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Ancient helium and tungsten isotopic signatures preserved in mantle domains least modified by crustal recycling

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4 **Ancient helium and tungsten isotopic signatures**
5 **preserved in mantle domains least modified by crustal**
6 **recycling**

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51 **Abstract.** Rare high- $^3\text{He}/^4\text{He}$ signatures in ocean island basalts (OIB) erupted at volcanic hotspots derive
52 from deep-seated domains preserved in Earth's interior. Only high- $^3\text{He}/^4\text{He}$ OIB exhibit anomalous- ^{182}W —
53 an isotopic signature inherited during the earliest history of the Earth—supporting an ancient origin of
54 high- $^3\text{He}/^4\text{He}$. However, it is not understood why some OIB host anomalous- ^{182}W while others do not. We
55 provide new geochemical data for the highest- $^3\text{He}/^4\text{He}$ lavas from Iceland (up to 42.9 times atmospheric)
56 with anomalous- ^{182}W , and examine how Sr-Nd-Hf-Pb isotopic variations—useful for tracing subducted,
57 recycled crust—relate to high- $^3\text{He}/^4\text{He}$ and anomalous- ^{182}W . These data, together with data on global
58 OIB, show that the highest- $^3\text{He}/^4\text{He}$ and the largest magnitude ^{182}W anomalies are found only in
59 geochemically depleted mantle domains—with high- $^{143}\text{Nd}/^{144}\text{Nd}$ and low- $^{206}\text{Pb}/^{204}\text{Pb}$ —lacking strong
60 signatures of recycled materials. In contrast, OIB with the strongest signatures associated with recycled
61 materials have low- $^3\text{He}/^4\text{He}$ and lack anomalous- ^{182}W . These observations provide important clues
62 regarding the survival of the ancient He and W signatures in Earth's mantle. We show that high- $^3\text{He}/^4\text{He}$
63 mantle domains with anomalous- ^{182}W have low W and ^4He concentrations compared to recycled
64 materials and are therefore highly susceptible to being overprinted with low- $^3\text{He}/^4\text{He}$ and normal (not
65 anomalous) ^{182}W characteristic of subducted crust. Thus, high- $^3\text{He}/^4\text{He}$ and anomalous- ^{182}W are
66 preserved exclusively in mantle domains least modified by recycled crust. This model places the long-
67 term preservation of ancient high- $^3\text{He}/^4\text{He}$ and anomalous- ^{182}W in the geodynamic context of crustal
68 subduction and recycling, and informs on survival of other early-formed heterogeneities in Earth's interior.

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72 **Keywords:** Mantle geochemistry, hotspot volcanism, ^{182}W , $^3\text{He}/^4\text{He}$, Hadean, core, mantle

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75 **Significance Statement.** The recent discovery of anomalous- ^{182}W signatures in modern, plume-derived
76 hotspot lavas provides evidence for survival of domains in Earth's interior since the early Hadean.
77 Critically, only lavas with ancient, high- $^3\text{He}/^4\text{He}$ signatures preserve anomalous- ^{182}W signatures.
78 However, it is still not known why only high- $^3\text{He}/^4\text{He}$ lavas have anomalous- ^{182}W , while other hotspot
79 lavas do not. Here we show that only hotspot lavas lacking strong recycled crust signatures exhibit high-
80 $^3\text{He}/^4\text{He}$ and anomalous- ^{182}W . This observation is explained by subduction of W- and ^4He -rich crust that
81 has low- $^3\text{He}/^4\text{He}$ and normal- ^{182}W : following subduction, the crust masks high- $^3\text{He}/^4\text{He}$ and anomalous-
82 ^{182}W signatures characteristic of ancient mantle domains. Thus, our model links destruction of Hadean
83 geochemical signatures with the well-established geodynamic process of plate subduction and crustal
84 recycling.

85

86 Introduction

87 Plate tectonic motions deliver oceanic and continental crust into the mantle at subduction zones.
88 Following residence times of ~2 Ga, subducted crustal material is returned to the shallow mantle in
89 buoyantly upwelling conduits of hot mantle material, known as mantle plumes, where they are partially
90 melted and erupted as OIB at volcanic hotspots (1-3). Recycled materials contribute to a variety of
91 geochemically-distinct mantle components often referred to as EM1 (enriched mantle I), EM2 (enriched
92 mantle II), and HIMU (high 'μ' or high $^{238}\text{U}/^{204}\text{Pb}$) (1-3). While there is debate surrounding the exact nature
93 of the recycled protoliths responsible for these mantle endmembers (4-7), they are likely generated by
94 input of shallow geochemical reservoirs into the deep Earth. The three mantle components have $^{87}\text{Sr}/^{86}\text{Sr}$,
95 $^{143}\text{Nd}/^{144}\text{Nd}$, $^{176}\text{Hf}/^{177}\text{Hf}$, and $^{206}\text{Pb}/^{204}\text{Pb}$ distinct from the depleted mantle (DM) sampled by basalts
96 erupted at mid-ocean ridges (MORB). In three-dimensional Sr-Nd-Pb isotopic space, these four
97 geochemical endmembers can be used to define the apices of a tetrahedron (8) (Figure 1), and have low
98 $^3\text{He}/^4\text{He}$, similar to, or lower than, MORB ($8 \pm 2 \text{ Ra}$, ratio to atmosphere) (9). An additional mantle
99 component is found internal to the other four and, like DM, is characterized by geochemically depleted
100 compositions that relate to long-term crustal extraction from the mantle, which changes the parent-
101 daughter ratios that govern key radiogenic isotopic systems, resulting in high $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{176}\text{Hf}/^{177}\text{Hf}$
102 coupled with low $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{206}\text{Pb}/^{204}\text{Pb}$. Existing long-lived radiogenic isotopic data show that this fifth
103 component is geochemically depleted (8,10), and has been variously referred to as FOZO (Focus Zone
104 (8)), PHEM (Primordial Helium Mantle (11)), or C (Common (12)). In contrast to DM, this geochemically
105 depleted mantle composition was argued to exhibit elevated $^3\text{He}/^4\text{He}$ (8), signaling the preservation of an
106 ancient reservoir in the deep Earth (13).

107 Recent discoveries employing the short-lived ^{182}Hf - ^{182}W isotopic systematics have demonstrated
108 the presence of anomalous, negative $\mu^{182}\text{W}$ (where μ is the deviation of $^{182}\text{W}/^{184}\text{W}$ from the terrestrial
109 standard in parts per million) in modern OIB (14-17), and the anomalies are found uniquely in lavas with
110 high $^3\text{He}/^4\text{He}$ (14-16). ^{182}W anomalies were generated only while the parent nuclide ($^{182}\text{Hf} \rightarrow ^{182}\text{W} + 2\beta^-$,
111 $t_{1/2} = 8.9 \text{ Ma}$) was extant (i.e., $< 60 \text{ Ma}$ following Solar System formation), so the observation of
112 anomalous ^{182}W signals in the modern mantle implies that these signatures have been preserved in the
113 Earth since the early Hadean. This is consistent with the discovery of heterogeneous ^{129}Xe and ^{142}Nd —

114 which are products of the short-lived isotopes ^{129}I and ^{146}Sm , respectively, extant only in the Hadean
115 (18,19)—in the modern mantle, further supporting the observation of chemical heterogeneities preserved
116 in the Earth's interior since the Hadean.

117 An important recent observation is that ^{182}W and $^3\text{He}/^4\text{He}$ data from oceanic basalts define a
118 negatively sloping array, with anomalous ^{182}W signatures in OIB uniquely associated with lavas that
119 exhibit high $^3\text{He}/^4\text{He}$ (14-16). This fundamental advance relates primordial high $^3\text{He}/^4\text{He}$ with an early-
120 formed isotopic heterogeneity. However, several outstanding questions regarding ^{182}W systematics in the
121 modern mantle remain: 1) What is the mechanism responsible for the unique association of anomalous
122 $\mu^{182}\text{W}$ with high- $^3\text{He}/^4\text{He}$ hotspots lavas, and why do low- $^3\text{He}/^4\text{He}$ lavas lack ^{182}W anomalies? 2) Do the
123 geodynamic processes of subduction and recycling impact the ^{182}W - $^3\text{He}/^4\text{He}$ mantle array? 3) Do ^{182}W
124 systematics exhibit relationships with isotopic tracers (i.e., Sr-Nd-Pb) sensitive to crustal subduction and
125 recycling, or are ^{182}W systematics somehow insensitive to these geodynamic processes such that Sr-Nd-
126 Pb isotope systematics are decoupled from ^{182}W ?

127 In order to address these outstanding questions, we provide a new large geochemical dataset on
128 high- $^3\text{He}/^4\text{He}$ lavas from the Iceland hotspot, and examine the data in the context of new global OIB
129 datasets combining He, Sr, Nd, Hf, and Pb isotopes with W isotopes. The data and analysis provide new
130 insights into the mechanisms for preservation of anomalous ^{182}W and high- $^3\text{He}/^4\text{He}$ signatures in the
131 mantle.

132

133 **Results**

134 We present new He-Sr-Nd-Hf-Pb-W isotopic data (see Materials and Methods) on a suite of 28
135 basalt samples from all Icelandic localities known to have high $^3\text{He}/^4\text{He}$ ($> 25 \text{ Ra}$): mid-Miocene lavas
136 from northwest Iceland (Vestfirðir), as well as Neovolcanic zone lavas from the Eastern Rift Zone (ERZ),
137 the South Iceland Volcanic Zone (SIVZ), and Vaðalda shield volcano in the Northern Rift Zone (NRZ) (20-
138 22) (SI Appendix, Figure S1, Datasets S1, S2, and S3). These data are presented together with published
139 data (23) from ~60 Ma Baffin Island and West Greenland (BIWG) flood basalt lavas, an additional high-
140 $^3\text{He}/^4\text{He}$ (up to 50 Ra (23-26)) locality on the Iceland hotspot track. He, Pb, and W isotopic data on a
141 small subset ($N = 8, 9,$ and $10,$ respectively) of the Iceland samples examined here (14,15), as well as

142 $^3\text{He}/^4\text{He}$ data on a different subset of ten samples (20,22), were published previously. The dataset
143 presented here includes new $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ for all 28 samples, and new $^{176}\text{Hf}/^{177}\text{Hf}$ and Pb
144 isotopic measurements for 27 and 19 samples, respectively. We also present new $^3\text{He}/^4\text{He}$
145 measurements by coupled vacuum crushing and fusion of olivine and clinopyroxene crystals on ten
146 samples (including one sample with the highest $^3\text{He}/^4\text{He}$ in the dataset, 42.9 Ra; Figure 2; SI Appendix,
147 Figure S2), and new $^3\text{He}/^4\text{He}$ measurements by fusion on the same powders from nine samples with
148 previously published $^3\text{He}/^4\text{He}$ by crushing (15). We present a new W isotopic measurement on the
149 highest $^3\text{He}/^4\text{He}$ ERZ lava, complementing 10 prior measurements made on this suite of 28 samples (15)
150 (additional ^{182}W analyses on a different suite of 6 Icelandic lavas are also available (15)). Additionally, we
151 present new major and trace element data for all of the mid-Miocene Vestfirðir lavas and a subset of the
152 Neovolcanic zone lavas (SI Appendix, Figure S3). The new and published data on the 28 samples of this
153 study represent the first full geochemical characterization of all known high- $^3\text{He}/^4\text{He}$ localities in Iceland.

154 The new Iceland dataset, combined with Sr-Nd-Pb isotope data from the literature, expands the
155 number of OIB samples characterized for both ^{182}W (14-17) and Sr-Nd-Pb isotopic compositions by nearly
156 60% (SI Appendix, Dataset S4). This augmented dataset allows examination of relationships between Sr-
157 Nd-Pb isotopes, traditionally used to detect signatures of crustal recycling, and $^3\text{He}/^4\text{He}$ and ^{182}W , which
158 can trace early-formed reservoirs in the Earth.

159 In Figure 2 and SI Appendix Figure S4, the He-Sr-Nd-Hf-Pb isotopic compositions for most
160 (~75%) of the Iceland lavas with $^3\text{He}/^4\text{He} > 21$ Ra are presented here for the first time and show that the
161 highest $^3\text{He}/^4\text{He}$ lavas sample four high- $^3\text{He}/^4\text{He}$ components with distinct Sr-Nd-Hf-Pb isotopic
162 compositions. The Vestfirðir (up to 42.9 Ra) and Vaðalda (33.6 Ra) lavas are similar geochemically and
163 are grouped together. The Vaðalda lava shows the recent (Pleistocene) reappearance of a geochemical
164 component not known in Iceland since the mid-Miocene (Figure 2; SI Appendix, Figure S4). The ERZ
165 lavas host a second Iceland high- $^3\text{He}/^4\text{He}$ component (25.9 Ra; SI Appendix, Dataset S1) with Sr-Nd-Hf
166 isotopic ratios more geochemically depleted than Vestfirðir and Vaðalda lavas. The SIVZ lavas represent
167 a third Iceland high- $^3\text{He}/^4\text{He}$ component (25.7 to 26.2 Ra (20); SI Appendix, Dataset S1) with $^{206}\text{Pb}/^{204}\text{Pb}$
168 higher than the other two components. A fourth high- $^3\text{He}/^4\text{He}$ Iceland component is evident in previously
169 published geochemical datasets from BIWG, which have $^3\text{He}/^4\text{He}$ up to 50 Ra, the highest known

170 terrestrial mantle-derived $^3\text{He}/^4\text{He}$ (23-26)): after applying major and trace element filters for continental
171 crust assimilation (23), the Baffin Island lavas exhibit the lowest $^{206}\text{Pb}/^{204}\text{Pb}$ identified along the Iceland
172 hotspot track (Figure 2).

173 The four geochemically-distinct high- $^3\text{He}/^4\text{He}$ Iceland components are clearly resolved in Sr-Nd-
174 Hf-Pb isotope space (Figure 2; SI Appendix, Figure S4), and the range in $^{206}\text{Pb}/^{204}\text{Pb}$ across the four
175 Iceland high- $^3\text{He}/^4\text{He}$ components found at five geographic locations—Vestfirðir/Vaðalda, ERZ, SIVZ, and
176 BIWG—spans a remarkable ~40% of the total variability found in global OIB (Figure 2; SI Appendix,
177 Figure S4). The wide range of Sr-Nd-Hf-Pb isotopic compositions in Icelandic, and global (including
178 Hawaii, Samoa, and Galapagos), high- $^3\text{He}/^4\text{He}$ ($> 25 \text{ Ra}$) lavas is notable. Nonetheless, high- $^3\text{He}/^4\text{He}$
179 lavas globally exhibit geochemically depleted Sr-Nd-Hf isotopic signatures and relatively unradiogenic
180 $^{206}\text{Pb}/^{204}\text{Pb}$ compositions ($^{206}\text{Pb}/^{204}\text{Pb} < 19.5$) (Figure 2; SI Appendix, Figure S4), where the chondritic
181 reference frame for $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{176}\text{Hf}/^{177}\text{Hf}$ serves as reference point for geochemical depletion:
182 geochemically depleted lavas have $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{176}\text{Hf}/^{177}\text{Hf}$ ratios higher than chondrites (i.e., as
183 shown for $^{143}\text{Nd}/^{144}\text{Nd}$ in Figure 3). Complementing negative $\mu^{182}\text{W}$ anomalies observed in high- $^3\text{He}/^4\text{He}$
184 hotspot lavas globally (14-16), Vestfirðir and SIVZ lavas have negative $\mu^{182}\text{W}$ anomalies (15), and we
185 further report here another resolvable negative $\mu^{182}\text{W}$ anomaly (-10.0 ± 3.8) in the highest $^3\text{He}/^4\text{He}$ ERZ
186 lava (SI Appendix, Dataset S2). However, recently published ^{182}W measurements of the high- $^3\text{He}/^4\text{He}$
187 BIWG lavas do not reveal $\mu^{182}\text{W}$ anomalies (15), which is discussed below.

188 In Figure 1, Sr-Nd-Pb isotopic compositions of the lavas that define the four Iceland high- $^3\text{He}/^4\text{He}$
189 components, and the highest $^3\text{He}/^4\text{He}$ lavas from other hotspots, are compared with the common mantle
190 component. This component is shown as an ellipsoid and represents the 95% confidence interval for the
191 volume encompassing the best-fit intersections of the trendlines that were fit to OIB arrays formed by
192 individual hotspots (27), as this component (i.e., FOZO) was originally defined by the intersection of
193 global OIB arrays (8) (but we note that not all OIB arrays extend into the common component region). In
194 Iceland, only the highest $^3\text{He}/^4\text{He}$ lava from the SIVZ plots within the common mantle component
195 ellipsoid, while the highest $^3\text{He}/^4\text{He}$ ERZ, Vestfirðir, and Baffin Island lavas are shifted toward
196 compositions more geochemically depleted (and in the case of Baffin Island, more similar to MORB) than
197 the common mantle component. Among the other hotspots globally with $^3\text{He}/^4\text{He} > 25 \text{ Ra}$, only the

198 Galapagos high- $^3\text{He}/^4\text{He}$ lava (30.3 Ra) plots within the ellipsoid, while Hawaiian (32.3 Ra) and Samoan
199 (33.4 Ra) high- $^3\text{He}/^4\text{He}$ lavas do not (Figure 1). In summary, most of the highest $^3\text{He}/^4\text{He}$ OIB globally are
200 not necessarily associated with the common mantle component in Sr-Nd-Pb isotopic space, as previously
201 defined. Due to this isotopic variability, we do not refer to the high- $^3\text{He}/^4\text{He}$ mantle domain with
202 anomalous ^{182}W as FOZO, C, or PHEM, all of which share depleted characteristics.

203 The geochemically depleted high- $^3\text{He}/^4\text{He}$ mantle is most clearly distinguished from the
204 geochemically depleted DM by having high $^3\text{He}/^4\text{He}$ and anomalous ^{182}W . However, due to extreme
205 geochemical depletion in some high- $^3\text{He}/^4\text{He}$ lavas (in particular the BIWG lavas), we are not able to
206 specify which mantle domain, DM or high- $^3\text{He}/^4\text{He}$, extends to more geochemically depleted
207 compositions. Nonetheless, we show for the first time that only geochemically depleted mantle domains
208 have the largest magnitude $\mu^{182}\text{W}$ anomalies. The association of anomalous $\mu^{182}\text{W}$ with geochemical
209 depleted mantle (i.e., superchondritic $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{176}\text{Hf}/^{177}\text{Hf}$) provides key insights into processes
210 responsible for the preservation, and destruction, of ^{182}W anomalies in Earth's interior.

211

212 **Discussion**

213 **Influence of recycled crust on high- $^3\text{He}/^4\text{He}$ and anomalous ^{182}W signatures in the mantle.** An
214 important issue in chemical geodynamics is how short-lived radiogenic isotopic signatures generated in
215 the Hadean Earth— ^{182}W , ^{142}Nd , and ^{129}Xe —have survived in the convecting mantle to be sampled by
216 modern OIB (16-19). The presence of high $^3\text{He}/^4\text{He}$ and anomalous ^{182}W in Iceland hotspot lavas require
217 a mechanism for preserving these ancient signatures in the OIB mantle. We argue that the survival of
218 high- $^3\text{He}/^4\text{He}$ and anomalous ^{182}W signatures in the convecting mantle will depend on whether
219 anomalous ^{182}W and high- $^3\text{He}/^4\text{He}$ mantle domains mix with W- and ^4He -rich recycled materials, or with
220 DM, which also has low $^3\text{He}/^4\text{He}$ and normal ^{182}W . Compared to domains with anomalous ^{182}W and high
221 $^3\text{He}/^4\text{He}$, the mantle endmembers sampling recycled materials—EM1, EM2, and HIMU—lack anomalous
222 $\mu^{182}\text{W}$. Subducted oceanic and continental crust have high W concentrations and will not necessarily host
223 anomalous $\mu^{182}\text{W}$ (if derived from reservoirs that formed after the early Hadean), and they will evolve high
224 ^4He concentrations and low $^3\text{He}/^4\text{He}$ over time (due to having higher U and Th concentrations and to loss
225 of He during subduction) (see SI Appendix, Datasets S5 and S6 for data sources). Thus, one hypothesis

226 is that subducted crust can overprint high- $^3\text{He}/^4\text{He}$ and anomalous $\mu^{182}\text{W}$ signatures with low $^3\text{He}/^4\text{He}$ and
227 normal $\mu^{182}\text{W}$, explaining why mantle domains similar to EM1, EM2, and HIMU have low $^3\text{He}/^4\text{He}$ and lack
228 anomalous $\mu^{182}\text{W}$, while domains with high $^3\text{He}/^4\text{He}$ and anomalous $\mu^{182}\text{W}$ do not extend to extreme EM
229 (low $^{143}\text{Nd}/^{144}\text{Nd}$) or HIMU (high $^{143}\text{Nd}/^{144}\text{Nd}$ and high $^{206}\text{Pb}/^{204}\text{Pb}$) compositions that reflect large
230 contributions from W- and ^4He -rich recycled materials (Figure 3; SI Appendix, Figure S5).

231 The addition of variable quantities of subducted materials to mantle domains with anomalous
232 ^{182}W is a potential mechanism to explain variability in the magnitude of recently discovered ^{182}W
233 anomalies in modern hotspot lavas. Before the discovery of ^{182}W anomalies in the present-day mantle,
234 subduction and recycling of W-rich crust was considered as a possible mechanism to explain the
235 complete absence of detectable core-derived ^{182}W anomalies in the Earth's mantle (28,29). However, the
236 discovery of ^{182}W variability in the modern mantle indicates that ^{182}W heterogeneity has not been entirely
237 overprinted with subducted crust. Thus, we explore a more nuanced version of this early model to gain
238 new insights into the coupled ^{182}W - $^3\text{He}/^4\text{He}$ systematics of the mantle. We quantitatively examine the
239 influence of recycled crust and DM on mantle domains that host anomalous $\mu^{182}\text{W}$ and high- $^3\text{He}/^4\text{He}$
240 compositions (Figure 4).

241 In $\mu^{182}\text{W}$ versus $^3\text{He}/^4\text{He}$ space, the data form a negatively-sloping array that is composed of
242 subrends formed by different hotspots (14-16), and the strength of the overall global trend is supported
243 by a high correlation (Spearman rank coefficient $r_s = -0.67 \pm 0.07$) (Figure 4). Assuming an average age
244 for recycled crust of 2 Ga (an age consistent with the slope of the Northern Hemisphere Reference Line
245 (NHRL) in Pb isotopic space (30)), we show that mixing ancient subducted oceanic and continental crust
246 with high- $^3\text{He}/^4\text{He}$ mantle sources of OIB with anomalous $\mu^{182}\text{W}$ forms mixing trends with negatively
247 sloping arrays that describe the coupled $\mu^{182}\text{W}$ and $^3\text{He}/^4\text{He}$ systematics of global OIB (Figure 4).

248 Absolute He and W concentrations in the mantle are not well constrained, but the concentration
249 contrast between high- $^3\text{He}/^4\text{He}$ mantle domains and subducted materials can be inferred to construct the
250 mixing models in Figure 4. All W and He isotopic compositions and mantle source concentrations for the
251 model in Figure 4 are provided in SI Appendix, Dataset S5. Compared to the high- $^3\text{He}/^4\text{He}$ mantle,
252 oceanic and continental crust is expected to have high concentrations of W (0.12 and 1.9 ppm,
253 respectively) (31, 32) and ^4He , the latter due to the decay of Th and U (which are elevated in oceanic and

254 continental crust relative to the high- $^3\text{He}/^4\text{He}$ mantle). High- $^3\text{He}/^4\text{He}$ lavas with anomalous ^{182}W from
255 Hawaii (Loihi) provide a window into the W concentration of the high- $^3\text{He}/^4\text{He}$ mantle, which is calculated
256 by melt models to have W concentrations (0.011 ± 0.005 ppm W (33)) that are approximately 11 and 170
257 times lower than subducted oceanic and continental crust, respectively. The mantle sources of high-
258 $^3\text{He}/^4\text{He}$ lavas have He concentrations (34) that are estimated to be approximately >9 and >80 times
259 lower than subducted oceanic and continental crust, respectively (see SI Appendix, Dataset S5). Taking
260 these W and He concentrations as representative of the high- $^3\text{He}/^4\text{He}$ mantle with anomalous ^{182}W ,
261 subducted crust can overwhelm the W and He budgets of high- $^3\text{He}/^4\text{He}$ mantle domains, overprinting
262 them with low $^3\text{He}/^4\text{He}$ and normal $\mu^{182}\text{W}$, and thus generating the negatively sloping global OIB array in
263 Figure 4. Finally, adding DM—which likewise has low $^3\text{He}/^4\text{He}$ (8 Ra) and normal $\mu^{182}\text{W}$ —to the high-
264 $^3\text{He}/^4\text{He}$ mantle also generates a negatively sloping array that follows the slope of the OIB field in $\mu^{182}\text{W}$ -
265 $^3\text{He}/^4\text{He}$ space (Figure 4; DM He (34) and W (33) concentrations are provided in SI Appendix, Dataset
266 S5). The takeaway message is that anomalous ^{182}W and high $^3\text{He}/^4\text{He}$ in plume-fed hotspots, such as
267 Iceland, are derived from mantle domains in which these ancient signatures are not completely
268 overprinted by recycled materials, or mixing with DM.

269 This model for overprinting of the high- $^3\text{He}/^4\text{He}$ mantle by mixing with recycled oceanic and
270 continental crust is also consistent with the generation of low $^3\text{He}/^4\text{He}$ and normal ^{182}W observed in OIB
271 endmembers EM1, EM2, and HIMU (Figure 4, SI Appendix, text, Dataset S5). Other recycled materials
272 could overprint domains with high $^3\text{He}/^4\text{He}$ and anomalous ^{182}W , but such materials would also need to be
273 1) relatively enriched in W (to effectively mask anomalous ^{182}W domains) and 2) degassed in He and
274 enriched in Th and U (to generate both the high ^4He concentrations and low $^3\text{He}/^4\text{He}$ that are necessary
275 to mask high- $^3\text{He}/^4\text{He}$ domains). Nonetheless, mixing ancient subducted continental and oceanic crust, as
276 done in Figure 4, serves to illustrate how recycled materials efficiently overprint high- $^3\text{He}/^4\text{He}$ and
277 anomalous ^{182}W domains in the mantle.

278

279 **Quantifying the relationship between ^{182}W and crustal recycling.** It has long been assumed that
280 addition of recycled materials to the mantle results in mantle domains with low $^3\text{He}/^4\text{He}$ (13,35,36), which
281 is supported by the modelling exercise in Figure 4, and the observation that mantle endmembers hosting

282 the strongest recycling signatures in Sr-Nd-Pb isotopic space—EM1, EM2, and HIMU—have low $^3\text{He}/^4\text{He}$
283 ($\leq 8 \text{ Ra}$). However, the relationships between $\mu^{182}\text{W}$ and Sr-Nd-Pb isotopes, and the impact of subducted
284 materials on $\mu^{182}\text{W}$ signatures in OIB, remain to be explored in detail. Armed with a dataset of Sr-Nd-Pb-
285 W isotopes on global OIB (SI Appendix, Dataset S4), we identify clear relationships between $\mu^{182}\text{W}$ and
286 recycled signatures in OIB (Figure 5).

287 Based on the modelling in Figure 4, we propose that, if mixed with a high- $^3\text{He}/^4\text{He}$ component
288 having anomalous ^{182}W , mantle domains lacking input of recycled crust will have the most extreme
289 negative ^{182}W anomalies, and that addition of progressively larger quantities of recycled crust will cause a
290 reduction in the magnitude of the ^{182}W anomaly. This hypothesis is difficult to test because no single
291 isotopic ratio is indicative of recycled materials that contribute to HIMU *and* recycled materials that
292 contribute to EM1 and EM2: high $^{206}\text{Pb}/^{204}\text{Pb}$ can be used to identify a contribution from a HIMU
293 component, but the $^{206}\text{Pb}/^{204}\text{Pb}$ ratio is less effective in identifying EM components; low $^{143}\text{Nd}/^{144}\text{Nd}$ and
294 high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios can be used to identify EM component contributions, but are less effective in
295 identifying HIMU contributions. A single parameter is needed that indicates the magnitude of contribution
296 from the various mantle endmembers. Therefore, we propose a “mantle endmember index” that draws on
297 $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$, and $^{206}\text{Pb}/^{204}\text{Pb}$ to quantify the contribution of EM1-EM2-HIMU mantle endmember
298 melt compositions to OIB lavas. This index allows evaluating how a fourth isotope system (e.g., $\mu^{182}\text{W}$)
299 varies in lavas as a function of the magnitude of the contribution from any of the mantle endmembers
300 (Figure 5). Central to this index are two assumptions: 1) Lavas sampling geochemically depleted mantle
301 (DM)—which has low $^{206}\text{Pb}/^{204}\text{Pb}$, low $^{87}\text{Sr}/^{86}\text{Sr}$, and high $^{143}\text{Nd}/^{144}\text{Nd}$ —lack a significant component of
302 recycled materials, and 2) an increase in the contribution of recycled material results in a shift away from
303 the depleted mantle composition in Sr-Nd-Pb isotopic space towards HIMU and EM compositions (Figure
304 5). Larger shifts correspond to greater recycled contributions.

305 This shift away from the geochemically depleted mantle (DM) can be measured as a distance
306 ($D^{\text{Sr-Nd-Pb}}$) in three-dimensional $^{87}\text{Sr}/^{86}\text{Sr}$ - $^{143}\text{Nd}/^{144}\text{Nd}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ space, using the following equation:

307

$$308 \quad D^{\text{Sr-Nd-Pb}} = [((^{87}\text{Sr}/^{86}\text{Sr}_\text{O} - ^{87}\text{Sr}/^{86}\text{Sr}_\text{D})/X)^2 + ((^{143}\text{Nd}/^{144}\text{Nd}_\text{O} - ^{143}\text{Nd}/^{144}\text{Nd}_\text{D})/Y)^2 + ((^{206}\text{Pb}/^{204}\text{Pb}_\text{O} - ^{206}\text{Pb}/^{204}\text{Pb}_\text{D})/Z)^2]^{0.5}$$

309

310 where the subscripts O and D indicate the isotopic composition of any “OIB” sample and the
311 geochemically “depleted” reference composition, respectively; the depleted reference composition has Sr-
312 Nd-Pb isotopes of 0.7025, 0.5132, and 17.5, respectively; X, Y, and Z represent the Sr-Nd-Pb isotopic
313 differences between the geochemically depleted reference composition and the most extreme crustal
314 recycling signatures in OIB considered in $^{87}\text{Sr}/^{86}\text{Sr}$ (0.7186 - 0.7025), $^{143}\text{Nd}/^{144}\text{Nd}$ (0.5132 - 0.5123), and
315 $^{206}\text{Pb}/^{204}\text{Pb}$ (22.0 - 17.5) isotopic spaces, respectively. Higher values of $D^{\text{Sr-Nd-Pb}}$ for an OIB sample reflect
316 a greater contribution from EM1, EM2, and/or HIMU (and therefore more recycled materials), while lower
317 values indicate that lavas are more similar to the DM component and indicate a smaller contribution from
318 these three mantle endmembers (and therefore less recycled material) (see right panel in Figure 5). A
319 similar parameter for measuring distance in three-dimensional Pb isotope multispace (i.e., $D^{208/207/206\text{Pb}}$)
320 was developed elsewhere (37) and serves as the basis for the $D^{\text{Sr-Nd-Pb}}$ parameter presented here.

321 Figure 5 shows $\mu^{182}\text{W}$ in OIB as a function of $D^{\text{Sr-Nd-Pb}}$ and demonstrates that all OIB data with the
322 most extreme negative $\mu^{182}\text{W}$ have low $D^{\text{Sr-Nd-Pb}}$ values (< 0.65), consistent with the hypothesis that only
323 lavas sampling geochemically depleted mantle domains (i.e., lacking significant recycled materials), host
324 the most extreme negative $\mu^{182}\text{W}$ anomalies. However, many geochemically depleted lavas with low $D^{\text{Sr-}}$
325 Nd-Pb values lack anomalous ^{182}W , and this is accounted for by dilution of the anomalous ^{182}W by
326 entrainment of a depleted mantle (DM) component lacking anomalous ^{182}W , which melts in the shallow
327 mantle with the host plume, and subsequently mixes with melts of the high- $^3\text{He}/^4\text{He}$ plume component.
328 This process is described by the binary mixing line to MORB in Figure 5. For example, the subset of lavas
329 from Iceland with normal $\mu^{182}\text{W}$ may result from addition of a MORB component, consistent with the
330 mixing line between high- $^3\text{He}/^4\text{He}$ mantle melts and MORB in Figure 5. While a depleted component has
331 been suggested to be inherent to the Iceland plume (38,39), it is also possible that the plume has
332 incorporated ambient depleted upper mantle during ascent (40), consistent with our hypothesis for
333 reduction in the magnitude of ^{182}W anomalies in the Iceland plume by mixing with DM melts.

334 At higher $D^{\text{Sr-Nd-Pb}}$ values, and thus increasing fractions of recycled materials, the magnitude of
335 the $\mu^{182}\text{W}$ anomalies diminishes in lavas at hotspots that host anomalous $\mu^{182}\text{W}$. For example, at
336 maximum contributions of recycled materials, lavas sampling endmember EM1 (Pitcairn, $D^{\text{Sr-Nd-Pb}} = 0.85$),
337 EM2 (Samoa, $D^{\text{Sr-Nd-Pb}} = 1.44$), and HIMU (Mangaia Island on the Macdonald hotspot track, $D^{\text{Sr-Nd-Pb}} =$

338 0.99) all lack $\mu^{182}\text{W}$ anomalies. Critically, the magnitude of the recycled crust signature appears to
339 modulate the magnitude of the anomalous ^{182}W signature: OIB samples with progressively higher values
340 of $D^{\text{Sr-Nd-Pb}}$ at the Pitcairn, Samoa, and Macdonald hotspots show diminished $\mu^{182}\text{W}$ anomalies (Figure 5).
341 In contrast, input of less recycled material (i.e., lower $D^{\text{Sr-Nd-Pb}}$) corresponds with increasingly negative
342 $\mu^{182}\text{W}$ anomalies. For example, the single Pitcairn lava with a slightly negative $\mu^{182}\text{W}$ anomaly has the
343 lowest $D^{\text{Sr-Nd-Pb}}$ in the sample suite and also the highest $^3\text{He}/^4\text{He}$ among Pitcairn lavas. This pattern is also
344 reflected in Macdonald and Samoan hotspot samples, where lavas with the lowest $D^{\text{Sr-Nd-Pb}}$ also have the
345 lowest $\mu^{182}\text{W}$ and the highest $^3\text{He}/^4\text{He}$ (Figure 5). Figure 5 further shows that the relationship between $D^{\text{Sr-}}$
346 Nd-Pb and $\mu^{182}\text{W}$ at these three hotspots, which sample the three canonical mantle endmembers, is
347 consistent with binary mixing trends between the most anomalous $\mu^{182}\text{W}$ component and the recycled
348 crust endmembers (EM1, EM2, and HIMU). The mixing trends are consistent with an increasing
349 contribution of recycled materials being responsible for the dilution of anomalous $\mu^{182}\text{W}$ signatures
350 (mixing model endmembers are in SI Appendix, Dataset S6). Thus, while the exact mechanism for the
351 Hadean origin of $\mu^{182}\text{W}$ anomalies is not known (see below), it is clear that crustal subduction and
352 recycling play a central role in modulating the expression of these anomalies in OIB. Finally, we anchor
353 the mixing models in Figure 5 to an extreme low $\mu^{182}\text{W}$ composition (i.e., from Fernandina, Galapagos)
354 only because this composition represents the most negative value (-22.7) in the existing OIB dataset. As
355 the OIB $\mu^{182}\text{W}$ dataset expands in the future, we might expect highly negative $\mu^{182}\text{W}$ anomalies to be
356 present in OIB over a range of Sr-Nd-Pb isotopic compositions and therefore over a range of $D^{\text{Sr-Nd-Pb}}$
357 values, just as high- $^3\text{He}/^4\text{He}$ signatures are found in lavas with heterogeneous Sr-Nd-Pb isotopic
358 compositions and $D^{\text{Sr-Nd-Pb}}$ values (SI Appendix, Figure S5). However, like high- $^3\text{He}/^4\text{He}$ lavas, we
359 anticipate that lavas with highly negative $\mu^{182}\text{W}$ will be limited to those with geochemically depleted
360 isotopic compositions and low $D^{\text{Sr-Nd-Pb}}$ values, due to the masking influence of recycled materials on both
361 high- $^3\text{He}/^4\text{He}$ and anomalous $\mu^{182}\text{W}$ signatures in the mantle.

362 Some hotspot localities with existing $\mu^{182}\text{W}$ data lack Sr-Nd-Pb isotopic data, and thus $D^{\text{Sr-Nd-Pb}}$
363 cannot be calculated. For example, Canary hotspot lavas from La Palma lack $\mu^{182}\text{W}$ anomalies (14,41,42)
364 (SI Appendix, Dataset S4) and the lavas analyzed for $\mu^{182}\text{W}$ do not have complete Sr-Nd-Pb isotopic
365 datasets. The lack of $\mu^{182}\text{W}$ anomalies may be consistent with contribution from a HIMU component

366 present in some La Palma lavas (43), which would be associated with high $D^{\text{Sr-Nd-Pb}}$ values that are linked
367 to recycled crust that masks anomalous $\mu^{182}\text{W}$. If the strength of the global relationship between $\mu^{182}\text{W}$
368 and $^3\text{He}/^4\text{He}$ serves as a guide to which OIB host anomalous $\mu^{182}\text{W}$ (Figure 4), then the lack of high-
369 $^3\text{He}/^4\text{He}$ at the Canaries (the highest- $^3\text{He}/^4\text{He}$ value is 9.7 Ra (44)) is consistent with the absence of
370 anomalous ^{182}W signatures.

371 The presence of extreme negative $\mu^{182}\text{W}$ only in global OIB with $^{143}\text{Nd}/^{144}\text{Nd} > 0.5127$ (Figure 3)
372 is explained by the fact that lavas with low $^{143}\text{Nd}/^{144}\text{Nd}$ values (< 0.5127) may be impacted by subducted
373 continental crust (as suggested for the low $^{143}\text{Nd}/^{144}\text{Nd}$ in EM1 (6) and EM2 (36 lavas), a mechanism that
374 both reduces $^{143}\text{Nd}/^{144}\text{Nd}$ and masks anomalous $\mu^{182}\text{W}$. At the more geochemically depleted end of the
375 spectrum, where additional $\mu^{182}\text{W}$ anomalies may be found in the future, there is currently a lack of OIB
376 with existing $\mu^{182}\text{W}$ analyses. Figure 5 calls particular attention to the lack of data among the most
377 geochemically depleted OIB, as there is a total absence of ^{182}W -characterized lavas with $D^{\text{Sr-Nd-Pb}} < 0.25$.
378 Given their geochemically depleted isotopic signatures (with $^{143}\text{Nd}/^{144}\text{Nd}$ to ~ 0.5131) (16,24,25), and their
379 extreme high $^3\text{He}/^4\text{He}$ (up to 50 Ra) (23-26), the proto-Iceland plume BIWG continental flood basalts
380 (CFB) offer an important opportunity in this regard, and is discussed below. For example, the least
381 crustally-contaminated lavas among the Baffin Island suite (23) have $D^{\text{Sr-Nd-Pb}}$ ranging from 0.11 to 0.17.
382 However, continental crust contamination represents a challenge for identifying mantle source $\mu^{182}\text{W}$
383 signatures in these lavas.

384
385 **The case of the BIWG high- $^3\text{He}/^4\text{He}$ CFB.** While all high- $^3\text{He}/^4\text{He}$ OIB have anomalous $\mu^{182}\text{W}$ (14-16),
386 the high- $^3\text{He}/^4\text{He}$ West Greenland CFB lavas lack resolvable $\mu^{182}\text{W}$ anomalies in lavas with $^3\text{He}/^4\text{He}$ up to
387 48 Ra (15). This contrasts with results from an earlier study, which reported positive $\mu^{182}\text{W}$ (up to $\mu^{182}\text{W} =$
388 $+48.4$) for lavas from Baffin Island and the Ontong Java Plateau (OJP) ($\mu^{182}\text{W} = +23.9$) (45). However,
389 $\mu^{182}\text{W}$ anomalies were not identified in analyses of OJP lavas in other studies (41,46). Until the positive
390 $\mu^{182}\text{W}$ for OJP and Baffin Island (15) are replicated, we will not include them in further discussion in this
391 paper. In Figures 3 and 5 (and SI Appendix, Figure S5), we show OJP as lacking anomalous ^{182}W
392 (41,46). Additionally, we rely on recent ^{182}W analyses from West Greenland (15) to constrain the
393 composition of the proto-Iceland plume.

394 The absence of negative $\mu^{182}\text{W}$ anomalies in the West Greenland picrites might reflect an
395 absence of anomalous $\mu^{182}\text{W}$ in the mantle source (15). Alternatively, it could result from shallow level
396 continental crust contamination that overprinted mantle-derived $\mu^{182}\text{W}$ anomalies. High- $^3\text{He}/^4\text{He}$ ratios in
397 the lavas with $\mu^{182}\text{W}$ measurements do not necessarily exclude crustal assimilation influence on the Sr-
398 Nd-Pb-W isotopic compositions. For example, a crustally-contaminated Baffin Island lava—sample
399 BI/CS/7—has $^{87}\text{Sr}/^{86}\text{Sr}_{60}=0.704912$ and $^{143}\text{Nd}/^{144}\text{Nd}_{60}=0.512730$ compositions consistent with a
400 continental crust contribution, yet the olivines preserve high $^3\text{He}/^4\text{He}$ (43.9 Ra) (24). These data suggest
401 that crustal contamination may not impact $^3\text{He}/^4\text{He}$ in the same manner it affects the heavy radiogenic
402 isotopic systems, possibly because $^3\text{He}/^4\text{He}$ is measured in gas trapped and preserved in olivines formed
403 at depth, prior to significant shallow crustal assimilation that influences the whole rock Sr-Nd-Pb-W
404 isotopic compositions.

405 Significant crustal assimilation appears unlikely in a subset of the BIWG lavas examined in
406 several key studies (23-25). Nonetheless, the extreme enrichment of W in upper continental crust (1.9
407 ppm W) (31) relative to West Greenland lavas (down to 0.022 ppm) (15) suggests that small degrees of
408 continental crust assimilation can dramatically shift $\mu^{182}\text{W}$ in the lavas, while leaving the long-lived
409 radiogenic isotopic systems relatively unmodified, as demonstrated in the SI Appendix text. Thus, the
410 presence of $\mu^{182}\text{W}$ anomalies in the mantle source of West Greenland lavas cannot be ruled out, since
411 they may have been obscured by small degrees of crustal assimilation. For this reason, $\mu^{182}\text{W}$ for the
412 West Greenland picrites are not considered further here nor shown in the figures (see discussion in the SI
413 Appendix, text). The reason the West Greenland lavas are the only modern mantle-derived lavas to plot
414 off the global OIB $\mu^{182}\text{W}$ - $^3\text{He}/^4\text{He}$ array may relate to the fact that they are the only high- $^3\text{He}/^4\text{He}$ lavas
415 with $\mu^{182}\text{W}$ analyses that erupted in a continental setting, where continental crust assimilation is a
416 concern.

417
418 **Origin of anomalous $\mu^{182}\text{W}$ signatures in oceanic hotspots.** The discovery of negative $\mu^{182}\text{W}$ values in
419 volcanic hotspot lavas has been interpreted to reflect a geochemical influence from the Earth's core
420 (16,17), which should have low $\mu^{182}\text{W}$ (~ -220) (42)). Similarly, ancient high $^3\text{He}/^4\text{He}$ has been suggested
421 to reside in Earth's core (24, 47-49). Given the relationship between high $^3\text{He}/^4\text{He}$ and anomalous $\mu^{182}\text{W}$

422 in modern OIB, it is tempting to suggest a model where both high $^3\text{He}/^4\text{He}$ and anomalous $\mu^{182}\text{W}$ are
423 supplied to the mantle by the core. While there are limited high-precision ^{182}W data on MORB (14), such a
424 model could potentially explain why depleted upper mantle does not host clear ^{182}W anomalies, as the
425 geochemically depleted upper mantle is less likely to host a component that came from or interacted with
426 the core than lower mantle domains sampled by upwelling plumes. An advantage of this model is that it
427 can potentially explain our observation that only geochemically depleted lavas have negative $\mu^{182}\text{W}$
428 anomalies and high $^3\text{He}/^4\text{He}$, as any W and He emerging from the core will have a proportionally greater
429 impact on the $^3\text{He}/^4\text{He}$ and $\mu^{182}\text{W}$ compositions of geochemically depleted mantle domains (which have
430 low W and He concentrations) compared to mantle domains overprinted by recycled crust materials
431 (which have high W and He concentrations and therefore are more immune to He and W isotopic
432 signatures from the core). Furthermore, the non-primitive Pb-isotopic composition of lavas with the most
433 anomalous $\mu^{182}\text{W}$ compositions suggests that low $\mu^{182}\text{W}$ is not due to preservation of an ancient
434 component in the mantle, but instead mixing between a depleted mantle domain and a component with
435 low $\mu^{182}\text{W}$ and high $^3\text{He}/^4\text{He}$, a process that is consistent with a core contribution to a depleted mantle
436 domain. However, preservation of high $^3\text{He}/^4\text{He}$ in the core is controversial, and a lower mantle origin for
437 high $^3\text{He}/^4\text{He}$ has also been suggested (8,10,13). Thus, the analysis and modelling proposed here focus
438 on the modern mantle sources and eruptive products of OIB without any assumptions regarding specific
439 mechanisms for the origin of high $^3\text{He}/^4\text{He}$ and anomalous $\mu^{182}\text{W}$ in the mantle. Looking forward, models
440 invoked to generate negative $\mu^{182}\text{W}$ in the silicate Earth—core addition, late accretion, or silicate Earth
441 differentiation (16,17,41,50,51)—must also explain the link between anomalous $\mu^{182}\text{W}$ and high $^3\text{He}/^4\text{He}$
442 in geochemically depleted mantle sources of OIB.

443
444 **Geodynamic implications for high $^3\text{He}/^4\text{He}$ and anomalous ^{182}W in the mantle.** Under very restrictive
445 conditions—in geochemically depleted mantle domains lacking recycled crust—early Earth high- $^3\text{He}/^4\text{He}$
446 and anomalous $\mu^{182}\text{W}$ signatures have survived convective mixing and interaction with subducted crust to
447 be sampled by upwelling mantle plumes and erupted at modern hotspot volcanoes. The geodynamic
448 process of crustal subduction and recycling plays an outsized role in the destruction of both anomalous
449 $\mu^{182}\text{W}$ and high $^3\text{He}/^4\text{He}$ because W and ^4He concentrations in recycled oceanic and continental crust

450 may be higher than the high- $^3\text{He}/^4\text{He}$ mantle (by approximately one to two orders of magnitude, for
451 oceanic and continental crust, respectively; SI Appendix, Dataset S5). Given evidence provided by high
452 $^3\text{He}/^4\text{He}$ and anomalous ^{182}W , the Iceland mantle plume, together with a number of other plumes globally,
453 sample mantle domains that are relatively unmodified by crustal recycling.

454 This geodynamic framework, which relies on the well-established mechanism of crustal
455 subduction and recycling in the mantle, can be used to evaluate the preservation and distribution of
456 ancient isotopic signatures in the mantle. Like anomalous ^{182}W , nucleosynthetic and short-lived
457 radiogenic isotopic anomalies of elements enriched in Earth's crust (31,32) relative to the mantle (52)
458 (e.g., like ^{142}Nd (18), or even nucleosynthetic ^{92}Mo if heterogeneities are detected in the modern mantle
459 (53)) may be expected to survive primarily in geochemically depleted mantle domains least influenced by
460 recycling. Similarly, nucleosynthetic isotopic signatures for ^{100}Ru (54), if found in the modern mantle (53),
461 would be expected to be less impacted by crustal subduction and recycling than W, because Ru is
462 concentrated in the mantle (52) relative to the crust (31). This would also be the case for elements that
463 are concentrated in the crust or atmosphere relative to the mantle, but are not as efficiently subducted
464 into the mantle as W (e.g., ^{129}Xe (55)). Furthermore, preservation of ancient high- $^3\text{He}/^4\text{He}$ and anomalous
465 $\mu^{182}\text{W}$ signatures in the mantle requires isolation of the high- $^3\text{He}/^4\text{He}$ mantle from subducted crust (56).
466 Due to having higher viscosity, which slows convective motions, the deep lower mantle is an ideal
467 location to preserve ancient domains. The Ultra-Low Velocity Zones (ULVZs) may host anomalous $\mu^{182}\text{W}$
468 (16), and the large low shear wave velocity provinces (LLSVPs) are suggested to host high $^3\text{He}/^4\text{He}$ (57).
469 If the LLSVPs (58-60) or ULVZs (61) are denser than ambient mantle, and thus more difficult to entrain,
470 the observation that high $^3\text{He}/^4\text{He}$ is entrained only by the hottest, most buoyant plumes is more easily
471 explained (62) (Figure S6). Expanded datasets covering a greater geographic range of hotspots with
472 lower buoyancy fluxes will be important for extending these geophysical relationships to $\mu^{182}\text{W}$.

473

474 **Materials and Methods**

475 We present new whole rock Sr-Nd-Hf-Pb-W isotopic and major and trace element data, $^3\text{He}/^4\text{He}$ data
476 obtained on olivines and clinopyroxenes (by crushing and fusion), and olivine major element data for a
477 suite of 18 mid-Miocene lavas from northwest Iceland (Vestfirðir) (SI Appendix, Datasets S1, S2, S3, and
478 S7). Vestfirðir sample locations, constraints on sample ages, descriptions, and age corrections for Sr-Nd-

479 Hf-Pb isotopic compositions are also provided (SI Appendix, Datasets S1, S8, and S9). Additionally, we
480 present new Sr-Nd-Hf-Pb isotopic data (as well as W isotopic data for ERZ sample A24) for five samples
481 from the SIVZ (20.7 to 25.7 Ra), four samples from the ERZ (20.6 to 25.9 Ra), and one NRZ sample from
482 Vaðalda (33.6 Ra) (SI Appendix, Dataset S1 and S3). Protocols for wet chemistry and radiogenic isotopic
483 analyses (Sr-Nd-Hf-Pb) follow standard techniques and were carried out at four different institutions by
484 TIMS (thermal ionization mass spectrometry) or MC-ICP-MS (multi-collector inductively coupled plasma
485 mass spectrometry) following methods detailed in the SI Appendix, Methods: ENS Lyon (Ecole Normale
486 Supérieure de Lyon: Hf and Pb isotopes by MC-ICP-MS), UCSB (University of California, Santa Barbara:
487 Sr and Nd isotopes by TIMS), UNC (University of North Carolina: Sr and Nd isotopes by TIMS), and USC
488 (University of South Carolina: Sr, Nd, Hf, and Pb isotopes by MC-ICP-MS). Methods for analysis of W
489 isotopes at the University of Vienna are provided elsewhere (SI Appendix, Methods). Measurements of
490 He isotopes, carried out at WHOI (Woods Hole Oceanographic Institution), follow standard protocols (SI
491 Appendix, Methods). Whole rock major and trace element analyses were carried out at the Geoanalytical
492 Lab at Washington State University following standard protocols (SI Appendix, Methods). All new data
493 and protocols associated with this work are provided in the SI Appendix.
494

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505
506

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657
658
659 **Figure Legends**

660 **Figure 1. Mantle tetrahedron showing that most of the highest $^3\text{He}/^4\text{He}$ lavas in Iceland, and global**
661 **hotspots, do not overlap with the common mantle component (shown as ellipsoid (27)).** Four
662 different high- $^3\text{He}/^4\text{He}$ ($> 25 \text{ Ra}$) components in Iceland (Vestfirðir, ERZ, SIVZ, Baffin Island) are shown.
663 The highest $^3\text{He}/^4\text{He}$ ($> 30 \text{ Ra}$) samples from each global oceanic hotspot with available Sr-Nd-Pb
664 isotopes—Galapagos, Hawaii, Samoa—are also shown (data sources for these additional lavas are in SI
665 Appendix, Figure S4). EM1, EM2, and HIMU apices of the tetrahedron are published elsewhere (27), as
666 is MORB (63), but EM2 extends to more extreme compositions (36) (indicated by arrow). Data for the
667 highest $^3\text{He}/^4\text{He}$ lavas with available Sr-Nd-Pb isotopes from the different Iceland high- $^3\text{He}/^4\text{He}$ ($> 25 \text{ Ra}$)
668 localities are from SI Appendix, Dataset S1; the Baffin Island lava is the highest $^3\text{He}/^4\text{He}$ (39.9 Ra) lava
669 that is the least crustally contaminated (23). Measured values are shown for all lavas except for Baffin
670 Island, where both age-corrected [c] and measured compositions are shown: the calculated present-day
671 Baffin Island mantle source is indistinguishable from the measured composition in the figure (see SI
672 Appendix, Figure S4 caption for details of the age correction and present-day source calculation).
673

674
675 **Figure 2. Relationships between $^3\text{He}/^4\text{He}$, $^{87}\text{Sr}/^{86}\text{Sr}$, and $^{206}\text{Pb}/^{204}\text{Pb}$ in Iceland hotspot lavas**
676 **showing four geochemically distinct high- $^3\text{He}/^4\text{He}$ Iceland components (Vestfirðir, ERZ, SIVZ, and**
677 **Baffin Island).** The highest $^3\text{He}/^4\text{He}$ lavas from Hawaii, Galapagos, and Samoa also exhibit
678 heterogeneous Sr-Nd-Pb isotopic compositions. Data sources for these lavas are in SI Appendix, Figure
679 S4. Only the least crustally contaminated Baffin Island lavas are shown (23). Measured heavy radiogenic
680 isotopic compositions are shown for all samples except for Baffin Island, where present-day mantle

681 source values are plotted (23) (see SI Appendix, Figure S4 caption for details of present-day source
682 calculation). For Iceland hotspot lavas, only high-precision Pb isotopic data (MC-ICP-MS and double-
683 spike TIMS) are shown. Upper Panel: The colored symbols represent lavas with $^3\text{He}/^4\text{He} > 21 \text{ Ra}$
684 sampling all known high- $^3\text{He}/^4\text{He}$ regions of the Iceland hotspot—ERZ, SIVZ, NRZ (Vaðalda), Vestfirðir,
685 and BIWG—and white symbols represent Iceland lavas (all of which have $^3\text{He}/^4\text{He} < 21 \text{ Ra}$) from the
686 other regions of Iceland. Lower Panel: Colored symbols represent lavas with $^3\text{He}/^4\text{He} > 21 \text{ Ra}$ sampling
687 all known high- $^3\text{He}/^4\text{He}$ regions of the Iceland hotspot, and the white symbols represent Iceland lavas with
688 $^3\text{He}/^4\text{He} \leq 21 \text{ Ra}$ (or lavas that have not been characterized for $^3\text{He}/^4\text{He}$).

689

690 **Figure 3. Relationships between $^3\text{He}/^4\text{He}$, $\mu^{182}\text{W}$, $^{143}\text{Nd}/^{144}\text{Nd}$, and $^{206}\text{Pb}/^{204}\text{Pb}$ in global OIB showing**
691 **that only geochemically-depleted OIB (i.e., high $^{143}\text{Nd}/^{144}\text{Nd}$ and low $^{206}\text{Pb}/^{204}\text{Pb}$) lacking recycled**
692 **crust signatures host the highest $^3\text{He}/^4\text{He}$ and largest magnitude $\mu^{182}\text{W}$ anomalies.** The compilation
693 of He-Sr-Nd-Pb isotopes (from this study and prior studies; SI Appendix, Dataset S4) for lavas with
694 available $\mu^{182}\text{W}$ shows that only lavas with low $^{206}\text{Pb}/^{204}\text{Pb}$ (top panel) or high $^{143}\text{Nd}/^{144}\text{Nd}$ (middle panel)
695 host the most extreme negative $\mu^{182}\text{W}$ anomalies. Lavas with the most extreme negative $\mu^{182}\text{W}$ anomalies
696 also exhibit high $^3\text{He}/^4\text{He}$ (bottom panel, where only lavas with available $\mu^{182}\text{W}$ data are shown). In
697 contrast, the mantle endmembers EM1, EM2, and HIMU have low $^3\text{He}/^4\text{He}$ and lack anomalous $\mu^{182}\text{W}$, as
698 does DM. The chondritic reference $^{143}\text{Nd}/^{144}\text{Nd}$ is published elsewhere (64). All data and error bars (2σ)
699 are provided in SI Appendix, Dataset S4. The $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ for Ontong Java Plateau (OJP)
700 is an average of Kroenke- and Kwaimbaita-type lavas (65); present-day mantle source compositions (23)
701 are show in lieu of measured or age-corrected ages due to the 120 Ma eruption age. The OJP $\mu^{182}\text{W}$ data
702 shown lack of anomalous signatures (41,46).

703

704 **Figure 4. Impact of mixing subducted crust and depleted mantle (DM) on OIB mantle domains with**
705 **high- $^3\text{He}/^4\text{He}$ and anomalous $\mu^{182}\text{W}$ compositions.** The mixing models demonstrate how subducted
706 crustal materials, and DM, reduce $^3\text{He}/^4\text{He}$ and overprints resolvable $\mu^{182}\text{W}$ anomalies in the mantle,
707 thereby generating the negatively sloping OIB array. Sediment (large grey box), oceanic crust (large grey
708 triangle), and depleted mantle (DM, large black box) are each mixed with four OIB mantle sources with

709 high $^3\text{He}/^4\text{He}$ and anomalous $\mu^{182}\text{W}$. Endmember compositions used are in SI Appendix, Dataset S5.
710 Samoa and Hawaii have indistinguishable trends (14-16), but Samoan $\mu^{182}\text{W}$ is characterized to higher
711 $^3\text{He}/^4\text{He}$. Intervals are marked at 10% for mixtures with DM; for mixtures with sediment and oceanic crust,
712 intervals are marked at 1% for the first 10% addition of subducted material, thereafter every 20%.
713 Sediment (derived from continental crust) and oceanic crust generate compositions similar to the extreme
714 Samoan EM2 lava (5% subducted sediment), the extreme Pitcairn EM1 lava (3% sediment), and the
715 extreme Mangaia HIMU lavas (25% oceanic crust) (see SI Appendix, Datasets S4 and S5). OIB data
716 symbols are the same as in Figure 3. Symbols are the same as in Figure 3; OIB data symbols and error
717 bars (2σ) are provided in SI Appendix, Dataset S4.

718

719 **Figure 5. $\mu^{182}\text{W}$ as a function of $D^{\text{Sr-Nd-Pb}}$ showing that OIB sampling significant recycled crust**
720 **(represented by extreme EM1, EM2, and HIMU lavas), or MORB, lack $\mu^{182}\text{W}$ anomalies.** Left panel:
721 Samples with the lowest $D^{\text{Sr-Nd-Pb}}$ (defined in main text) have the smallest contributions from the EM1,
722 EM2, and HIMU mantle endmembers (and the smallest recycled crust contributions). Samples with the
723 highest $D^{\text{Sr-Nd-Pb}}$ plot closest to EM1, EM2, and HIMU in Sr-Nd-Pb isotopic space (and host the greatest
724 recycled crust contributions). Binary mixing lines are shown between a melt with anomalous $\mu^{182}\text{W}$ and
725 melts of EM1 (blue line), EM2 (pink line), HIMU (purple line), and MORB melts of DM (black mixing line)
726 (see SI Appendix, Dataset S6 for model compositions); 10% mixing increments are shown. Symbols are
727 the same as Figure 3 and data (and 2σ errors) are from SI Appendix Dataset S4 (except for OJP data,
728 which are indicated in Figure 3). Right panel: Illustrates calculation of $D^{\text{Sr-Nd-Pb}}$ values and shows
729 examples of three large $D^{\text{Sr-Nd-Pb}}$ values, one medium $D^{\text{Sr-Nd-Pb}}$ value, and one small $D^{\text{Sr-Nd-Pb}}$ value; all $D^{\text{Sr-Nd-Pb}}$
730 $D^{\text{Sr-Nd-Pb}}$ values are distances in $^{87}\text{Sr}/^{86}\text{Sr}$ - $^{143}\text{Nd}/^{144}\text{Nd}$ - $^{206}\text{Pb}/^{204}\text{Pb}$ isotope space calculated between the
731 depleted reference composition (Depl. Ref. Comp.) and OIB samples.

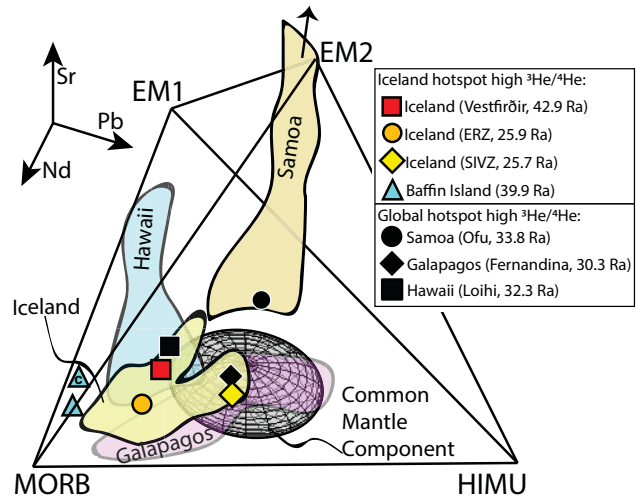


Figure 1

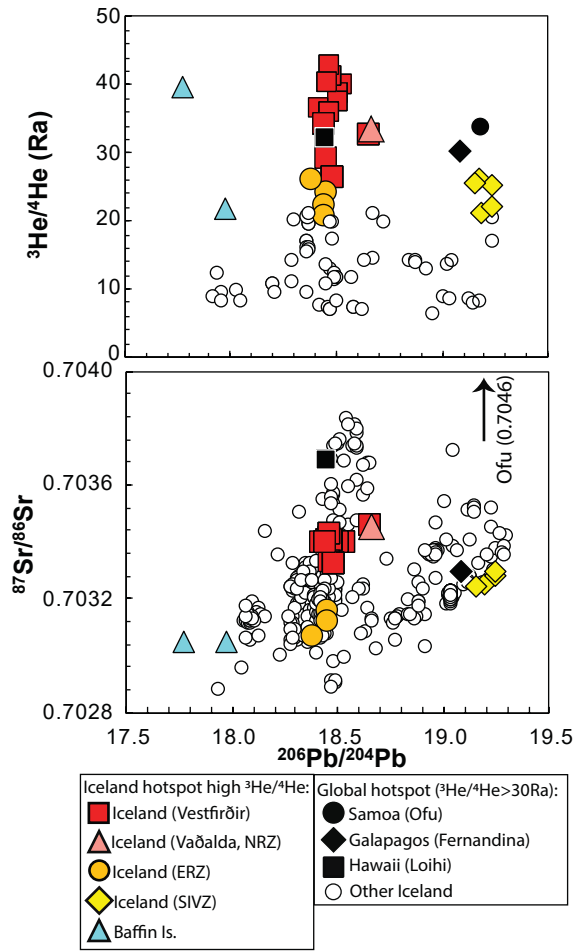


Figure 2

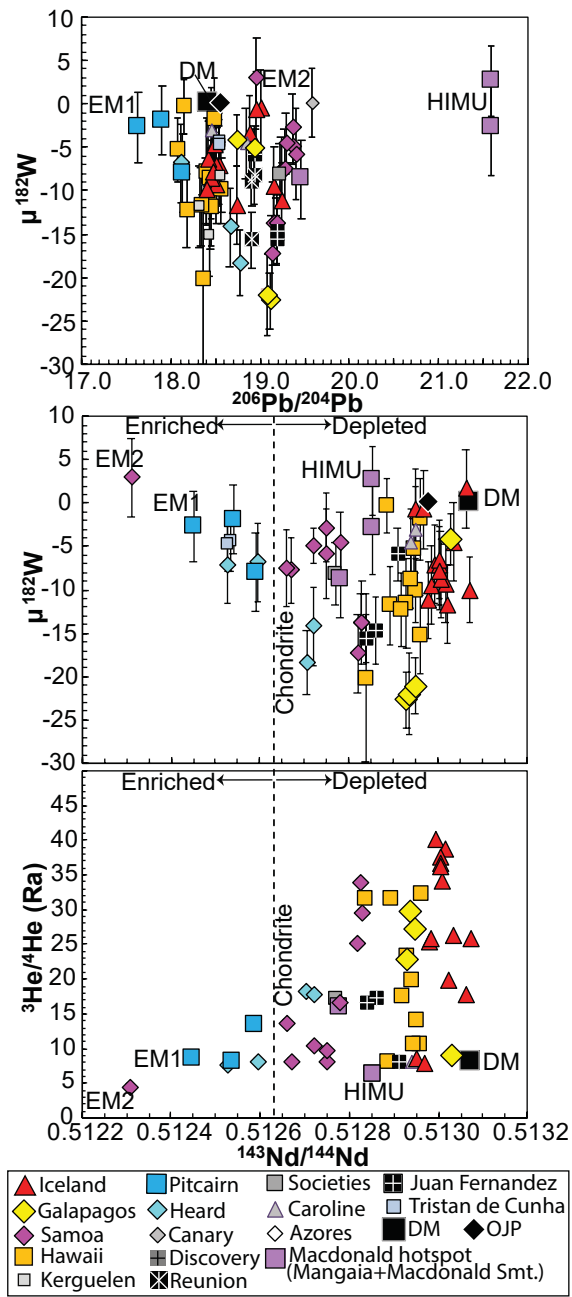


Figure 3

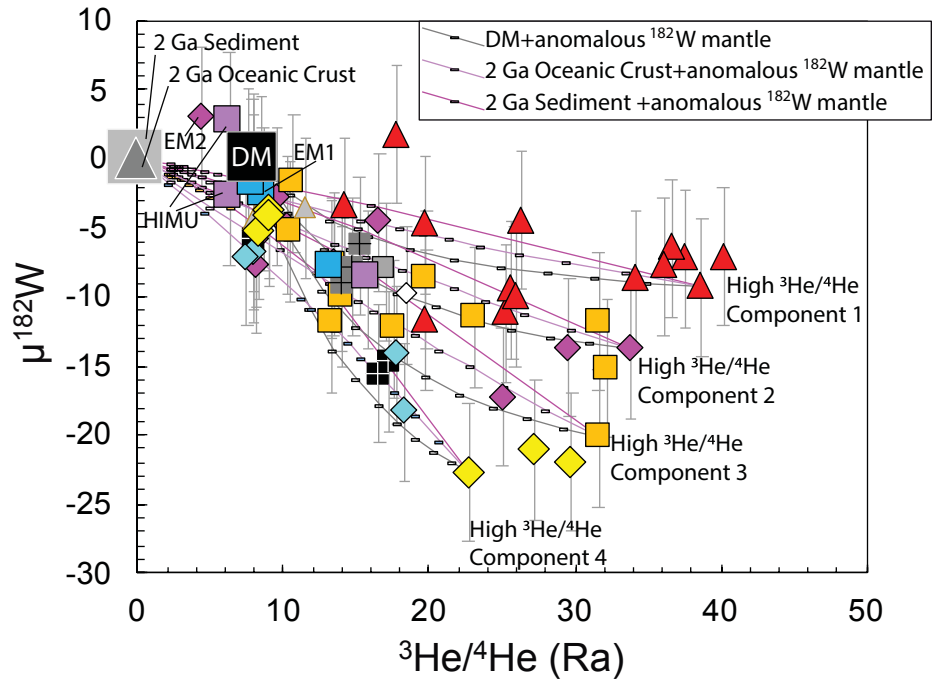


Figure 4

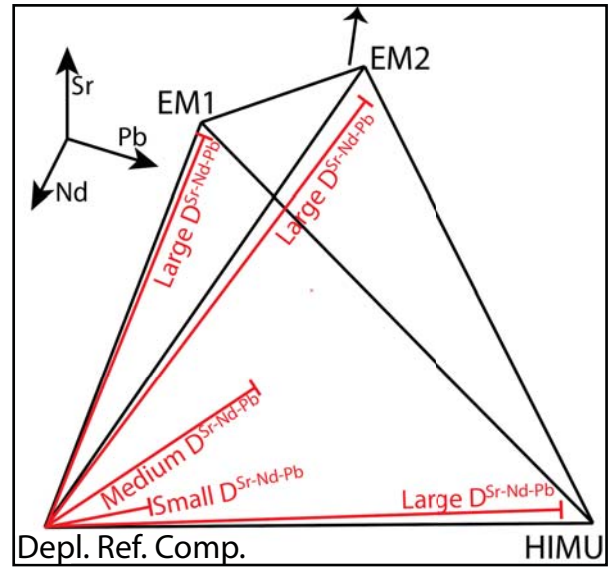
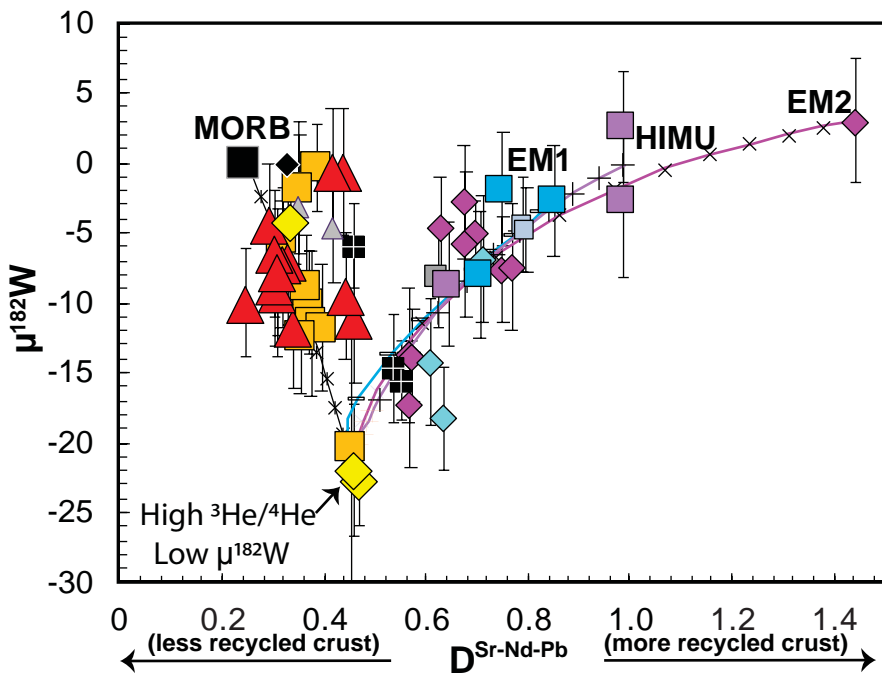


Figure 5