Geochemical Perspectives Letters

Tungsten isotopes in Baffin Island lavas: Evidence of Iceland plume evolution

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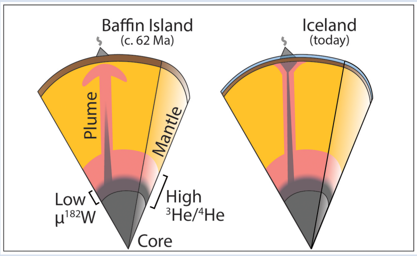
Abstract



Tungsten and helium isotope ratios in lavas derived from deeply rooted mantle plumes are tracers of lower mantle compositional heterogeneity or core–mantle exchange. We measured the tungsten isotopic compositions of lavas with exceptionally high-$^3He/^4He$ ratios that erupted above the head of the Iceland plume on Baffin Island. These lavas have $^{182}W/^{184}W$ ratios that are indistinguishable from the convecting upper mantle, unlike younger lavas in Iceland that have lower $^{182}W/^{184}W$ ratios. This implies that only the Iceland plume tail was infused with low-$^{182}W/^{184}W$ material, likely from the core. If high-$^3He/^4He$ helium also comes from the core, then diffusion across the core–mantle boundary may stratify plume-source mantle domains, with elevated $^3He/^4He$ travelling farther into the lower mantle than $^{182}W/^{184}W$ anomalies. Over Earth history, tungsten diffusion from the core can explain the decline of $^{182}W/^{184}W$ in the convecting mantle. We speculate that the uneven pace of this decline corresponds with evolving lower mantle dynamics.

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Introduction

Geochemical heterogeneities preserved in Earth since its formation place fundamental constraints on planetary accretion and long-term evolution. Mantle plumes that sample the deepest portions of the mantle contain isotopic evidence of ancient, >4.5 Gyr, geochemical reservoirs that survived the mixing caused by giant impacts during the final stages of planetary accretion and billions of years of mantle convection (Mundl-Petermeier *et al.*, 2019). Two competing models have emerged that might explain the preservation of these ancient heterogeneities: (1) the preservation of ancient gas-rich mantle domains (*e.g.*, Kurz *et al.*, 1982) and (2) core–mantle chemical exchange since planetary accretion (*e.g.*, Rizo *et al.*, 2019). To test these hypotheses, we measured the tungsten (W) isotopic composition of Baffin Island lavas erupted above the Iceland mantle plume, which contains high-$^3He/^4He$ ratios that have been uniquely well preserved since planetary formation.

The ^{182}Hf-^{182}W isotope system is a sensitive tracer of core–mantle interaction. During the first ~60 Myr of solar system history, ^{182}W was produced by the decay of the now extinct radionuclide ^{182}Hf ($t_{1/2} = 8.9$ Myr; Vockenhuber *et al.*, 2004). The upper terrestrial mantle has $\mu^{182}W \approx 0$ (where $\mu^{182}W = [(^{182}W/^{184}W)_{sample}/(^{182}W/^{184}W)_{standard} - 1] \times 10^6$), which is substantially higher than the average $\mu^{182}W$ of chondrites of approximately −190 (Kleine *et al.*, 2009). Because Hf is lithophile while W is moderately siderophile under reducing conditions (*e.g.*, Wade *et al.*, 2013), core formation increased the Hf/W of the mantle and left the metallic core with Hf/W near zero. The superchondritic $\mu^{182}W$ of the mantle thus most likely reflects core formation during the lifetime of ^{182}Hf, in which case the core has $\mu^{182}W$ lower than −190. Therefore, negative $\mu^{182}W$ values observed in some mantle plume-related magmas may be evidence of core–mantle exchange (*e.g.*, Rizo *et al.*, 2019). Alternatively, the lowermost mantle could host ancient isotopic heterogeneities, either resulting from early silicate differentiation events (*e.g.*, Touboul *et al.*, 2012) or introduced during the late accretion of chondritic material with low $\mu^{182}W$ relative to the terrestrial mantle (*e.g.*, Willbold *et al.*, 2011).

Intriguingly, some of the lowest $\mu^{182}W$ values have been measured in lavas with elevated $^3He/^4He$ compared to upper mantle values (greater than ~8 Ra, where Ra is the atmospheric ratio; *e.g.*, Mundl-Petermeier *et al.*, 2020). This suggests that $\mu^{182}W$ anomalies are associated with geochemical reservoirs that retain primordial ^3He trapped during planetary accretion before nebular gases dispersed, or ^3He that was accreted later from solar wind irradiated meteoritic material (Mukhopadhyay and Parai, 2019). Traditionally, high $^3He/^4He$ has been attributed to the preservation of primordial mantle domains, either in the entire lower mantle (*e.g.*, Kurz *et al.*, 1982) or within certain regions in the lower mantle (*e.g.*, Rizo *et al.*, 2016). Alternatively, high-$^3He/^4He$ helium concentrated in the core might escape and become entrained in mantle plumes (Bouhifd *et al.*, 2013). If so, core–mantle exchange may explain the observation that

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high-^{3}He/^{4}He ratios correlate with low μ^{182}W in some mantle plumes (Mundl-Petermeier *et al.*, 2019, 2020). As more data become available, however, the correlation between W and He isotopic compositions in modern ocean island basalts (OIBs) seems less universal.

Lavas erupted at c. 62 Ma on Baffin Island above the Iceland mantle plume have the highest ^{3}He/^{4}He of any measured terrestrial igneous rock (Horton *et al.*, 2023) and therefore contain an unusually pure primordial helium component. Thus, if high ^{3}He/^{4}He is sourced from the core, it might reasonably be expected that these lavas also exhibit low μ^{182}W. In this study, we reassess μ^{182}W in high-^{3}He/^{4}He lavas from Baffin Island because previous attempts to measure their W isotopic compositions have produced inconsistent results (Rizo *et al.*, 2016; Jansen *et al.*, 2022).

Results

We analysed glass picked from five Baffin Island pillow lavas containing olivine phenocrysts. Tungsten concentrations (20.9–107.0 ng g^{-1}) correlate with other highly incompatible and immobile elements, such as Th, but do not vary systematically with Sr, Nd, or Hf isotopic compositions (Fig. S-4). Weighted average μ^{182}W and μ^{183}W are -2.7 ± 6.6 and $+2.8 \pm 6.6$, respectively (2 s.d., $n = 5$; Table 1, Fig. 1). All μ^{182}W and μ^{183}W from individual samples are indistinguishable at the 2 s.e. confidence level from the Alfa Aesar and NIST SRM 3163 standards (Table S-1, Fig. S-5).

The lack of resolvable μ^{182}W anomalies in Baffin Island lavas agrees with the results published by Jansen *et al.* (2022) and from the stratigraphically similar lavas from West Greenland (Mundl-Petermeier *et al.*, 2019) but differs from the positive μ^{182}W anomalies reported by Rizo *et al.* (2016). Improvements in N-TIMS techniques, including the ability to quantify the oxygen isotopic compositions during tungsten oxide measurements (see Supplementary Information), give us confidence that these results are more representative of the Baffin Island mantle source than results published in Rizo *et al.* (2016), which should be interpreted with caution. Also, the absence of μ^{183}W anomalies in data reported here alleviates concerns raised by the Jansen *et al.* (2022) dataset about contamination and mass-independent fractionation.

Table 1 Tungsten isotopic compositions of the Baffin Island samples. μ^{182}W and μ^{183}W are reported as deviations from the Alfa Aesar standard (^{182}W/^{184}W = 0.864888 ± 0.000006 and ^{183}W/^{184}W = 0.467151 ± 0.000004, 2 s.e., $n = 8$) and normalised to ^{186}W/^{184}W, denoted by subscript 6/4. 2 s.e. represents the internal run precision of each individual analyses.

Sample	$\mu^{182}W_{6/4}$	2 s.e.	$\mu^{183}W_{6/4}$	2 s.e.
PING18-H16	−7.3	5.0	−0.6	4.5
PING18-H2	−0.6	4.2	2.2	3.5
PING18-H20	0.5	3.8	6.9	3.3
DURB18-H11	−1.7	6.0	4.3	5.1
RB18-H3	−5.3	4.8	−0.8	3.9

The helium isotopic compositions of Baffin Island lavas are well characterised (*e.g.*, Horton *et al.*, 2023; Table S-2) and have been reported for two samples analysed in this study: olivine crushing experiments for RB18-H3 and PING18-H2 imply minimum magmatic ^{3}He/^{4}He ratios of 36.8 ± 2.0 and 55.1 ± 1.3 Ra, respectively (Horton *et al.*, 2023). Olivine separates from the remaining three samples yielded insufficient helium for isotopic characterisation. These samples contained smaller olivines (<1 mm) than the helium-rich samples (>3 mm); we suspect that the low helium contents of the former reflect olivine growth after magma degassing. Nonetheless, the μ^{182}W results reported here are unambiguously associated with high-^{3}He/^{4}He lavas.

Discussion

The origins of the tungsten and helium in the Iceland plume. Our μ^{182}W results are unresolvable from the mantle and therefore do not require a core component in Baffin Island lavas source. Yet, the high-^{3}He/^{4}He helium and solar-like neon (Horton *et al.*, 2023) in these rocks and other lavas from the Iceland plume have presumably been preserved in Earth since the late stages of planetary accretion. On a global scale, rock samples from all 15 hotspots with anomalously low μ^{182}W also have anomalously high ^{3}He/^{4}He (Mundl-Petermeier *et al.*, 2020). This suggests a common origin of both elements in mantle plumes, such as primordial or ancient mantle, late accreted material, or the core.

Given the high W concentrations and positive μ^{182}W of Archean crust, small amounts of crustal assimilation could mask a core μ^{182}W signature. Assimilation modelling (see Supplementary Information) predicts correlations between μ^{182}W, trace elements, and long-lived radiogenic isotope ratios that are not observed in our data. This suggests that crustal assimilation is unlikely to have significantly influenced the W isotopic compositions of the Baffin Island lavas. Rather, the lack of μ^{182}W anomalies in Baffin Island high-^{3}He/^{4}He lavas indicates that helium and tungsten in plumes either (a) derive from a common source but are decoupled in the Iceland plume, or (b) have different origins. Either way, plumes

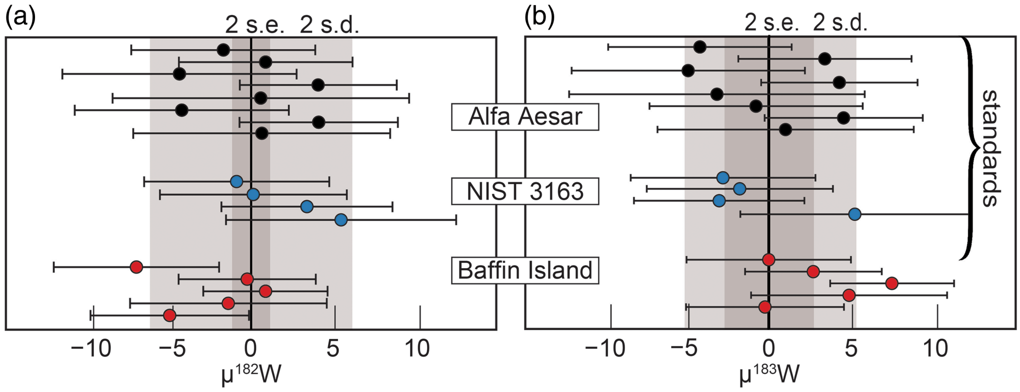

Figure 1 **(a)** μ^{182}W and **(b)** μ^{183}W for Alfa Aesar standard, NIST 3163 standard, and Baffin Island lavas.

appear to form in ways that produce systematic He-W correlations in many cases, but not universally.

The hypothesis that the high-^{3}He/^{4}He ratios are derived from primordial, non-degassed mantle in the Baffin Island lavas is inconsistent with other geochemical constraints, including our new μ^{182}W data. The primordial mantle likely had positive μ^{182}W, based on the positive μ^{182}W compositions of the Moon (Kruijer *et al.*, 2012; Touboul *et al.*, 2015) and mantle-derived rocks in the Archean (*e.g.*, Reimink *et al.*, 2020, and references therein). However, modern mantle plumes do not have positive μ^{182}W. Furthermore, the Baffin Island lavas have superchondritic ^{143}Nd/^{144}Nd and ^{176}Hf/^{177}Hf (Willhite *et al.*, 2019), suggesting they are not derived from a primordial mantle component but a differentiated mantle reservoir. These observations, combined with correlations between high ^{3}He/^{4}He and low μ^{182}W in many hotspots globally, imply the latter likely derive from a common deep Earth reservoir that is not primordial mantle.

Ancient differentiated mantle reservoirs that formed while ^{182}Hf was extant are similarly difficult to reconcile with coupled He-W isotopic compositions. Magma ocean silicate cumulates generated in the aftermath of the Moon-forming giant impact should be depleted in incompatible trace elements and might host high-^{3}He/^{4}He helium (Coltice *et al.*, 2011). If formed while ^{182}Hf was extant, cumulates would presumably acquire a positive μ^{182}W composition because W is more incompatible than Hf in silicate minerals (Righter and Shearer, 2003). Therefore, an ancient depleted mantle with high ^{3}He/^{4}He would be expected to have higher μ^{182}W than primordial mantle. Furthermore, silicate differentiation would have fractionated Sm from Nd, thereby influencing the abundances of ^{142}Nd—the product of ^{146}Sm decay ($t_{1/2} \approx 100$ Myr)—in the segregates. However, like most post-Archean mantle-derived rocks, Baffin Island (de Leeuw *et al.*, 2017) and Iceland lavas (Murphy *et al.*, 2010) lack ^{142}Nd/^{144}Nd anomalies. This indicates that the Iceland plume did not derive from mantle differentiated during the lifetimes of ^{146}Sm or ^{182}Hf.

Silicate differentiation within Hadean crust might have produced restite with low μ^{182}W decoupled from ^{142}Nd/^{144}Nd (Tusch *et al.*, 2022). However, the formation of, and subsequent magmatic differentiation within, Hadean crust would have caused extensive degassing of primordial gases. If so, Hadean crustal restites that foundered into the mantle would acquire low ^{3}He/^{4}He over time. Mantle plumes incorporating such material would acquire positively correlated ^{3}He/^{4}He and μ^{182}W, which is not observed.

Alternatively, hidden low-μ^{182}W mantle domains—perhaps formed during late accretion—are potential hosts of primordial ^{3}He. The μ^{182}W of the convecting mantle may have decreased by ~27 since Moon formation, based on lunar and Eoarchean terrestrial rock compositions (*e.g.*, Willbold *et al.*, 2011). About 0.5 wt. % of late accreting chondritic material with high W and highly siderophile element (HSE) concentrations but low μ^{182}W might explain the decline in mantle μ^{182}W (Willbold *et al.*, 2011). Mantle domains that contain an above average amount of late accreting material have been proposed as alternative sources of negative μ^{182}W (Marchi *et al.*, 2018). However, late accretion seems an unlikely common source of W and He because most late accreting material is expected to have high ^{3}He/^{4}He and to be HSE-rich, yet high-^{3}He/^{4}He helium in late accreting materials would not necessarily enter the mantle, and HSE abundances do not correlate with μ^{182}W or ^{3}He/^{4}He in mantle plumes (*e.g.*, Rizo *et al.*, 2019). Although estimating the mantle source HSE abundances from the HSE contents of erupted magmas is difficult, to our knowledge, no clear correlation between μ^{182}W and the HSE content has been demonstrated in spite of the wide range in μ^{182}W seen in OIBs.

Given the above discussion, combined with W isotope evidence from other mantle plumes, the negative μ^{182}W anomalies (as low as −12.9) in some Iceland high-^{3}He/^{4}He lavas might derive from the core (Mundl-Petermeier *et al.*, 2019). Even though the Baffin Island lavas do not have a μ^{182}W anomaly outside the analytical uncertainty, the external reproducibility of these samples still allow a small amount of bulk core material (~1.08 %) mixed with ambient mid-ocean ridge basalt (MORB)-like depleted mantle (see Supplementary Information, Fig. S-6). However, bulk mixing of this much core into the mantle is mechanically improbable and inconsistent with the Os concentrations in Iceland and Baffin Island lavas (Rizo *et al.*, 2016; Mundl-Petermeier *et al.*, 2019), which limit the amount of bulk core contribution to <0.1 %, assuming 2.83 μg g^{-1} Os in the core (Day, 2013).

Importantly, ^{3}He/^{4}He appears decoupled from not only W but also from lithophile elements, HSEs, and heavier noble gases in the Iceland plume (*e.g.*, Mundl-Petermeier *et al.*, 2019). This observation is consistent with helium diffusion into Iceland

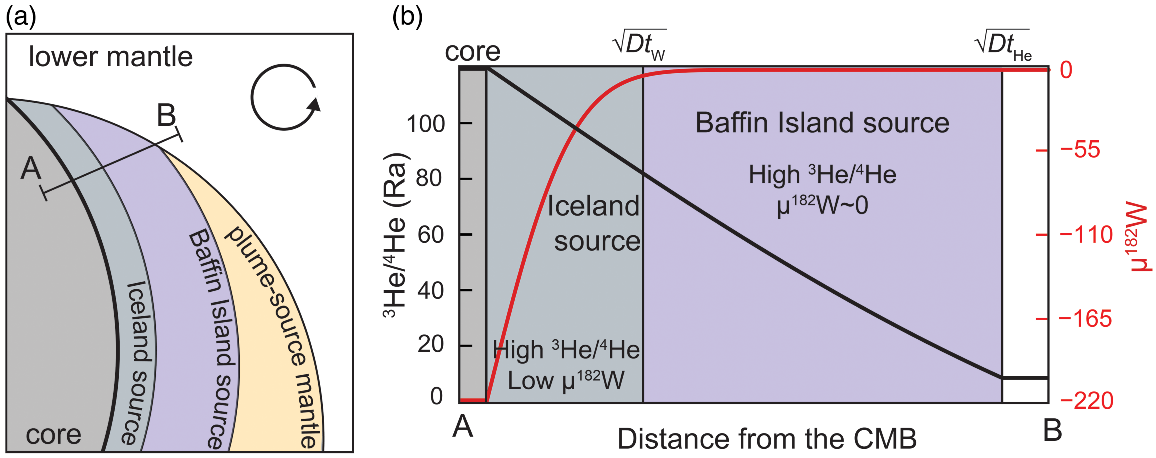

Figure 2 Conceptual model of a stratified Iceland plume source region. **(a)** On Gyr timescales, diffusion across the CMB may lower μ^{182}W and raise ^{3}He/^{4}He in the lowermost mantle on different length scales. **(b)** In such a scenario, there would be isotopic gradients in the lowermost mantle from the core ($\mu^{182}\text{W} = -200$, $^3\text{He}/^4\text{He} = 120$ Ra) to ambient mantle ($\mu^{182}\text{W} = 0$, $^3\text{He}/^4\text{He} = 8$ Ra). Iceland and Baffin Island mantle sources may derive from within and beyond the region impacted by W isotopic diffusion, respectively. Calculations for the isotopic composition curves are in the Supplementary Information and assume a diffusion timescale of 1 Gyr.

mantle from—rather than bulk mixing with—a ^{3}He-rich reservoir. Theoretically, He concentration gradients (Horton *et al.*, 2023) and W isotopic gradients (Ferrick and Korenaga, 2023) exist across the core–mantle boundary (CMB) that could drive diffusion into the mantle (Fig. 2). Alternatively, He and W diffused into the Iceland plume source from different reservoirs, such as ancient differentiated mantle and the core, respectively.

Helium may diffuse farther into mantle plume sources than W. Assuming the mantle plume source is stable on Gyr timescales (as expected for large low-shear wave velocity provinces, LLSVPs; *e.g.*, Ferrick and Korenaga, 2023), the characteristic length scales of diffusion ($\sqrt{Dt}$) for helium might be ~40 km, if theoretical diffusion rates for the upper mantle (Wang *et al.*, 2015) are extrapolated to lower mantle temperatures. For W, this length scale may be only 5–10 km (Ferrick and Korenaga, 2023), suggesting that mantle domains farther from the CMB may be characterised by He but not W isotopic anomalies. If anomalous W and He in the Iceland plume diffused from the core, our results imply that the plume head originated farther from the CMB than the plume tail.

Baffin Island mantle may have originated from the periphery of a helium-infused zone in the lowermost mantle (10–40 km from the core), whereas low-μ^{182}W Iceland mantle might have resided nearer the core (Fig. 2). Negative μ^{182}W anomalies may only exist in the Iceland plume tail, which may have preferentially entrained denser material proximal to the core–mantle boundary (Jones *et al.*, 2019). The plume head, from which Baffin Island lavas derived, may have instead entrained primarily portions of the lowermost mantle beyond the diffusion limit of W but still infused with ^{3}He from the core. Thus, plume tails might be the most efficient conveyors of material from the CMB.

This model predicts that ^{3}He/^{4}He is highest in lavas with the most negative μ^{182}W, a trend observed for all high-^{3}He/^{4}He hotspots, except the Iceland plume (Jackson *et al.*, 2020). Maximum ^{3}He/^{4}He in Iceland plume lavas apparently declined from >65 Ra at 62 Ma (Horton *et al.*, 2023) to <26 Ra in the neovolcanic zones of Iceland (Harðardóttir *et al.*, 2018). This decline could be due to the incorporation of convecting upper mantle into the Iceland plume—enough to explain up to a 40 % MORB component in modern Iceland lavas—as a result of the ridge-centred plume position (Shorttle and Maclennan, 2011). The addition of this much material from the convecting upper mantle would have moderated ^{3}He/^{4}He and μ^{182}W, implying that the Iceland plume itself currently exhibits μ^{182}W closer to −21.

Implications for planetary accretion and convecting mantle evolution. By combining μ^{142}Nd and μ^{182}W constraints with our diffusion model, an early Earth chronology emerges. Some Eoarchean rocks have ^{142}Nd anomalies (*e.g.*, Caro *et al.*, 2003) produced by igneous differentiation that fractionated Sm and Nd prior to 4 Ga. Such differentiation could be expected to also fractionate Hf from W, so any differentiation that occurred before ^{182}Hf became extinct would have produced μ^{182}W anomalies that would correlate with μ^{142}Nd variations (Touboul *et al.*, 2012). However, correlations between μ^{142}Nd and μ^{182}W are rarely observed. Instead, these observations seem consistent with: (i) a Moon-forming impact after ^{182}Hf was extinct (~4.5 Ga) that homogenised the silicate portions of the Earth–Moon system, (ii) generation of ^{142}Nd/^{144}Nd anomalies in the mantle by differentiation events that post-dated the Moon-forming impact that were subsequently erased by mixing after the extinction of ^{146}Sm around 4 Ga, and (iii) μ^{182}W decline in the mantle until present caused by CMB diffusion.

The inferred mantle μ^{182}W decline since the Hadean requires that the average residence time (τ) of material diffused from the core at the CMB was ≤30 Myr (Fig. 3a). Due to the higher temperatures in early Earth, early mantle convection may have been rapid. Fast convection would also have efficiently homogenised the mantle and, hence, efficient W isotopic transfer across the CMB (Hadean rapid convection path, Fig. 3b). However, μ^{142}Nd and positive μ^{182}W heterogeneities throughout the mantle persisted at least until the end of the Archean, and potentially even for longer (Slowing mantle convection path, Fig. 3b). Perhaps the μ^{182}W of the mantle rapidly decreased during the late Archean to early Proterozoic, coinciding with development of continents and therefore a liminal stage of mantle dynamics. Alternatively, CMB cover by long-term stable structures may have increased in the late Archean (*i.e.* increasing ξ), inhibiting transfer of core-derived W to the convecting mantle. Either way, this transition suggests a link between continent formation and lower mantle dynamics during the initiation of modern plate tectonics.

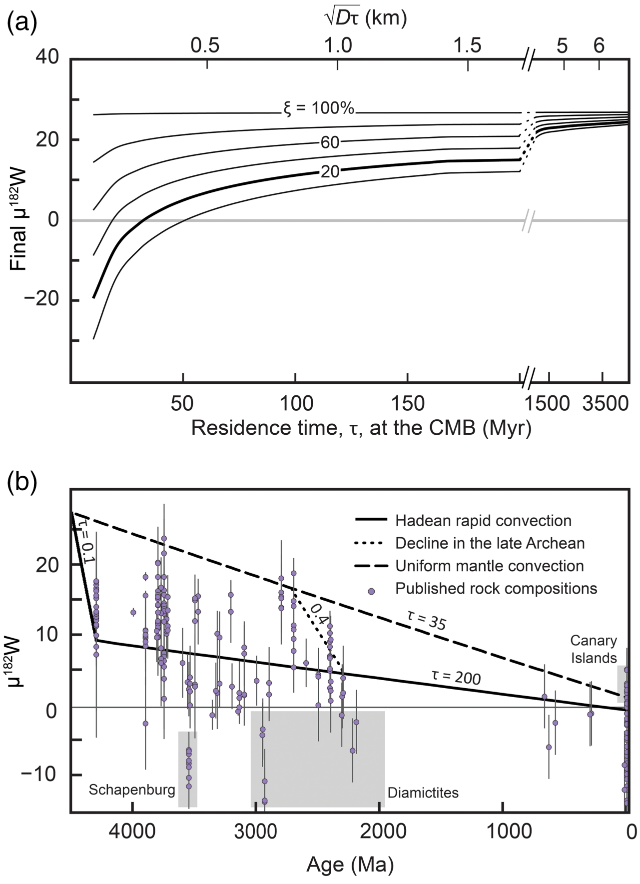

Figure 3 **(a)** Modern mantle μ^{182}W may be a function of the mantle residence time, τ, at the CMB and the percentage of the core surface area, ξ, insulated by long-term stable structures. We assume that (i) the early mantle had μ^{182}W of the Moon (+27); (ii) basal mantle acquired core-like μ^{182}W corresponding to the characteristic length scale of diffusion ($\sqrt{D\tau}$), where *D* is the diffusivity ($D = 4.62 \times 10^{-10}$ m^2 s^{-1}; Ferrick and Korenaga, 2023); and (iii) core-infused mantle parcels mixed efficiently into the bulk mantle. Diffusion can explain a μ^{182}W decline to zero if τ is short (<30 Myr) and the CMB is <20 % insulated. **(b)** A compilation of μ^{182}W data (Reimink *et al.*, 2020; Nakanishi *et al.*, 2023) and three potential mantle μ^{182}W trajectories: (i) constant τ = 35 Myr; (ii) fast Hadean decline (τ = 0.1 Myr) corresponding to rapid Hadean convection followed by slower decline (τ = 200 Myr); and (iii) fast early Paleoproterozoic decline (τ = 0.4 Myr) due to a change in the style of plate tectonics.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2337.

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