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Tungsten isotopic constraints on the origin and evolution of the Moon

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1 **Origin and evolution of the Moon, Tungsten**
2 **isotopic constraints**

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22 **1 Introduction**

23 The Moon most likely formed as the result of a collision between the proto-Earth and a
24 differentiated body possibly the size of Mars (Cameron and Benz, 1991; Hartmann and Davis, 1975).
25 The enormous amount of energy released by this giant impact caused widespread melting on the proto-
26 Earth and the ejection of material into Earth's orbit from which the Moon subsequently accreted. Upon
27 accretion of the Moon, the lunar mantle underwent global silicate differentiation most likely facilitated
28 by a lunar magma ocean (e.g. Wood et al., 1970). Magma ocean crystallization likely produced the wide
29 diversity of lunar source rocks and involved the successive crystallization of mafic cumulates consisting
30 of olivine and pyroxene, followed by crystallization of plagioclase which floated to the lunar surface to
31 form the lunar crust consisting of ferroan anorthosites (FAN). Finally, the residual liquid of the lunar
32 magma ocean represents a separate component within the Moon termed KREEP (enriched in Potassium
33 K, Rare Earth Elements, and Phosphorous). Re-melting of these magma ocean crystallization products
34 of the low-Ti and high-Ti mare basalt source regions as well as the formation of Mg-suite lunar highland
35 rocks.

36 Although a giant impact followed by magma ocean differentiation can account for most
37 geochemical observations of lunar rocks, the origin and subsequent evolution of the Moon remain hotly
38 debated. Nevertheless, our understanding of lunar evolution has been greatly enhanced by isotopic
39 studies of lunar samples. In this chapter, we will review key insights about the evolution of the Moon
40 as provided by isotope studies of lunar samples based on the element tungsten (W). Below we will first
41 revisit the general principles of the short-lived ^{182}Hf - ^{182}W system and then present a brief historical
42 overview of W isotope studies on lunar samples. Finally, we will present several key findings based on
43 W isotopes regarding the timescales of lunar magma ocean differentiation, late accretion onto the Earth
44 and Moon, and the origin of the Moon.

45 **2 Key principles: Tungsten isotopes and the Hf-W chronometer**

46 Tungsten ($Z=74$) is a refractory element with five stable isotopes (^{180}W , ^{182}W , ^{183}W , ^{184}W and ^{186}W).
47 The radioactive decay of now extinct ^{182}Hf ($t_{1/2} \sim 8.9$ million years, Ma) to ^{182}W provides a chronometer
48 for dating chemical processes that fractionate Hf and W during the first ~ 60 Ma of solar system history
49 (see e.g. Kleine and Walker, 2017 and references therein). This time interval is particularly suited for
50 studying the accretion and differentiation histories of planetary bodies in the inner solar system. Given
51 that W, a moderately siderophile element, is largely partitioned in the metallic core of a planet, and Hf,
52 a lithophile element, is retained in the silicate mantle, strong Hf/W fractionations occur during metal-
53 silicate separation. Thus, following metal-silicate separation, the mantle and core of a planetary body
54 develop markedly distinct ^{182}W signatures over time (Fig. 1). As such, the Hf-W system is ideally suited
55 for examining the timescales of planetary core formation. Moreover, because of the different
56 partitioning of Hf and W during silicate differentiation (Righter and Shearer, 2003), additional ^{182}W
57 variations can be generated during early mantle differentiation processes such as magma ocean
58 crystallization and crust formation (see e.g. Kleine and Walker, 2017). Finally, variations in ^{182}W may
59 not only be used to study the timescales of core formation and silicate differentiation on planetary
60 bodies, but, as will be shown below, can also provide key insights into the origin and evolution of the
61 Moon.

62 **3 Tungsten isotope studies of lunar samples**

63 Over the past decades several studies have focused on determining the W isotope composition of lunar
64 samples. The first of these found large and variable ^{182}W excesses in different lunar source lithologies
65 (Lee et al., 1997). These ^{182}W variations were initially interpreted to result from radioactive decay of

68 ^{182}Hf within the Moon, in which case the Moon would have to have formed and differentiated within
69 the lifetime of ^{182}Hf , that is, within the first ~ 60 Ma of the solar system. However, subsequently it was
70 recognized that ^{182}W variations among lunar samples at least to some degree are caused by
71 superimposed secondary neutron capture effects resulting from the interaction of the lunar samples with
72 galactic cosmic rays (Leya et al., 2000). As most lunar samples have been exposed to galactic cosmic
73 rays for an extended time period, these effects can potentially be very large. The governing neutron
74 capture reaction, $^{181}\text{Ta}(\text{n},\gamma)^{182}\text{Ta}(\beta^-)^{182}\text{W}$, illustrates that such neutron capture effects not only depend
75 on the neutron fluence of a sample but also on its Ta/W, leading to overall excesses in $^{182}\text{W}/^{184}\text{W}$. To
76 account for these cosmogenic effects, subsequent W isotope studies of lunar samples employed
77 different approaches to account for the effects of neutron capture (Kleine et al., 2005; Lee et al., 2002).
78 One successful approach has been the analyses of lunar metal samples, which do not contain any Ta-
79 derived cosmogenic ^{182}W (Kleine et al., 2005; Touboul et al., 2007). Using this approach applied to
80 metal separates from mare basalts and KREEP-rich breccias, Touboul et al. (2007) found that there are
81 no resolvable ^{182}W variations within the Moon, and that the ^{182}W composition of the Moon is
82 indistinguishable from that of terrestrial rock standards (Fig. 2).

83 In recent years analytical improvements invoked another revival of W isotope studies in lunar
84 samples. Tungsten isotope analyses utilizing multi-collector inductively coupled plasma mass
85 spectrometry (MC-ICPMS) or thermal ionization mass spectrometry (TIMS) now routinely provide a
86 measurement precision on W isotope ratios of ~ 5 parts-per-million (2σ); this is a five to ten-fold
87 improvement in measurement precision compared to earlier studies. Accordingly, recent studies on
88 lunar samples have employed high-precision W isotope measurements combined with novel ways to
89 quantify the effects of secondary neutron capture. In particular, Kruijer et al. (2015) performed high-
90 precision W isotope measurements on KREEP-rich samples by MC-ICPMS and used Hf isotopes as an
91 empirical dosimeter to quantify the effects of secondary neutron capture. Touboul et al. (2015)
92 performed high-precision W isotope measurements by NTIMS and analyzed large metal fractions of
93 KREEP-rich samples to circumvent cosmogenic effects. Despite using different measurement
94 techniques and approaches to quantify cosmogenic effects, both studies identified a small excess in
95 $\mu^{182}\text{W}$ of *ca.* +26 ppm for KREEP-rich samples relative to the Earth's mantle (where $\mu^{182}\text{W}$ is the parts-
96 per- 10^6 deviation in $^{182}\text{W}/^{184}\text{W}$ from terrestrial standard values) (Fig. 2). Note that these new $\mu^{182}\text{W}$
97 results are consistent with the earlier metal data from Touboul et al. (2007), but are significantly more
98 precise, explaining why such ^{182}W excesses were not detected in the earlier study. In a subsequent study,
99 Kruijer and Kleine (2017) found that low-Ti and high-Ti mare basalts, as well as Mg suite norite 77215
100 and lunar meteorite Kalahari 009 also exhibit similar excesses in $\mu^{182}\text{W}$ (Fig. 3). Taken together, these
101 data therefore imply that (i) the bulk silicate Moon exhibits a uniform excess in $\mu^{182}\text{W}$ of *ca.* +26 ppm
102 relative to the present-day bulk silicate Earth (Fig. 3), and (ii) that there is currently no evidence for
103 resolvable ^{182}W variations within the lunar mantle, even at a very high level of precision.
104

105 4 Key research findings

106 4.1 Timescales of lunar differentiation

107 Crystallization of the lunar magma ocean led to distinct mantle reservoirs with markedly different
108 Hf/W, where the source of high-Ti mare basalts is thought to be characterized by the highest (Hf/W of
109 ~ 40 –80) and KREEP by the lowest ratios (Hf/W ~ 10 –20) (e.g., Kleine et al., 2005; Righter and Shearer,
110 2003; Touboul et al., 2007). Hence, if magma ocean crystallization occurred within the lifetime of ^{182}Hf ,
111 then these reservoirs should have evolved to distinct $\mu^{182}\text{W}$ over time. As such, the $\mu^{182}\text{W}$ compositions
112 of different lunar source lithologies can be used to shed light on the timescales for the solidification of
113 the magma ocean. However, all lunar samples for which cosmogenic ^{182}W effects have been quantified,

114 including low-Ti and high-Ti mare basalts, KREEP, Mg-suite norites and lunar meteorites, have
115 indistinguishable pre-exposure $\mu^{182}\text{W}$ values (Fig. 3). The homogeneous $\mu^{182}\text{W}$ of lunar rocks despite
116 the large range of source Hf/W ratios demonstrates that lunar differentiation occurred after the effective
117 lifetime of ^{182}Hf , more than ~ 70 Ma after solar system formation, *i.e.* later than ~ 4.5 Ga (Kruijer and
118 Kleine, 2017). This timescale is consistent with the 4.35–4.37 Ga ages derived for the major period of
119 lunar differentiation using other dating methods. For instance, both the Sm–Nd isochron ages for lunar
120 crustal rocks, the Sm–Nd and Lu–Hf model ages of KREEP, and the average Sm–Nd model age of the
121 mare basalt sources, as well as a peak in Pb–Pb ages observed in lunar zircons all appear to converge
122 at 4.35–4.37 Ga (see summary in Borg et al., 2015 and references therein). Collectively, these relatively
123 young ages, combined with the lack of ^{182}W heterogeneity in the Moon, support the idea that the major
124 period of differentiation on the Moon occurred relatively late. Note that such a late differentiation of
125 the Moon only marginally overlaps the results of a recent Hf isotope study on lunar zircons which
126 suggested that lunar differentiation may have occurred earlier, within *ca.* 60 Ma after CAIs (Barboni et
127 al., 2017). The exact reason for this apparent discrepancy remains unclear, and resolving this issue will
128 require a better understanding of the significance of the old inferred ages for individual lunar zircon
129 grains versus the model ages inferred for large geochemical reservoirs on the Moon.
130

131 4.2 Origin of ^{182}W excess in the Moon

132 All lunar rock types investigated so far exhibit a uniform ^{182}W excess of *ca.* +26 ppm relative to the
133 present-day bulk silicate Earth. This ^{182}W difference can in principle have three different origins. One
134 option is that the ^{182}W excess is radiogenic in origin. This would require that the Earth and the Moon
135 have different Hf/W and that the Moon formed within the lifetime of ^{182}Hf . However, the Hf/W of both
136 the terrestrial and lunar mantles are not well defined, and as such it is currently unclear if these ratios
137 are sufficiently different to produce a resolvable ^{182}W difference. Moreover, independent estimates for
138 the age of the Moon strongly suggest that the Moon formed after the life-time of ^{182}Hf , making it quite
139 unlikely that the ^{182}W excess of the Moon is radiogenic in origin. The second possibility is that the ^{182}W
140 difference was generated during the giant impact and reflects a larger fraction of impactor material
141 within the Moon. As discussed below (Section 4.3), such an ^{182}W excess is in fact predicted in the giant
142 impact model for lunar origin. A corollary of this is that identifying any potential radiogenic ^{182}W excess
143 in the Moon is merely impossible, because there is currently no way to distinguish such a signature
144 from an ^{182}W difference generated during the giant impact.

145 The third possibility to explain the ^{182}W difference between the Moon and the present-day bulk
146 silicate Earth is disproportional late accretion. Late accretion is defined as the ‘late’ addition of on
147 average broadly chondritic material to the lunar and terrestrial mantles following the formation of the
148 Moon and the end of core formation on Earth. Evidence for this ‘late veneer’ comes from the inferred
149 abundances of highly siderophile elements (HSE) in the mantles of the Earth and the Moon. For both
150 bodies, the HSE mantle abundances are higher than expected for metal-silicate equilibration during core
151 formation. If the late veneer hypothesis is correct, then addition of such a late veneer would have led to
152 a $\mu^{182}\text{W}$ difference between the Earth and the Moon, because (i) the late veneer material had a different
153 $\mu^{182}\text{W}$ value to that of the bulk silicate Earth, and (ii) HSE evidence suggests that proportionally more
154 late veneer material was added to the Earth than to the Moon (e.g. Day and Walker, 2015). The studies
155 by Kruijer et al. (2015) and Touboul et al. (2015) both showed that the $\sim +26$ ppm ^{182}W excess in the
156 Moon is remarkably consistent with the expected ^{182}W difference resulting from disproportional late
157 accretion, with a total mass and composition inferred from HSE systematics (Fig. 4). Thus, the $\mu^{182}\text{W}$
158 anomaly in the Moon does not only provide strong support for the late veneer hypothesis, but also
159 implies that the HSE budget of the bulk silicate Earth was set after the giant impact and core formation,
160 implying that any previously accumulated HSE in the proto-Earth’s mantle have been removed by metal

161 segregation during the giant impact. In detail, the magnitude of the effect of late accretion on $\mu^{182}\text{W}$
162 depends on the mass and composition assumed for the late veneer as well as on the W concentration of
163 the pre-late veneer BSE. As such the calculated pre-late veneer $\mu^{182}\text{W}$ of the BSE may have been
164 between *ca.* +10 and +50 ppm (Kleine and Walker, 2017). Thus, the \sim +26 ppm ^{182}W excess of the Moon
165 is well within the range of expected compositions for the pre-late veneer BSE, meaning that no
166 resolvable $\mu^{182}\text{W}$ difference between the pre-late veneer BSE and the Moon remains once the effects of
167 late accretion are taken into account. As a result, there is currently neither strong evidence for a $\mu^{182}\text{W}$ difference between
168 the Earth and the Moon.
169

171 **4.3 Constraints on the origin of the Moon**

172 The close agreement between the predicted late-veneer-induced $\mu^{182}\text{W}$ shift and that observed
173 between the present-day BSE and the Moon implies that, prior to addition of the late veneer, the Earth's
174 mantle and the Moon had indistinguishable $\mu^{182}\text{W}$ values. This $\mu^{182}\text{W}$ similarity between the Earth and
175 the Moon is consistent with the isotopic homogeneity observed for other elements such as Ti, Si, and
176 O (e.g., Wiechert et al., 2001; Zhang et al., 2012; Armytage et al., 2012). The latter might reflect that
177 the proto-Earth and the impactor derive from an isotopically homogeneous reservoir (e.g., Dauphas et
178 al., 2014) or that the Moon formed either from proto-Earth mantle material (e.g., Ćuk and Stewart,
179 2012), or from equal portions of the mantles of two colliding half-Earths (Canup, 2012). However, such
180 mechanisms cannot easily account for the similarity in $\mu^{182}\text{W}$. This is because unlike Ti, Si and O, $\mu^{182}\text{W}$
181 variations do not reflect a particular mix of presolar and solar nebula components in the precursor
182 materials of the Earth and impactor, but instead reflect radiogenic ingrowth in the mantles of the proto-
183 Earth and impactor following core formation in these bodies. As the accretion and core formation
184 histories of the proto-Earth and impactor were likely different, the mantles of these two bodies should,
185 therefore, have different ^{182}W compositions. In the context of the giant impact model the Moon can be
186 considered a three-component mixture consisting of (i) impactor mantle, (ii) impactor core, and (iii)
187 proto-Earth mantle. Given that these components likely each have distinct $\mu^{182}\text{W}$ and are mixed in
188 random proportions during the giant impact, producing similar $\mu^{182}\text{W}$ between the Earth's mantle and
189 the Moon seems very unlikely (Kruijer et al., 2015). Nevertheless, Dauphas et al. (2014) used an
190 inversion model to illustrate that the lunar $\mu^{182}\text{W}$ composition can be reproduced for $\mu^{182}\text{W}$ and Hf/W
191 values of the impactor mantle that are expected for planetary embryos that formed early in Solar System
192 history. Similarly, Wade and Wood (2016) showed that, provided that the Moon predominantly derives
193 from the proto-Earth's mantle, the lunar $\mu^{182}\text{W}$ composition can be explained by a giant impact of a
194 strongly reduced impactor onto an oxidized proto-Earth. Nevertheless, while the above two studies
195 demonstrate that the $\mu^{182}\text{W}$ similarity between the Earth and the Moon can in principle be produced in
196 a giant impact, they have to resort to happenstance to account for the close similarity in $\mu^{182}\text{W}$ between
197 the Earth and Moon.

198 To better assess the significance of the nearly identical ^{182}W compositions of Earth's mantle and the
199 Moon immediately after the giant impact, Kruijer and Kleine (2017) calculated the expected ^{182}W
200 composition of the Moon in various giant impact scenarios using a mixing model. This was done by
201 randomly varying several key parameters, including the metal-silicate partition coefficients for W in
202 the Earth and the impactor, the time of core formation in the impactor, the impactor-to-Earth mass ratio
203 and the fraction of impactor core material in the Moon. The results of these calculations demonstrate
204 that the Earth–Moon ^{182}W similarity is an unlikely outcome of any giant impact (Fig. 5), which
205 regardless of the amount of impactor material incorporated into the Moon should have generated a
206 significant ^{182}W excess in the Moon (most likely more than +100 ppm). Conversely, the probability of
207 producing indistinguishable $\mu^{182}\text{W}$ is very small, only \sim 5% if the Moon predominantly consists of proto-

208 Earth material and ~1% if the Moon largely derives from the impactor (Fig. 5). As a result, regardless
209 of which giant impact scenario is assumed, the $\mu^{182}\text{W}$ similarity is an unlikely outcome of the giant
210 impact. Consequently, post-giant impact processes modifying the $\mu^{182}\text{W}$ value of the Moon seem to be
211 required. This can be accomplished if the post giant impact state of the Earth–Moon system facilitated
212 efficient isotopic homogenization, even for refractory elements like W (Pahlevan and Stevenson, 2007;
213 Zhang et al. 2012), as may be the case for high-energy, high angular momentum giant impacts (e.g.
214 Lock and Stewart, 2017). Alternatively, the lunar $\mu^{182}\text{W}$ value was modified by large, secondary
215 impact(s) on the Moon after the giant impact.

217 5 Conclusions and outlook

218 Tungsten isotope studies on lunar samples demonstrate that there are no resolvable ^{182}W within the
219 Moon, implying that lunar differentiation occurred later than ~70 Ma after Solar System formation.
220 Nevertheless, the Moon is characterized by a uniform ^{182}W excess over the present-day BSE, which is
221 most easily explained by disproportional late-accretion onto the Earth and Moon. After accounting for
222 this effect, the ^{182}W signatures of the pre-late veneer BSE and Moon are indistinguishable, however.
223 This ^{182}W similarity is an unlikely outcome of the giant impact, which should have generated a
224 significant ^{182}W difference between the Earth and Moon. Thus, post giant impact processes appear
225 required that modified the lunar ^{182}W composition.

226 To better understand the ^{182}W similarity between the Earth and the Moon future studies should focus
227 on better constraining the composition of the late veneer as well as the relative masses of late accretion
228 added to the Earth and Moon. For instance, it will be important to assess whether the low HSE
229 abundances inferred for the lunar mantle truly reflect a low influx of late accreted material or instead
230 internal processes on the Moon. Another research avenue that should be explored are secondary
231 processes that may have modified the ^{182}W value of the Moon such as large secondary impacts. If the
232 core of such an impactor directly merged with the lunar core, then such secondary impacts may have
233 modified the ^{182}W of the lunar mantle without disturbing its HSE record. Finally, obtaining independent
234 estimates for the pre-late veneer ^{182}W composition of the Moon would be desirable. Such estimates can
235 potentially be derived by studying the ^{182}W systematics of terrestrial rocks with variable contributions
236 of late veneer material.

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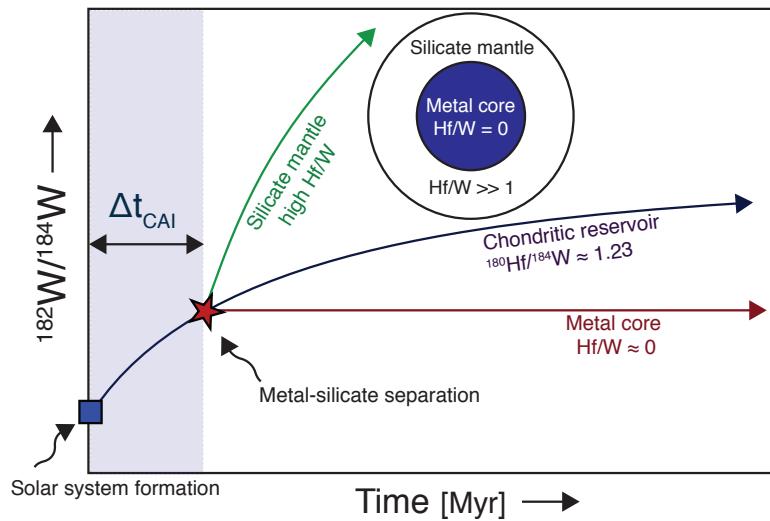
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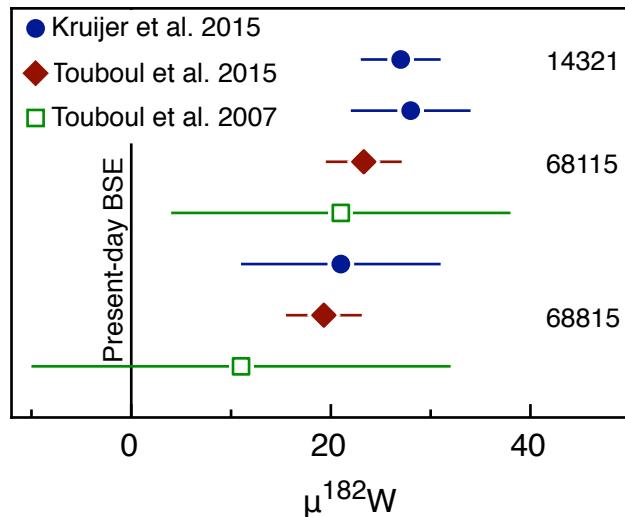
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Figures:



298 **Fig. 1:** Explanatory diagram showing $^{182}\text{W}/^{184}\text{W}$ vs. time with evolution lines for reservoirs with distinct
299 $^{180}\text{Hf}/^{184}\text{W}$. Upon metal-silicate separation (red star) in a planetary body at a given time after Solar
300 System formation (blue square), the silicate mantle (high Hf/W) follows a steeper trajectory than the
301 chondritic reservoir, whereas the metal core ($\text{Hf/W} \approx 0$) retains the $^{182}\text{W}/^{184}\text{W}$ of the bulk body acquired
302 at the time core formation. Shaded area denotes the time interval between a metal-silicate separation
303 event and Solar System formation, as defined by the formation of Ca-Al-rich inclusions (CAIs).

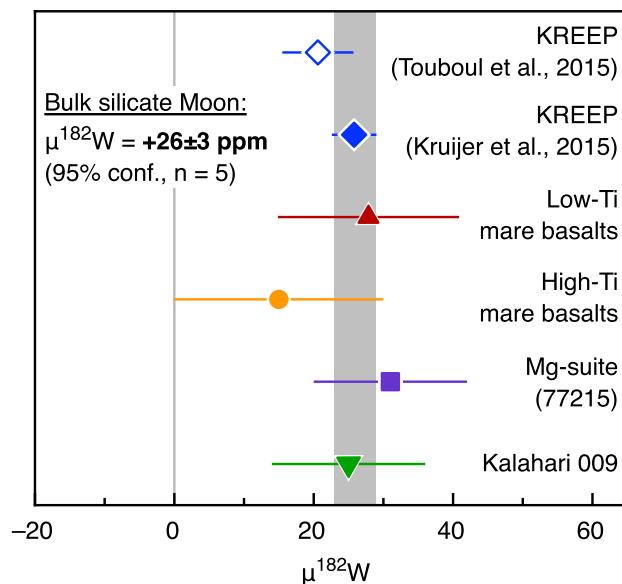
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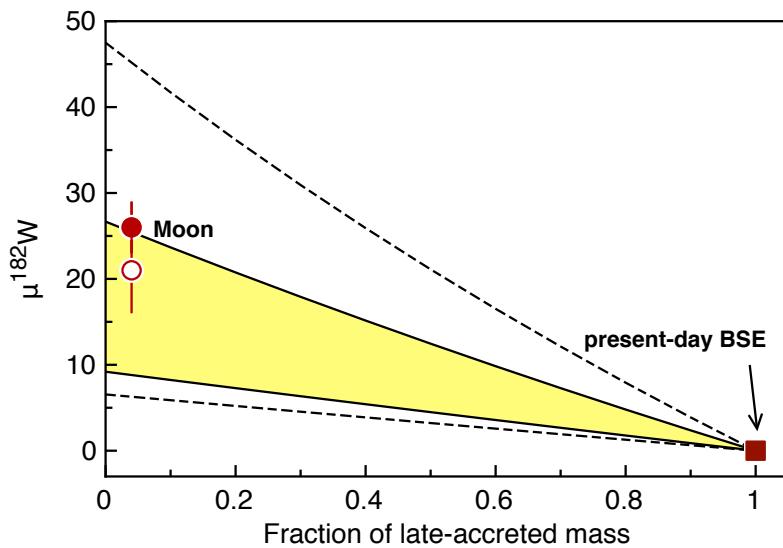
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Fig 2: Comparison of $\mu^{182}\text{W}$ data for KREEP-rich samples 14321, 68115, and 68815 obtained in three different W isotope studies. These three samples are devoid of neutron capture effects as reflected by the absence of Hf isotope anomalies in these samples (Kruijer et al., 2015).

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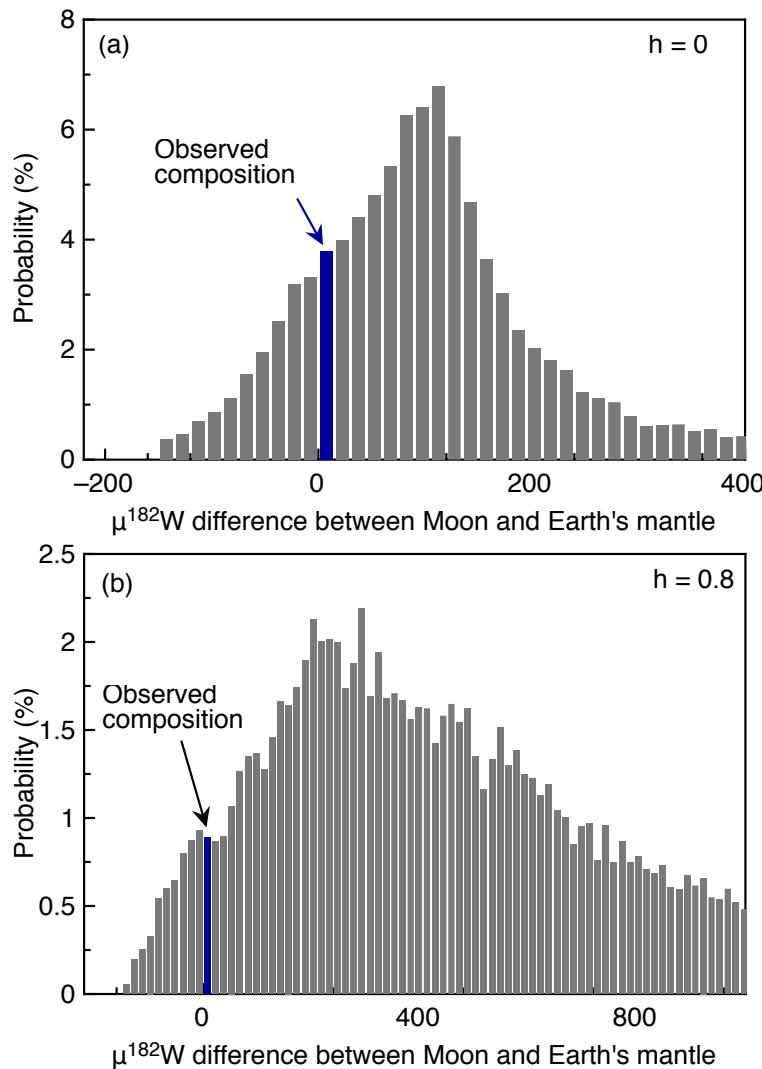


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313 **Fig. 3.** Pre-exposure $\mu^{182}\text{W}$ of different lunar rock types. Error bars denote external uncertainties (95%
314 conf.) on pre-exposure $\mu^{182}\text{W}$. Hashed area shows the weighted mean $\mu^{182}\text{W}$ value and the associated
315 95% conf. limits. Date sources: Touboul et al. (2015), Kruijer et al. (2015), and Kruijer and Kleine
316 (2017). Figure adopted from Kruijer and Kleine (2017).
317



320 **Fig. 4:** $\mu^{182}\text{W}$ versus the mass fraction of the late veneer on Earth. Mass balance calculations (yellow
 321 envelope) predict a positive $\mu^{182}\text{W}$ signature if the full complement of late accretion is subtracted from
 322 the present-day bulk silicate Earth (BSE; right red square) [assuming a W concentration of 200 ppb
 323 and $\mu^{182}\text{W} = -190$ for chondrites; and a late veneer complement in the present-day BSE between 3 and
 324 8 wt%]. Dashed lines illustrate the additional uncertainty introduced when also considering the
 325 uncertainty on the W concentration of the BSE of 13 ± 5 ppb. The predicted $\mu^{182}\text{W}$ is consistent with the
 326 observed $\mu^{182}\text{W}$ value of the Moon [solid red circle, value for bulk silicate Moon from Kruijer and
 327 Kleine (2017) and Kruijer et al. (2015) and open red circle, value for KREEP from Touboul et al.
 328 (2015)]. Figure modified after Kruijer et al. (2015) and Kleine and Walker (2017).
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334 **Fig. 5:** Histograms showing the predicted $\mu^{182}\text{W}$ difference between the Moon and Earth's mantle.
335 Shown are the result for two different giant impact scenarios (a, b), each involving a different mass
336 fraction of impactor mantle (h) within the Moon. The observed ^{182}W composition of the Moon is shown
337 for comparison. Figure modified after Kruijer and Kleine (2017).
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