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Highly siderophile element and  $^{182}\text{W}$  evidence for a partial late veneer in the source of 3.8 Ga rocks from Isua, GreenlandChristopher W. Dale<sup>a,\*</sup>, Thomas S. Kruijer<sup>b</sup>, Kevin W. Burton<sup>a</sup><sup>a</sup> Department of Earth Sciences, Durham University, Durham, DH1 3LE, UK<sup>b</sup> Institut für Planetologie, University of Münster, Wilhelm-Klemm-Str. 10, 48149 Münster, Germany

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## ABSTRACT

The higher-than-expected concentrations of highly siderophile elements (HSE) in Earth's mantle most likely indicate that Earth received a small amount of late accreted mass after core formation had ceased, known as the 'late veneer'. Small  $^{182}\text{W}$  excesses in the Moon and in some Archaean rocks – such as the source of 3.8 billion-year-old Isua magmatics – also appear consistent with the late veneer hypothesis, with a lower proportion received. However,  $^{182}\text{W}$  anomalies can also relate to other processes, including early mantle differentiation. To better assess the origin of these W isotope anomalies – and specifically whether they relate to the late veneer – we have determined the HSE abundances and  $^{182}\text{W}$  compositions of a suite of mafic to ultramafic rocks from Isua, from which we estimate HSE abundances in the source mantle and ultimately constrain the  $^{182}\text{W}$  composition of the pre-late veneer mantle.

Our data suggest that the Isua source mantle had HSE abundances at around 50–65% of the present-day mantle, consistent with partial, but not complete, isolation from the late veneer. These data also indicate that at least part of the late veneer had been added and mixed into the mantle at the time the Isua source formed, prior to 3.8 Ga. For the same Isua samples we obtained a  $13 \pm 4$  ppm  $^{182}\text{W}$  excess, compared to the modern terrestrial mantle, in excellent agreement with previous data. Using combined  $^{182}\text{W}$  and HSE data we show that the Moon, Isua, and the present-day bulk silicate Earth (BSE) produce a well-defined co-variation between  $^{182}\text{W}$  composition and the mass fraction of late-accreted mass, as inferred from HSE abundances. This co-variation is consistent with the calculated effects of various late accretion compositions on the HSE and  $^{182}\text{W}$  signatures of Earth's mantle. The empirical relationship, therefore, implies that the Moon, Isua source and BSE received increasing proportions of late-accreted mass, supporting the idea of disproportional late accretion to the Earth and Moon, and consistent with the interpretation that the lunar  $^{182}\text{W}$  value of  $27 \pm 4$  ppm represents the composition of Earth's mantle before the late veneer was added. In this case, the Isua source can represent ambient mantle after the giant moon-forming impact, into which only a part of Earth's full late veneer was mixed, rather than an isotopically distinct mantle domain produced by early differentiation, which would probably require survival through the giant Moon-forming impact.

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## 1. Introduction

Late accretion is the period of material addition thought to follow the giant Moon-forming impact and the completion of core formation on Earth (e.g., Walker, 2009). The principal evidence for such late material addition comes from the concentrations of highly siderophile elements (HSE: e.g. Os, Pt, Re, Au) in Earth's mantle, which are higher than expected for metal–silicate equilibration during core formation (Brenan and McDonough, 2009;

Mann et al., 2012). Moreover, the HSE in the mantle occur in broadly chondritic relative proportions, which is unexpected for metal–silicate partitioning (Meisel et al., 1996; Becker et al., 2006). These observations have led to the idea that the elevated HSE abundances in Earth's mantle reflect the late addition of broadly chondritic material (the 'late veneer') to the mantle after core formation was complete (e.g. Kimura et al., 1974).

Recently, the extinct  $^{182}\text{Hf}$ – $^{182}\text{W}$  decay system ( $t_{1/2} = 8.9$  Ma), which so far has mainly been used as a chronometer of core formation (Kleine et al., 2009), has emerged as a new tool with which to investigate late accretion (Willbold et al., 2011, 2015; Kruijer et al., 2015; Touboul et al., 2015). During core formation, lithophile Hf separated from moderately siderophile W, causing

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Hf/W fractionation which, following decay of  $^{182}\text{Hf}$ , led to distinct  $^{182}\text{W}$  signatures in the mantle and core of planetary bodies. For instance, the present-day bulk silicate Earth (BSE) exhibits a higher  $^{182}\text{W}/^{184}\text{W}$  by  $\sim 2\varepsilon$ -unit ( $1\varepsilon = 0.01\%$ ) relative to chondrites, indicating that at least some part of Earth's core formed during the lifetime of  $^{182}\text{Hf}$  (Kleine et al., 2002; Schoenberg et al., 2002; Yin et al., 2002; Kleine et al., 2004). The  $^{182}\text{W}/^{184}\text{W}$  difference between Earth's mantle and chondrites not only provides constraints on the timing of core formation, but can also be used as a tracer of late-accreted material. This is because the addition of a late veneer with a composition and mass as inferred from HSE systematics lowered the  $^{182}\text{W}/^{184}\text{W}$  of Earth's mantle by  $\sim 0.1$  to  $\sim 0.4\varepsilon$ -units (Willbold et al., 2011; Touboul et al., 2012, 2015; Kruijjer et al., 2015). Several recent studies have identified  $^{182}\text{W}$  excesses of this magnitude in some Archaean samples and, in some cases, these signatures have been attributed to derivation of these samples from sources that lack some portion of the late veneer (Willbold et al., 2011, 2015). However, the origin of these  $^{182}\text{W}$  excesses is not fully understood and in other cases have been ascribed to very early differentiation events within Earth's mantle, rather than late accretion (Touboul et al., 2012, 2014; Rizo et al., 2016b).

One problem with the interpretation of the  $^{182}\text{W}$  data is that until now no sample has been identified for which  $^{182}\text{W}$  and HSE data provide consistent estimates for the amount of late-accreted material in the sample source. Elevated  $\varepsilon^{182}\text{W}$  of  $\sim 0.13$  ( $\varepsilon^{182}\text{W} = (^{182}\text{W}/^{184}\text{W}_{\text{sample}}/^{182}\text{W}/^{184}\text{W}_{\text{terrestrial standard}} - 1) \times 10^4$ ) in metabasalts and gneisses from the Isua supracrustal belt (ISB), Greenland, were attributed by Willbold et al. (2011) to an absence of late veneer, but HSE data, which would provide an independent estimate, were not available. More recently, Rizo et al. (2016b) used combined  $^{182}\text{W}$  and HSE data for mafic and ultramafic rocks to argue that the  $^{182}\text{W}$  enrichments in the Isua region were more likely produced by an early mantle differentiation event. Kostomuksha komatiites (Baltic Shield, Russia) have an  $\varepsilon^{182}\text{W}$  excess of  $\sim 0.15$ , while Komati komatiites (Barberton greenstone belt, South Africa) have no resolvable excess, yet the HSE contents of these samples suggest that the fraction of late-accreted material in the Kostomuksha source is substantially higher than in the Komati source, inconsistent with a late accretion origin for the  $^{182}\text{W}$  anomalies (Touboul et al., 2012). One possibility is that the HSE and  $^{182}\text{W}$  systematics are decoupled, with W and HSE derived from different sources. For samples from the Nuvvuagittuq greenstone belt (Quebec, Canada), Touboul et al. (2014) argued for subduction-triggered addition of  $^{182}\text{W}$ -enriched fluids to a mantle source containing the full complement of late-accreted material. The origin of the  $^{182}\text{W}$  enrichment in these fluids remains unclear, however, and could be related to either a lower proportion of late veneer or early differentiation. In contrast, the constancy of  $^{182}\text{W}$  excesses ( $\sim +0.15$ ; Isua, Kostomuksha, Nuvvuagittuq and also Acasta) has been interpreted to reflect a global signature of a pre-late veneer mantle, enriched in HSE by giant impactor core material (Willbold et al., 2015). Recently, negative  $\varepsilon^{182}\text{W}$  has been reported – the first such finding – in Schapenburg komatiites (Barberton greenstone belt), and this appears coupled with depletions in  $^{142}\text{Nd}$ , suggesting a signature of early mantle differentiation (Puchtel et al., 2016). Finally, elevated  $\varepsilon^{182}\text{W}$  has been found for the first time in Phanerozoic samples (Rizo et al., 2016a), but the much larger magnitude of the anomaly (up to  $\varepsilon^{182}\text{W} = +0.5$  in Baffin Island picrites) is difficult to reconcile with any currently existing interpretations. Thus, overall, the origin of  $^{182}\text{W}$  enrichments in terrestrial samples remains unclear and could either relate to late accretion, early differentiation, or both, or possibly even reflect heterogeneities produced during the main phase of accretion itself. Yet, while it is likely that at least some of the  $^{182}\text{W}$  excesses observed for Archaean samples are due to late accretion, so

far there is no single sample set for which this has been independently demonstrated.

Two recent studies have shown that the Moon exhibits an  $\varepsilon^{182}\text{W}$  excess of  $\sim 0.22$  to  $\sim 0.27$  compared to the present-day BSE (Kruijjer et al., 2015; Touboul et al., 2015). This excess is in good agreement with the  $^{182}\text{W}$  difference expected for disproportional late accretion to the Earth and Moon and with the overall calculated effect of late accretion on the  $^{182}\text{W}$  composition of Earth's mantle. These two studies, therefore, concluded that the  $^{182}\text{W}$  composition of the Moon is indistinguishable from that of the pre-late veneer terrestrial mantle. This  $^{182}\text{W}$  homogeneity is unexpected, however, given that the Moon is thought to consist of a mixture of impactor and proto-Earth material (e.g., Canup, 2004), both of which should be characterised by distinct  $^{182}\text{W}$  compositions compared to the composition of Earth's mantle immediately after the giant impact (Kruijjer et al., 2015). As such it is important to constrain the pre-late veneer  $^{182}\text{W}$  composition of Earth's mantle without relying solely on the analyses of lunar samples. For example, if the lunar  $^{182}\text{W}$  excess indeed represents the composition of the pre-late veneer BSE, then terrestrial samples with  $\varepsilon^{182}\text{W} \sim +0.15$  should derive from mantle sources that contain about half of the late veneer.

Here we present combined HSE concentrations (Os, Ir, Ru, Pt, Pd, Re),  $^{182}\text{W}$  and  $^{187}\text{Os}$  data for 3.7–3.8 Ga samples from the Itsaq gneiss complex (south-west Greenland). The samples include pillow basalts and amphibolites from, respectively, the Isua supracrustal belt (ISB) and the adjacent orthogneiss terrain to the south (see Nutman and Friend, 2009) including several which have previously been identified as possessing  $^{182}\text{W}$  excesses (Willbold et al., 2011). In addition,  $^{182}\text{W}$  data and HSE concentrations are also presented for previously unstudied ultramafic rocks from the orthogneiss terrain, which arguably provide the most reliable estimate of mantle source HSE concentrations to date, as their major element composition and HSE proportions most closely approach those of the mantle itself. From these data we use the HSE concentrations to determine the proportion of late accretion received by the Isua mantle source, and combine this with W isotope data in order to place constraints on the formation of  $^{182}\text{W}$  anomalies and the history of late accretion to Earth.

## 2. Samples and methods

### 2.1. Samples

Pillow basalts, amphibolites and ultramafic samples were collected from the early Archaean 3.7–3.8 Ga Itsaq gneiss complex, south-west Greenland, by Stephen Moorbath in 1998 and 2000. Pillow basalts were derived from the eastern arm of the Isua supracrustal belt (ISB) itself, while amphibolites and ultramafic rocks were sampled from numerous enclaves or pods in the adjacent  $\sim 3.8$  Ga orthogneiss terrain to the south. Specifically, the ultramafic samples came from several enclaves just to the south of Lake 682 m (see Fig. 8 in Nutman and Friend, 2009), from the same region postulated to contain mantle dunites and harzburgites (Friend et al., 2002; Rollinson, 2007), although their exact origin remains unresolved. They are distinct from previously studied ultramafics in that they do not come from the ISB itself (cf. Szilas et al., 2015; Rizo et al., 2016b), and they arguably provide the most robust mantle estimate due to their mantle-like HSE patterns. Three pillow basalts and one amphibolite have previously been analysed for  $^{182}\text{W}$ , but not HSE (Willbold et al., 2011). In addition, data is also presented for two samples of younger 3.4 Ga Ameralik metadolerite dykes from within the Itsaq gneiss complex, collected by Robin Gill (localities RG24 and RG247). This provides a temporal constraint on the evolution of  $^{182}\text{W}$  anomalies in this region.

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