are not resolvable from the $\mu^{142}\text{Nd}$ of standards (Fig. 1b), they suggest the presence of Hadean mantle heterogeneities in the DMM, given that ^{142}Nd variability can only be produced by Sm/Nd fractionation occurring before the extinction of ^{146}Sm (i.e. before ~ 4 Ga). Recent studies propose that Earth's building blocks may have been characterised by $\mu^{142}\text{Nd}$ of ~ -7.5 (e.g., Frossard *et al.*, 2022; Johnston *et al.*, 2022), and a $^{147}\text{Sm}/^{144}\text{Nd}$ ratio of ~ 0.201 — approximately 2.5 % higher than the chondritic reference value of 0.1960 (to evolve from initial $\mu^{142}\text{Nd} \sim -7.5$ to 0 today). Consequently, the low $\mu^{142}\text{Nd}$ values (≤ -5) detected in certain MORB and mantle peridotite samples might be relics of these primordial components in the DMM. Preservation of primordial heterogeneities in the DMM is, however, difficult to explain. Not only must they survive the mixing of Earth's magma ocean stage, but must also evolve with a near-chondritic Sm/Nd ratio during the Hadean to preserve their $\mu^{142}\text{Nd}$ (≤ -5) measured today. This early, near-chondritic Sm/Nd ratio contrasts with the later, superchondritic Sm/Nd ratio required to explain the $\epsilon^{143}\text{Nd} > +8$ of all analysed PAR MORB samples, even including those exhibiting low $\mu^{142}\text{Nd}$.

Intriguingly, the weighted mean $\mu^{142}\text{Nd}$ of all available data for MORB and mantle peridotites ($\mu^{142}\text{Nd} = -1.6 \pm 0.9$; $n = 27$), and OIB ($\mu^{142}\text{Nd} = +2.0 \pm 0.6$; $n = 66$) (Fig. 2a) reveals slight differences, suggesting distinct mantle sources and evolutionary histories. Despite the small ^{142}Nd difference of ~ 3.6 ppm falling within the range of current analytical precision, a student's t-test comparing the datasets indicates a 99.99 % probability of significant difference (p-value of 9.45×10^{-5}). While t-tests are valid statistical methods for distinguishing datasets, they do not consider uncertainties associated with individual data points. To obtain the most accurate $\mu^{142}\text{Nd}$ estimates for the DMM and the OIB sources, Monte Carlo bootstrap simulations were

Table 1 $\mu^{182}\text{W}$ and $\mu^{142}\text{Nd}$ of MORBs from the Pacific-Antarctic Ridge. Detailed isotope compositions can be found in the Supplementary Information (Data Tables S-2 and S-3).

Sample	$\mu^{182}\text{W}$	± 2 s.e.	$\mu^{142}\text{Nd}$	± 2 s.e.
PAC2DR01-1	-1.7	3.2	-7.3	2.4
PAC2DR01-1 duplicate 1			-3.6	3.6
PAC2DR01-1 duplicate 2			-4.0	3.3
PAC2DR01-1 average			-5.0	2.4
PAC2DR02-1	1.0	2.9	0.2	2.5
PAC2DR04-2			-1.8	2.4
PAC2DR05-2g	-3.0	4.8	1.4	2.4
PAC2DR06-6	-1.1	3.5	0.5	1.6
PAC2DR08-1	-3.7	5.8	-2.8	2.2
PAC2DR20-1	-0.8	3.4	-1.0	2.4
PAC2DR22-1			-5.7	2.3
PAC2DR27-1	-3.9	4.1	-1.7	2.4
PAC2DR30-1			-0.04	3.2
PAC2DR31-3			2.8	2.4
PAC2DR32-1			-5.3	2.3
PAC2DR33-1			-1.8	2.4
PAC2DR36-1			-1.6	2.4
Mean ± 2 s.d.	-1.9	3.5	-1.6	5.0



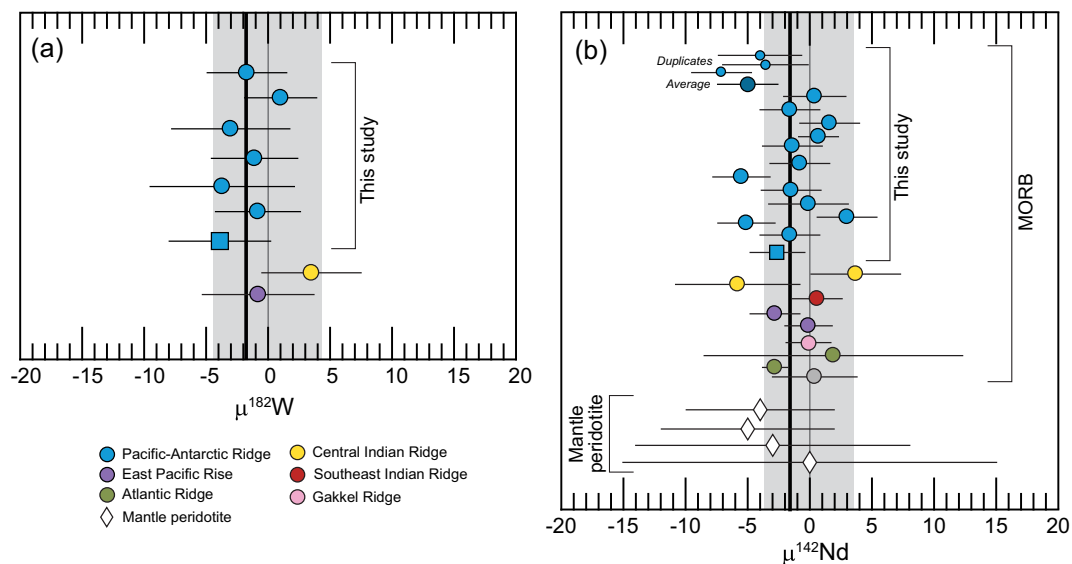


Figure 1 (a) $\mu^{182}\text{W}$ and (b) $\mu^{142}\text{Nd}$ for PAR MORBs of this study. Vertical thin grey lines with shaded areas show average standards ($\mu^{182}\text{W} = 0$ and $\mu^{142}\text{Nd} = 0$) with external reproducibility (2 s.d.). Vertical thick black lines show the mean for all MORBs analysed here ($\mu^{182}\text{W} = -1.9 \pm 3.5$; $\mu^{142}\text{Nd} = -1.6 \pm 5.0$). Smaller symbols in (b) are duplicate analyses with the average value and 2 s.d. in darker shade. MORB $\mu^{182}\text{W}$ of East Pacific Rise and Central Indian Ridge are, respectively, from Rizo *et al.* (2016) and Mundl *et al.* (2017). The $\mu^{142}\text{Nd}$ for other MORBs are from Boyet and Carlson (2006), Caro *et al.* (2006), Jackson and Carlson (2012), and Hyung and Jacobsen (2020), and mantle peridotite data from Cipriani *et al.* (2011).

conducted, accounting for uncertainties associated with each individual $\mu^{142}\text{Nd}$ data point. Figure 2b shows the resulting probability density plots, implying statistically distinguishable $\mu^{142}\text{Nd}$ values for the DMM and the OIB sources.

The lower average $\mu^{142}\text{Nd}$ value of the DMM compared to the OIB sources indicates differentiation before ~ 4 Ga. This early differentiation is consistent with the distinct $^{129}\text{Xe}/^{130}\text{Xe}$ ratios observed in MORB and OIB, also implying mantle differentiation around 4.45 Ga (Mukhopadhyay, 2012). Notably, a low Sm/Nd fractionation factor of ~ 0.02 at 4.45 Ga (defined as $^{147}\text{Sm}/^{144}\text{Nd}_{[\text{newly formed reservoir}]} / ^{147}\text{Sm}/^{144}\text{Nd}_{[\text{parent reservoir}]} - 1$) can explain the observed ~ 3.6 ppm $\mu^{142}\text{Nd}$ difference between DMM and OIB sources. This scenario, however, requires the DMM to have evolved with a lower Sm/Nd ratio compared to the OIB sources, implying that the DMM was originally *less* depleted in incompatible elements compared to OIB sources. This contradicts the evidence that the DMM is *more* depleted, based on its present day $\epsilon^{143}\text{Nd}$ of nearly +9 of MORB (Gale *et al.*, 2013). Oceanic crust sequestration and preferential slab subduction to the lower mantle have been proposed as a plausible model to deplete the lower mantle to a greater extent than the upper mantle (Tucker *et al.*, 2020). The seemingly opposite time-averaged Sm/Nd required to explain MORB $\mu^{142}\text{Nd}$ and $\epsilon^{143}\text{Nd}$ requires Sm/Nd fractionation that could result from extraction of continental crust occurring after extinction of ^{146}Sm (after 4 Ga).

Modern Mantle $\mu^{182}\text{W}$ Heterogeneities – Established Early or Acquired Through Time?

A significant observation derived from our results is that the average $\mu^{182}\text{W}$ of the modern DMM (-1.9) is approximately 10 to 20 ppm lower than the average Hadean-Archean mantle (e.g., Rizo *et al.*, 2019). This difference in $\mu^{182}\text{W}$ suggests the incorporation of a negative $\mu^{182}\text{W}$ component to the DMM, in order to decrease the $\mu^{182}\text{W}$ of the Archean mantle to the present

day composition. Component candidates with negative $\mu^{182}\text{W}$ include: 1) an early-formed enriched crust or mantle domain, 2) late accretionary chondritic material, and 3) mantle plume material.

Negative $\mu^{182}\text{W}$ in the earliest oceanic crust, or differentiated mantle domain formed after Earth's magma ocean solidification, can result from Hf/W fractionation during the lifetime of ^{182}Hf (i.e. first ~ 50 Ma of Solar System history). The remixing of this negative $\mu^{182}\text{W}$ material into the mantle could lead to the observed decrease in $\mu^{182}\text{W}$ from the Archean to the modern mantle. If characterised by negative $\mu^{182}\text{W}$, such material would also exhibit negative $\mu^{142}\text{Nd}$. However, combined negative $\mu^{182}\text{W}$ and $\mu^{142}\text{Nd}$ have currently only been observed in ~ 3.6 Ga komatiites (Puchtel *et al.*, 2016). Although early mantle differentiation models capable of explaining decoupled ^{182}W and ^{142}Nd have been proposed (e.g., Tusch *et al.*, 2022), these still require an initial ~ 4.35 Ga source with low $\mu^{182}\text{W}$ and low $\mu^{142}\text{Nd}$, which existence has yet to be proven. Furthermore, although some negative $\mu^{182}\text{W}$ has been detected in Paleoproterozoic rocks and diamictites from South Africa (Puchtel *et al.*, 2016; Mundl *et al.*, 2018; Tusch *et al.*, 2022), more than 95 % of ancient rocks currently analysed instead have positive $\mu^{182}\text{W}$ between +10 and +20 (e.g., Rizo *et al.*, 2019). The scarce evidence for negative $\mu^{182}\text{W}$ in ancient rocks, and the rather constant positive $\mu^{182}\text{W}$ in throughout the Archean, suggests that if abundant negative- $\mu^{182}\text{W}$ material existed, either early crustal subduction was deep enough to escape subsequent sampling, or most of that negative $\mu^{182}\text{W}$ crust has now been recycled into the mantle.

Alternatively, the late accretion of materials with chondritic bulk compositions after core formation is a hypothesis proposed to account for the abundance of highly siderophile elements (HSE) in the upper mantle. This accretion could also explain the decrease of $\mu^{182}\text{W}$ of the DMM (e.g., Willbold *et al.*, 2011), given the $\mu^{182}\text{W}$ of ~ -200 of chondrites (e.g., Kleine and Walker, 2017), and would be unrelated to $\mu^{142}\text{Nd}$. The common absence of correlations between $\mu^{182}\text{W}$ and HSE abundances (e.g., Rizo *et al.*, 2019), however, suggests that late accretion is not the predominant process explaining the decrease of $\mu^{182}\text{W}$

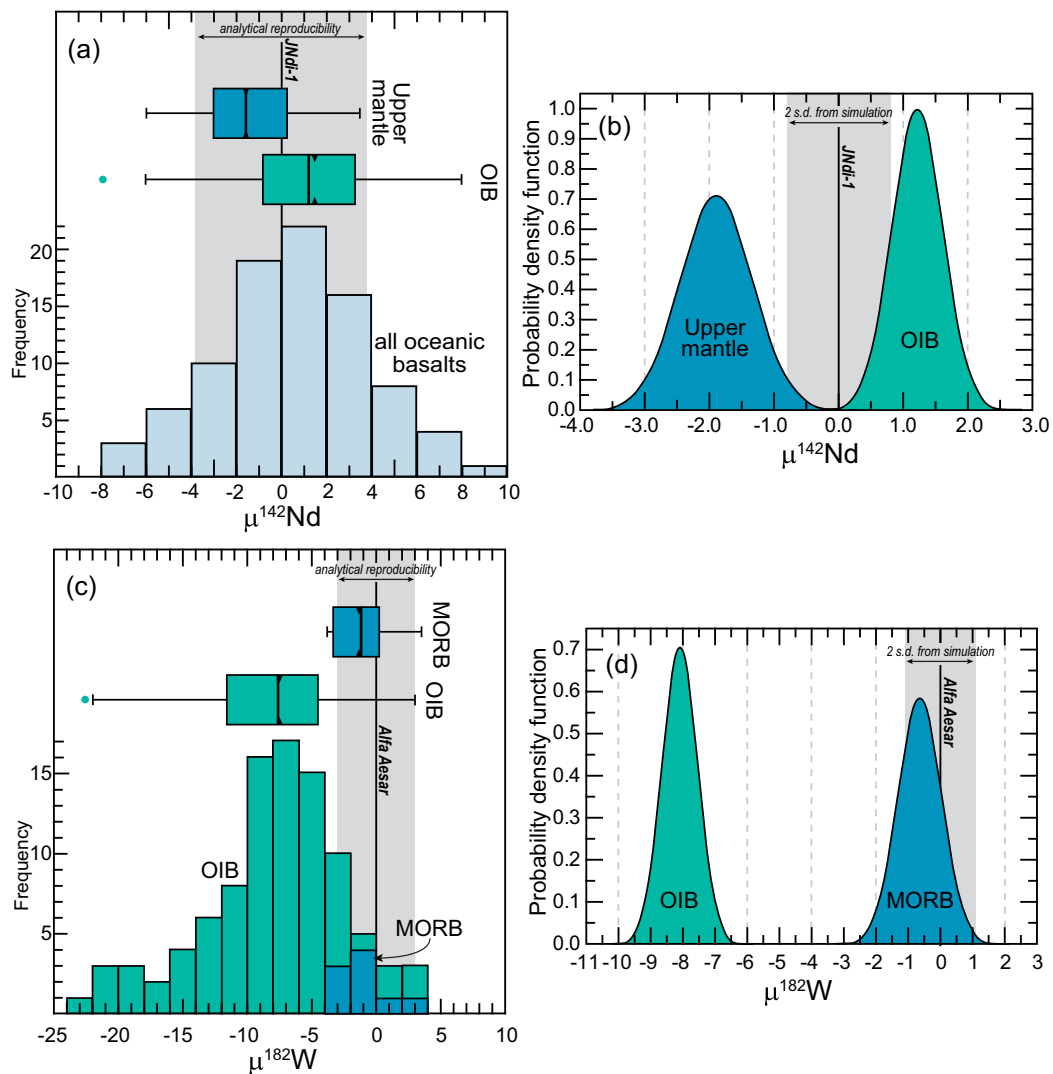


Figure 2 Compilation and computed probability density of (a, b) $\mu^{142}\text{Nd}$ and (c, d) $\mu^{182}\text{W}$ for MORB, OIB and mantle peridotites. Upper mantle (a) includes MORB and mantle peridotite samples. Left side plots (a and c) present histograms of all data; grey bands show the analytical reproducibility on the standards obtained during this study; thick bars on box plots present the median, and the triangle notches show the weighted mean values. Right side plots (b, d) show compositions generated by Monte Carlo bootstrap simulations (10,000 runs). Vertical lines and grey areas in b and d show the mean and 2 s.d. envelope of the Nd and W standards derived from the simulations. Database and references provided in the Supplementary Information Data Table S-4.

in the mantle. Furthermore, late accretion cannot explain $\mu^{182}\text{W}$ -He correlations found in some OIB (e.g., Mundl *et al.*, 2017), since noble gas characteristics are most likely not preserved after impacts.

The model that would best explain the $\mu^{182}\text{W}$ observations, supported by a wide range of evidence such as seismology, multiple radiogenic isotope systems, and trace element geochemistry, is some mass transfer from mantle plumes into the upper mantle. Negative $\mu^{182}\text{W}$ in modern rocks is currently exclusively associated with plume-derived magmas, showing $\mu^{182}\text{W}$ values as low as -22.7 (Mundl *et al.*, 2017). Ocean island basalts, the lavas derived from mantle plumes, also display a range of Sr, Nd and Pb isotope compositions (e.g., Zindler and Hart, 1986), believed to result from the incorporation of recycled components (sediments, crust) into the plume sources over time (e.g., Hofmann and White, 1982). Although the $\mu^{182}\text{W}$ variability of the PAR MORB is only within 5 ppm, $\mu^{182}\text{W}$ values appear to be correlated with $(\text{La}/\text{Gd})_N$ and Sr, Pb, and Nd isotopic compositions (Fig. 3a–d). For example, sample PAC2DR27-1, characterised as a Transitional-MORB (T-MORB),

presents the lowest measured $\mu^{182}\text{W}$ and is the most trace element enriched basalt, with the most radiogenic $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, and the lowest $^{143}\text{Nd}/^{144}\text{Nd}$ (Fig. 3a–d, and Fig. S-2). The correlations between $\mu^{182}\text{W}$ vs. $(\text{La}/\text{Gd})_N$ and Sr, Pb, and Nd isotopic compositions could be evidence of the progressive incorporation of mantle plume material into the upper mantle. Although the $\mu^{182}\text{W}$ variability observed in the PAR MORB is small, mantle melting averages the isotope heterogeneity present in the mantle sources (e.g., Stracke, 2021), and thus the $\mu^{182}\text{W}$ variability shown in Figures 3a–d likely represent a minimum estimate for the total range of $\mu^{182}\text{W}$ variability of their mantle sources.

Different Causes for ^{142}W and ^{142}Nd Variability, but Shared Process for Their Decrease

The ultimate source of negative $\mu^{182}\text{W}$ in plume-derived magmas, and the 10–20 ppm $\mu^{182}\text{W}$ decrease from the Archean

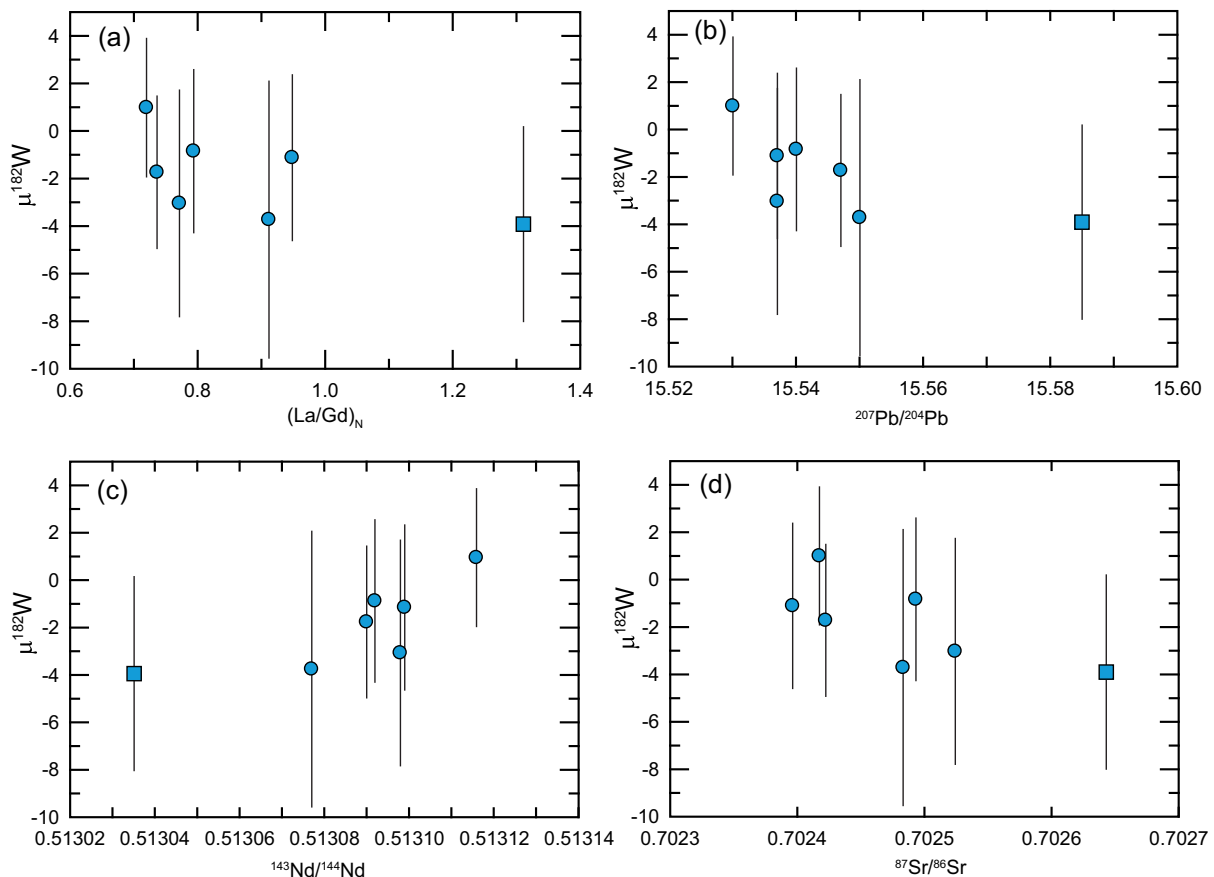


Figure 3 $\mu^{182}\text{W}$ vs. (a) normalised La/Gd ratio, and radiogenic (b) Pb, (c) Nd and (d) Sr isotope compositions, for the PAR MORB analysed here. Blue circles are N-MORB and square is T-MORB sample PAC2DR27-1. Tungsten isotope data from this study. Trace element concentrations and radiogenic isotope compositions from Hamelin *et al.* (2011).

to the modern mantle, has recently been the subject of active research. For example, chemical interaction between the core and the base of the mantle has been proposed to explain the negative $\mu^{182}\text{W}$ of plume-derived magmas (*e.g.*, Rizo *et al.*, 2019), given that the core is characterised by $\mu^{182}\text{W}$ of ~ -200 and W concentration of ~ 0.5 ppm (*e.g.*, Kleine and Walker, 2017). While the decrease in $\mu^{182}\text{W}$ was found to require unrealistically high time-integrated plume flux or unrealistically low $\mu^{182}\text{W}$ in the plumes (Peters *et al.*, 2021), recent studies propose viable models for the $\mu^{182}\text{W}$ decline taking into account W diffusion from the core into the mantle and the residence time of W at the core-mantle boundary (Kaare-Rasmussen *et al.*, 2023).

The $\mu^{142}\text{Nd}$ difference between the DMM and OIB sources (Fig. 2) shows that these reservoirs could have separated ~ 4.45 Gyr ago, consistent with the I-Xe interpretation (Mukhopadhyay, 2012). The coupled low $\mu^{182}\text{W}$ and low $\mu^{142}\text{Nd}$ in some ~ 3.6 Ga komatiites could represent remnant fingerprints of this differentiation (Puchtel *et al.*, 2016). Most ^{182}W and ^{142}Nd comparative studies, however, imply different causes for the isotope variability in mantle-derived magmas. Nevertheless, shared underlying processes could have driven the decrease in magnitude of ^{182}W and ^{142}Nd variations. Given the available data, the period of $\mu^{182}\text{W}$ decrease between 3 and 2.4 Ga (Tusch *et al.*, 2021; Nakanishi *et al.*, 2023) seems to coincide with the timing of the homogenisation of $\mu^{142}\text{Nd}$ heterogeneities due to mantle mixing (*e.g.*, Carlson *et al.*, 2019). The onset of deep cold slab subduction in the early Earth (Klein *et al.*, 2017) might have generated upwellings from the core-mantle boundary, carrying negative $\mu^{182}\text{W}$, which gets remixed into the convective mantle. The number of combined ^{182}W and ^{142}Nd isotope studies in the same rock samples is

limited. However, conducting similar additional investigations in Proterozoic rocks is crucial for enhancing our understanding of the timescales of mantle stirring and the geodynamic processes that have shaped Earth's modern mantle composition.

Acknowledgements

We acknowledge the generous support from the Carnegie Institution for Science Tuve Fellow Visiting Scientists program to H.R. and to J.O., allowing the writing of this manuscript. We thank Mike Walter, Peter van Keken and James W. Dotti III for helpful discussions, as well as the comments from Bradley Peters and an anonymous reviewer, which contributed to enhancing this manuscript. We are grateful to Pierre-Luc Lacroix for generating the code for the Monte Carlo bootstrap simulations. We thank the technical support provided by Shuangquan Zhang, Nimal DeSilva and Smita Mohanty at the IGGRC of Carleton University and the University of Ottawa Geochemistry Laboratory. This research was funded by the Natural Sciences and Engineering Research Council of Canada Discovery grant to H.R. (RGPIN-477144-2015) to J.O. (RGPIN 435589-2013), and the Ontario Early Researcher Award to H.R.

Editor: Helen Williams

Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2412>.





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Cite this letter as: Peters, D., Rizo, H., O'Neil, J., Hamelin, C., Shirey, S.B. (2024) Comparative ^{142}Nd and ^{182}W study of MORBs and the 4.5 Gyr evolution of the upper mantle. *Geochem. Persp. Let.* 29, 51–56. <https://doi.org/10.7185/geochemlet.2412>

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