182W Evidence for Long-Term Preservation of Early Mantle Differentiation Products
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Over the past two decades, the short-lived $^{182}\text{Hf}-{^{182}\text{W}}$ isotopic system ($^{182}\text{Hf} \rightarrow ^{182}\text{W} + \beta^-$, half-life = 8.9 million years) has been widely used for dating early solar system processes because of the unique geochemical properties of the system (1–3). As a moderately siderophile element (MSE), W is largely, but not completely, extracted from the silicate portions of planetary bodies during segregation of metallic cores [e.g., (4–6)]. Hafnium, in contrast, is lithophile and is essentially retained in silicate portions of planetary bodies. Therefore, determination of the abundance of the daughter nuclide $^{182}\text{W}$, relative to other stable, nonradiogenic isotopes (e.g., $^{184}\text{W}$), is of special interest for constraining the timing of planetary core formation. For example, although Hf and W are both refractory elements and are presumed to occur in chondritic relative abundances in bulk planetary objects, most terrestrial rocks have $^{182}\text{W}/^{184}\text{W}$ ratios that are approximately 200 parts per million (ppm) higher than bulk chondrites (7–9). The higher than chondritic $^{182}\text{W}/^{184}\text{W}$ ratio of the terrestrial mantle has been interpreted to reflect core segregation and generation of suprachondritic Hf/W in the mantle during the first ~30 million years of solar system history, while $^{182}\text{Hf}$ was still extant (9). The isotopic difference between the mantle and chondritic meteorites, together with mass balance constraints, also implies that Earth’s core is a W-rich reservoir characterized by a $^{182}\text{W}/^{184}\text{W}$ ratio that is ~220 ppm lower than the terrestrial mantle.

After the cessation of substantial core segregation, the Hf/W ratio of Earth’s mantle would likely have been further modified by post-core formation events, such as the crystallization of transient magma oceans, partial melting of the mantle, or subsequent crystal-liquid fractionation processes (10). If such events occurred before $^{182}\text{Hf}$ became extinct, additional $^{182}\text{W}$ isotopic variations would have been generated in the mantle. It is even likely that $^{182}\text{W}$ isotopic heterogeneities were created in the mantle after $^{182}\text{Hf}$ was no longer extant—for example, as a result of late accretion. The term “late accretion” is used to describe the process of addition of ~0.3 to 0.8% of the total mass of the mantle (11) to Earth by continued accretion of materials with bulk chondritic compositions, subsequent to cessation of core formation. It is a process that has been commonly invoked to account for the relatively high absolute and chondritic relative abundances of the highly siderophile elements (HSEs: including Re, Os, Ir, Ru, Rh, Pt, Pd, and Au) present in the mantle (12–15). Late accretion would have lowered the $^{182}\text{W}^{184}\text{W}$ ratio of the mantle by 10 to 30 ppm, as materials with comparatively $^{182}\text{W}$-depleted compositions, such as planetesimals with bulk chondritic compositions, were accreted to Earth. Tungsten isotopic anomalies in mantle domains could also have been generated throughout Earth’s history as a result of interactions with the core.

Given these possibilities for heterogeneity, the isotopic composition of W in the mantle may complement data from other radiogenic isotope systems, such as the short-lived $^{146}\text{Sm}-{^{142}\text{Nd}}$ isotope system [e.g., (16–18)], for use in discerning the timing and nature of early Earth differentiation processes, as well as the detection of core-mantle exchange. The possibility of identifying these important processes has led to intensive W isotopic analysis of diverse terrestrial materials (19–21). Until recently, these studies have reported no resolved $^{182}\text{W}$ isotopic heterogeneities among terrestrial rocks. Willbold et al. (22), however, reported $13 \pm 4$ ppm $^{182}\text{W}$ enrichments, relative to younger terrestrial rocks and standards, in rocks dated at 3.8 billion years ago (Ga) from the Isua greenstone belt, Greenland. This study provided the first evidence for W isotopic heterogeneity in terrestrial materials. The rocks analyzed were also characterized by enrichments in $^{142}\text{Nd}$.

**Tungsten isotope heterogeneities in the Archean komatiites.** We recently developed a technique to measure $^{182}\text{W}/^{184}\text{W}$ to a 2 σ (SD) precision of ±4.6 ppm by means of negative thermal ionization mass spectrometry (23). This technique has enabled us to begin a high-resolution search for W isotopic anomalies in diverse terrestrial rocks. Here, we present W concentration and isotope composition data for 3.47 Ga komatiites from the Kostomuksha Formation of the Barberton greenstone belt, South Africa, and 2.82 Ga komatiites from the Kostomuksha greenstone belt, Baltic Shield, Russia (Table 1) (24).

For the Komati and Kostomuksha komatiites, W concentrations vary from 28 to 102 ppb and from 45 to 106 ppb, respectively. In neither suite do W concentrations correlate well with chemical indicators of crystal-liquid fractionation typical of komatiitic systems, such as the MgO content (fig. S2). This may indicate complex behavior of W within crystallizing flows and/or minor mobility of W within the flows at some point after lava solidification.

The $\mu^{182}\text{W}$ values (where $\mu$ is the deviation, in ppm, from terrestrial reference standards) for four Komati samples average $+2.7 \pm 4.1$ (2σ SD) and are indistinguishable from the average for the terrestrial standards and the modern La Palma basalt (Canary Islands) that we repeatedly analyzed during the analytical campaign (Fig. 1). In contrast, the 18 Komati komatiite samples analyzed (including one to five replicate digestions of the same samples) have an average $\mu^{182}\text{W}$ of $+15.0 \pm 4.8$ (2σ SD), which is well resolved from the terrestrial standard average (Fig. 1). In addition to differences in location, age, and W isotopic composition, the mantle sources of these two komatiite suites were also characterized by widely differing HSE concentration and Os isotopic systematics. The mantle source of the Komati komatiites has been estimated (25) to have had ~50% of the total HSE abundances of the modern primitive mantle (26). This is consistent with the observations of Maier et al. (27), who reported Pt and Ru contents in komatiites ranging in age between 3.5 and 2.9 Ga and discovered what appears to be a progressive increase in Pt concentrations in lavas from the early to late Archean. They attributed this increase to be the result of sluggish downward mixing of a HSE-rich veneer of late-accreted...
materials to Earth. The Komati source was also characterized by an essentially chondritic initial $\gamma^{187}$Os of $+0.23 \pm 0.12$ (24) (where $\gamma$ denotes the percent deviation of the initial Os isotopic composition of the source of the lava from that of a chondritic reference at that time), consistent with common chemical evolution models for Os in the early upper mantle [e.g., (28)]. The initial $\varepsilon^{143}$Nd of $+1.9 \pm 0.7$ [compiled from (29)] for the Kostomuksha suite, (where $\varepsilon$ denotes the deviation in parts per 10,000 of the isotopic composition of the source of the lava from that of a chondritic reference at that time) suggests a source characterized by long-term depletion in Nd relative to Sm. This isotopic composition is generally consistent with standard models of the evolution of $^{146}$Nd in the early Earth [e.g., (30, 31)]. No $^{142}$Nd data are currently available for the Kostomuksha komatiites.

In contrast to the Komati source, the mantle source of the Kostomuksha komatiites has been estimated to have had total HSE abundances that are ~80% of those present in the modern primitive mantle [32]. Puchtel et al. (33) reported coupled enrichments in both $^{187}$Os (initial $\varepsilon^{187}$Os = $+2.55 \pm 0.13$) and $^{186}$Os (initial $\mu^{186}$Os = $+2.8 \pm 6$). Thus, in comparison to the Komati komatiites, the Kostomuksha komatiites were derived from a mantle source characterized by long-term suprachondritic Hf/W, Re/Os, and Pt/Re ratios. The Os isotopic characteristics were interpreted by Puchtel et al. to reflect an early onset of Earth’s inner-core crystallization. This resulted in suprachondritic Pt/Os and Re/Os in the liquid outer core, creation of coupled $^{186,187}$Os enrichments over time, and transfer of this outer-core Os isotopic signature to the mantle source subsequently tapped by the Kostomuksha plume. The Sm-Nd isotopic systematics of the Kostomuksha komatiites, however, are not anomalous, with initial $\varepsilon^{143}$Nd of $+2.8 \pm 0.2$ (34) and initial $\mu^{142}$Nd of $-0.5 \pm 4.0$ (17). Such values are generally consistent with common chemical evolution models for the early upper mantle [e.g., (17)].

The combined W and HSE abundance data, as well as W, Os, and Nd isotopic systematics of the Komati and Kostomuksha komatiites, can be used to place some important constraints on the origin of the MHE and HSE in the deep sources of the two komatiite suites. First, given the consistency of the W isotopic data and W concentrations in the analyzed multiple lava flows from the Kostomuksha suite, it is unlikely that the enrichment in $^{182}$W is the result of contamination of parental magmas with the crust or secondary alteration of the lavas, even if we assume that the $^{182}$W-enriched crust could somehow have been formed. Further, the lithophile trace element data and Nd- and Pb-isotope systematics indicate no detectable crustal contribution to either komatiite system (29, 34). Hence, we interpret both the Komati and Kostomuksha komatiites to have W isotope compositions representative of their respective, deep mantle sources.

Second, the positive $\mu^{182}$W anomaly in the mantle source of the Kostomuksha komatiites cannot be due to the contribution of an outer-core component, as was initially suggested to explain the coupled $^{186,187}$Os enrichments present in the system (33). The observed W anomaly in the Kostomuksha komatiites is in the opposite direction ($\mu^{182}$W = $+15.0 \pm 4.8$) to that of a core component ($\mu^{182}$W $\approx -220$).

Third, Willbold et al. (22) concluded that the $^{182}$W-enriched isotopic composition of the Isua suite preserves the composition of the mantle prior to a final stage of late accretion to Earth and the Moon, termed the terminal cataclysm, or late heavy bombardment (35). This event has been previously hypothesized to have occurred between ~3.9 and 3.8 Ga, as assessed from ages of lunar impact melt rocks associated with the major impact basins [e.g., (36)]. Willbold et al. noted that the addition of materials of chondritic bulk composition ($\mu^{182}$W = 200) that were rich in W, as well as HSE, would have lowered the W isotopic composition of the bulk mantle from an older, more radiogenic composition. Although the 13 ppm enrichment in $^{182}$W in rocks from Isua is similar in magnitude to that in the source of the Kostomuksha komatiites, the interpretation that Willbold et al. proposed for the Isua rocks is problematic for the Kostomuksha rocks, and below we show why this is the case.

**Coupled late accretion and mantle mixing model.** In order to explain the observed W, Os, and Nd isotopic systematics for the Komati and Kostomuksha sources, we model mixing between modern mantle and a primordial, $^{182}$W-enriched, yet HSE-depleted, reservoir (Fig. 2). Total HSE content is expressed as the deviation of content in the mantle source, relative to the concentration in the modern mantle. The effects of contributions of late-accreted materials ranging from 0.3 to 0.8% of the total mass of the mantle are calculated, assuming that the HSE present in the silicate Earth today were derived entirely from late-accreted materials. Beginning with a radiogenic $^{182}$W value and zero total HSE content prior to late accretion, this model shows that Earth’s mantle would have evolved toward its present W isotopic composition, as increasingly more chondritic materials were accreted and the total HSE content increased to the present-day values. This

<table>
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<th>Table 1. Tungsten concentrations (in ppb) and isotopic compositions of 3.47 Ga Komati (South Africa) and 2.82 Ga Kostomuksha (Russia) komatiites, and the modern La Palma basalt LP15 (Canary Islands).</th>
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<tbody>
<tr>
<td><strong>Sample</strong></td>
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<td>Average (2σ SD, n = 4)</td>
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<td>La Palma basalt</td>
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<td>Average (2σ SD, n = 3)</td>
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<td>Alfa Aesar W standard (2σ SD, n = 40)</td>
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scenario is broadly similar to those advocated by Willbold et al. (22) and Maier et al. (27). Given the estimates that the Komatit and Kostomuksha komatiite sources contained ~50 and ~80%, respectively, of the late accretionary component present in the modern mantle (25, 32), the older Komatit komatiites should show more radiogenic $^{182}\text{W}$ than the Kostomuksha komatiites (Fig. 2). This is not observed, which suggests that the Kostomuksha source cannot simply be primordial mantle that had been largely stripped of HSE by core formation.

**Early metal-silicate equilibration model.** We next present two approaches for explaining the $^{182}\text{W}$ enrichments, as well as other isotopic and elemental characteristics, of the Kostomuksha komatiites. For the first approach, we assume that $^{182}\text{W}$, $^{186}\text{Os}$, and $^{187}\text{Os}$ excesses were generated by the same process. This would mean that the Kostomuksha source included an early-formed component that was characterized by high Hf/W, as well as high Re/Os and Pt/Os, relative to the present-day mantle. One possible mechanism for the attainment of these chemical characteristics would be metal-silicate partitioning at high temperatures and pressures, such as might be found at the base of a primordial transient magma ocean or in a partially molten zone at or near the core-mantle boundary. Transient magma oceans, as well as more limited molten regions at the core-mantle boundary, have been proposed to explain both geochemical and geophysical observations (e.g., [37]). Here, we envision a very early, deep mantle location where silicate melt could potentially equilibrate with metal that is either the growing core, or metal that is passing through the molten region on its way to the core. This approach (24) is based on experimental observations of substantially reduced metal-silicate partitioning for Re and Pt, relative to Os, at high temperatures and pressures, compared to relatively low temperatures and pressures of the upper mantle or crust. It is also based on the more siderophile behavior of W under moderately reducing conditions with respect to the present upper mantle.

In contrast to simple downward mixing of late-accreted materials proposed by Maier et al. (27), our model requires the long-term survival of some portion of the mantle whose HSE and MRE were set by metal-silicate partitioning under reducing conditions at high temperature and pressure. In order to achieve the isotopic and elemental characteristics of the Kostomuksha source, we envision a plume tapping this liquid or solid reservoir and mixing material from this reservoir with overlying mantle characterized by HSE concentrations and Os isotopic characteristics similar to those in estimates for primitive mantle. The HSE characteristics of the dominant mantle reservoir would be best explained by late accretion. This reservoir would also serve as an appropriate source for the Komatit komatiites. A good fit to the $^{187}\text{Os}$ and $^{182}\text{W}$ isotopic characteristics of the Kostomuksha komatiites can be achieved by mixing ~53% material of the early-formed reservoir with 47% material of the overlying mantle (Fig. 3 and table S6). Because the major fractionations that occur in this model result from metal-silicate interaction, the processes envisioned would have had no predictable effect on the lithophile element isotopic systems, including the Sm-Nd system.

**Early magmatic differentiation model.** A second approach to account for the $^{182}\text{W}$ enrichment in the mantle is to focus entirely on late differentiation processes, such as magma ocean crystallization or partial melting (24). Silicate crystal-liquid fractionation processes can induce Hf/W fractionation, and hence potentially could have resulted in the production of mantle reservoirs with $^{182}\text{W}$ excesses or deficits. In general, both Hf and W behave as incompatible elements and concentrate into silicate melts. In the upper mantle, Hf and W abundances are largely controlled by the presence of clinopyroxene and garnet, both of which preferentially incorporate Hf relative to W (10). Therefore, fractional crystallization of silicate melts and partial melting of
silicate rocks at upper-mantle conditions produce solids with high Hf/W ratios relative to melts. Strong Hf/W fractionation is also expected in the lower mantle, as Hf is less incompatible than W in Mg-perovskite, a lower-mantle phase (38). Thus, Mg-perovskite would be characterized by higher Hf/W relative to equilibrium melts. Hence, any cumulate reservoir formed as a result of magma ocean crystallization, or any mantle depleted by melting and crustal extraction, would be characterized by high Hf/W ratios—and would develop positive $^{182}$W anomalies—if isolated from the convecting portion of the mantle while $^{182}$Hf was still extant.

Clinopyroxene, garnet, and Mg-perovskite are also phases that control rare earth element (REE) fractionation in Earth’s mantle (table S7) (24). All of these mineral phases are typically depleted in Nd relative to Sm, such that, in addition to their higher Hf/W ratio, solids in equilibrium with melts at lower- and upper-mantle conditions would also have had a higher Sm/Nd ratio. Therefore, it might be expected that if the Kostomuksha W isotopic anomaly is related to early silicate differentiation, the enriched $^{182}$W should also be accompanied by an excess in $^{142}$Nd. Yet, although Kostomuksha komatiites have $^{142}$Nd values that are identical to Earth’s modern mantle within analytical uncertainty (17), a contribution from an early differentiated mantle component cannot be summarily excluded. This is because the magnitude of Hf-W fractionation induced by partial melting and crystallization is likely much larger than the magnitude of Sm-Nd fractionation. Hence, early igneous processes could potentially result in substantial $^{182}$W anomalies without a resolvable effect on $^{142}$Nd.

To model this type of scenario, we consider two early differentiation end members. The first, similar to the differentiation model considered in Willbold et al. (22) to account for the Isua data, assumes that Hf-W fractionation is coupled to the global differentiation event advocated by Boyet and Carlson (17) for explaining the 20 ppm enrichment in $^{142}$Nd of the accessible silicate Earth relative to chondrites. At ~4.53 Ga, bulk mantle characterized by chondritic Sm/Nd and supra-chondritic Hf/W would have differentiated into two complementary reservoirs: an early enriched reservoir (EER), characterized by lower Sm/Nd and Hf/W ratios, and an early depleted reservoir (EDR) with higher ratios. Partial remixing of the EER back into the EDR could have resulted in the generation of the modern accessible mantle with appropriate Sm/Nd and Hf/W ratios that translate into the observed present-day $^{142}$Nd and $^{182}$W values of 0. Consistent with this model, positive $^{182}$W anomalies in early Archean samples from Isua [7 to 15 ppm (16, 18)] are accompanied by a $^{182}$W anomaly of +13 ± 4 ppm (22). Hence, if both isotopic anomalies reflect the involvement of remnants of the EER, a $^{182}$W anomaly that is considerably smaller than 15 ppm would be expected when there is no resolvable $^{142}$Nd enrichment. Within analytical uncertainties, Kostomuksha komatiites have identical $^{182}$W to the Isua samples, so if this interpretation is correct for the Isua samples, this global mantle differentiation model is difficult to reconcile with the combined data for the Kostomuksha komatiites.

A second mantle differentiation model explores the effects of early partial melting of the mantle with a present-day $^{182}$W = 0 and $^{142}$Nd = 0, as opposed to the parental reservoir with chondritic $^{142}$Nd = −20, considered in the early global differentiation model. This parental reservoir, with current $^{142}$Nd isotope compositions that are identical to the modern upper mantle, corresponds to bulk mantle, assuming an initial Earth with supra-chondritic Sm/Nd (16), or to an already depleted mantle, if Earth was instead characterized by chondritic Sm/Nd and experienced early global mantle differentiation, as proposed by Boyet and Carlson (17). The Hf-W and Sm-Nd fractionations induced by partial melting are estimated using a batch melting model and published partition coefficients for these elements between constituent mantle phases and silicate melt (table S7) (24). For this model, the $^{182}$W and $^{142}$Nd values of the Kostomuksha komatiites are most consistent with a mantle source that was residual to partial melting of the lower mantle earlier than ~4.52 Ga, assuming that the degree of melting remained lower than ~4% (Fig. 4) (24).

![Fig. 3.](image)

Fig. 3. (A and B) Plot of $^{182}$W versus (A) $^{186}$Os and (B) $^{187}$Os, illustrating mixing between the modern primitive mantle and a mantle reservoir preserved from an early magma ocean, where Re/Os, Pt/Os, and Hf/W ratios were established by metal-silicate equilibration at high pressure and temperature, resulting in extreme $^{187,186}$Os and $^{182}$W enrichments at 2.82 Ga. For this model, the coupled $^{186}$Os and $^{182}$W excesses in the source of the Kostomuksha komatiites (solid circle) can be explained by a 53% contribution from the isotopically enriched reservoir to the ambient mantle source (open circle).
Fig. 4. Coupled $^{142}$Nd-$^{182}$W evolution model of depleted reservoirs formed from a parental reservoir with $^{182}$W and $^{142}$Nd values of 0, corresponding to the isotopic compositions of the modern upper mantle (open circle). Dashed lines are loci of equal differentiation time after the formation of calcium and aluminum–rich inclusions (CAI). $\Delta$m$_{CAI}$ [in millions of years (Myr)], marking the beginning of the solar system. Solid curves are the loci of depletion with identical degrees of partial melting, F (in percent). This model assumes partial melting of a lower mantle with 80% Mg-perovskite, 15% magnesiowüstite, and 5% Ca-perovskite. Partition coefficients for Hf, W, Sm, and Nd in Mg- and Ca-perovskite used are reported in table S7. Note that mantle depletion earlier than 4.52 Ga can result in the generation of a resolvable $^{182}$W anomaly without a detectable $^{142}$Nd anomaly.

dilution factor of $\sim$5, as necessary for explaining the high HSE abundances in the source of the Kostomuksha komatiites, the most consistent mantle differentiation model involves $\sim$2% partial melting of the lower mantle at $\sim$4.55 Ga, which then produced a reservoir with $^{182}$W of $\sim$60 and $^{142}$Nd of $\sim$2.

For Isua rocks, the model favored by Willbold et al. (22), whereby the mantle source corresponds to a portion of the mantle preserved from late accretion, cannot be excluded at present because the HSE concentrations in the source of these rocks are not yet determined. We note, however, that our models of metal-silicate equilibration and magmatic differentiation could also successfully account for the apparent coupling of the positive $^{182}$W anomaly of Isua rocks with, respectively, the $^{187}$Os [initial $^{187}$Os/$^{186}$Os = +4.2 ± 1.2 (40)] and $^{142}$Nd excess ($^{142}$Nd = +7 to +15 (16, 18)) also present in the Isua rocks. Although the mantle sources of the Isua rocks and Kostomuksha komatiites could have experienced similar early processes, they must sample distinct mantle reservoirs, which formed under different conditions (as they are characterized by different $^{187}$Os and $^{142}$Nd excesses), despite having similar $^{182}$W anomalies.

Implications for early mantle dynamics. We conclude that the positive $^{182}$W anomaly of the Kostomuksha komatiite source does not correspond to the composition of the dominant Hadean mantle prior to the addition of extraterrestrial materials via late accretion. Instead, it must record a very early differentiation event, which could be related to either metal-silicate equilibration or mantle differentiation processes. Combined investigations of HSE abundances and $^{186,187}$Os, $^{182}$W, and $^{142}$Nd systematics of other komatiites derived from mantle sources with variable Sm/Nd ratios may ultimately permit discrimination between the different models presented here.

Regardless of the true cause of the W anomaly, the generation of these komatiites at 2.8 Ga attests to the long-lived nature of an early $^{182}$W-enriched mantle reservoir, and highlights the evident sluggish mixing of at least some portions of the mantle throughout the Hadean and Archean. Dense cumulate piles, crystallized from a basal magma ocean [e.g., (37)], may constitute good candidates for storage and preservation of $^{182}$W heterogeneities over at least 1.7 billion years. If generation of this primordial component predated the formation of the Moon (in excess of 4.60 million years after formation of the solar system (39)), it can be concluded that the putative Moon-forming giant impact, although highly energetic, did not induce complete homogenization of Earth’s mantle. This would most likely mean that Earth was not completely melted by the event.

References and Notes

24. See supporting material on Science Online.

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Supporting Online Material

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SOM Text
Figs. S1 to S4
Tables S1 to S7
References (43–57)

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