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# Differentiation mechanisms of the early Hadean mantle: Insights from combined $^{176}\text{Hf}$ - $^{142,143}\text{Nd}$ signatures of Archean rocks from the Saglek Block

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1           **Differentiation mechanism of the early Hadean mantle: insights from**  
2 **combined  $^{176}\text{Hf}$ - $^{142,143}\text{Nd}$  signatures of Archean rocks from the Saglek Block.**

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13

14

15 *Abstract*

16 *The presence of positive  $^{142}\text{Nd}$  anomalies in Eoarchean rocks provides evidence for early ( $4.40\pm 0.03$*   
17 *Ga; Morino et al., 2017) depletion of Earth's mantle. This model age lies within errors of the Pb-Pb*  
18 *"age of the Earth" (Connelly and Bizzarro, 2016), and is similar to model ages inferred for*  
19 *crystallization of the lunar mantle (McLeod et al., 2014), implying that this global event may reflect*  
20 *crystallization of a global magma ocean following the Moon-forming impact. However, the*  
21 *differentiation mechanism responsible for the formation of this early depleted reservoir and the depth*  
22 *at which it formed cannot be constrained from the Sm-Nd isotope system alone, because the magnitude*  
23 *of Sm/Nd fractionation during partial melting or fractional crystallization shows little dependence on*  
24 *pressure-controlled changes in mantle mineralogy. In contrast, the Lu-Hf isotope system is highly*  
25 *dependent on mineralogy, notably the presence or absence of garnet, an upper mantle phase, and thus*

26 may be used to constrain the pressure of fractionation. This study provides the first  $^{176}\text{Lu}$ - $^{176}\text{Hf}$   
27 isotopic results on mafic and ultramafic rocks belonging to the Eoarchean (Nulliak) and Mesoarchean  
28 suites of the Saglek Block (northern Labrador, 3.2-3.9 Ga). The  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  dating confirms the  
29 distinction between these two groups of rocks and provides ages consistent with those obtained from  
30  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  dating. The whole rock  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  errorchrons yield ages and initial epsilon values of  
31  $3766\pm 140$  Ma,  $\epsilon^{176}\text{Hf}_i=6.0\pm 2.5$  and  $3023\pm 390$  Ma,  $\epsilon^{176}\text{Hf}_i=-0.3\pm 2.5$  for the Nulliak suite and the  
32 Mesoarchean suite respectively. The time-integrated  $^{176}\text{Lu}/^{177}\text{Hf}$  for the sources of the Nulliak and the  
33 Mesoarchean suites considering a time of differentiation at  $4.40\pm 0.03$  Ga are estimated to be  
34  $0.047\pm 0.005$  and  $0.033\pm 0.005$ , respectively. For the Mesoarchean samples, the combined  $^{146,147}\text{Sm}$ -  
35  $^{142,143}\text{Nd}$  and  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  data are consistent with a near-chondritic mantle source. On the other hand,  
36 Nulliak ultramafic rocks were derived from a mantle reservoir with superchondritic Lu/Hf and Sm/Nd.  
37 The Nulliak parent reservoir, however, does not plot on the  $\epsilon^{176}\text{Hf}$ - $\epsilon^{143}\text{Nd}$  mantle array defined by  
38 modern oceanic basalts. Instead, the Nulliak source more likely belongs to a distinct array defined by  
39 Eo- and Meso-Archean komatiites. These results are interpreted in the framework of a simple model of  
40 crystallization of a primordial magma ocean. It appears that the fractionation observed in the mantle  
41 source of Nulliak was most likely generated by crystallization of a garnet-bearing assemblage in the  
42 shallow mantle, above the transition zone rather than by perovskite fractionation in the lower mantle.  
43 To preserve this depleted reservoir from the rest of the hot and vigorously convecting mantle, the  
44 Nulliak mantle source may have been isolated either at the top of the mantle in a buoyant lithosphere  
45 or near the core-mantle boundary, with the latter setting being more consistent with the komatiitic  
46 nature of the erupted rocks. Given that the garnet signature argues for differentiation of the Nulliak  
47 source at relatively shallow depth, its isolation in the deep mantle would require a cumulate overturn  
48 following crystallization of the magma ocean.

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## 1. Introduction

Models of planet formation indicate that the final stage of Earth's accretion was punctuated by highly energetic collisions with planetary embryos (Chambers and Wetherill, 1998), following their rapid growth from a swarm of planetesimals (Chambers, 2004; Morbidelli et al., 2012). The last of these collisions (Canup and Asphaug, 2001), likely the Moon-forming impact, is believed to have induced large-scale melting of the proto-Earth's mantle, generating a global magma ocean that may have extended down to the core-mantle boundary (CMB) (Canup, 2004). Magma ocean crystallization could potentially have resulted in gross chemical stratification of Earth's mantle (Ohtani, 1985), sequestration of a basal molten layer near the CMB (Labrosse et al., 2007; Coltice et al., 2011) and the extraction of a primordial crust by upward migration of residual melts (Caro et al., 2005; Bourdon and Caro, 2007). Nevertheless, the long-term consequences of this event on the structure and composition of Earth's mantle, as well as the ultimate fate of primordial silicate reservoirs remain speculative, obscured by 4.5 Gyr of geodynamic activity and complete rejuvenation of Earth's surface. Our understanding of primary differentiation processes thus mainly relies on indirect observations based on short-lived radioactive decay systems (Caro et al., 2003; Touboul et al., 2012; Mukhopadhyay, 2012; Caracausi et al., 2016; Rizo et al., 2016; Mundl et al., 2017; Peters et al., 2018), which provide temporal and compositional information on primordial silicate reservoirs. One of these observations is that positive  $^{142}\text{Nd}$  anomalies (i.e. high  $^{142}\text{Nd}/^{144}\text{Nd}$  compared to the homogeneous value of the present-day mantle) are present in Archean terranes of West Greenland and Northern Canada (Caro et al., 2006; Bennett et al., 2007; Rizo et al., 2012; Debaille et al., 2013; Morino et al., 2017). Such anomalies were produced by decay of now extinct  $^{146}\text{Sm}$  ( $T_{1/2}=103$  Myr) in a mantle reservoir that was depleted in incompatible elements near the end of terrestrial accretion, at  $4.40\pm 0.03$  Ga (Morino et al., 2017). This reservoir (hereafter referred to as Early Depleted Mantle or EDM) was episodically sampled by Archean magmatism at least until 2.7 Gyr ago (Debaille et al., 2013). Model age calculations using coupled  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  chronometry indicate that the differentiation event responsible for the formation of the EDM predated late-stage crystallization of the lunar magma ocean by  $<50$  Ma (Solomon and Longhi, 1977; Boyet and Carlson, 2007; Borg et al., 2011; Elkins-Tanton et al., 2011; McLeod et al., 2014). This time interval is consistent with slower cooling of the residual

79 lunar magma ocean under the blanketing effect of a thick anorthositic crust (Elkins-Tanton et al.,  
80 2011). The  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  age of the EDM also lies within error of the Pb-Pb age of the Earth, at  
81 ca.  $4.42\pm 0.01$  Ga (Connelly and Bizzarro, 2016), which was suggested to record Pb loss by  
82 devolatilization or by sequestration in the core during the giant impact phase of accretion (Oversby  
83 and Ringwood, 1971; Wood et al., 2006; Lagos et al., 2008; Wood et al., 2010; Connelly and Bizzarro,  
84 2016). Collectively, these chronological constraints point towards a major differentiation event at ca.  
85 4.40 Ga, possibly triggered by the Moon-forming impact, and ultimately resulting in global chemical  
86 differentiation of the mantle-crust system.

87 While the chronological aspects of early mantle differentiation are well established from  
88 coupled  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  systematics, fundamental uncertainties still remain as to the physical  
89 process that produced the depleted reservoir carrying positive  $^{142}\text{Nd}$  anomalies. Preservation of  
90 primordial mantle domains on a billion year timescale requires long-term isolation from the  
91 convective system (Morino et al., 2017). This observation is consistent with creation of positive  $^{142}\text{Nd}$   
92 anomalies in a chemically buoyant lithospheric mantle, which may have been preserved from  
93 rehomogenization as part of a long-lived Hadean stagnant lid (Debaille et al., 2013; Caro et al., 2017).  
94 Alternatively, the presence of positive  $^{142}\text{Nd}$  anomalies in Eoarchean rocks may reflect entrainment  
95 within hot plumes of perovskitic cumulates formed by fractional crystallization of a basal magma  
96 ocean (Labrosse et al., 2007). The  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  systems alone cannot provide definitive  
97 constraints on this issue because the magnitude of Sm/Nd fractionation during partial melting or  
98 fractional crystallization shows little dependence on pressure-controlled changes in mantle  
99 mineralogy, i.e., Sm is less incompatible than Nd throughout the mantle, so cumulates with  
100 moderately elevated Sm/Nd would be produced at all mantle depths. To circumvent this limitation,  
101 Caro et al. (2005) proposed an approach based on coupled  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  and  $^{176}\text{Lu}$ - $^{176}\text{Hf}$   
102 systematics, with the aim of fingerprinting the crystallization of Mg-perovskite (bridgmanite) at lower  
103 mantle pressures. Unlike Sm/Nd, Lu/Hf partitioning is strongly dependent on mantle mineralogy (e.g.  
104 Salters and White, 1998; Corgne and Wood, 2004). At upper mantle pressures pertaining to partial  
105 melting and/or fractional crystallization in the presence of garnet, Lu is less incompatible than Hf.

106 Crystallization of a shallow (<660 km) magma ocean is thus expected to generate positively correlated  
107 Lu/Hf and Sm/Nd fractionations, provided that melt segregation proceeded faster than upward  
108 migration of the solidification front (Solomatov and Moresi, 1996; Solomatov and Louis, 2007). In  
109 contrast, at lower mantle pressures in the presence of bridgmanite, Hf becomes more compatible than  
110 Lu (Corgne et al., 2004), so that crystallization of a deep magma ocean is expected to produce  
111 negatively correlated Lu/Hf and Sm/Nd fractionations (Caro et al., 2005). The  $^{176}\text{Hf}$ - $^{143}\text{Nd}$  signatures  
112 of Eoarchean rocks carrying  $^{142}\text{Nd}$  anomalies, which ultimately reflect the time-integrated Lu/Hf and  
113 Sm/Nd of their sources, thus have the potential to provide important constraints on the depth and  
114 processes pertaining to differentiation of the EDM.

115 The approach outlined above requires determination of both the Sm/Nd and Lu/Hf of the  
116 mantle reservoir hosting the positive  $^{142}\text{Nd}$  anomaly. While the former can be precisely determined  
117 from coupled  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  systematics (Caro et al., 2003; Caro et al., 2006; Bennett et al., 2007;  
118 Rizo et al., 2011; Rizo et al., 2012; O'Neil et al., 2016), previous attempts to estimate  $(\text{Lu}/\text{Hf})_{\text{EDM}}$  from  
119 Eoarchean mafic rocks of the Isua Supracrustal Belt (ISB, West Greenland; 3.7-3.8 Ga) yielded  
120 conflicting results. Rizo et al. (2011) and Hoffmann et al. (2011) reported unradiogenic initial  
121  $^{176}\text{Hf}/^{177}\text{Hf}$  signatures in amphibolites of tholeiitic affinity sampled in the northwestern section of the  
122 ISB. This result, indicative of a subchondritic  $(\text{Lu}/\text{Hf})_{\text{EDM}}$ , is consistent with melting of cumulates  
123 mainly composed of bridgmanite, suggesting derivation of ISB metabasalts from a deep mantle source  
124 (Rizo et al., 2011). At odds with this interpretation, Hoffmann et al. (2010) reported highly radiogenic  
125 initial  $^{176}\text{Hf}/^{177}\text{Hf}$  in boninite-like metabasalts situated in the Eastern section of the ISB. This  
126 radiogenic signature requires a mantle source with superchondritic Lu/Hf, seemingly more consistent  
127 with its prior depletion in the garnet stability field. Remarkably, both magmatic series are  
128 characterized by a positive  $^{142}\text{Nd}$  anomaly averaging +10 ppm (Caro et al., 2006; Hoffmann et al.,  
129 2010; Rizo et al., 2011; O'Neil et al., 2016), and yielded similar time-integrated  $^{147}\text{Sm}/^{144}\text{Nd}$  source  
130 ratios (ca. 0.210-0.215). Hoffmann et al. (2011) suggested that decoupling of  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  and  $^{147}\text{Sm}$ -  
131  $^{143}\text{Nd}$  systems in Isua meta-tholeiites may reflect contamination of their source by detrital  
132 metasediments with low Lu/Hf recycled in a proto-subduction zone, rather than a pristine signature

133 from a deep perovskitic reservoir. Conversely, the highly radiogenic  $^{176}\text{Hf}/^{177}\text{Hf}$  measured in boninite-  
134 like amphibolites may indicate contamination of their source by LREE-poor chemical sediments  
135 characterized by high Lu/Hf (e.g. cherts), as suggested by Blichert-Toft et al. (2015) to explain the  
136 apparent decoupling of  $^{176}\text{Hf}$ - $^{143}\text{Nd}$  signatures in South African komatiites.

137 The Eoarchean Nulliak suite of the Saglek Block (Labrador, Canada), constituted of mafic and  
138 ultramafic rocks, provides an opportunity to test these scenarios and better constrain the geodynamic  
139 evolution of the EDM following solidification of the magma ocean. Unlike many occurrences of  
140 ultramafic rocks in Eoarchean terranes, the Nulliak suite underwent only partial serpentinization, and  
141 yielded a precise  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  isochron age and initial  $^{143}\text{Nd}/^{144}\text{Nd}$  (Morino et al., 2017). All mafic and  
142 ultramafic rocks of the Nulliak assemblage exhibit a homogeneous  $^{142}\text{Nd}$  anomaly of  $8\pm 3$  ppm,  
143 allowing calculation of a model age of differentiation for their mantle source at  $4.40\pm 0.03$  Ga with a  
144 time-integrated  $^{147}\text{Sm}/^{144}\text{Nd}$  of 0.211. In this study, we present the first  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  isotopic data for  
145 mafic and ultramafic rocks of the Saglek block, using the two sample suites previously analyzed for  
146  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  by Morino et al. (2017). We show that the Nulliak source composition inferred from  
147  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  and  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  systematics can be reproduced by crystallization of a shallow  
148 magma ocean in the garnet stability field.

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## 150 **2. Samples and Geologic Setting**

151 The samples analyzed in this study were collected during fieldwork in the Hebron-Saglek fjord  
152 region of the Saglek block (3.2-3.9 Ga, northern Labrador, Canada). Regional geology and prior  
153 geochronological work were summarized in several previous studies (e.g. Schiøtte et al., 1989,  
154 Bridgwater and Schiøtte, 1991; Schiøtte et al., 1992; Komiya et al., 2015; Morino et al., 2017).  
155 Geographic locations, as well as major and trace element compositions of our samples, are provided in  
156 Morino et al. (2017). The Saglek block (Fig 1) is located near the northern extremity of the Labrador  
157 Peninsula, across from the Akuleq terrane of Southwest Greenland, and represents the westernmost  
158 part of the North Atlantic craton. The Saglek block is divided into two tectonic domains separated by

159 the NS-trending, subvertical Handy Fault (Ryan and Martineau, 2012). The western domain  
160 underwent high grade metamorphism in the granulite facies at ca. 2.7-2.8 Ga, while the eastern domain  
161 is primarily characterized by amphibolite facies metamorphism (Ryan and Martineau, 2012). Both  
162 domains are dominated by Eoarchean quartzofeldspathic gneisses of the Uivak suite (3.6-3.9 Ga) with a  
163 subordinate Mesoarchean component (the Lister gneisses, 3.2 Ga) and late (ca. 2.4 Ga) intrusions of  
164 post-tectonic granitoids (Komiya et al., 2015; Shimojo et al., 2016). Numerous volcano-sedimentary  
165 enclaves are present in the area, embedded in Eoarchean gneisses of the Uivak suite (Ryan and  
166 Martineau, 2012). On the basis of field relationships of supracrustal rocks with the Mesoarchean  
167 Saglek dyke swarm, Bridgwater and Schiøtte (1991) and Ryan and Martineau (2012) proposed a  
168 subdivision of these units into an Eoarchean group called the Nulliak assemblage, and a Mesoarchean  
169 group called the Upernavik formation. On the basis of whole-rock  $^{147}\text{Sm}$ - $^{143}\text{Nd}$ ,  $^{187}\text{Re}$ - $^{187}\text{Os}$  and U-Pb  
170 zircon dating, the emplacement age of the Nulliak assemblage was estimated to be 3.6-4.0 Ga  
171 (Collerson et al., 1991, Schiøtte et al., 1992, Ishikawa et al., 2017, Morino et al., 2017). The  
172 emplacement age of Upernavik supracrustal rocks remains poorly constrained but is likely between 3.1  
173 and 3.4 Ga (Schiøtte et al., 1992, Morino et al., 2017, Ishikawa et al., 2017).

174 Our sample set includes 15 ultramafic rocks (4 metadunites, 11 metapyroxenites) and 5  
175 amphibolites collected east of the Handy fault, in the vicinity of Jerusalem Harbour and along the  
176 coast north of Iterugnek fjord. In the field, mafic/ultramafic rocks occur either as monolithological  
177 meter-sized enclaves (LA9 series) or as larger (50-500 m) mafic/ultramafic units (LA11 and LA8  
178 series) enclosed in granitoid gneisses of the Uivak suite. The samples were further divided into two  
179 groups on the basis of their  $^{146}\text{Sm}$ - $^{142}\text{Nd}$  and  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  systematics (Morino et al 2017). A first  
180 group, which displays a positive  $^{142}\text{Nd}$  anomaly of  $+8\pm 3$  ppm, yielded a  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  whole-rock  
181 isochron age of  $3.78\pm 0.09$  Ga and was thus assigned to the Eoarchean Nulliak assemblage. The second  
182 group, without  $^{142}\text{Nd}$  anomaly, yielded a  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  isochron age of  $3.36\pm 0.10$  Ga and was  
183 tentatively assigned to the Mesoarchean Upernavik formation.

184 Although primary textures indicative of a volcanic origin were not preserved, ultramafic rocks  
185 from both the Meso- and Eoarchean suites exhibit geochemical characteristics consistent with



186 differentiation of komatiitic lavas (Morino et al 2017). Using a previously established komatiite  
187 classification scheme (Nesbitt and Su, 1979), ultramafic rocks were therefore divided into Al-depleted,  
188 Al-undepleted and Al-enriched subgroups according to their major and trace element compositions  
189 (Morino et al 2017).

### 190 **3. Metasomatic alteration and crustal contamination**

191 Supracrustal rocks of the Saglek block have undergone poly-phase metamorphism in the  
192 amphibolite to granulite facies, as well as metasomatic alteration. These events resulted in extensive  
193 recrystallization, serpentinization and redistribution of fluid mobile elements (e.g. U, Ba, Rb, K) from  
194 the surrounding granitoid gneisses to the ultramafic protoliths (Liu et al., 2016, Morino et al, 2017).  
195 The geochemical effects of post-emplacement alteration are broadly similar to those reported in mafic  
196 rocks (amphibolites) of the Isua Supracrustal Belt (Polat and Hofmann, 2003). In plots of selected  
197 REE (Rare Earth Elements) and HFSE (High Field Strength Elements) versus immobile trace elements  
198 such as Zr, samples of the Meso- and Eoarchean suites display systematic variations (Fig 2 A-D)  
199 reflecting preserved magmatic differentiation trends. This observation is consistent with the limited  
200 mobility of HFSE and REE during metamorphism. For the Eoarchean suite, excluding possibly  
201 contaminated samples (see below), correlation coefficients (R) in binary plots versus Zr tend to  
202 decrease from the heavy and intermediate REE (Lu: R=0.87, Sm: R=0.97) to the light REE (Nd:  
203 R=0.73, La: R=0.22) reflecting the higher mobility of LREE during metamorphism. This effect is  
204 however less pronounced for the Mesoarchean suite (R>0.95 for Sm, Lu, Nd; R=0.83 for La). All  
205 HFSE are tightly correlated with Zr (Hf: R>0.99 for both the Meso- and Eoarchean suites) indicating  
206 little post-magmatic mobility. In contrast, secondary enrichments in Th and U are evident in both  
207 Meso- and Eoarchean samples (Fig. 2 E-F), consistent with the previously reported mobility of these  
208 elements in high grade metamorphic terranes (Polat and Hofmann, 2003).

209 The effects of crustal contamination on the trace element composition of Saglek ultramafic  
210 rocks are illustrated in Figure 3, using  $(Ce/Yb)_N$  as a measure of LREE enrichment (the subscript N  
211 indicates normalization to primitive mantle values of McDonough and Sun (1995)). The extent of  
212 crustal assimilation is difficult to assess with confidence due to superimposed chemical variability

213 induced by metasomatism, fractionation during melt extraction and intrinsic source heterogeneities.  
214 Based on literature data, uncontaminated komatiites, nevertheless, define compositional domains in  
215 plots of selected trace element ratios versus  $(\text{Ce}/\text{Yb})_N$ , which can be used to screen for anomalous  
216 LREE enrichment (Fig 3). These komatiites exhibit flat to depleted trace element patterns, with  
217  $(\text{Ce}/\text{Yb})_N < 2$ ,  $(\text{Sm}/\text{Nd})_N = 0.8-2$ ,  $(\text{Th}/\text{La})_N = 0.5-1$  and  $(\text{Nb}/\text{La})_N \approx 1$ . Crustal contamination is expected to  
218 translate into LREE enrichment, pronounced negative Nb anomalies and variable enrichment in highly  
219 incompatible elements such as Th and U (e.g. Puchtel et al., 2016). On the basis of their Ce/Yb ratio,  
220 and the binary plots of Figure 2, it appears that two samples of the Eoarchean suite (LA9-18 and LA9-  
221 19) and one sample of the Mesoarchean suite (LA11-14) may have been affected by crustal  
222 contamination. Both Eoarchean samples display a marked enrichment in LREE with  $(\text{Ce}/\text{Yb})_N = 3-4$   
223 and  $(\text{Sm}/\text{Nd})_N \approx 0.7$  (Fig. 3) as well as anomalously high Zr (and other incompatible elements) contents,  
224 consistent with assimilation of felsic material. Both samples also plot above magmatic differentiation  
225 trends in diagrams of Hf, Sm and Nd versus MgO. Aside from these two samples, however, ultramafic  
226 rocks from the Nulliak group display highly depleted LREE signatures (Fig 3A) and therefore show no  
227 compelling evidence of crustal assimilation. Mesoarchean samples exhibit slightly higher  $(\text{Ce}/\text{Yb})_N$ ,  
228 typically between 1.5 and 2, but, with the exception of LA11-14, all samples remain within the  
229 compositional range defined by Barberton and Abitibi komatiites. Both the Meso- and Eoarchean  
230 suites also exhibit variable but generally high  $(\text{Th}/\text{La})_N$  and low  $(\text{Nb}/\text{La})_N$  compared to typical  
231 komatiite values (Fig 3). However, neither ratio correlate with  $(\text{Ce}/\text{Yb})_N$ . Given the poor correlations  
232 between Zr and La and Th (Fig. 2E), we interpret these anomalous signatures to reflect Th and La  
233 redistribution during metasomatism associated with Neoproterozoic metamorphism rather than pervasive  
234 crustal contamination.

235 The variability of Lu/Hf values in Saglek ultramafic rocks is similar to that observed in Archean  
236 komatiites (Fig 4) and can thus be interpreted in terms of trace element fractionation during melt  
237 extraction. As shown in Figure 4, Lu/Hf in the Meso- and Eoarchean suites displays a positive  
238 correlation with  $\text{Al}_2\text{O}_3/\text{TiO}_2$  (and a negative correlation with Gd/Yb), consistent with a primary control  
239 by the garnet component of their mantle source. The Al-depleted samples ( $\text{Al}_2\text{O}_3/\text{TiO}_2 \approx 5-11$ ) have the

240 lowest (Lu/Hf)<sub>N</sub> values (0.2-0.7) because garnet is not exhausted from the mantle source during partial  
241 melting, and strongly retains Lu relative to Hf (e.g. Robin-Popieul et al., 2012). For Eoarchean Al-  
242 depleted samples, this primary fractionation may have been accentuated by assimilation of crustal  
243 material with low Lu/Hf. In contrast, Al-enriched samples (Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub>≈30-50), derived from garnet-  
244 rich residues produced by prior extraction of low Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> melts, have the highest (Lu/Hf)<sub>N</sub> values  
245 (1.5-3). The Al-undepleted type (Al<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub>≈15-25) is characterized by near-primitive (Lu/Hf)<sub>N</sub>,  
246 reflecting first-stage melting above the garnet solidus.

#### 247 **4. Analytical procedure**

248 Lutetium and Hafnium isotopic analyses were performed on 15 ultramafic and 5 mafic rocks  
249 previously analyzed for <sup>146,147</sup>Sm-<sup>142,143</sup>Nd by Morino et al (2017). Different digestion aliquots from  
250 the same bulk powder were used for <sup>176</sup>Lu-<sup>176</sup>Hf and <sup>147</sup>Sm-<sup>143</sup>Nd analyses. Chemical separation of  
251 these elements was done in a controlled-air clean lab under a HEPA filtered laminar flow hood using  
252 equipment previously cleaned in HF. A mixed <sup>176</sup>Lu-<sup>180</sup>Hf spike was added to 100-300 mg whole rock  
253 powder and dissolved in a HNO<sub>3</sub>-HF mixture (1:1). The digestion was carried out in high-pressure  
254 PTFE/TFM BOLA bombs heated in an oven during 3 days at 150°C (Luais et al., 2009). The solutions  
255 were then slowly evaporated on a hot plate and the residues dissolved in 5 ml of 6 N HCl, after which  
256 the bombs were replaced in the oven for 3 days at 150°C. Lastly, the dissolved samples were  
257 transferred into Savillex vials and dried down then redissolved in 4 N HCl to break down residual  
258 fluorides. This final step was repeated several times until a perfectly clear solution was obtained.

259 Hafnium and Lutetium were separated from the rock matrix using a single-column procedure  
260 (Table 1) adapted from Munker et al. (2001), Wimpenny et al. (2013) and Yang et al. (2010). Upon  
261 complete dissolution of fluoride precipitates, samples were dried, dissolved in 5 mL of 3 N HCl + 0.1  
262 N ascorbic acid and introduced onto 1.8 mL Bio-Rad Poly-Prep columns filled with LN resin (50-100  
263 mesh, Eichrom technologies). The matrix and most of the REE were first eluted in 3 N HCl, after  
264 which Yb and Lu were eluted in 10 mL of 3.4 N HCl and 10 mL of 6 N HCl, respectively. Titanium  
265 was then rinsed off as a peroxide complex with up to 50 mL of a 3 N HNO<sub>3</sub> + 0.5% H<sub>2</sub>O<sub>2</sub> mixture,  
266 after which Hf and Zr were collected in 2 mL of 2 N HF. This protocol allows separation of the bulk

267 of Yb from the Lu fraction, thereby reducing uncertainties related to isobaric interference correction  
268 on  $^{176}\text{Lu}$ . The Ti/Hf in the final Hf cut was typically between 0.3 and 1, down from 300-600 in  
269 ultramafic rocks and 270-400 in mafic rocks. Tests performed using synthetic solutions with variable  
270 Ti/Hf ( $\approx 0.5$  to 200) confirmed that at such low levels, the presence of Ti has no significant effect on  
271 the accuracy and reproducibility of  $^{176}\text{Hf}/^{177}\text{Hf}$  measurements. The analysis of mixed standard  
272 solutions with Zr/Hf=35 also showed that the presence of Zr has no effect on measured  $^{176}\text{Hf}/^{177}\text{Hf}$   
273 values. Total procedural blanks measured for each purification were <50 pg for both Lu and Hf and  
274 chemistry yields were 80-90% for Lu and >90% for Hf.

275 Hafnium isotopes were analyzed on a Thermo Scientific *Neptune Plus* MC-ICPMS equipped  
276 with an Apex-HF inlet system (Elemental Scientific) and standard skimmer and sampler cones. Prior  
277 to analysis, the Hf separate was dissolved in 1 mL of 0.5 N  $\text{HNO}_3$  - 0.3 N HF. An aliquot of this  
278 solution was used to estimate the approximate Hf concentration of the sample, after which the solution  
279 was diluted to a concentration of 100 ppb, typically yielding a  $^{177}\text{Hf}$  signal of 4 V on  $10^{11} \Omega$  amplifiers.  
280 Hafnium isotopes were measured in static multicollection in low resolution mode (Table 2), collecting  
281 50 cycles with an integration time of 4.194 s. per cycle. Baselines were measured prior to each  
282 analysis with an integration time of 30 s. Potential isobaric interferences at mass 176 were monitored  
283 by collecting  $^{172}\text{Yb}$  and  $^{175}\text{Lu}$  in cups L4 and L2, respectively, and corrected offline using  
284  $^{172}\text{Yb}/^{176}\text{Yb}=1.711$  and  $^{175}\text{Lu}/^{176}\text{Lu}=37.666$  (Vervoort, 2013; Vervoort et al., 2004). However, neither  
285 Lu nor Yb were found in significant amounts ( $^{172}\text{Yb}/^{176}\text{Hf} < 2.5 \times 10^{-4}$  and  $^{175}\text{Lu}/^{176}\text{Hf} < 5 \times 10^{-4}$  in all  
286 cases) in the Hf separates following the chemical procedure and these corrections were always  
287 negligible. Likewise, neither W nor Ta were detected in Hf separates after chemical purification and  
288 thus no correction was applied for potential interferences at mass 180. The instrumental blank was  
289 measured for 40 s. prior to each sample or standard analysis, following a 6 minute wash period in 0.6  
290 N HF-1 N  $\text{HNO}_3$  (3 min.) and in 0.3 N HF-0.5 N  $\text{HNO}_3$  (3 min.), and was subtracted offline from the  
291 following sample/standard analysis. Instrumental mass discrimination was corrected by bracketing  
292 using the Nancy Ames standard, whose isotopic composition ( $^{176}\text{Hf}/^{177}\text{Hf}_{\text{Ames}}=0.282243 \pm 0.0000013$   
293 2S.D., n=30) was anchored to a  $^{176}\text{Hf}/^{177}\text{Hf}$  value of  $0.282156 \pm 0.000008$  (2S.D., n=21) for the inter-

294 laboratory JMC-475 standard. The  $^{176}\text{Hf}/^{177}\text{Hf}$  value for the JMC-475 standard is consistent with the  
295 accepted value of 0.282160 determined by Blichert-toft (2001).

296 The analytical protocol for Lu isotope measurements is similar to that used for Hafnium  
297 (Table 2). The Lu separate was dissolved in 2%  $\text{HNO}_3$  and diluted to a concentration of 20 ppb,  
298 equivalent to a 3.5 V signal on  $^{175}\text{Lu}$ . Instrumental mass fractionation was corrected by bracketing  
299 using the NIST standard reference material 3132a, which yielded an average  $^{175}\text{Lu}/^{176}\text{Lu}$  value of  
300  $0.0265732 \pm 0.0000055$  (2SD, n=15) similar within errors to the natural value obtained by Blichert-Toft  
301 et al. (1997). The Yb-Lu interference at mass 176 was monitored by collecting  $^{173}\text{Yb}$  and corrected  
302 offline using  $^{176}\text{Yb}/^{173}\text{Yb}=0.79639$ . The potential isobaric interference from  $^{176}\text{Hf}$  was monitored using  
303  $^{177}\text{Hf}$  and corrected using  $^{176}\text{Hf}/^{177}\text{Hf}=0.282160$  (Vervoort, 2015). However, no detectable amount of  
304 Hf was found in the Lu separates following chemistry. Instrumental blanks were measured for 40  
305 seconds prior to sample analysis following a 6 minute wash in 2%  $\text{HNO}_3$ .

306 The precision and accuracy of  $^{176}\text{Hf}/^{177}\text{Hf}$  and  $^{176}\text{Lu}/^{177}\text{Hf}$  determination was monitored by  
307 measuring spiked aliquots of the BIR-1 and BHVO-2 geostandards. The results of 7 analyses of  
308 BHVO-2 yielded an average  $^{176}\text{Hf}/^{177}\text{Hf}$  value of  $0.283094 \pm 0.000030$  (2 S.D.) and a  $^{176}\text{Lu}/^{177}\text{Hf}$  ratio  
309 of  $0.0086 \pm 0.0001$  (2 S.D.). Analyses of the BIR-1 geostandard (N=5) yielded  
310  $^{176}\text{Hf}/^{177}\text{Hf}=0.283256 \pm 0.000070$  and  $^{176}\text{Lu}/^{177}\text{Hf}=0.0584 \pm 0.0022$ . The  $^{176}\text{Hf}/^{177}\text{Hf}$  values obtained for  
311 both geostandards are within error of those previously reported (e.g. Weis et al., 2007, and references  
312 therein), confirming the accuracy of the spike corrected ratios.

313

314

## 5. Results

315 The  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  results from this study and previous  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  data for mafic and  
316 ultramafic rocks from the Saglek block are presented in Table 3 and summarized in Figure 5. By  
317 convention,  $^{176}\text{Hf}/^{177}\text{Hf}$  and  $^{143}\text{Nd}/^{144}\text{Nd}$  isotopic ratios are expressed by their relative deviation in parts  
318 per 10,000 from the chondritic value (Bouvier et al., 2008) at the time of interest, using the  
319 conventional epsilon ( $\epsilon$ ) notation. Variations of  $^{142}\text{Nd}/^{144}\text{Nd}$ , noted  $\mu^{142}\text{Nd}$ , are expressed by their

320 deviation (in ppm) from the JNdi standard, which is considered representative of the Bulk Silicate  
321 Earth.

322 Measured  $^{176}\text{Hf}/^{177}\text{Hf}$  values range from 0.281785 to 0.283040 among the Mesoarchean  
323 samples and from 0.281152 to 0.287879 among the Eoarchean samples (Table 3). Regressions of these  
324 values as a function of  $^{176}\text{Lu}/^{177}\text{Hf}$  were calculated using the Isoplot/Ex software (version 4.15;  
325 Ludwig, 2012), and initial  $^{176}\text{Hf}/^{177}\text{Hf}$  values were calculated from the whole-rock errorchrons using  
326 the method of Fletcher and Rosman (1982). **Uncertainties on  $^{176}\text{Hf}/^{177}\text{Hf}$  values are taken to  $\pm 35$  ppm  
327 and are based on the 2 S.D. variation of repeated analyses of the JMC-475 Hf standard.** Where two or  
328 more analyses were available for an individual sample, the mean value was used. Age calculations  
329 were performed using a  $^{176}\text{Lu}$  decay constant of  $1.867 \cdot 10^{-11}$  (Scherer et al., 2001; Söderlund et al.,  
330 2004). Errors on ages and initial  $^{176}\text{Hf}/^{177}\text{Hf}$  calculated by linear regression are quoted as 2 S.D. The  
331 results (Fig. 2) confirm the presence of two mafic/ultramafic suites in the Saglek block, as previously  
332 inferred from their  $^{142,143}\text{Nd}$  systematics (Morino et al 2017). Regression of the Mesoarchean group,  
333 without  $^{142}\text{Nd}$  anomaly, yields an age of  $3023 \pm 390$  Ma (MSWD=84) with an initial  
334  $^{176}\text{Hf}/^{177}\text{Hf} = 0.280825 \pm 0.000071$ , corresponding to  $\epsilon^{176}\text{Hf}_{(3.0)} = -0.31 \pm 2.5$  (Fig. 2). This age is consistent  
335 with the  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  age of  $3365 \pm 100$  Ma obtained by Morino et al (2017) but remains poorly defined  
336 owing to limited spread in Lu/Hf among the analyzed samples. The slope of the regression line is also  
337 largely determined by sample LA11-14, which may have been affected by crustal contamination  
338 (Section 3). Despite the large uncertainty associated with the whole-rock regression, the near-  
339 chondritic  $\epsilon^{176}\text{Hf}$  inferred for the Mesoarchean suite appears robust, showing only weak dependence  
340 on actual emplacement age. Initial  $\epsilon^{176}\text{Hf}$  values for individual samples based on the more precisely  
341 defined  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  age remain within error of the chondritic value, with a mean  $\epsilon^{176}\text{Hf}_{(3.36)} = 1.4 \pm 1.7$   
342 (2 S.E., Standard Error). Considered together with the previously determined  $\epsilon^{143}\text{Nd}_{(3.36)}$  of  $0.4 \pm 0.4$   
343 (Morino et al 2017), this result suggests derivation of the Mesoarchean ultramafic suite from an  
344 undepleted mantle source with near-chondritic Lu/Hf and Sm/Nd.

345 Regression of Nulliak mafic and ultramafic samples carrying a positive  $^{142}\text{Nd}$  anomaly yields a  
346 slope corresponding to an age of  $3794 \pm 130$  Ma (MSWD=142) with an initial  $^{176}\text{Hf}/^{177}\text{Hf}$

347  $=0.280461\pm0.000064$ ,  $\varepsilon^{176}\text{Hf}_{(3.79)}=5.1\pm2.3$  (Fig. 5). This age remains unchanged when considering only  
348 ultramafic samples in the regression ( $T=3766\pm140$  Ma; MSWD=118;  $^{176}\text{Hf}/^{177}\text{Hf}$   
349  $=0.280504\pm0.000072$ ,  $\varepsilon^{176}\text{Hf}_{(3.77)}=6.0\pm2.5$ ). Discarding two samples with high Ce/Yb, possibly  
350 reflecting crustal assimilation (section 3), also yields a similar age and initial  $^{176}\text{Hf}/^{177}\text{Hf}$  ( $T=3818\pm170$   
351 Ma,  $^{176}\text{Hf}/^{177}\text{Hf}=0.280466\pm0.000080$ , MSWD=95,  $\varepsilon^{176}\text{Hf}_{(3.82)}=5.8\pm2.9$ ). All regression lines thus yield  
352  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  ages consistent with the previously determined  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  age of  $3782\pm93$  Ma (Morino et  
353 al., 2017). Both the  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  and  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  ages are also indistinguishable from the U-Pb zircon  
354 date of  $3776\pm8$  Ma obtained by Schiøtte (1989) from a metavolcanic unit of the Nulliak assemblage  
355 situated on Bluebell island (Saglek Bay). This excellent agreement argues that the Lu-Hf dates have  
356 geologic significance despite the elevated MSWD values of the regressions. We thus interpret the  
357 initial  $^{176}\text{Hf}/^{177}\text{Hf}$  of the ultramafic rocks ( $0.280504\pm0.000072$ ;  $\varepsilon^{176}\text{Hf}_{(3.77)}=6.0\pm2.5$ ) to reflect  
358 derivation of the Nulliak ultramafic suite from a mantle source with super-chondritic Lu/Hf.

## 359 6. Discussion

### 360 5.1. Lu/Hf and Sm/Nd composition of the Early Depleted Mantle

361 The knowledge of  $^{142,143}\text{Nd}$  and  $^{176}\text{Hf}$  signatures of Nulliak ultramafic rocks offers the opportunity  
362 to precisely determine the time-integrated  $^{147}\text{Sm}/^{144}\text{Nd}$  and  $^{176}\text{Lu}/^{177}\text{Hf}$  of the EDM. Using coupled  
363  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  systematics, Morino et al (2017) estimated the former to be  $0.211\pm0.007$ , with a  
364 corresponding model age of differentiation of 4.4 Ga. Following this simple approach, the  $^{176}\text{Lu}/^{177}\text{Hf}$   
365 of the Nulliak source is calculated using a two-stage evolution model with an instantaneous  
366 differentiation event at  $T_d=4.4$  Ga. The mantle is assumed to evolve with a chondritic Lu/Hf prior to  
367 this date and a fractionated Lu/Hf afterwards. This differentiation event creates the early depleted  
368 mantle (EDM) considered here as the Nulliak source. Evolution of the  $^{176}\text{Lu}$ - $^{177}\text{Hf}$  system in this two-  
369 stage model is described by the following equation:

$$370 \left( \frac{^{176}\text{Hf}}{^{177}\text{Hf}} \right)_{T_e}^{\text{EDM}} = \left( \frac{^{176}\text{Hf}}{^{177}\text{Hf}} \right)_{T_p}^{\text{CHUR}} + \left( \frac{^{176}\text{Lu}}{^{177}\text{Hf}} \right)_{T_p}^{\text{CHUR}} \left[ 1 - e^{-\lambda_{176}T_e} \right] + \left( \frac{^{176}\text{Lu}}{^{177}\text{Hf}} \right)_{T_p}^{\text{EDM}} \left[ e^{-\lambda_{176}T_d} - e^{-\lambda_{176}T_e} \right] \quad (1)$$

371 Where  $\lambda_{176}$  is the  $^{176}\text{Lu}$  decay constant ( $1.867 \cdot 10^{-11}$ ; Scherer et al., 2001; Söderlund et al., 2004),  
372  $(^{176}\text{Lu}/^{177}\text{Hf})_{\text{CHUR}}=0.0336$  and  $(^{176}\text{Hf}/^{177}\text{Hf})_{\text{CHUR}}=0.282785$  at  $T_p$  is the present-day composition of the  
373 Chondritic Uniform Reservoir (Bouvier et al, 2008),  $T_d$  is the differentiation age of the EDM obtained  
374 from coupled  $^{146,147}\text{Sm}-^{142,143}\text{Nd}$  systematics (Morino et al 2017) and  $T_e$  is the time at which the  
375 parental melts of the Nulliak suite were extracted from the EDM. Application of Equation (1) with  
376  $T_e=3.77$  Ga and  $(^{176}\text{Hf}/^{177}\text{Hf})_{T_e}=0.280504\pm 0.000072$  yields a time-integrated  $(^{176}\text{Lu}/^{177}\text{Hf})_{\text{EDM}}$  of  
377  $0.047\pm 0.005$  for a corresponding  $(^{147}\text{Sm}/^{144}\text{Nd})_{\text{EDM}}$  of  $0.211\pm 0.007$ . By comparison, the time-  
378 integrated  $^{176}\text{Lu}/^{177}\text{Hf}$  and  $^{147}\text{Sm}/^{144}\text{Nd}$  values of the present-day depleted mantle are estimated to be  
379  $0.042-0.047$  and  $0.22-0.24$ , respectively, assuming a mean age of  $1.8-2.8$  Ga for mantle depletion from  
380 a chondritic source (Salters and Stracke, 2004) and chondritic  $^{176}\text{Lu}/^{177}\text{Hf}$  and  $^{147}\text{Sm}/^{144}\text{Nd}$  values of  
381  $0.0336$  and  $0.1960$  (Bouvier et al, 2008). The EDM and the MORB source thus exhibit similar Lu/Hf  
382 but differ in that the EDM was characterized by a less fractionated LREE pattern.

383 The  $^{147}\text{Sm}/^{144}\text{Nd}$  and  $^{176}\text{Lu}/^{177}\text{Hf}$  obtained for the sources of the Eo- and Mesoarchean ultramafic  
384 suites of the Saglek block are summarized in Figure 6, together with those obtained for the sources of  
385 Isua amphibolites (Hoffmann et al., 2010; Rizo et al., 2012) and several Mesoarchean komatiites of  
386 the Kaapvaal craton (Puchtel et al., 2013; Hoffmann and Wilson, 2016; Puchtel et al., 2016). The  
387  $(^{176}\text{Lu}/^{177}\text{Hf})_{\text{EDM}}$  inferred from Nulliak ultramafic rocks is similar to that estimated for the source of  
388 boninite-like metabasalts of the ISB (Hoffmann et al., 2010) but differs markedly from the slightly  
389 subchondritic value inferred from ISB meta-tholeiites (Rizo et al., 2012). This observation could imply  
390 a heterogeneous composition of the EDM with respect to Hf isotopes, or, alternatively, decoupling of  
391  $^{176}\text{Lu}-^{176}\text{Hf}$  and  $^{147}\text{Sm}-^{143}\text{Nd}$  systematics in the source of ISB meta-tholeiites, possibly related to the  
392 recycling of detrital components (Hoffmann et al., 2010; Hoffmann et al., 2011). In any case, our  
393 results show that the slightly subchondritic Lu/Hf inferred for the source of some ISB meta-tholeiites  
394 is not fully representative of the EDM. Rather, the concordant results obtained from Nulliak ultramafic  
395 rocks and ISB boninite-like metabasalts indicate that their positive  $^{142}\text{Nd}$  anomalies were produced in a  
396 mantle reservoir having superchondritic Sm/Nd and Lu/Hf.



397 Collectively, Eo- and Mesoarchean komatiites of the Saglek block and Kaapvaal craton define an  
398  $\epsilon^{176}\text{Hf}-\epsilon^{143}\text{Nd}$  array with a slope of  $\approx 4$  that differs significantly from the present-day mantle array  
399 ( $\epsilon^{176}\text{Hf} \approx 1.6 \times \epsilon^{143}\text{Nd}$ , Chauvel et al., 2008). This observation demonstrates that mantle differentiation  
400 processes in the early Earth differed markedly from those that operated throughout much of Earth's  
401 history to produce the present-day depleted mantle. A simple interpretation could be that both the  
402 modern Hf-Nd array and the komatiite array illustrated in Figure 6 formed as a result of melt  
403 extraction, but that the latter reflects deeper melting in the early Earth. Partial melting in a hotter  
404 mantle would involve larger extents of fractionation in the garnet stability field, which in turn could  
405 produce a residual mantle with elevated Lu/Hf. It is important to recognize, however, that the high  
406 Lu/Hf characterizing the EDM is not representative of the Archean depleted mantle, but appears to  
407 represent a single component in the source of Eo- and Mesoarchean komatiites. By contrast, protoliths  
408 of Archean TTGs (tonalite-trondhjemite-granodiorite) consistently sampled a mantle reservoir with  
409 only slightly superchondritic Lu/Hf (0.032-0.038, Guitreau et al., 2012). The Hf-Nd signature of the  
410 EDM also differs from that observed in Neoproterozoic komatiites, which plot on the present-day mantle  
411 array (Blichert-Toft and Puchtel, 2010; Puchtel et al., 2016). This observation suggests that the  
412 composition of the depleted reservoir sampled by komatiite magmatism evolved with time, in response  
413 to changes in global mantle-crust differentiation processes. The Hf-Nd signature of Neoproterozoic  
414 komatiites is consistent with their extraction from a mantle reservoir depleted by formation of the  
415 continental crust, while the Nulliak Hf-Nd signature likely records trace element fractionation  
416 associated with primordial mantle differentiation during the final stage of terrestrial accretion. In the  
417 following sections, we further evaluate this hypothesis in the framework of a simplified model of  
418 magma ocean crystallization. Our goal is to constrain the depth at which differentiation of the EDM  
419 reservoir occurred and to explore the possible geodynamic implications.

## 420 *5.2. The magma ocean crystallization model*

421 Two scenarios for solidification of the terrestrial magma ocean are considered in the following  
422 sections. In Model 1, we investigate Lu/Hf and Sm/Nd fractionation at lower mantle pressures, in the  
423 presence of an assemblage composed of ferropericlase, bridgmanite and Ca-perovskite. Following

424 Elkins-Tanton (2008), we assume that the magma ocean reached a depth of 2000 km. Crystallization  
425 most likely started from the bottom and proceeded upward because the thermal adiabat crosses the  
426 liquidus curve at the base of the mantle (e.g. Solomatov and Louis, 2007). Trace element partition  
427 coefficients in perovskite estimated at 23-25 GPa are assumed to remain valid throughout the lower  
428 mantle, which is treated as a single homogeneous reservoir. Model 2 considers the sequential  
429 crystallization of a shallow magma ocean from the transition zone to the surface (Caro et al., 2005). In  
430 this model, Earth's upper mantle is subdivided into 6 layers (22-18 GPa, 18-15 GPa, 15-12 GPa, 12-7  
431 GPa, 7-4.5 GPa, 4.5-2 GPa) to account for changes in mineralogy and garnet/melt partition  
432 coefficients. Crystallization is assumed to proceed from the bottom up, and the residual liquid  
433 remaining after the full crystallization of each layer is used as the starting liquid composition for the  
434 overlying layer (Caro et al., 2005). The impact of trapped liquid on Lu/Hf and Sm/Nd fractionation  
435 will be also considered. The mineral proportions for each layer, derived from Elkins-Tanton (2008)  
436 and Irifune (1994), are consistent with a pyrolitic mantle composition (Fig 7). For the sake of  
437 simplicity, it is assumed that all mineral phases grow synchronously, that the solid phases equilibrate  
438 with all of the remaining liquid during the growth of each layer (ie, that the liquid composition is  
439 homogeneous at each point in time), and that the modal proportions remain constant during  
440 crystallization within each of the layers. We acknowledge that these assumptions are unlikely to be  
441 valid throughout the entire crystallization sequence. Nevertheless, this simplified model provides an  
442 adequate framework to investigate the relationships between Lu/Hf and Sm/Nd fractionation expected  
443 to occur at various depths in the terrestrial magma ocean.

444 In both models, we assume a homogeneous initial melt with a pyrolitic composition and  
445 chondritic Lu/Hf and Sm/Nd (McDonough and Sun, 1995). During crystallization, elemental  
446 fractionation is controlled by the partition coefficient  $D$  of the bulk mineral assemblage relative to the  
447 melt, which is calculated according to the following equation;

448 
$$D = \sum y_j K_{d_j} \quad (2)$$

449 Where  $j$  represents the mineral,  $y$  its proportion in the mineralogical assemblage and  $K_d$  the partition  
450 coefficient between the mineral and the melt. The chosen partition coefficients for this model are  
451 compiled from the literature and reported in Table 4 (Salters and Longhi, 1999; Draper et al., 2003;  
452 Corgne, 2004; Corgne et al., 2004; Corgne and Wood, 2004; Corgne et al., 2012). Whenever possible,  
453 partition coefficients were selected from experiments using pyrolitic or chondritic starting  
454 compositions at the appropriate pressure range (i.e. 23-25 GPa for bridgmanite, ferropericlase and Ca-  
455 perovskite, 15-25 GPa for majorite, 2.8-9 GPa for garnet). When high pressure data were unavailable  
456 (clinopyroxene, orthopyroxene), partition coefficients at 2.8 GPa from Salters and Longhi (1999) were  
457 used. At lower mantle pressures, Lu/Hf and Sm/Nd fractionation are controlled by bridgmanite and  
458 Ca-perovskite, whereas at upper mantle pressure, garnet and clinopyroxene have the strongest  
459 influence. All elements of interest are highly incompatible in olivine (e.g. Salters and Longhi, 1999).  
460 Partition coefficients in olivine and its high pressure polymorphs were therefore assumed to be zero  
461 for all elements of interest. It is assumed that within each layer, partition coefficients do not vary with  
462 pressure, composition or temperature.

463 The evolution of the of bulk partition coefficients (noted  $D_{Lu}/D_{Hf}$  and  $D_{Sm}/D_{Nd}$ ) across the  
464 mantle illustrates the strong pressure dependence of Lu/Hf fractionation (Fig. 7); As  $P$  increases,  $D_{Lu}$   
465 in the bulk mineral assemblage decreases from 7 at 3 GPa to 1 at >15 GPa, while  $D_{Hf}$  remains roughly  
466 constant, at ca. 0.03-0.07. The largest Lu/Hf fractionation is thus expected to occur at pressures  
467 between 2 and 5 GPa (Pressures less than ~2 GPa are not considered because garnet is not stable.) At  
468 lower mantle pressures, Hf is expected to become more compatible than Lu ( $D_{Lu}/D_{Hf}<1$ ), while  
469  $D_{Sm}/D_{Nd}$  remains above 1 throughout the upper and lower mantle.

470 The extent of chemical differentiation in a magma ocean depends on whether melt can  
471 segregate from the solid phase during crystallization. This in turn depends on several factors  
472 (Solomatov and Louis, 2007), such as the crystal size (which controls the sedimentation rate), the  
473 cooling rate, the rate of melt percolation and the density contrast between melt and solid. Solomatov  
474 and Louis (2007) estimated that solidification of the lower mantle proceeded rapidly ( $10^3$  yr),  
475 effectively hampering chemical differentiation, while the upper mantle would have crystallized on  $10^7$ -

476  $10^8$  yr timescales compatible with some degree of melt extraction. In any case, rapid progression of the  
 477 solidification front is likely to trap a substantial amount of liquid in the partially molten regions,  
 478 ultimately resulting in subdued chemical fractionation compared to that expected from a pure  
 479 cumulate. To account for this effect, the model includes a trapped liquid component, modelled as a  
 480 mineral phase with partition coefficients of 1 for all elements of interest. It is assumed that the fraction  
 481 of trapped liquid is the same in the different layers.

482 Chemical differentiation in the magma ocean may have proceeded through either batch or  
 483 fractional crystallization. During the early stages of solidification, turbulent convection likely favored  
 484 batch crystallization, while fractional crystallization may have been the predominant mechanism at  
 485 small (<20%) fractions of residual melt (Snyder et al., 1992; Borg and Draper, 2003; Debaille et al.,  
 486 2008). As the transition from batch to fractional processes is poorly constrained, the effects of these  
 487 two modes of crystallization on Lu/Hf and Sm/Nd fractionation were investigated throughout the  
 488 entire crystallization sequence. The results are very similar for batch and fractional crystallization (see  
 489 online appendix) so these processes are not discussed separately in the following sections.

490 Elemental concentrations were calculated from the following equations, for batch (Eq. 3-4)  
 491 and fractional (Eq. 5-6) crystallization:

$$492 \quad \left\{ \begin{array}{l} \left( \frac{C_i^l}{C_i^0} \right) = \frac{1}{DX + (1-X)} \\ \left( \frac{C_i^s}{C_i^0} \right) = \frac{D}{DX + (1-X)} \end{array} \right. \quad (3-4)$$

$$493 \quad \left\{ \begin{array}{l} \left( \frac{C_i^l}{C_i^0} \right) = (1-X)^{D-1} \\ \left( \frac{C_i^s}{C_i^0} \right) = \frac{1-(1-X)^D}{X} \end{array} \right. \quad (5-6)$$

494 Where  $C_i^l$  is the concentration of element  $i$  remaining in the liquid,  $C_i^s$  is the concentration of element  
 495  $i$  in the cumulate and  $C_i^0$  is the initial concentration in the melt,  $X$  corresponds to the extent of

496 crystallization within a given layer and  $D$  is the partition coefficient of the crystallizing mineralogical  
497 assemblage. Sequential crystallization of the upper mantle is modelled according to the following  
498 procedure. The deepest reservoir (22-18 GPa), here composed of majorite and olivine, crystallizes  
499 from a pyrolytic melt until its total solidification, and the residual melt is taken as the starting  
500 composition for the next step of crystallization. This calculation is then repeated for the next 5 layers  
501 until the upper mantle reaches 93% solidification. The composition of the final liquid thus  
502 corresponds to the last 7% of residual melt, which is equivalent to 3% of the mass of the BSE.

503

### 504 *5.3. Lu/Hf and Sm/Nd fractionation in the terrestrial magma ocean*

505 Results of both models are presented in Figure 8, for the batch crystallization case. Results  
506 corresponding to the fractional crystallization case are provided in supplementary material. Similar to  
507 previous studies, Model 1 predicts a strong decoupling of Lu/Hf and Sm/Nd at lower mantle pressures  
508 (Caro et al., 2005, Rizo et al., 2011). Depending on the relative proportions of bridgmanite and Ca-  
509 perovskite, batch and fractional crystallization yield cumulates with  $^{176}\text{Lu}/^{177}\text{Hf}$  ranging from 0.028 to  
510 0.035 and  $^{147}\text{Sm}/^{144}\text{Nd}$  ranging from 0.20 to 0.26 (compared to the chondritic values of 0.0336 and  
511 0.196, respectively) after complete crystallization of the lower mantle, representing ~62% of the total  
512 volume of the magma ocean. Crystallization of the lower mantle in roughly modal proportions, (7%  
513 Ca-perovskite; 75% bridgmanite, 18% ferropericlasite) would produce a cumulate with a near-  
514 chondritic Lu/Hf (0.034) but superchondritic Sm/Nd, leading with time to slightly positive  $\epsilon^{176}\text{Hf}$   
515 ( $\sim 0.2$  at  $T=3.75$  Ga) and positive  $\epsilon^{142,143}\text{Nd}$  (Fig. 8A). The decoupling of Sm/Nd and Lu/Hf  
516 fractionation induced by crystallization of a perovskitic reservoir is consistent with the signature  
517 reported by Rizo et al. (2011) for Isua metatholeiites, but cannot explain the signature of the Nulliak  
518 source ( $^{176}\text{Lu}/^{177}\text{Hf}=0.047$ ,  $^{147}\text{Sm}/^{144}\text{Nd}=0.211$ ), and, by extension, the slope of the Hf-Nd komatiite  
519 array (Fig. 6).

520 For the upper mantle model (Model 2), all cumulates generated from the transition zone to the  
521 surface exhibit highly fractionated Lu/Hf and Sm/Nd, predominantly induced by the garnet component

522 (Fig. 8B). The  $^{176}\text{Lu}/^{177}\text{Hf}$  and  $^{147}\text{Sm}/^{144}\text{Nd}$  ratios in upper mantle cumulates range from 0.10 to 0.14  
523 and from 0.30 to 0.52, respectively. Crystallization of a shallow magma ocean is therefore expected to  
524 produce a depleted upper mantle with positive  $\epsilon^{176}\text{Hf}$  and  $\epsilon^{142,143}\text{Nd}$ , generally consistent with the  
525 signature inferred for the Nulliak source. However, while crystallization at upper mantle pressures can  
526 explain the direction of displacement of the calculated  $(\text{Lu}/\text{Hf})_{\text{EDM}}$  and  $(\text{Sm}/\text{Nd})_{\text{EDM}}$  relative to a  
527 chondritic composition, the model produces cumulates with Sm/Nd and Lu/Hf markedly higher than  
528 those estimated for the Nulliak source. It follows that crystallization of a shallow magma ocean cannot  
529 reproduce the composition of the EDM unless chemical fractionation was buffered by trapping of  
530 liquid during solidification.

531 Figure 9 shows the composition of magma ocean cumulates at the different pressure ranges considered  
532 in Model 2, assuming a fraction of trapped liquid between 5% and 30%. The results show little  
533 dependence upon pressure or mineralogical assemblage and the produced trends all fit the Nulliak  
534 source composition within the calculated uncertainties. Nevertheless, the best fit to the Nulliak  
535 composition is obtained for a depth range of 60-400 km, and a fraction of trapped liquid of 5-15%.  
536 Radiogenic decay in these reservoirs would translate into an approximately 4:1 relationship between  
537  $\epsilon^{176}\text{Hf}$  and  $\epsilon^{143}\text{Nd}$  consistent with the initial ratios deduced from the Nulliak Sm-Nd and Lu-Hf  
538 errorchrons. We thus interpret the  $^{176}\text{Hf}$ - $^{142,143}\text{Nd}$  composition of the Nulliak ultramafic rocks to reflect  
539 differentiation of the EDM at relatively low pressure in the upper mantle. Our results also show that  
540 Eo- and Mesoarchean komatiites, while often viewed as originating from the deep mantle, carry  $^{176}\text{Hf}$ -  
541  $^{143}\text{Nd}$  signatures consistent with prior differentiation of their source in the garnet stability field (Fig.  
542 6).

543

#### 544 *5.4. Geodynamic evolution of the EDM*

545 In absence of a clear perovskite signature, the interpretation that the EDM formed by magma  
546 ocean crystallization rather than by partial melting of a solidified mantle relies solely on chronological  
547 constraints. The synchronicity of the  $^{146,147}\text{Sm}$ - $^{142,143}\text{Nd}$  model age for differentiation of the silicate

548 Earth with the Pb-Pb age thought to reflect impact-provoked Pb loss (Connelly and Bizzarro, 2016)  
549 suggests that the EDM was produced near the end of terrestrial accretion, most likely as a result of  
550 extensive melting triggered by the Moon-forming impact. However, whether the EDM represents  
551 magma ocean cumulates or the residue of post-crystallization melting remains uncertain. Regardless of  
552 the exact mechanism by which the EDM formed, our results indicate that differentiation must have  
553 taken place at relatively low pressure within the garnet stability field. This shallow depleted reservoir  
554 must have remained at least partially isolated from the convective system, so as to account for the  
555 presence of positive  $^{142}\text{Nd}$  anomalies in Neoproterozoic lavas of the Abitibi belt (Debaille et al., 2013).  
556 Long-term preservation of primordial mantle domains could imply inefficient convective stirring in  
557 the early Earth, perhaps due to the presence of a stagnant-lid regime (Debaille et al., 2013). However,  
558 several lines of evidence suggest that ancient mantle domains remained isolated on a billion year  
559 timescale near the core-mantle boundary, and are still sampled by modern plume magmatism  
560 (Mukhopadhyay, 2012; Caracausi et al., 2016; Rizo et al., 2016; Mundl et al., 2017; Peters et al.,  
561 2018). In this regard, an important observation is that while the EDM (or mantle domains with similar  
562 Lu/Hf and Sm/Nd) contributed to the source of Eo- and some MesoArchean komatiites, as shown by  
563 the correlation between the Lu/Hf vs. Sm/Nd values of their sources (Figure 6), this reservoir was not  
564 commonly tapped by magmatism involved in Archean crustal growth (Guitreau et al., 2012).

565 Magma ocean overturn provides a scenario which can potentially account for these  
566 observations. Overturn would occur during or shortly after solidification, reorganizing the  
567 stratification according to cumulate density (Elkins-Tanton et al., 2003; Elkins-Tanton et al., 2005;  
568 Elkins-Tanton, 2008; Brown et al., 2014). This would result in the sinking of shallow and dense  
569 cumulates to depths near the core-mantle boundary, which would trigger a return flow of deep  
570 undifferentiated material into the upper mantle. The storage of dense upper mantle cumulates at the  
571 core mantle boundary would favor their long-term preservation, consistent with the apparent closed  
572 system evolution of the EDM, while the transfer of deep undifferentiated material into the upper  
573 mantle may account for the near-chondritic Lu/Hf ratio characterizing the mantle source of the  
574 protoliths of Archean TTGs (Guitreau et al., 2012). Alternatively, it is entirely plausible that chemical

575 differentiation in the magma ocean remained marginal until the ultimate stage of solidification,  
576 hampered by inefficient crystal settling and rapid cooling of the mantle compared to the rate of melt  
577 percolation. Effective chemical fractionation may then only be possible in the shallowest part of the  
578 upper mantle (Solomatov and Louis, 2007). In this case, the shallow depleted reservoir would be  
579 chemically buoyant, favoring its long-term preservation as part of a long-lived stagnant lid (O'Neill  
580 and Debaille, 2014; Caro et al., 2017). Recycling of this primordial lithosphere would then provide a  
581 means of transferring both enriched and depleted material into the deep mantle where they may have  
582 contributed in various proportions to plume-related magmatism.

## 583 7. Conclusions

584 Because of its sensitivity to pressure, the  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  system provides a means of constraining  
585 the depths of differentiation of the mantle sources of volcanic rocks. This approach was undertaken on  
586 mafic and ultramafic rocks of the Saglek block (northern Labrador, Canada) in order to assess the  
587 composition and differentiation mechanism of mantle domains carrying positive  $^{142}\text{Nd}$  anomalies. The  
588 results of this study can be summarized as follows:

589 1) The two suites of mafic and ultramafic rocks of the Saglek Block, identified by Morino et al. (2017)  
590 on the basis of their  $^{142}\text{Nd}$  compositions, yielded  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  whole-rock errorchrons with ages  
591 consistent with those previously determined by  $^{147}\text{Sm}$ - $^{143}\text{Nd}$  dating. This finding confirms the  
592 existence of the two-distinct series, the Eoarchean Nulliak suite with a  $^{176}\text{Lu}$ - $^{176}\text{Hf}$  age of  $3766\pm 140$   
593 Ma ( $3782\pm 93$  Ma by  $^{147}\text{Sm}$ - $^{143}\text{Nd}$ ), and a Mesoarchean suite, dated at  $3023\pm 390$  Ma (compared to  
594  $3365\pm 100$  Ma by  $^{147}\text{Sm}$ - $^{143}\text{Nd}$ ). These age constraints are consistent with U-Pb dating of magmatic and  
595 detrital zircons from the Nulliak and Upernavik formations (Schiotte, 1989; Schiotte et al, 1989, 1992)  
596 and with the  $^{187}\text{Re}$ - $^{187}\text{Os}$  study from Ishikawa et al. (2017), who reported minimum model ages ( $T_{\text{RD}}$ )  
597 of 3.4-3.6 Ga and 2.8-3.3 Ga in two ultramafic suites of the Saglek block.

598 2) The model age ( $4.40\pm 0.03$  Ga) obtained by Morino et al. (2017) for the formation of the EDM was  
599 used to calculate the time-integrated Lu/Hf source ratios for each of the suites, yielding  $^{176}\text{Lu}/^{177}\text{Hf}$   
600 ratios of 0.047 and 0.033 for the Eoarchean and Mesoarchean mantle sources respectively. The



601 superchondritic  $^{176}\text{Lu}/^{177}\text{Hf}$  of the Nulliak source is coupled with a superchondritic  $^{147}\text{Sm}/^{144}\text{Nd}$   
602 ( $\approx 0.21$ ). In order to investigate the origin of this coupled signature, we developed a simple model of  
603 magma ocean crystallization. Results show that garnet played a critical role in the fractionation of  
604 Lu/Hf and Sm/Nd of the Nulliak mantle source. More specifically, our modeling results imply that the  
605 isotopic signature of the Nulliak suite was generated by crystallization of its parent mantle reservoir  
606 above the transition zone, in a depth range of 400 to 60 km. Substantial quantities of trapped liquid are  
607 required in the cumulate pile, but the proportions cannot be estimated unambiguously given the  
608 simplifying assumptions made in the model.

609 3) When the parameters for the Nulliak source as well as for the mantle sources of other Eo- and  
610 Mesoarchean komatiites are plotted in a diagram of Sm/Nd versus Lu/Hf, a well-defined trend is  
611 observed, suggesting a single formation mechanism for the sources of all of these komatiites. This  
612 trend differs from the Hf-Nd mantle array defined by modern basalts, having more radiogenic Hf  
613 isotopic compositions for a given Nd isotopic ratio. This observation may be consistent with our  
614 interpretation that the source of Eo- and Mesoarchean komatiites formed by melting or crystallization  
615 processes involving garnet at depths of 400 to 60 km. This contrasts with the likely origin of the  
616 modern Hf-Nd array, which would involve melt extraction at mostly shallower depths, including a  
617 large component in the spinel facies in the absence of garnet.

618 4) The conclusion that the differentiation of Eo- and Mesoarchean komatiite sources occurred in the  
619 upper mantle could appear surprising considering that these magmas are thought to be derived from  
620 the deep mantle. This suggests that a mantle overturn mechanism brought the komatiite source  
621 reservoirs, which differentiated in the garnet stability field, down into the deep mantle, where they  
622 were effectively isolated for hundreds of millions of years. This deep mantle source was then  
623 sporadically sampled by plume mechanisms.

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633

### Figure captions

634 **Fig. 1.** Geological location and simplified geological map of the Hebron fjord - Saglek fjord region of  
635 the Saglek block, showing the location of the main sampling sites (from Morino et al., 2017 Modified  
636 from Ryan and Martineau (2012)). Large ( $\approx 100$ -1000 m) enclaves composed of both mafic and  
637 ultramafic rocks are referred to as units A, B and C. The dotted rectangle indicates the general area  
638 where smaller (meter-sized) enclaves were collected. Correlated sample numbers and GPS coordinates  
639 are provided in Morino et al. (2017).

640 **Fig. 2.** (A-H) Selected trace elements versus Zr in ultramafic rocks of the Eo- and Mesoarchean suites  
641 of the Saglek block. Full symbols represent samples with  $(\text{Ce}/\text{Yb})_N$  within the range of  
642 uncontaminated komatiites (Cf. Fig. 3). Open symbols represent samples with anomalous  $(\text{Ce}/\text{Yb})_N$ ,  
643 possibly reflecting crustal contamination.

644 **Fig. 3.** (A-C) Selected trace element ratios versus  $(\text{Ce}/\text{Yb})_N$  in ultramafic rocks from the Saglek block.  
645 Komatiites of the Barberton and Abitibi greenstone belts, and Gorgona Island are shown for  
646 comparison. Komatiite data are compiled from the GEOROC database ([http://georoc.mpch-](http://georoc.mpch-mainz.gwdg.de)  
647 [mainz.gwdg.de](http://georoc.mpch-mainz.gwdg.de)). Average Archean TTGs (tonalite-trondjemite-granodiorite) have  $(\text{Ce}/\text{Yb})_N \approx 15$ ,  
648  $(\text{Th}/\text{La})_N \approx 2$ ,  $(\text{Nb}/\text{La})_N \approx 0.25$ ,  $(\text{Sm}/\text{Nd})_N \approx 0.5$  (Condie, 1993). Note that three samples from the  
649 Eoarchean group and one from the Mesoarchean group have Th concentrations below the detection  
650 limit of 0.06 ppm. The corresponding datapoints (marked by descending arrows in panel B) represent  
651 maximum  $(\text{Th}/\text{La})_N$  values.

652 **Fig. 4.**  $(\text{Lu}/\text{Hf})_N$  versus  $\text{Al}_2\text{O}_3/\text{TiO}_2$  in ultramafic rocks of the Saglek block. Data for Barberton,  
653 Abitibi and Gorgona komatiites are compiled from the GEOROC database ([http://georoc.mpch-](http://georoc.mpch-mainz.gwdg.de)  
654 [mainz.gwdg.de](http://georoc.mpch-mainz.gwdg.de)).

655 **Fig. 5.** (A-B) Lu-Hf isochron diagrams for mafic/ultramafic enclaves of the Saglek Block. The  
656 regression for the panel A includes rocks that were affiliated with a Mesoarchean event by Morino et  
657 al. (2017). The regression in panel B includes samples previously defined as Eoarchean. (C-D)  $^{147}\text{Sm}$ -  
658  $^{143}\text{Nd}$  isochron diagrams for mafic/ultramafic enclaves of the Saglek block from Morino et al. (2017).  
659 Uncertainties on ages and initial ratios are 2 S.D. Regressions were performed using Isoplot/Ex  
660 version 4.15 (Ludwig, 2012), assuming a typical uncertainty of 0.5% on  $^{147}\text{Sm}/^{144}\text{Nd}$  and  $^{176}\text{Lu}/^{177}\text{Hf}$ .

661

662 **Fig. 6.**  $^{176}\text{Lu}/^{177}\text{Hf}$  versus  $^{147}\text{Sm}/^{144}\text{Nd}$  of the sources of the Saglek ultramafic suites compared to the  
663 sources of Mesoarchean komatiites and Isua mafic rocks. The grey field represents the modern  
664 terrestrial array (Chauvel et al., 2008).  $^{147}\text{Sm}/^{144}\text{Nd}$  and  $^{176}\text{Lu}/^{177}\text{Hf}$  source ratios are calculated

665 according to Eq. (1), assuming a differentiation event at 4.4 Ga and considering 2 S.D. uncertainties  
666 on initial  $\varepsilon^{176}\text{Hf}$  values from the Isoplot regression. For nearly all localities, the parameters of the  
667 mantle source were calculated using the initial ratios of the corresponding isochrons. The sole  
668 exception concerns the Isua boninites, for which the mean  $^{147}\text{Sm}/^{144}\text{Nd}$  and  $^{176}\text{Lu}/^{177}\text{Hf}$  values were  
669 determined from the mantle source parameters calculated individually for each sample; the  
670 uncertainties shown represent the 2 S.D. variations of these mean values. Note that model ages of  
671 differentiation for the mantle sources of Kaapvaal komatiites (i.e.  $T_d$  in Equation (1)) cannot be  
672 inferred from  $^{146,147}\text{Sm}-^{142,143}\text{Nd}$ , either due to absence of  $^{142}\text{Nd}$  data or due to an apparent decoupling  
673 of the  $^{146}\text{Sm}-^{142}\text{Nd}$  and  $^{147}\text{Sm}-^{143}\text{Nd}$  systems (Puchtel et al., 2016). Accordingly, the time-integrated  
674 Lu/Hf and Sm/Nd values of their mantle sources cannot be estimated with as much confidence as for  
675 the Isua and Nulliak sources and should be considered first-order estimates. The blue and yellow  
676 envelopes represent the calculated compositions of magma ocean cumulates predicted to crystallize at  
677 lower and upper mantle pressures, respectively (See Section 5 and Figures 8-9).

678 **Fig. 7. A.** Modal compositions of mineral assemblages considered for models 1 and 2 of  
679 crystallization of a terrestrial magma ocean (Opx: Orthopyroxene, Cpx: Clinopyroxene; Ca-Pv: Ca-  
680 perovskite, Bg: Bridgmanite, Fp: Ferropericlasite). Model 1 considers the crystallization of the deep  
681 mantle as an assemblage of Ca-perovskite, bridgmanite and ferropericlasite. Model 2 considers the  
682 sequential crystallization of the upper mantle from the transition zone to the surface. Modal  
683 compositions are from Elkins-Tanton (2008). **B.** Variation of  $D^{\text{Lu}}/D^{\text{Hf}}$  and  $D^{\text{Sm}}/D^{\text{Nd}}$  with pressure and  
684 thus mineral assemblage. Open diamonds reflect the results of experimental studies at various  
685 pressures, with partition coefficients for individual phases combined in the shown mineral proportions  
686 to obtain the bulk D value. For simplicity, constant  $D^{\text{Lu}}/D^{\text{Hf}}$  and  $D^{\text{Sm}}/D^{\text{Nd}}$  values were used in the  
687 calculations for each interval; these are shown as vertical bars. Partitioning data are from Corgne et al.  
688 (2005, 2012), Draper et al. (2003) and Salters & Longhi (1999).

689 **Fig. 8.**  $^{147}\text{Sm}/^{144}\text{Nd}$  versus  $^{176}\text{Lu}/^{177}\text{Hf}$  produced by batch crystallization processes for Models 1 and 2.  
690 Squares are for cumulates and circles are for the residual melt. These results are compared with the  
691 Nulliak depleted mantle source (red star). **(A)** Results for model 1. Each color represents a proportion  
692 of Ca-Perovskite (CaPv) in the mineralogical assemblage. The impact of trapped liquid (TL) in the  
693 assemblage is presented in this Figure. The proportion of trapped liquid varies from 0 to 30% with an  
694 incremental step of 5%. **(B)** Results for model 2. For each layer, the  $^{147}\text{Sm}/^{144}\text{Nd}$  versus  $^{176}\text{Lu}/^{177}\text{Hf}$  of  
695 the starting liquid are those of the residual liquid produced by total crystallization of the underlying  
696 layer (with the exception of the bottom layer, which starts with a liquid having chondritic ratios Lu/Hf  
697 and Sm/Nd). This diagram shows only the results for pure cumulates generated by batch  
698 crystallization, because the high Sm/Nd and Lu/Hf of these cumulates require large scales for the x  
699 and y-axes.

700

701 **Fig. 9.** Effect of retention of trapped liquid on  $^{147}\text{Sm}/^{144}\text{Nd}$  versus  $^{176}\text{Lu}/^{177}\text{Hf}$  compositions of  
702 cumulates formed by batch sequential crystallization of the upper mantle (Model 2). The proportion of  
703 trapped liquid considered ranges between 5 and 30% (with 5% increments), and is assumed to remain  
704 constant during the crystallization process. Pressure ranges of the corresponding layer are indicated in  
705 each of the six panels.

706

707

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FIGURE .1

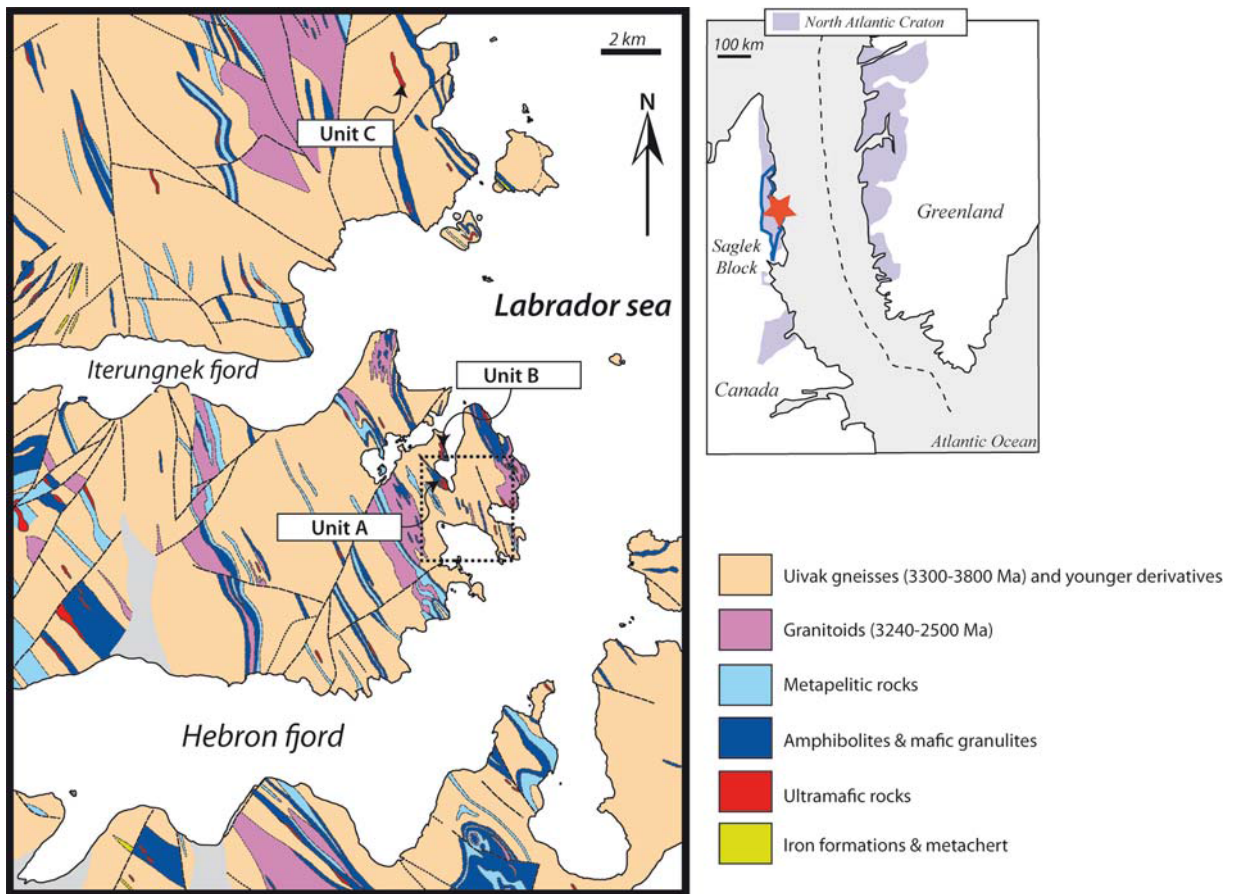


FIGURE 2

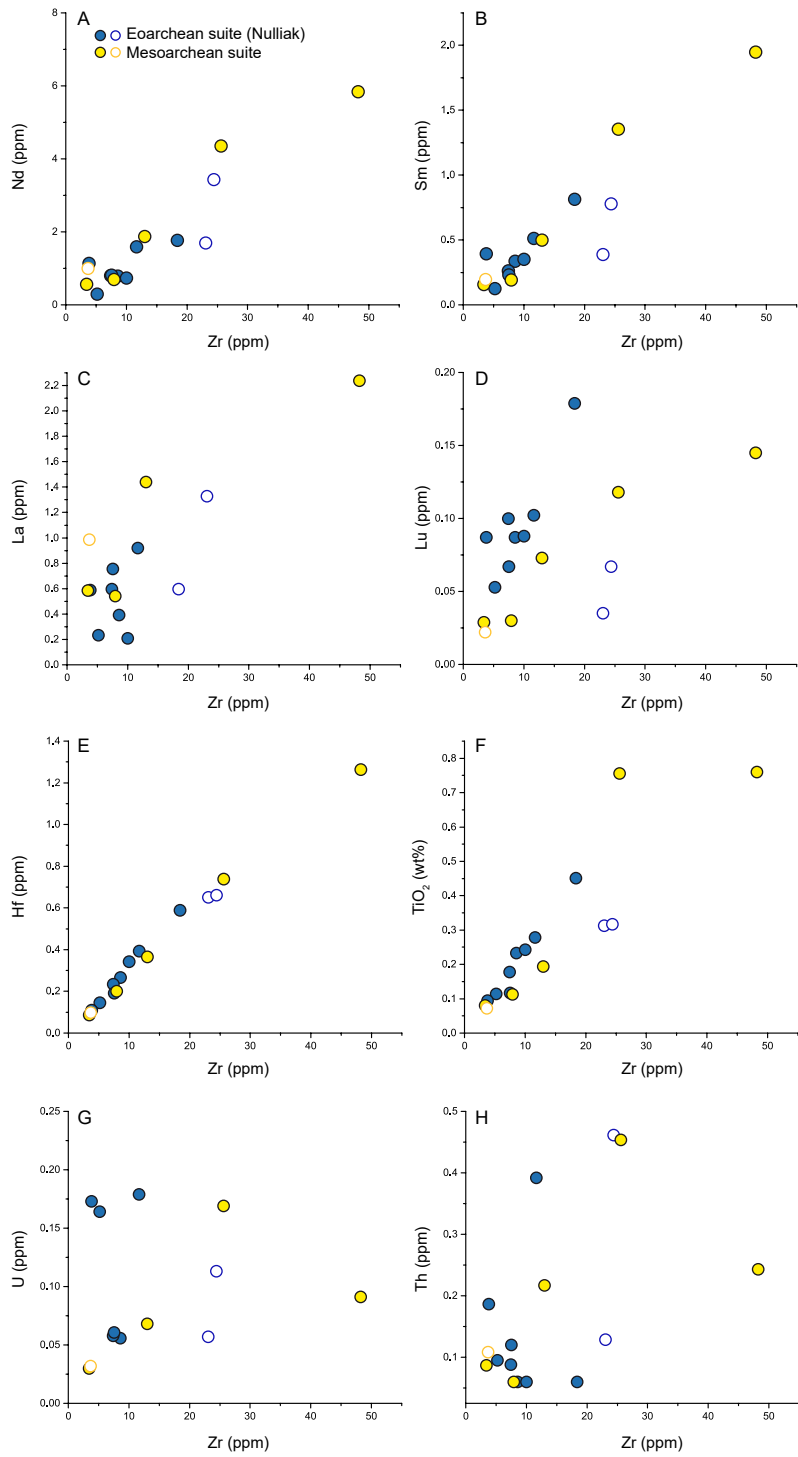


FIGURE 3

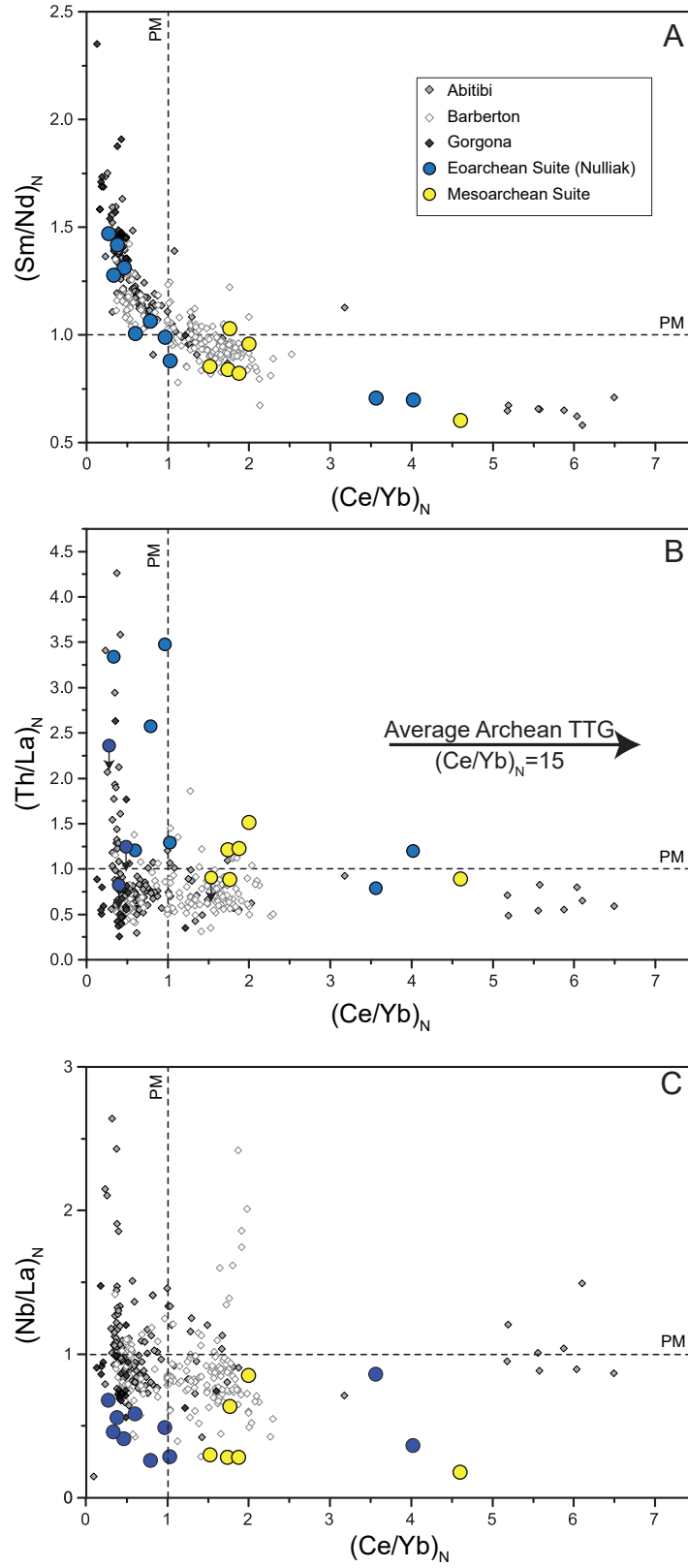


FIGURE 4

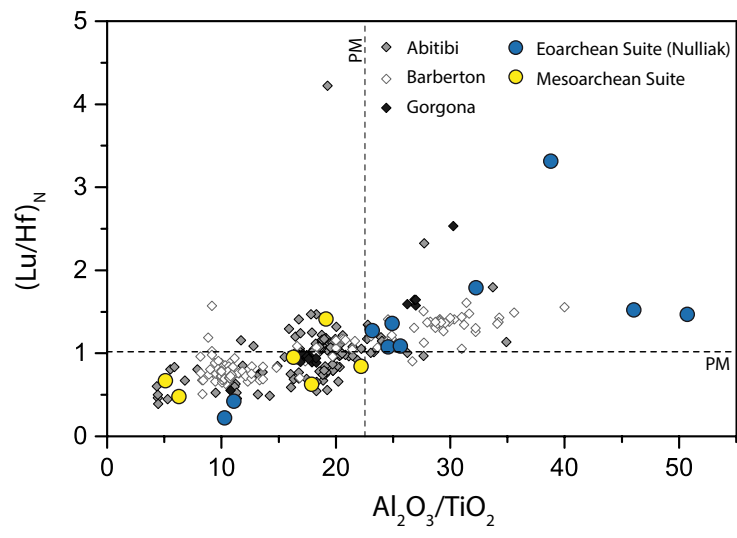


FIGURE 5.

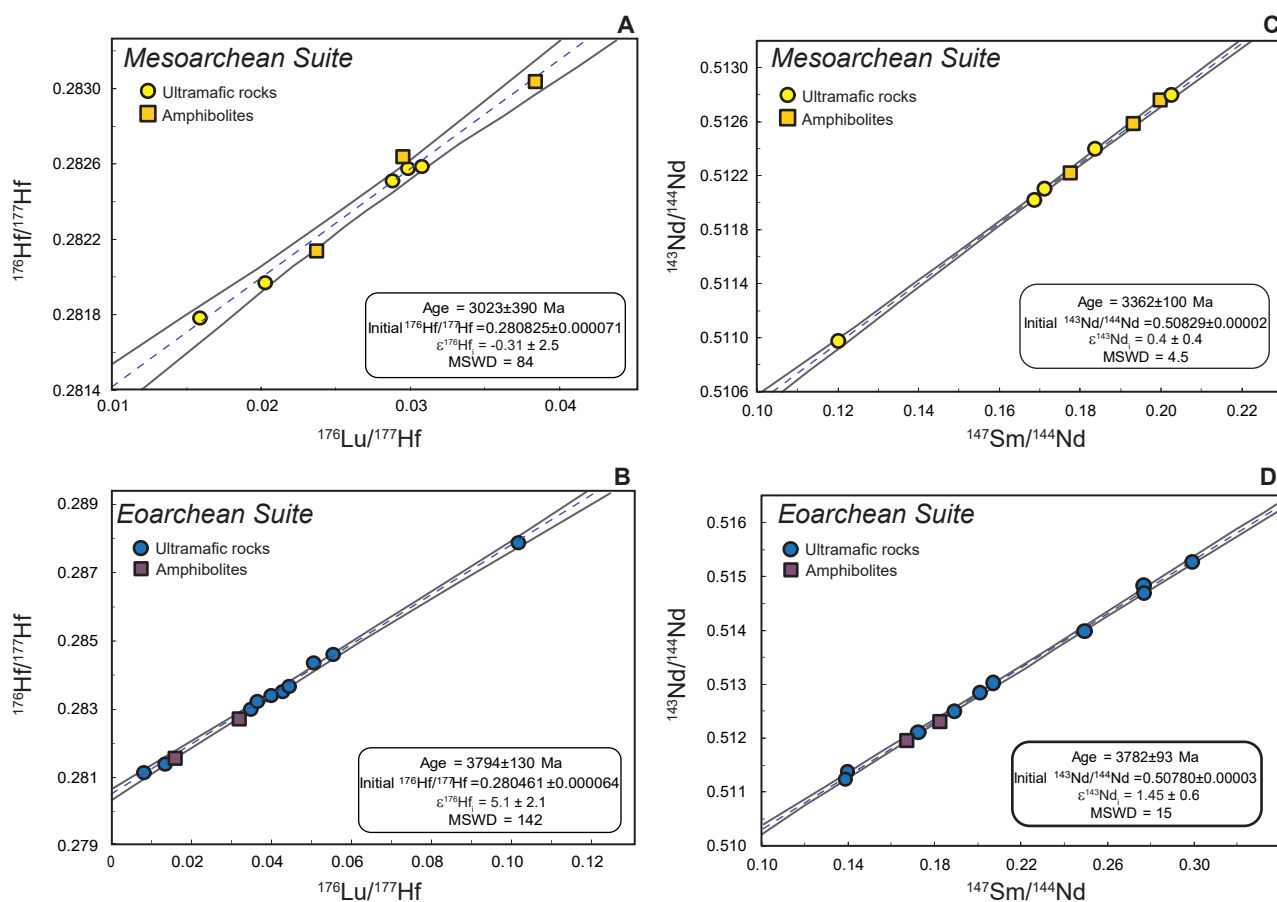


FIGURE 6.

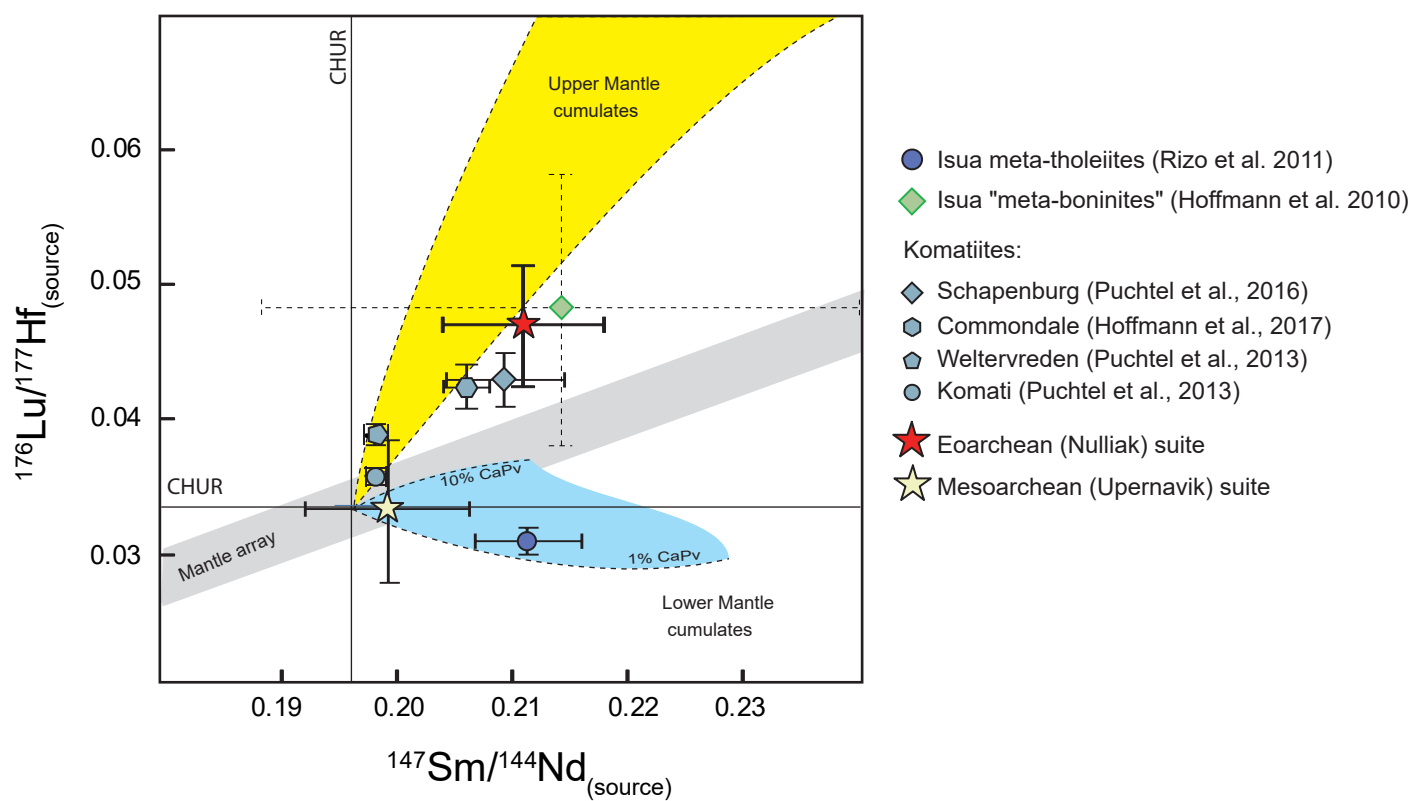




FIGURE 7.

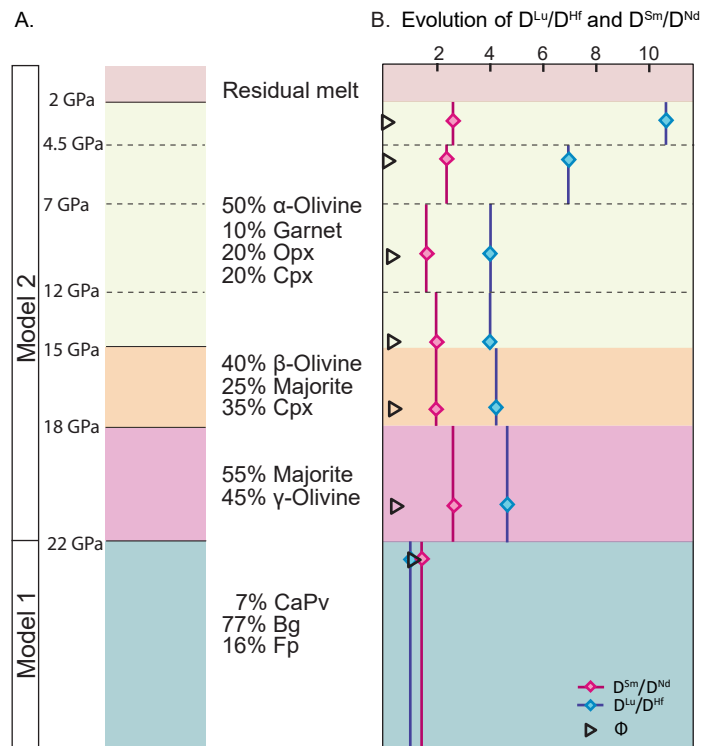


FIGURE 8.

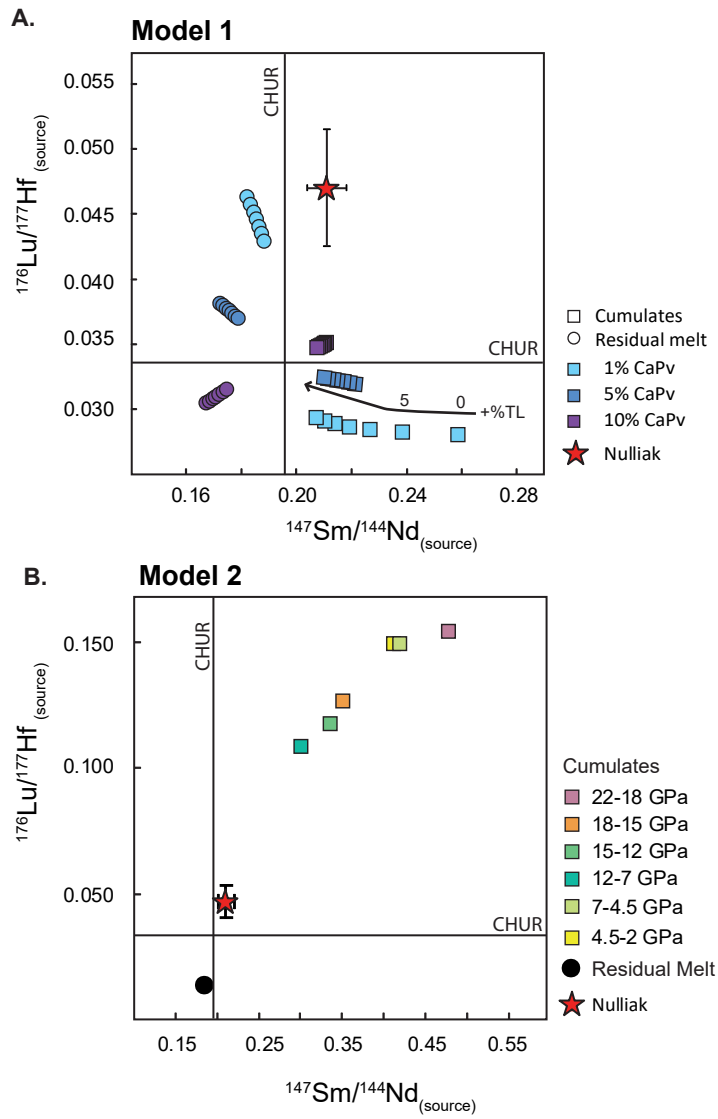


FIGURE 9

