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1	The Komatiite Testimony to Ancient Mantle Heterogeneity
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36 Abstract

Komatiites are crystallized samples of high-temperature, high-MgO lavas that were common during the Archean, but became increasingly rarer in the Proterozoic and Phanerozoic. Although the origin of komatiites remains a subject of debate, all komatiites included in this review, ranging in age from 3.6 to 2.0 Ga, are interpreted to have most likely been derived from anhydrous melting in mantle plumes. These plumes are estimated to have been initiated at different depths in the mantle, thus, providing important information about the chemical evolution of the early Earth.

The ^{142,143}Nd, ¹⁷⁶Hf, ¹⁸²W, ¹⁸⁷Os, and ¹⁸⁶Os systematics and trace- and highly siderophile 44 element (HSE) abundances of these komatiites provide strong evidence for the presence of 45 isotopic and chemical heterogeneities in the mantle during the first half of Earth history. 46 47 These heterogeneities likely reflect the combined effects of: (1) the co-existence of diverse 48 post-magma ocean silicate domains that were characterized by variably-fractionated lithophile 49 and siderophile element abundances; (2) the presence of distinct reservoirs that included 50 mantles and cores of late accreted, differentiated planetesimals; and (3) isotopic exchange 51 across the core-mantle boundary. These data highlight the complexity of komatiite mantle 52 sources, none of which were similar in composition to estimates for the modern bulk silicate 53 Earth (BSE). Moreover, no single petrogenetic model can account for the remarkably diverse 54 chemical and isotopic compositions of komatiites.

The disappearance of resolvable positive and negative ¹⁴²Nd anomalies, as well as 55 decoupled ¹⁴³Nd-¹⁷⁶Hf isotopic signatures, in the mafic-ultramafic rock record by ~2.5 Ga 56 57 indicate that, by the end of the Archean, the earliest silicate reservoirs, formed through 58 primordial magma ocean crystallization, had been largely destroyed as a result of vigorous 59 convective mantle mixing. This implies that, during the Hadean and Archean, it took the mantle ~1.5 Ga to mix away the early formed ¹⁴²Nd heterogeneities via wholesale mantle 60 convection. Similar to ¹⁴²Nd systematics, there appears to have been a shift from mostly 61 positive ¹⁸²W anomalies in pre-2.5 Ga komatiite mantle sources to no ¹⁸²W offsets in post-2.5 62 Ga komatiite mantle sources. Coupled with the disapperance of projected HSE depletions in 63 64 komatiite mantle sources at ~2.5 Ga, relative to the modern BSE, this shift may indicate that by the end of the Archean, late accreted planetesimals had become largely homogenized 65 within the mantle, and core-mantle interaction took over as the main driving force of creating 66 ¹⁸²W isotope anomalies, possibly coincident with the timing of the onset of modern-style plate 67 68 tectonics on Earth.

69

70 **1. Introduction**

71 Accurately determining how the distribution of chemical elements within the Earth has changed over time has been, and remains, one of the most fundamental challenges in Earth 72 73 science; it has far-reaching implications not only for the long-standing debate of how 74 terrestrial planets formed and evolved, but also for understanding ongoing processes on Earth. 75 The information pertaining to the origin and early evolution of Earth comes largely from the geological rock record between ~4.0 and 2.0 Ga, which harbors isotopic and elemental 76 77 signatures generated in the terrestrial reservoirs via early chemical fractionation processes and the radioactive decay of short- and long-lived refractory nuclides, including ^{146,147}Sm, ¹⁷⁶Lu, 78 ¹⁸²Hf, ¹⁸⁷Re, and ¹⁹⁰Pt. 79

80 The mantle is the largest of all the terrestrial reservoirs. Its chemically and isotopically 81 heterogeneous nature has long been established by numerous pioneering studies of the Earth's 82 rock record (e.g., Hart and Brooks, 1977; Hofmann and Hart, 1978; Zindler et al., 1982; Hart 83 and Zindler, 1986; Zindler and Hart, 1986; Jacobsen, 1988; Galer and Goldstein, 1991). Some 84 of the heterogeneities have been argued to be primordial in nature, reflecting initial planetary 85 accretion/differentiation and magma ocean crystallization processes (Goldstein and Galer, 86 1992; Albarède et al., 2000; Drake, 2000; Boyet and Carlson, 2005; Frost et al., 2008; Caro, 87 2011; Touboul et al., 2012; Carlson et al., 2015; Jacobsen and Yu, 2015; Puchtel et al., 2016a; 88 Rizo et al., 2016; Boyet et al., 2021). Others likely were formed as a result of a protracted 89 terrestrial accretion history (e.g., Willbold et al., 2011; 2015; Dale et al., 2017; Puchtel et al., 90 2018; Archer et al., 2019) or later processes associated with the dynamic regime of the planet, 91 especially crustal recycling (e.g., DePaolo, 1980; Armstrong, 1981; Hofmann and White, 92 1982; Patchett et al., 1984; Shirey and Hanson, 1986; Chase and Patchett, 1988; Galer et al., 93 1989; Bowring and Housh, 1995; Bennett et al., 1996; Salters and White, 1998; Blichert-Toft 94 et al., 2015).

95 Despite these decades-long research efforts, still limited insights into the terrestrial mantle 96 mixing history mean that the nature, origin, scale, and longevity of early mantle 97 heterogeneities, in terms of different elements and isotopic systems, remain contentious. This 98 review is intended to fill some of the existing gaps in our understanding of early Earth 99 chemical evolution by synthesizing the existing short- and long-lived refractory radiogenic 100 isotopic and elemental abundance data for a representative set of well-preserved and well-101 characterized komatiite and basalt systems that are especially advantageous for studying 102 processes that occurred within the first 2.5 billion years of Earth history. In parallel, we

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address the question of the complex origins of komatiites and their usefulness as proxies forthe chemical composition of the early Earth mantle.

105 Here, we consider komatiite-basalt systems from 13 different localities around the globe: 106 the 3.55 Ga Schapenburg, 3.48 Ga Komati, and 3.26 Ga Weltevreden komatiites in the 107 Kaapvaal Craton of South Africa; the 3.53 Ga Coonterunah, 3.34 Ga Kelly, and 3.18 Ga Ruth 108 Well and Regal komatiites and basalts in the Pilbara Craton of Western Australia; the 2.82 Ga 109 Kostomuksha, 2.41 Ga Vetreny, and 2.05 Ga Lapland komatiites in the Fennoscandian Shield 110 of northern Europe; the 2.72 Ga Pyke Hill-Alexo and Boston Creek komatiites in the Superior 111 Craton of Canada, and the 2.7 Ga Belingwe komatiites in the Rhodesian Craton of South 112 Africa. These komatiite-basalt systems were chosen because of good preservation of their 113 primary mineralogical and textural features and chemical and isotopic compositions, and 114 because complete sets of published high-precision chemical and isotope data, obtained on the 115 same sample sets, are available for these systems. A schematic world map showing the 116 location of these komatiite-basalt systems is provided in the *Electronic Supplement*.

117 **2.** Komatiites as probes of the early Earth mantle

118 Komatiites are ultramafic lavas containing more than 18% MgO in the parental liquid 119 (Arndt and Nisbet, 1982). To be classified as a komatiite, the crystallized lava must either 120 contain spinifex texture, or be genetically related to lavas containing spinifex texture. Spinifex 121 texture is a crucial feature that distinguishes komatiites from other ultramafic lavas, such as 122 picrites, boninites, and meimechites. It is characterized by the presence of large, skeletal 123 crystals of olivine that form during rapid, in situ crystallization of high-MgO, superheated 124 (*i.e.*, heated to above liquidus temperatures) silicate liquids. Due to the absence of nucleation 125 sites, superheated liquids display a reluctance to nucleate, and olivine crystals that eventually 126 form do so rapidly upon cooling below the liquidus and are typically large and skeletal, 127 leading to the formation of spinifex texture (e.g., Arndt, 1994; Arndt et al., 2008).

128 Ever since komatiites were first discovered and described in South Africa (Viljoen and 129 Viljoen, 1969), these rocks have served as a source of valuable information bearing on the 130 chemical and thermal evolution of the mantle (e.g., Arndt et al., 2008). Owing to formation 131 via the highest degrees of partial melting of all mantle-derived magmas, komatiites have 132 compositions approaching that of mantle peridotite and, thus, represent the closest available 133 approximation of the chemical characteristics of the mantle among volcanic rocks (e.g., 134 Arndt, 1977; Nesbitt et al., 1979; Herzberg, 1992). As high-degree, superheated, and low-135 viscosity melts, komatiites extracted large proportions of highly siderophile elements (HSE,

136 including Re, Os, Ir, Ru, Pt, and Pd) from the mantle, experienced little differentiation prior to 137 emplacement, and likely sampled mantle domains on the order of tens of millions of cubic 138 kilometers, as evidenced by the sheer volumes of komatiitic magmas erupted in a number of 139 Archean terrains. Some of these occurrences are argued to be similar in volume to recent and 140 modern oceanic plateaus, such as the Ontong Java plateau (e.g., Condie, 1975; Schubert and 141 Sandwell, 1989; Storey et al., 1991; Kusky and Kidd, 1992; Desrochers et al., 1993; Kimura et al., 1993; Abbott, 1996; Kent et al., 1996; Puchtel et al., 1998; Arndt et al., 2001). During 142 143 crystallization of komatiite lava flows, major and trace elements, including HSE, behaved in 144 predictable manners relative to indices of magmatic differentiation, making it possible to 145 obtain precise chronological information, estimate element mobility, and determine HSE 146 abundances in the mantle sources of most komatiites. The ability of komatiites to provide a 147 reliable record of the chemical and isotopic characteristics of mantle domains that existed 148 during the first half of Earth history is the key underlying factor for focusing on komatiites in 149 this review, which is intended to summarize what komatiites have taught us about the 150 evolution of early Earth. On the other hand, although basalts are less informative as probes of 151 the HSE characteristics of the mantle, because of their inherently higher Nd, Hf, and W 152 abundances, they are shown here to be reliable recorders of mantle Nd, Hf, and W isotopic 153 compositions and have also been studied in several komatiite-basalt systems.

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3. The analytical toolbox to study the chemical evolution of the mantle

155 The chemical evolution of the mantle has long been studied using radiogenic isotope 156 systems and lithophile and siderophile element abundances in mantle-derived rocks. The lithophile element-based isotopic systems, particularly ^{142,143}Nd and ¹⁷⁶Hf, are, by way of their 157 158 refractory nature and optimal half-lives, powerful tools for examining possible primordial 159 magma ocean processes, as well as subsequent mantle melting and differentiation processes 160 (e.g., DePaolo and Wasserburg, 1976; Jacobsen and Wasserburg, 1979; Patchett and Tatsumoto, 1980; Patchett et al., 1981; White and Patchett, 1984; Zindler and Hart, 1986; 161 162 Sivell and McCulloch, 1991). Silicate melts have lower Sm/Nd and Lu/Hf than co-existing silicate solids, and after separation, evolve to distinctive ¹⁴³Nd and ¹⁷⁶Hf compositions. If 163 Sm/Nd fractionated prior to ~4.0 Ga, while ¹⁴⁶Sm was still extant, variations in ¹⁴²Nd can also 164 be generated. Hence, application of the combined ^{142,143}Nd systems provides the temporal 165 166 resolution necessary for deciphering the timing and mechanisms of the earliest mantle 167 differentiation events (Goldstein and Galer, 1992; Harper and Jacobsen, 1992; Boyet et al., 168 2003; Caro et al., 2003, 2006, 2017; Boyet and Carlson, 2005, 2006; Bennett et al., 2007;

O'Neil et al., 2008, 2012, 2016; Rizo et al., 2012, 2013, 2016; Roth et al., 2013, 2014).
Further combining the ^{142,143}Nd with the ¹⁷⁶Hf systematics allows identification of specific
processes that may have controlled mantle differentiation, including fractionation of the highpressure bridgmanite (Mg-perovskite) and Ca-perovskite assemblage during crystallization of
a primordial magma ocean (Caro et al., 2005; Hoffmann et al., 2011; Rizo et al., 2011;
Puchtel et al., 2013; 2016a; Hoffmann and Wilson, 2017; Boyet et al., 2021).

The short-lived ¹⁸²Hf-¹⁸²W system can be used to detect fractionation of the lithophile 175 trace element Hf from the siderophile trace element W occurring within the first ~60 Ma of 176 177 Solar System history. This isotope system constitutes a high-resolution tool for identifying 178 magma ocean crystallization, the effects of late accretion, and, by virtue of its dual lithophile 179 and siderophile nature, potentially core-mantle interaction processes (Kleine et al., 2002, 180 2004, 2009; Yin et al., 2002, Jacobsen and Yu, 2015; Reimink et al., 2020; Willbold et al., 2011, 2015; Touboul et al., 2012; Puchtel et al., 2018, 2020, 2022; Rizo et al., 2019; Mundl-181 Petermeier et al., 2019, 2020). Further, application of the combined ¹⁴²Nd and ¹⁸²W tools can 182 183 help discriminate between early magma ocean differentiation processes and the effects of late 184 accretion (Harper and Jacobsen, 1996; Rizo et al., 2016; 2019; Puchtel et al., 2018; Peters et 185 al., 2021).

Abundances of HSE, combined with ¹⁸⁶Os and ¹⁸⁷Os systematics, provide additional 186 187 information about the chemical evolution of the mantle and the processes that operated early 188 in Earth history (e.g., Righter et al., 2000; Walker, 2009, 2016; Day, 2013; Puchtel et al., 189 2014; Day et al., 2016). The behavior of Re, Pt, and Os is governed by their strong 190 partitioning into metal or sulfide liquids relative to silicate melt (Crocket et al., 1997; Righter 191 and Drake, 1997). During mantle melting, Os is moderately to highly compatible with the 192 melting residue, whereas Pt and Re are incompatible to various degrees (e.g., Barnes et al., 193 1985; Rehkämper et al., 1999; Mallmann and O'Neill, 2007). Thus, the two Os isotopic 194 systems together with HSE abundances provide insight into some primordial processes to 195 which the lithophile element-based isotopic systems are not sensitive, including late-stage 196 planetary accretion and metal-silicate differentiation. Estimates of HSE abundances in 197 komatiite mantle sources can also be used to track the mixing rates of the mantle and the 198 timing of homogenization of late accreted materials within the mantle (e.g., Maier et al., 199 2009; Puchtel et al., 2018, 2020, 2022). In combination with Os isotope systematics and HSE abundances, the ¹⁸²Hf-¹⁸²W isotopic system can be used to discriminate between the effects of 200 201 early planetary differentiation, late accretion, and core-mantle interaction (e.g., Touboul et al.,

2012; Puchtel et al., 2016a, 2018, 2020, 2022; Archer et al., 2019; Rizo et al., 2019; Tusch et
al., 2021).

204 The water content of a komatiitic lava is also an important parameter for assessing where 205 in the mantle the parental melt originated. Water is highly incompatible during mantle melting 206 (e.g., Moore, 1970; Michael, 1995; Saal et al., 2002; Hauri et al., 2006; Cooper et al., 2012). 207 The water content of a given komatiite mantle source can be derived from that of a komatiite 208 lava provided that effects of secondary alteration can be corrected for. For example, 209 correlations between the abundances of water and other lithophile trace elements, e.g., Ce, in 210 submarine basalt glasses have been used in a number of studies to establish H₂O contents of 211 mantle-derived melts (e.g., Michael, 1995; Saal et al., 2002; Hauri et al., 2006; Cooper et al., 212 2012). Mid-ocean ridge basalt (MORB) and ocean island basalt (OIB) magmas have H₂O/Ce 213 \leq 370 (Hauri et al., 2006), similar to the mantle value of 200±50 (Hirschmann, 2018); the 214 rather large range is due to the fact that during anhydrous mantle melting, spinel lherzolite has 215 a bulk solid/melt $D_{\text{H2O}} \approx D_{\text{Ce}}$, whereas garnet lherzolite has a bulk solid/melt $D_{\text{H2O}} = 0.5 \times D_{\text{Ce}}$. 216 By contrast, arc magmas have H₂O/Ce of 2,000 to 3,000, due to variable contributions of H₂O 217 from the subducting slab (Cooper et al., 2012).

218 The oxidation state of a komatiite magma and its source reflects important information 219 about the location and processes involved in its generation. For example, knowledge of fO_2 220 can potentially be used to distinguish between a hydrous (*i.e.*, subduction) versus anhydrous 221 (*i.e.*, plume) origin of komatiites. Canil (1997, 1999, 2002) and Canil and Fedortchouk (2001) 222 used V partitioning between liquidus olivine and komatiite liquid as a redox indicator. These 223 authors showed that the major factor controlling the distribution of V between olivine and komatiitic melt $(D_V^{ol/liq})$ was oxygen fugacity, fO_2 . More oxidized, water-rich island arc 224 magmas (higher fO_2) are invariably characterized by $D_V^{ol/liq} < 0.01$, whereas less oxidized, 225 anhydrous mantle plume-derived magmas (e.g., OIB and MORB) exhibit $D_V^{ol/liq} = 0.025$ -226 227 0.10.

Additional diagnostic features pointing to the origins of komatiites include estimates of potential temperatures of inferred mantle sources and depths of melting initiation. To compare mantle temperatures at various localities, McKenzie and Bickle (1988) proposed the concept of a mantle potential temperature (\mathbf{T}_p) as a point of reference. Mantle potential temperature is the temperature that the solid adiabatically-convecting asthenospheric mantle would have had at the surface if it ascended without undergoing melting. Mantle potential temperature determines the depth of melting initiation (*e.g.*, Richter, 1988; Herzberg, 1992, 1995; 235 Herzberg et al., 2007), which, in turn, governs the style of mantle melting and melt 236 segregation (fractional versus batch). The depth of melting initiation and subsequent melt 237 segregation determines the chemical composition of the resultant komatiite melts, including 238 MgO contents. Pressures >8 GPa (240 km depth) stabilize majorite garnet relative to olivine 239 and pyroxene in the source (e.g., Ohtani, 1984; Herzberg and Ohtani, 1988). If the 240 temperature of a plume core is sufficiently high to initiate melting in the majorite garnet 241 stability field, the melting results in the formation of Al-depleted komatiitic melts with 242 subchondritic Al₂O₃/TiO₂ and Gd/Yb, provided the melt separated from the melting residue 243 while still in the majorite garnet stability field. By contrast, melting and melt segregation at 244 lower pressures, within the spinel lherzolite stability field, produces Al-undepleted komatiitic 245 magmas with chondritic Al₂O₃/TiO₂ and Gd/Yb (e.g., Green, 1975; Ohtani, 1984; Kato et al., 246 1988; Herzberg, 1995).

247 **4.**

4. Which parts of the mantle do komatiites sample?

In order to assess the extents of early mantle isotopic and chemical heterogeneities sampled by komatiites, it is important to determine where in the mantle komatiites originate. The location of komatiite magma generation is almost certainly not the same for all komatiites, given the extent of chemical and isotopic heterogeneity inferred for their mantle sources. This, in turn, requires assessment of diverse komatiite formation models.

253 Komatiites were most common during the Archean and may, during this eon, have been 254 the volumetrically second most significant constituent among volcanic rocks, comprising as 255 much as 25% of the entire volume of volcanic sequences, the rest being composed mostly of 256 basalts (e.g., Condie, 1975, 1981, 1994; deWit and Ashwal, 1997). Some of these basalts were 257 also derived from komatiites via fractional crystallization processes, while others were lower-258 degree partial melts generated from the same sources as the spatially associated komatiites 259 (Arndt et al., 1977; Campbell et al., 1989; Condie, 2005). The abrupt decline in the abundance 260 of komatiites at the Archean-Proterozoic boundary, as well as the decrease in the MgO 261 contents of emplaced komatiite lavas, have been interpreted as evidence for a significant, up 262 to 300°C, secular cooling of the mantle over the course of Earth history (e.g., Bickle, 1982; 263 Nisbet et al., 1993; Herzberg et al., 2007; Herzberg and Gazel, 2009). Some authors have 264 linked this to a transition from drip to subduction tectonics (Campbell and Griffiths, 2014), 265 arguing that an increasing abundance of subducted slabs allowed for formation of the core-266 enveloping D" insulating layer, which they in turn associate with a step-like drop in maximum 267 komatiite magma MgO contents and mantle potential temperatures between 2.7 and 2.0 Ga.

268 The origin of komatilites has been a subject of studies for the past half century (e.g.,269 Green, 1975, 1981; Allègre, 1982; Arndt et al., 1998, 2008; Parman et al., 2001, 2004; Berry 270 et al., 2008; Sobolev et al., 2016, 2019; Sossi et al., 2016; Asafov et al., 2018). A common 271 interpretation is that the parental magmas to komatilites were produced in unusually hot 272 upwellings of mantle material, termed mantle plumes (e.g., Cawthorn, 1975; Campbell et al., 273 1989; Griffith and Campbell, 1990; Richards et al., 1991; Loper, 1991; Herzberg, 1995; 274 Richard et al., 1996). The plumes most likely initiated at one of the two major thermal 275 boundary layers in the mantle, *i.e.*, either the core-mantle boundary (CMB) at 2900 km, or the 276 660 km discontinuity, with the ensuing rise driven by a buoyancy contrast between the less 277 dense, hot plume material and the cooler and denser surrounding mantle (Campbell and 278 Griffiths, 1990, 1992, 1993; Farnetani and Richards, 1995; Farnetani, 1997). The initially 279 solid mantle plume material was partially melted upon reaching shallower depths as a result 280 of decompression. Based on experimental data, Campbell et al. (1989) argued that komatiites 281 were produced in the hotter plume core containing mostly materials derived from near the 282 thermal boundary layer, whereas spatially associated basalts were generated in the cooler 283 plume head which, upon arrival at the surface, would have contained a substantial amount of 284 entrained upper mantle material. However, later studies (e.g., Farnetani and Richards, 1995) 285 argued that such entrainment was unlikely to contribute significantly to the erupted melts.

286 Two major types of modern mantle plumes have been proposed based on the depth of 287 their initiation, determined using recent high-resolution seismic tomography imaging data 288 (e.g., Courtillot et al., 2003; Montelli et al., 2004; He et al., 2015; Bao et al., 2022). The first 289 type is "primary", or Morganian, plumes that are commonly thought to originate in the 290 deepest parts of the lower mantle, perhaps from seismically anomalous regions. Early 291 geophysical studies (e.g., Dziewonski et al., 1977) identified seismically anomalous zones 292 along the core-mantle boundary, the so-called Large Low Shear-Wave Velocity Provinces 293 (LLSVP) and Ultra-Low Velocity Zones (ULVZ). The origin of these regions in the mantle is 294 debated. It has been proposed that LLSVPs may be stratified into primordial bottom domains 295 near the core-mantle boundary, often referred to as the D" layer, and mafic shallow domains 296 that extend from ~1100 to 2300 km depths (e.g., Ballmer et al., 2016; Koppers et al., 2021). 297 These regions in the mantle may serve as nurseries for the majority of "primary" mantle 298 plumes (French and Romanovicz, 2015; Totsvik et al., 2016; Koppers et al., 2021). Ocean 299 island basalt systems that have been associated with this type of plume include Azores, 300 Canary, Coral Sea, Easter, Galapagos, Hawaii, Iceland, Samoa, and Tahiti (Courtillot et al., 301 2003).

The "secondary" type, or Andersonian, plumes have been proposed to originate at the 660 km discontinuity and, hence, would be an upper mantle feature. Ocean island basalt systems that have been associated with this type of plumes include Ascension, Caroline, McDonald, and Pitcairn islands (Courtillot et al., 2003).

306 Considering the difficulties in determining even present-day mantle plume characteristics 307 (e.g., Fletcher and Wyman, 2015, and references therein), it is not surprising that there are 308 few reliable estimates of Archean plume parameters (Arndt et al., 2008). This is due to the 309 fact that, unlike modern plumes, ancient plumes are extinct mantle features and, therefore, 310 cannot be examined by geophysical methods. Consequently, indirect approaches must be 311 taken, such as geological field observations. For example, the size of postulated plumes can 312 potentially be constrained by the lateral extent of contemporaneous magmatic activity within 313 a given region. Envisioned parental plume heads of up to 2500 km in diameter for large 314 systems, such as the Yilgarn Craton in Western Australia (Campbell and Hill, 1988; Mole et 315 al., 2014; Barnes and Van Kranendonk, 2014; Barnes et al., 2016), have been argued to be 316 indicative of plumes that originated at the CMB. Smaller plume heads, estimated to range 317 from 200 km to 300 km in diameter, and leading to less widespread volcanism, such as the 318 komatiite-basalt systems of the Pilbara Craton in Western Australia and the Superior Craton 319 in Canada (Arndt et al., 2001; Van Kranendonk, 2008; Hickman and Van Kranendonk, 2012; 320 Dostal and Mueller, 2013; Wyman, 2013, 2018, 2020), are likely to be more typical of upper 321 mantle plumes originated at the 660 km discountinuity.

Puchtel et al. (2009b, 2013, 2016a) calculated H₂O contents of <0.2 wt. % in the Pyke Hill, Belingwe, Schapenburg, Komati, and Weltevreden komatiites, and concluded that their parental magmas were formed under dry, high-temperature melting conditions. Similar estimates of water content have been obtained for the Belingwe komatiites on the basis of the oxidation state of iron in melt inclusions in olivine (*e.g.*, Berry et al., 2008). Thus, it is a common assumption that most komatiites originated from dry, deep-seated mantle sources (Arndt et al., 1998, 2008).

Consistent with the interpretation of an anhydrous, deep mantle origin for most komatiites, Nicklas et al. (2018, 2019) concluded, on the basis of a global survey of partitioning behavior of V between olivine and emplaced komatiitic and picritic melts, that the redox states of a variety of komatiite and OIB sources, ranging in age from 3.48 Ga to present day, were either similar to, or lower than, those of modern MORB.

Recent studies of komatiitic olivine melt inclusions (*e.g.*, Sobolev et al., 2016, 2019; Asafov et al., 2018), while maintaining an anhydrous, deep mantle plume origin for komatiites, have argued for relatively high water contents in some komatiite magmas, with water being added to rising, dry plume material from a thin hydrous layer located at the top of the mantle transition zone (MTZ), between the 410- and 660- km seismic discontinuities. The origin of these seismic interfaces is attributed to solid- solid phase transitions from olivine to wadsleyite and ringwoodite to perovskite+magnesiowüstite, respectively (*e.g.*, Bina and Helffrich, 1994). The hydrous layer is predicted by Bercovici and Karato (2003) in their MTZ water filter model to occur at the 410 km depth.

343 Alternatively, it has been argued that komatiltes in general, and Komati lavas from the 344 Kaapvaal Craton in South Africa in particular, were produced in a special type of Archean 345 subduction environment via hydrous melting of the mantle at shallow depths and at 346 temperatures similar to, or only slightly higher than, those of the ambient upper mantle (e.g., 347 Allègre, 1982; Grove et al., 1997; Parman et al., 1997, 2001, 2004). An important initial 348 condition of this hypothesis is the requirement that the Komati komatiites represent intrusive 349 bodies, since magmatic water escapes outgassing only under pressure (Grove et al., 1997). 350 According to the hydrous komatiite model of Parman et al. (1997, 2001), the presence of 351 water determined both the crystallization sequence and composition of the magmatic minerals 352 of the Komati komatiites. Parman et al. (1997) carried out a detailed experimental study 353 crystallizing material of komatiitic composition under both anhydrous and hydrous 354 conditions, paying particular attention to the composition of the crystallizing pyroxene. Using 355 the pyroxene compositions, these authors estimated that the original Komati komatiite magma 356 contained between 4 and 6 wt. % H₂O. Such a large amount of water would require the 357 temperature in the Komati source to be ~250°C lower than in anhydrous melting models. In 358 their follow-up review paper, Grove and Parman (2004) argued that the Archean mantle was $\leq 100^{\circ}$ C hotter than the modern mantle, hence, offering a view on the thermal evolution of the 359 360 Earth that contrasts with those that are based on the traditional assumption of a substantially hotter Archean mantle (e.g., Richter, 1985, 1988; Bickle, 1986; Nisbet et al., 1993; Herzberg 361 362 et al., 2007, 2010).

The summary of estimates of the mantle potential temperatures $(\mathbf{T}_p \circ \mathbf{C})$ for the sources of the komatiite systems considered here, as well as the liquidus temperatures of the emplaced komatiite magmas $(\mathbf{T}_{liq} \circ \mathbf{C})$ and depths of melting initiation in the respective mantle plumes $(\mathbf{D}_{melt\ init})$, are presented in **Table 1**. These estimates were generally derived from the studies for the particular komatiite systems referenced in **Table 1** and updated here on the basis of the more recent estimates of the emplaced komatiite lava compositions from Nicklas et al. (2018, 2019), where available, coupled with the previous and more recent mantle melting parameterization models of McKenzie and Bickle (1988), Herzberg and O'Hara (2002),
Herzberg and Asimow (2008), and Herzberg and Gazel (2009).

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5. Chemical and isotopic data

A summary of the available chemical and isotopic data for the komatiite systems
considered in this review are given in Tables 1 and 2.

375 5.1. Emplaced lava compositions, mantle potential temperatures, and depths of melting

Estimates of emplaced komatiite lava compositions (MgO_{liq}), their liquidus temperatures ($T_{liq}^{\circ}C$), mantle potential temperatures ($T_{p}^{\circ}C$), and depths of mantle melting initiation (D_{melt} init) are listed in **Table 1**. The details of the methodology used for calculating these parameters are provided in the *Electronic Supplement*.

380 The calculated depths of melting initiation range between 605 and 203 km, while liquidus 381 temperatures vary between 1606°C and 1498°C. There is an apparent tendency for a decrease 382 in depth of melting initiation between 3.6 and 2.0 Ga. This is also reflected in the ~100°C 383 decrease in mantle potential temperatures of komatiite sources from 1841 to 1748°C and a 384 corresponding decrease in MgO contents of emplaced komatiite lavas from ~31 to 25 wt.% 385 over the same period of time. Two exceptions are the Coonterunah and Kelly komatiite 386 systems that are less MgO-rich than the other early Archean systems. Therefore, the data 387 place the depths of onset of melting for all komatiite systems considered here within and 388 above the present-day MTZ and provide minimum estimates for the depths of mantle plume 389 initiation. In the following sections, more clues, as to which parts of the mantle the studied 390 komatiite systems sampled (*i.e.*, depths of mantle plume initiation, $D_{plume init}$), will be 391 considered using the available geochemical and isotopic data (Table 2).

392 **5.2.** Chemical composition and isotopic systematics

393

5.2.1. Major and lihophile trace elements

394 The petrologically most important chemical element in komatiites, along with Mg, is Al; 395 as a result, variations in Al₂O₃, and its rare earth element (REE) behavioral equivalent Yb, 396 form an integral part of all komatiite classifications (e.g., Nesbitt et al., 1979; Jahn et al., 397 1982). The komatiite systems reviewed here can be subdivided into two major petrological 398 groups on the basis of their Al₂O₃ and Yb contents at a given MgO content, namely Al-399 depleted/enriched and Al-undepleted. The majority of the early Archean komatiite systems, 400 including the Schapenburg, Komati, Weltevreden, Ruth Well, and Regal systems, belong to 401 the Al-depleted/enriched type of lavas characterized by either sub- or supra-chondritic Al/Ti 402 and Gd/Yb ratios (**Table 1**). Exceptions are the Coonterunah and Kelly systems that belong to 403 the Al-undepleted type of lavas with $Al_2O_3/TiO_2 = 22.8$ and 23.1 and $Gd/Yb_N = 0.96$ and 0.91, 404 respectively (where N denotes BSE-normalized values of Hofmann, 1988). By contrast, the 405 majority of the late Archean komatiite systems, including the Pyke Hill-Alexo, Belingwe, 406 Vetreny, and Lapland systems, belong to the Al-undepleted type of lavas characterized by 407 generally chondritic Al/Ti and Gd/Yb ratios. The Boston Creek (Puchtel et al., 2018) and 408 Kostomuksha komatiites (Puchtel et al., 1998), with $Al_2O_3/TiO_2 = 5.1$ and 17.2 and Gd/Yb_N = 409 2.0 and 1.2, respectively, are notable exceptions. Although the Lapland komatiites have low 410 $Al_2O_3/TiO_2 = 13.2$ and high $Gd/Yb_N = 1.5$, these features have been shown to be the result of 411 enrichment in Ti and middle REE (MREE), rather than depletions in Al and heavy REE 412 (Puchtel et al., 2020).

413 Owing to their high liquidus temperatures and low viscosities during their passage through 414 and/or emplacement onto the continental crust, komatiitic liquids are highly susceptible to 415 contamination by upper crustal rocks (Huppert et al., 1984; Huppert and Sparks, 1985). 416 Archean upper crustal rocks are strongly enriched in Th, U, and light REE (LREE), and 417 relatively depleted in Nb (Rudnick and Gao, 2014) as a result of subduction zone processes 418 involving retention of Nb-rich rutile in the eclogitic residue (Foley et al., 2000; Rudnick et al., 419 2000). Because of that, upper crustal rocks are characterized by negative Nb anomalies 420 relative to elements with similar incompatibility during mantle melting (Th and La), *i.e.*, their Nb/Nb* <<1.0 (where Nb/Nb* = Nb_N/ $\sqrt{(Th_N \times La_N)}$). By contrast, primary komatiitic magmas 421 are expected to have Nb/Nb* \geq 1.0 (e.g., Hofmann et al., 1986; Jochum et al., 1991). Due to 422 423 large differences in highly incompatible lithophile trace element concentrations between the 424 upper crust and komatiitic melts, crustal contamination usually leads to sharp increases in the 425 abundances of Th, U, W, and LREE, but much less so of Nb, in the hybrid melts, which 426 acquire negative Nb anomalies in the process. Hence, the Nb/Nb* ratio represents a 427 diagnostic tool that has been utilized to evaluate and mathematically correct for the effects of 428 crustal contamination (e.g., Puchtel et al., 2016b, 2020, 2022).

The majority of the komatiite systems considered here are variably depleted in highly incompatible lithophile trace elements, with the La/Sm_N ratios ranging between 0.14 (the 2.05 Ga Lapland system) and 0.97 (the 3.48 Ga Komati system). The 2.72 Ga Boston Creek lavas are the sole exception (La/Sm_N = 1.9). Furthermore, all komatiite systems examined here have Nb/Nb* \geq 1.0, reflecting either a lack of crustal contamination, or that the effects of crustal contamination have been mathematically corrected for, as indicated in **Table 2**. The details of the correction protocol are provided in the *Electronic Supplement*. 436 5.2.2. Isotopic systems of the lithophile elements

Table 2 presents the initial ε^{143} Nd and ε^{176} Hf values (defined as the deviation in parts per 437 10,000 of initial ¹⁴³Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf in a given sample from those in the chondritic 438 439 reference at the time of lava emplacement) for each komatiite system, which represent 440 compositions of the respective komatiite-basalt mantle sources. These values were derived 441 from the respective Sm-Nd and Lu-Hf isochrons obtained for each komatiite-basalt system 442 and, where appropriate, corrected for crustal contamination, as indicated in **Table 2**, using the 443 lithophile trace element systematics discussed in the previous section. The resultant initial 444 ϵ^{143} Nd and ϵ^{176} Hf values are all positive, ranging between +0.46 and +4.9 and +1.7 and +10.2, 445 respectively, indicating that all the komatiite-basalt systems considered were derived from mantle sources with time-integrated supra-chondritic Sm/Nd and Lu/Hf. 446

By contrast, Blichert-Toft et al. (2015) reported ¹⁴³Nd and ¹⁷⁶Hf data for a suite of heavily 447 altered Komati drill core samples from several flows. That study produced a much greater 448 scatter, and both positive and negative initial ϵ^{143} Nd and ϵ^{176} Hf values for whole-rock 449 450 samples, than for the initial values obtained from the individual Sm-Nd and Lu-Hf isochrons 451 for the remarkably well preserved Komati komatiite samples from the Puchtel et al. (2013) 452 study reported in Table 2. Blichert-Toft et al. (2015) also reported data for variably leached clinopyroxene separates, which yielded initial ϵ^{143} Nd and ϵ^{176} Hf values of -2 and +5, 453 454 respectively. This may indicate more complex magmatic and/or alteration history of the 455 samples studied by Blichert-Toft et al. (2015) than for the Komati samples studied by Puchtel 456 et al. (2013).

457 The time-integrated Sm/Nd and Lu/Hf ratios in the mantle sources of komatiite systems derived from the respective Sm-Nd and Lu-Hf isochron sets are plotted in Fig. 1. These ratios 458 459 were calculated by assuming the minimum-degree fractionation from either the chondritic values or the values defined by the combined ^{142,143}Nd systematics (where available) to those 460 required to bring the ϵ^{143} Nd and ϵ^{176} Hf to the measured initial ϵ^{143} Nd and ϵ^{176} Hf of the 461 respective mantle sources by the time of komatiite-basalt system formation. All of these 462 komatiite systems were derived from mantle sources having time-integrated ¹⁴⁷Sm/¹⁴⁴Nd and 463 ¹⁷⁶Lu/¹⁷⁷Hf ratios between those of CHUR and DMM. An important feature of the data is that, 464 465 while all komatiite-basalt systems younger than ~3.0 Ga follow the terrestrial Nd-Hf mantle 466 evolution array, indicating long-term "coupled", or congruent, behavior of the Nd and Hf 467 isotope systematics in the post-3.0 Ga komatiite mantle sources, the komatiite systems older 468 than ~3.0 Ga (with the exception of the Pilbara Craton komatiite-basalt systems) plot above the terrestrial evolution curve, indicating long-term "decoupled", or incongruent, behavior of the Nd and Hf isotope systematics in their mantle sources. All these early Archean systems are from the Kaapvaal Craton in South Africa. The processes that might have been responsible for producing such Nd-Hf isotope decoupling are discussed in **Section 6.1**.

Relatively limited ¹⁴²Nd data are available for the komatiite systems reviewed in this 473 474 study (Table 2). The komatiite systems, for which these data are available, mostly exhibit μ^{142} Nd values indistinguishable from the standard reference value, where μ^{142} Nd is defined as 475 the deviation in parts per million of ¹⁴²Nd/¹⁴⁴Nd in a given komatiite sample from the 476 laboratory reference materials (AMES or JNdi), which are considered to represent the 477 478 composition of the modern BSE. The Schapenburg and Komati komatiite systems have small 479 μ^{142} Nd deficits, averaging -5.0±2.8 (2SD; Puchtel et al., 2016a) and -4.0±4.1 (2SD; Boyet et 480 al., 2021), respectively.

481 <u>5.2.3. Tungsten abundance and isotope systematics</u>

The ¹⁸²W data are reported as μ^{182} W, which is the deviation in parts per million of 482 ¹⁸²W/¹⁸⁴W in a given sample from that of the in-house Alfa Aesar laboratory W reference 483 material. The μ^{182} W value of 0 for the reference material is considered to represent the 484 485 composition of the BSE. This is based on the observation that a limited number of data for 486 modern MORB (Willbold et al., 2011; Rizo et al., 2016; Mundl et al., 2017), numerous data 487 for OIB with low ³He/⁴He (Mundl-Petermeier et al., 2020), and data for Proterozoic and Phanerozoic glacial diamictites and granites (Mundl et al., 2018; Nakanishi et al., 2021) are 488 489 all characterized by μ^{182} W values of ~0.

In contrast to ¹⁴²Nd, the majority of komatiite systems examined here exhibit either positive (all the Pilbara, the Kostomuksha, and the Boston Creek komatiite systems), or negative (the Schapenburg and Lapland komatiite systems) ¹⁸²W anomalies, with only the Komati and Vetreny komatiite systems lacking an ¹⁸²W anomaly.

494 The behavior of W during magmatic processes is governed mainly by the redox state of 495 the magmatic system in question (Newsom et al., 1996; Arevalo and McDonough, 2008; 496 König et al., 2011). Under reducing conditions, such as those that likely existed during 497 metallic core formation, W behaves as a moderately siderophile element (MSE), 498 concentrating largely in the metal relative to the silicate melt. During mantle melting under 499 relatively oxidizing conditions in the absence of a metal phase, such as typical of the modern 500 upper mantle, W behaves as a highly incompatible element, concentrating in the silicate melt 501 relative to the melting residue, with a degree of incompatibility similar to that of Th and U.

15

502 The Archean mantle was recently shown to be only slightly (by ~1.3 Δ FMQ log units) less 503 oxidized than, whereas the Proterozoic mantle was found to be similarly oxidized to, the 504 modern mantle (Nicklas et al., 2018, 2019). Therefore, it is expected that during the mantle 505 melting that produced the komatiite-basalt systems considered in this review, W behaved 506 similarly to Th and U and, hence, that in undisturbed komatiite-basalt systems, W/Th \approx 507 W/Th_{BSE}, where W/Th_{BSE} = 0.19 (McDonough and Sun, 1995). Indeed, in modern OIB and 508 MORB, for which W is characterized by a primary, magmatic distribution, the W/Th ratios 509 are similar to the BSE value of 0.19 (e.g., König et al., 2008, 2011; Jenner and O'Neill, 2012). 510 Modern island arc lavas, on the other hand, show uniformly elevated W/Th ratios relative to 511 the BSE value attributed to various constributions of fluid-borne W to the arc mantle from the 512 subducted slab (König et al., 2008, 2011). Although some komatiite systems (e.g., the 513 Komati, Vetreny, and Lapland systems) have average W/Th ratios within the uncertainty 514 limits of the BSE value, and in that respect are similar to modern OIB and MORB, others are 515 variably either enriched (most common) or depleted in W over Th (Table 1). In addition, the 516 lack of correlation between W concentrations and indices of magmatic differentiation, such as 517 MgO content, indicates either W mobility after lava emplacement in most komatiite-basalt 518 systems (e.g., Touboul et al., 2012; Puchtel et al., 2016a, 2018, 2020, 2022; Tusch et al., 519 2021) or involvement of a yet-unaccounted-for magmatic process.

520 The issue of the origin of W in Archean and Proterozoic komatiite-basalt systems is, 521 therefore, controversial. Recent studies have shown that W abundance and isotopic 522 systematics can be affected by crustal contamination, hydrothermal alteration, and 523 metamorphism (e.g., Touboul et al., 2014; Liu et al., 2016; Puchtel et al., 2016a,b, 2018, 524 2020, 2022; Tusch et al., 2019, 2021), resulting mostly in W enrichments relative to immobile 525 trace elements with similar degree of incompatibility, e.g., Th. Two mechanisms, based on the 526 origin of W, have been proposed to account for the elevated W/Th ratios in komatiites and 527 basalts. The first mechanism advocates derivation of W from an endogenous source, whereby 528 W originated within the komatiite-basalt system. The second mechanism assumes an 529 exogeneous origin, whereby W is introduced into the komatiite-basalt systems from 530 genetically unrelated sources. We address these two mechanisms in more detail below.

531 *Endogenous* origin of W in komatiite-basalt systems have been advocated for the 532 Kostomuksha (Touboul et al., 2012), Schapenburg (Puchtel et al., 2016a), Boston Creek 533 (Puchtel et al., 2018), and the Coonterunah and Kelly systems (Puchtel et al., 2022). In case of 534 the two Pilbara komatiite-basalt systems, the komatiites are characterized by high W/Th 535 ratios, whereas the basalts, that are spatially associated and genetically related to komatiites, 536 are depleted to only slightly enriched in W relative to Th. In these komatiite-basalt systems, 537 and also in many other Archean komatiite-basalt systems around the globe, komatiites 538 constitute 5-10% of the sequence by volume, the rest consisting of basalts (Condie, 1981; de 539 Wit and Ashwal, 1997; Smithies et al., 2007). Originally, basalts contain significantly higher 540 abundances of all highly incompatible trace elements, including W, compared to komatiites. 541 During secondary alteration, komatiites are turned into serpentinites, whereas basalts are 542 transformed into amphibolites. Serpentine is a major host for W and can accommodate several 543 orders of magnitude more W than the precursor mineral olivine, whereas W shows a lower 544 affinity for amphibole (Liu et al., 2016). Therefore, if W is mobilized and redistributed within 545 a komatiite-basalt volcanic sequence, it is expected to be preferentially captured by komatiites 546 during serpentinization of the primary mineral assemblage. The effect of this relative W 547 enrichment is amplified by the low trace element concentrations in komatiites, resulting in 548 high W/Th ratios (Puchtel et al., 2022).

In the case of the Boston Creek system, samples collected across a differentiated lava flow have remarkably uniform ¹⁸²W compositions, but show a wide range of W/Th ratios, from enriched in the lower olivine cumulate part to BSE-like in the low-Mg basaltic layer, with strong inverse correlation between MgO content (*i.e.*, amount of modal serpentine) and W/Th (Puchtel et al., 2018). The above features were argued by these authors to reflect an endogenous nature of W in the Boston Creek system, with isotopically uniform W having been redistributed within the lava flow and hence reflecting the primary magmatic signature.

556 The above scenarios have been argued to be applicable to several other komatiite-basalt 557 systems, including the Kostomuksha (Touboul et al., 2012), and the Schapenburg (Puchtel et 558 al., 2016a) systems.

559 *Exogenous* W in the komatiite-basalt systems considered in this review has been shown 560 to be derived from upper crustal rocks either during passage of mafic-ultramafic magmas 561 through the continental crust *en route* to the surface (the Vetreny and Lapland systems: 562 Puchtel et al., 2016b, 2020), or as a result of interaction with upper crustal rocks during both 563 the magmatic stage *via* crustal contamination, and the post-magmatic stage *via* fluid-rock 564 interaction (the Ruth Well and Regal systems: Puchtel et al., 2022).

565 Due to the generally high W abundances in upper crustal rocks (*e.g.*, Rudnick and Gao, 566 2014), crustal contamination has the potential to modify both W concentrations and ¹⁸²W 567 compositions of komatiite-basalt magmas during ascent, as has been argued to be the case for 568 the Vetreny, Lapland, Ruth Well, and Regal systems on the basis of studies of lithophile trace 569 element systematics (Puchtel et al., 2016b, 2020, 2022). In order to mathematically correct for 570 the effects of magmatic crustal contamination, these authors performed un-mixing 571 calculations using the degrees of crustal contamination estimated on the basis of trace element 572 systematics and assumptions about the W isotopic composition and abundances in the most 573 likely potential crustal contaminant.

574 Being a highly fluid-mobile element (e.g., König et al., 2008), W can be transferred from 575 spatially associated granitic rocks to komatiite-basalt sequences via W-rich fluids. Such a 576 mechanism has been proposed to affect the W isotopic composition of the Ruth Well and 577 Regal komatiite systems on the basis of the high W/Th ratios in both komatiites and basalts and the similarity in ¹⁸²W compositions between the most W-enriched komatiites and the 578 579 spatially associated 3.5 Ga tonalites of the Pilbara Craton (Puchtel et al., 2022). These authors 580 were able to evaluate the W isotopic composition of the primary lavas by assuming that their ¹⁸²W composition was similar to that of the least affected basalts from Tusch et al. (2021) and 581 582 Puchtel et al. (2022), with W/Th ratios closest to that in the BSE estimate.

583 After corrections have been applied where needed, the μ^{182} W values reported in **Table 2** 584 are considered to reflect those in the sources of the respective komatiite-basalt systems.

585 5.3. Os isotope systematics and HSE abundances

The ^{186,187}Os data are listed in **Table 2** and plotted in **Fig. 2**. The majority of the komatiite 586 systems have γ^{187} Os values (defined as percent deviation of the 187 Os/ 188 Os ratio in a given 587 588 komatiite sample from a chondritic reference value at the time of komatiite lava 589 emplacement) plotting within the chondritic range, indicating evolution of their mantle 590 sources with time-integrated near-chondritic Re/Os ratios. The only exceptions are the Schapenburg and Kostomuksha komatiite systems that have more radiogenic γ^{187} Os values of 591 592 +3.7 and +2.5, respectively, implying that their mantle sources evolved with long-term 593 slightly suprachondritic Re/Os.

The limited ¹⁸⁶Os data available indicate that, out of the seven systems studied, only three 594 (the Pyke Hill, Belingwe, and Vetreny systems) have initial μ^{186} Os values (defined as the 595 deviation in ppm of the ¹⁸⁶Os/¹⁸⁸Os ratio in a given komatiite sample from a chondritic 596 597 reference value at the time of the komatiite lava emplacement) that plot within the chondritic 598 range, indicating that their mantle sources evolved with time-integrated near-chondritic Pt/Os ratios. The other four komatiite systems have non-chondritic initial μ^{186} Os values and, thus, 599 600 evolved with either suprachondritic (the Weltevreden, Kostomuksha, and Lapland systems) or 601 subchondritic (the Komati system) time-integrated Pt/Os ratios. Importantly, among the 602 mantle sources with non-chondritic Re-Os and/or Pt-Os systematics, only in the Kostomuksha komatiite source are these isotope systematics coupled, whereas in the others, they aredecoupled.

The total HSE abundances in the komatiite mantle sources have been calculated using the bootstrap method of Puchtel et al. (2004b) that was refined in subsequent studies (*e.g.*, Puchtel et al., 2020, 2022). This method combines the ^{186,187}Os isotopic and HSE abundance data for a suite of komatiitic lavas derived from a common primary magma and related by fractional crystallization of olivine after lava emplacement. The details of the analytical protocol used in the bootstrap method of Puchtel et al. (2004b) are provided in the *Electronic*

611 Supplement.

612 The total HSE abundances in the sources of the komatiite systems considered in this 613 review, calculated as percent of those in the estimates of the modern BSE of Becker et al. 614 (2006), are presented in **Table 2** and plotted as a function of their age in **Fig. 3**. The 615 calculated total HSE abundances range from ~30% in the 3.55 Ga Schapenburg and 3.53 Ga 616 Coonterunah systems to ~120% in the 2.05 Ga Lapland system. Overall, the entire dataset, 617 except for the obvious outliers (e.g., the 2.72 Ga Boston Creek, 2.69 Ga Belingwe, and 2.41 618 Ga Vetreny systems), exhibits a broad trend of increasing HSE abundances in the komatiite 619 mantle sources with time. This trend is generally consistent with that defined by the much 620 more scattered data of Maier et al. (2009).

In the following sections, the data reviewed in **Section 5** will be discussed in light of the possible mechanisms responsible for creating these early chemical and isotopic heterogeneities in the mantle.

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625 **6.1.** Primordial differentiation of the mantle

6. Mechanisms for creating early mantle heterogeneities

626 Crystallization of a primordial magma ocean(s) is one possible mechanism for generating 627 early chemical and isotopic heterogeneities in the mantle (e.g., Boyet et al., 2003, 2021; Boyet 628 and Carlson, 2005, 2006; Caro et al., 2005, 2017; Brown et al., 2014; Puchtel et al., 2016a; 629 Byerly et al., 2017; Morino et al., 2017). Periodic magma oceans are likely a direct 630 consequence of planetary growth due to gravitational energy release from core formation and 631 high-energy collisions during final stages of accretion (e.g., Tonks and Melosh, 1993; Bottke 632 et al., 2010; Rubie et al., 2011). Crystallization of a magma ocean results in stratification 633 controlled by fractionation of mineral assemblages, the composition of which depends on the 634 depth of the magma ocean and the composition of the melted domain. Some studies have 635 argued for occasional, near-complete melting of the silicate Earth, such as may have occurred

636 during the putative giant impact that led to formation of the Moon (Rubie et al., 2011; Canup, 637 2012; Elkins-Tanton, 2012; Young et al., 2016). The pressures and temperatures near the 638 bottom of such a magma ocean would have stabilized bridgmanite (Mg-perovskite) and Ca-639 perovskite (Kato et al., 1988; Abe, 1997; Walter et al., 2004). Because most trace elements 640 are variably compatible in Ca-perovskite, with the degree of compatibility decreasing in the 641 order Lu>Hf>Sm>Nd (Corgne et al., 2005), the presence of Ca-perovskite in the fractionating 642 assemblage would result in stronger fractionation of Lu/Hf relative to Sm/Nd than that 643 observed under upper mantle conditions. Over time, this would result in Nd-Hf isotopic 644 decoupling in the mantle domains derived from magma ocean differentiation products, 645 relative to the coupling of these two isotope systems observed in the sources of modern 646 MORB and most OIB, which together define the modern Nd-Hf mantle array, or terrestrial 647 evolution curve (e.g., Salters and White, 1998).

648 The Nd-Hf mantle array owes its origin to the coupled, or congruent, Nd-Hf isotope 649 systematics of most terrestrial rocks that arise from similar partitioning behavior of the parent (¹⁴⁷Sm, ¹⁷⁶Lu) and daughter (¹⁴³Nd, ¹⁷⁶Hf) isotopes of the Sm-Nd and Lu-Hf systems during 650 651 upper mantle differentiation and crust-forming processes (e.g., Patchett et al., 1981; Salters 652 and White, 1998; Vervoort and Blichert-Toft, 1999; Vervoort et al., 1999, 2000). During 653 mantle melting, both parent isotopes are less incompatible with the melt fraction than the 654 daughter isotopes, which results in higher Sm/Nd and Lu/Hf ratios in the melting residue 655 compared to the melt; over time, the residual mantle evolves to more radiogenic Nd and Hf 656 isotopic compositions than the extracted melts. Both isotope systems are lithophile, refractory, 657 insensitive to redox conditions, and largely immobile during alteration and metamorphism. 658 These properties make them useful for investigating the interplay between the formation and recycling of oceanic and continental crust in driving the geochemical evolution of the mantle. 659

660 The decoupling of the Nd and Hf isotope systematics (and, thus, deviation from the 661 terrestrial Nd-Hf array) in the early Archean komatiite-basalt systems from the Kaapvaal 662 Craton, such as Schapenburg, Weltevreden, and Komati, have been interpreted to reflect the 663 involvement of primordial magma ocean processes.

Based on the combined ^{142,143}Nd and ¹⁷⁶Hf systematics, Puchtel et al. (2013) developed a model of formation of the mantle silicate reservoirs that gave rise to the Komati and Weltevreden komatiites *via* crystallization of a primordial magma ocean. According to their model, during magma ocean differentiation, the lower three quarters of the magma column (from depths of ~2900 to ~700 km) were largely dominated by bridgmanite, with subordinate amounts of ferropericlase and Ca-perovskite, Ca-Pv (Trønnes and Frost, 2002; Ito et al., 2004; 670 Caro et al., 2005; Elkins-Tanton, 2008). Puchtel et al. (2013) used an equilibrium 671 crystallization model based on the high-pressure and -temperature partitioning studies of 672 Corgne and Wood (2002; 2004), Corgne et al. (2005), Liebske et al. (2005), and Walter et al. 673 (2004), and assumed a completely molten mantle with a composition corresponding to that of 674 the early depleted reservoir (EDR) of Boyet and Carlson (2006) that crystallized within the 675 first 150 Ma of the Solar System history, i.e., by 4400 Ma.

676 Since the study of Puchtel et al. (2013) has been published, the EDR model of Boyet et al. (2006) has been proven untenable. This model was based on the data available at the time that 677 the accessible Earth has a greater ¹⁴²Nd/¹⁴⁴Nd ratio than chondrites (Carlson et al., 2007). This 678 679 ¹⁴²Nd difference required a higher-than-chondritic Sm/Nd ratio for the accessible Earth. This 680 ratio must have been acquired during global silicate differentiation within the first 30 million 681 years of Solar System formation (Boyet and Carlson, 2006) and implied the formation of a complementary ¹⁴²Nd-depleted reservoir that is either hidden in the deep Earth, or lost to 682 683 space by impact erosion (Caro et al., 2008). However, Burkhardt et al. (2016) showed that, compared to chondrites. Earth's building blocks were enriched in ¹⁴²Nd that was produced by 684 the s-process of nucleosynthesis, which leads to higher ¹⁴²Nd/¹⁴⁴Nd ratios. After these authors 685 corrected for this effect, the ¹⁴²Nd composition of chondrites and the accessible Earth were 686 687 shown to be indistinguishable. The results of Burkhardt et al. (2016), therefore, obviate the need for a hidden-reservoir or ¹⁴²Nd-superchondritic Earth models and imply a chondritic 688 689 Sm/Nd ratio for the BSE.

In view of the new data of Burkhardt et al. (2016), we have modified the model of Puchtel et al. (2013), as illustrated in **Fig. 4**; the methodology used in these model calculations are available as an *Electronic Supplement*. The composition of the primordial magma ocean, unlike in our previous model, is now represented by that of the chondritic uniform reservoir (CHUR).

Figure 4 shows the time-integrated ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁷⁶Lu/¹⁷⁷Hf ratios in the sources of 695 696 the Komati and Weltevreden komatiite systems formed as a result of the 4400 Ma magma 697 ocean differentiation and cryslallization event specified above, depending on the relative 698 proportions of fractionating bridgmanite and Ca-Pv. The upper panel of the diagram 699 represents aggregate compositions of the magma ocean cumulates, while the lower panel 700 represents the compositions of the residual liquids. As mentioned in the previous model, the 701 contrasting partitioning behavior of Lu and Hf between bridgmanite and Ca-Pv results in 702 differentiation trends that are highly affected by the relative proportions of these two phases. 703 According to this model, the Komati and Weltevreden mantle domains represent solidification products of a bridgmanite-Ca-Pv cumulate that was derived from ~20% and 12% accumulation of a bridgmanite:Ca-Pv assemblage in the proportions of 93:7 and 80:20, respectively. These proportions and the degrees of magma ocean crystallization are within the range of those envisaged for lower mantle conditions during the Hadean (*e.g.*, Caro et al., 2005). If the modified model of Puchtel et al. (2013) presented here is correct, the mantle plumes that gave rise to the Komati and Weltevreden komatiites must have originated in the lower mantle (**Table 1**).

Boyet et al. (2021) obtained a slightly negative average μ^{142} Nd value of -4.0 ± 4.1 for a set 711 712 of Komati drill core samples. As noted above, that study combined their ¹⁴²Nd results with the ¹⁴³Nd and ¹⁷⁶Hf data of Blichert-Toft et al. (2015) for clinopyroxene separates from the same 713 rocks. In order to explain the negative μ^{142} Nd and ϵ^{143} Nd values and the corresponding 714 715 positive ε^{176} Hf value, Boyet et al. (2021) proposed a four-stage model for the formation of 716 these lavas, whereby the negative μ^{142} Nd value and low Hf/Sm ratios developed during the 717 crystallization of a deep magma ocean soon after Earth accretion. The material that ultimately 718 became the source of the komatiites was a residual liquid produced by 50% crystallization of 719 a bridgmanite/ferropericlase/Ca-Pv mineral assemblage in the proportions 79:16:5, which 720 resulted in decoupling of the Nd and Hf isotope systematics. Thus, the model of Boyet et al. 721 (2021) also supports a lower mantle origin for the plume that gave rise to the Komati 722 komatiites.

723 Metal-silicate fractionation processes operating at the high temperatures and pressures 724 characterizing the first ~60 Ma of Solar System history at the base of an early transient 725 magma ocean, or in a partially molten zone at or near the core-mantle boundary (Labrosse et al., 2007), while ¹⁸²Hf was still extant, have also been argued to be capable of generating 726 coupled ¹⁸²W and ^{186,187}Os heterogeneities in the mantle (Touboul et al., 2012). To explain the 727 enriched ¹⁸²W and ^{186, 187}Os compositions of the Kostomuksha komatiite system, Touboul et 728 729 al. (2012) envisioned a lower mantle magma ocean reservoir where silicate melt could 730 potentially equilibrate with metal that represented either the growing core or the metal passing 731 through the molten region on its way to the core. This model is based on experimental data of 732 substantially reduced metal-silicate partitioning for Re and Pt, relative to Os, at high 733 temperatures and pressures (Righter and Drake, 1997; Cottrell and Walker, 2006; Fortenfant 734 et al., 2006; Yokoyama et al., 2009; Brenan and McDonough, 2009), and on the more 735 siderophile behavior of W under the somewhat more reducing conditions that likely existed in 736 the lower mantle during core formation compared to the present-day upper mantle (Wade and

737 Wood, 2005). The model of Touboul et al. (2012) requires the long-term survival of some 738 portion of the mantle in which HSE and MSE were set by high-temperature and -pressure metal-silicate partitioning under reducing conditions. In order to achieve the ^{186, 187}Os and 739 740 ¹⁸²W isotopic and elemental characteristics of the Kostomuksha komatiite source, Touboul et 741 al. (2012) envisioned a mantle plume tapping this reservoir and mixing material from it with 742 overlying mantle characterized by Os isotopic compositions and HSE concentrations similar 743 to those in estimates of the BSE of Meisel et al. (2001) and Becker et al. (2006), in the 744 proportions of ~1:1 (Fig. 5). If the model of Touboul et al. (2012) is correct, it implies that the 745 plume, which gave rise to the Kostomuksha komatiites, originated in the lowermost mantle.

746 Isotopic heterogeneities involving the HSE-based isotopic systems have also been argued 747 to be related to primordial magma ocean crystallization processes by Puchtel et al. (2014) in their study of the HSE abundances and ^{186, 187}Os compositions of the Komati and Weltevreden 748 749 komatiite systems considered above. These authors used the experimental partitioning data of 750 Borisov and Palme (1997) and Mallmann and O'Neill (2007) indicating that under the highly 751 reducing conditions that may have existed during crystallization of a primordial magma ocean 752 immediately following the main stage of core formation (Frost et al., 2008), solubility of Pt in 753 silicate melts, unlike the other HSE, may have been low and Pt, hence, would have had a 754 strong tendency to form alloys. Fractionation and subsequent heterogeneous dispersal of Pt 755 alloys within the mantle could have resulted in the formation of both Pt-enriched and -756 depleted domains. These would then have evolved with supra- and subchondritic Pt/Os ratios, respectively, which over time would have resulted in the decoupling of the ¹⁸⁶Os and ¹⁸⁷Os 757 758 systems in these domains (Puchtel et al., 2014).

759 A model of silicate-silicate fractionation during primordial mantle magma ocean 760 crystallization, involving early removal of bridgmanite and Ca-Pv, was also proposed by Puchtel et al. (2016a) to explain the coupled ¹⁸²W, ^{142,143}Nd, and ¹⁷⁶Hf isotope systematics of 761 the Schapenburg komatiite source. In their model, the authors recognized that crystal-liquid 762 763 fractionation in a purely silicate system of a global primordial magma ocean would lead to 764 high Hf/W, Sm/Nd, and Lu/Hf in early-formed cumulates and low Hf/W, Sm/Nd, and Lu/Hf in the residual liquid. If these fractionation processes occurred while ¹⁸²Hf and ¹⁴⁶Sm were 765 still extant, the residual silicate liquid would have developed a coupled deficit in ¹⁸²W and 766 ¹⁴²Nd, compared to the ambient mantle, which is what has been observed in the 3.55 Ga 767 768 Schapenburg komatiites. If the model of Puchtel et al. (2016a) is applicable, it places the 769 mantle domain that served as the sources for the Schapenburg mantle plume in the lowermost 770 mantle.

771 *6.2. Late accretion*

772 The term "late veneer" was first introduced by Turekian and Clark (1969) and later 773 elaborated on by Chou et al. (1978; 1983) to explain the excess of HSE abundances in the 774 terrestrial mantle compared to what would otherwise be expected after core formation on the 775 basis of the low pressure and -temperature metal-silicate experimental partitioning data 776 available at the time (e.g., Kimura et al., 1974). Late accretion is defined as the accretion of 777 0.5-1% (Walker, 2009) of Earth's mass of a chondritic component after cessation of core formation. In earlier studies, it was assumed that this chondritic component was added to the 778 779 surface of the Earth in the form of relatively small objects to create a thin layer initially 780 coating the entire planet (thus the term "veneer"). It has also been proposed that a slow 781 downward mixing of this putative thin layer into the mantle was responsible for the apparent 782 trend of increasing HSE abundances in Archean komatiites over time (Maier et al., 2009). 783 Studies by Bottke et al. (2002, 2007), however, have concluded that it is unlikely that late 784 accretion in the form of small objects could have achieved an Earth/Moon late accretionary 785 mass influx ratio of ~1200 required on the basis of the Re-Os isotopic and HSE abundance 786 studies of terrestrial (Meisel et al., 2001; Becker et al., 2006; Fischer-Gödde et al., 2011) and 787 lunar (Walker et al., 2004, 2014; Day et al., 2007; Day and Walker, 2015) samples. Bottke et 788 al. (2010) proposed that late accretion to Earth was dominated by addition of a few large 789 projectiles, with a mean diameter of ~2500 km, and that HSE were added to the Moon 790 through the accretion of much smaller bodies, ~250 km in diameter. Projectiles with a 791 diameter of 2500 km hitting the Earth likely have been differentiated into HSE-rich cores and 792 HSE-stripped silicate mantles (Marchi et al., 2018). As a result, mantle domains with low HSE abundances and positive ¹⁸²W anomalies, on the one hand, and high HSE abundances 793 794 and negative ¹⁸²W anomalies, on the other hand, would have been generated. In the absence of 795 modern-style plate tectonics prior to ~ 2.5 Ga (*e.g.*, Brown and Johnson, 2018), these mantle 796 domains would be expected to survive for extended periods of time before eventually 797 becoming homogenized within the mantle via wholesale mantle convection.

Heterogeneous distribution of HSE in the early Earth's mantle has been recognized from ^{186,187}Os and HSE abundance studies of komatiites (*e.g.*, Maier et al., 2009; Puchtel et al., 2004a,b, 2005, 2009a,b, 2014, 2016a,b, 2018, 2020, 2022). As noted in **Section 5**, available high-precision ¹⁸⁷Os data for komatiitic systems worldwide indicate that most komatiite sources were characterized by initial γ^{187} Os values ranging from only –0.1 to +1.3, implying that these komatiitic systems evolved with long-term Re/Os well within the range of chondritic meteorites (**Fig. 2a**). Only two exceptions have been identified so far, which are

the Schapenburg and Kostomuksha komatiite systems. The more limited ¹⁸⁶Os dataset 805 806 indicates a somewhat greater variability outside of the chondritic range, but, nevertheless, 807 suggests that the mantle sources of the late Archean Pyke Hill, Belingwe, and Vetreny 808 komatiite systems evolved with time-integrated Pt/Os within the chondritic range (Fig. 2b). By contrast, the high initial μ^{186} Os and γ^{187} Os values in the mantle source of the Kostomuksha 809 komatiites require long-term suprachondritic Pt/Os and Re/Os ratios. Originally, these 810 coupled ^{186,187}Os enrichments were interpreted to be the result of core-mantle interaction 811 processes (Puchtel et al., 2005). However, the ¹⁸²W data obtained by Touboul et al. (2012) for 812 these komatiites rendered this interpretation implausible and these authors instead proposed 813 that the combined ^{186,187}Os and ¹⁸²W systematics were the result of metal-silicate fractionation 814 in a primordial magma ocean, as discussed earlier. Additional contrasts are the Komati and 815 816 Weltevreden (Puchtel et al., 2014) and the Lapland (Puchtel et al., 2020) komatiite systems, 817 which evolved with non-chondritic time-integrated Pt/Os ratios and chondritic time-integrated Re/Os ratios, thereby displaying decoupling of the ¹⁸⁶Os and ¹⁸⁷Os systematics. For the 818 819 Komati and Weltevreden komatiite systems, this decoupled behavior has been interpreted to 820 be the result of fractionation of Pt-Fe alloys in a primordial magma ocean during the first 150 821 Ma of Earth history, followed by slow homogenization of the crystallized post-magma ocean 822 mantle domains characterized by fractionated Pt/Os ratios (Puchtel et al., 2014). For the 823 Lapland komatiite system, derivation from a mantle domain characterized by an excess of late 824 accreted, differentiated planetesimal core metal, the so-called "grainy" late accretion (Walker, 2016), were considered to be one of the two plausible models for explaining the decoupled 825 ¹⁸⁶Os and ¹⁸⁷Os systematics (Puchtel et al., 2020). 826

827 On the basis of variable Pt and Ru contents in komatiites of various ages, Maier et al. 828 (2009) argued for a gradual increase in HSE abundances in the presumed deep mantle sources of komatiites between ~3.5 and ~2.9 Ga due to slow downward mixing of a "late veneer" of 829 chondritic materials. However, the ^{186,187}Os and ¹⁸²W isotopic and HSE abundance studies of 830 831 Puchtel et al. (2020; 2022, and references therein) of the best preserved early Archean to early Proterozoic komatiite systems worldwide revealed a somewhat more complex picture. As 832 833 attested to by the plot of the calculated total HSE abundances in the sources of these komatiite 834 systems as percent of those in the estimates of the modern BSE of Becker et al. (2006) versus 835 age (Fig. 3), this trend has been argued unlikely to be the result of simple "slow downward 836 mixing of a late veneer of chondritic materials", although a broad trend of increasing HSE 837 abundances in komatiite sources over time is apparent (e.g., Puchtel et al., 2016a, 2018, 2020, 838 2022). The argument is two-fold. First, as is evident from Fig. 3, the total HSE abundances in the sources of some late Archean komatiite systems were even lower than those in some early Archean komatiite systems. Second, for some of the komatiite systems considered here, *e.g.*, the Schapenburg and Kostomuksha komatiites, the calculated total HSE abundances were shown to be inconsistent with the proportion of late accreted materials estimated to have been added to their sources based on the ^{186,187}Os and ¹⁸²W data (Touboul et al., 2012; Puchtel et al., 2016a).

These observations are illustrated in Fig. 6, where the μ^{182} W values of komatiite systems 845 846 are plotted against the calculated total HSE abundances in their mantle sources normalized to 847 those in the estimates for the modern BSE of Becker et al. (2006). This proportion 848 corresponds to the fraction of the total HSE budget of the modern BSE added during late 849 accretion, assuming an essentially HSE-free BSE immediately following core formation. According to this model, the HSE budgets, taking into account their respective uncertainties, 850 851 of those komatiitic systems that plot within the segment constrained by the blue lines in Fig. 852 6, are consistent with having been established *via* addition of late accreted materials to their 853 mantle source regions. By contrast, the HSE budgets of the komatiite systems that plot outside 854 this segment are presumed to have been affected by additional primordial processes, such as 855 metal-silicate (e.g., the Kostomuksha komatiites: Touboul et al., 2012) or silicate-silicate 856 (e.g., the Schapenburg komatiltes: Puchtel et al., 2016a) fractionation in a primordial magma ocean within the first 60 Ma of Solar System history. Furthermore, the combined ^{187,186}Os-857 ¹⁸²W and HSE abundance data for the Lapland komatiite system (Fig. 6) are consistent with 858 859 involvement of fractionated metal derived from cores of differentiated planetesimals during 860 late accretion processes and formation of Os-W isotope and HSE abundance heterogeneities 861 in the early mantle (Puchtel et al., 2020).

862 It has been argued that the impactors that created the largest impact basins on the Moon, 863 Mars, and presumably also the Earth, were hundreds of km in diameter (Ryder, 2002; Strom 864 et al., 2005; Bottke et al., 2010) and possibly dominated by differentiated planetesimals. Impacts of such bodies would be capable of generating large ¹⁸²W and HSE anomalies in 865 portions of the mantle as a result of such "grainy" late accretion (Walker, 2016; Marchi et al., 866 867 2018; 2020). Cores of differentiated planetesimals would have up to two orders of magnitude higher W abundances, and up to 350 ppm less radiogenic ¹⁸²W/¹⁸⁴W, than the modern BSE 868 (Kleine et al., 2002, 2004; Schoenberg et al., 2002; Yin et al., 2002). Hence, any mantle 869 870 domain to which an excess of this material was added would be ¹⁸²W-depleted and would also be expected to be enriched in HSE, compared to BSE, as is observed for the Lapland 871 872 komatiite system (Puchtel et al., 2020). On the other hand, the mantle domain to which less of a late accretionary HSE component was added, *e.g.*, in the form of the HSE-stripped silicate mantles of differentiated planetesimals, would be ¹⁸²W-enriched, compared to those mantle domains to which a full HSE complement of late accretionary material was added, and would also be depleted in HSE relative to BSE. A typical example of a komatiite system derived from such a mantle domain is the Boston Creek komatiites (Puchtel et al., 2018).

878 Using the data for the komatiite systems, for which late accretion has been argued to be the main cause for the observed ¹⁸²W versus source HSE abundance relationships (Fig. 6), 879 and assuming that the ¹⁸²W excesses were due entirely to a deficit of late accreted materials in 880 their mantle sources, Puchtel et al. (2022) estimated μ^{182} W of the pre-late accretionary BSE to 881 be +17 \pm 7 (**Fig. 6**). This estimate is consistent with the ¹⁸²W excess in the pre-late accretion 882 883 BSE of +18±9 ppm obtained by Kleine and Walker (2017) using an independent approach and to the estimates for the ¹⁸²W composition of the pre-late accretionary Moon of $+25\pm5$ 884 885 ppm (Kruijer et al., 2015; Touboul et al., 2015; Kruijer and Kleine, 2017). These similarities 886 provide further support to the hypothesis that the Earth and the Moon formed from material 887 with identical W isotopic compositions (Kleine and Walker, 2017), in concert with earlier 888 observations of similarity of O, Si, and Ti isotopic compositions between the Earth and the 889 Moon (e.g., Dauphas et al., 2014), as well as additional evidence that the Moon likely formed 890 mainly from terrestrial, rather than impactor, material.

891

6.3. Core-mantle boundary processes

892 Core-mantle exchange is yet another process that may have been responsible for creating large ¹⁸²W and ^{186, 187}Os isotope and HSE abundance heterogeneities in the mantle. Originally, 893 894 the idea was proposed by Walker et al. (1995) based on their studies of the Re, Pt, and Os 895 partitioning behavior in group IIAB magmatic iron meteorites. Walker et al. (1995) noted that 896 during magmatic differentiation, solid metal – liquid metal partition coefficients decrease in 897 the order D_{Os}>D_{Re}>D_{Pt}, so that after crystallization of the inner core, the liquid outer core 898 would end up with supra-chondritic Re/Os and Pt/Os and, given enough time, would develop coupled ^{186, 187}Os enrichments. Walker et al. (1997) observed a coupled radiogenic ^{186, 187}Os 899 900 signature in the Noril'sk sulfide ores, which they interpreted as having originated from 901 interaction of the starting 251 Ma Siberian superplume (e.g., Campbell, 2007) with the liquid 902 outer core at the CMB.

The concept of core-mantle interaction was further elaborated upon by Brandon et al. (1998, 1999, 2003) and Puchtel et al. (2005) in their studies of plume-derived ocean island basalts and komatiites. These authors proposed that the type of correlation between the initial 906 μ^{186} Os and γ^{187} Os values observed in plume-derived Hawaiian picrites and Gorgona Island 907 and Kostomuksha komatiites is unique to mixtures with fractionated liquid outer core metal 908 (Pt/Re ~100), whereas common crustal materials have Pt/Re <30.

909 The major drawback of the original core-mantle interaction model stems from the fact that 910 the onset of inner core crystallization is poorly constrained, ranging from <1.0 to 3.5 Ga given 911 the uncertainties in estimates of present-day heat flux at the CMB which vary from 3 to 10 912 TW (see review in Nakagawa, 2020, and references therein), as well as currently insufficient 913 knowledge of the partition coefficients of radioactive elements (e.g., K) under the CMB 914 conditions. Another issue concerns the fact that the core-mantle interaction model of Walker 915 et al. (1995) and Brandon et al. (1998, 1999) calls for physical addition of ~1% of the outer 916 core liquid metal to the silicate lower mantle. Because the liquid outer core has up to three orders of magnitude higher HSE abundances compared to the mantle, the ^{186, 187}Os 917 918 composition of the resulting hybrid mantle source would be dominated by that of the outer 919 core, and this source would also have had an order of magnitude higher HSE abundances 920 compared to those in the BSE. Most OIB, including Hawaiian picrites, are sulfur-saturated 921 melts and likely to have lost a significant part of their HSE inventory to either immiscible 922 sulfide liquid fractionation or to retaining residual sulfide in their melting source regions (e.g., 923 Shirey and Walker, 1998; Bennett et al., 2000; Jamais et al., 2008; Ireland et al., 2009). Such 924 loss of HSE during magma generation and ascent would not be the case for komatiites, 925 however, which are strongly sulfur-undersaturated magmas and, therefore, reliably record the 926 HSE abundances in their mantle source regions upon emplacement. Yet, those komatiites for which input from the outer core has been advocated to explain their coupled ^{186,187}Os 927 928 enrichments, e.g., Gorgona Island (Brandon et al., 2003) and Kostomuksha (Puchtel et al., 929 2005) komatilites, have calculated HSE abundances in their mantle source regions similar to 930 those in estimates for the modern BSE.

To reconcile the ^{186,187}Os and HSE abundance data for the Kostomuksha komatiites with 931 932 this apparent inconsistency, Puchtel and Humayun (2000) proposed a mechanism of core-933 mantle interaction via isotopic exchange at the core-mantle interface, without any significant 934 mass transfer from the core to the mantle. This idea has been further advanced by Humayun 935 (2011). The mechanism of core-mantle interaction of Humayun (2011) requires the outer core 936 liquid to be trapped in a cumulate pile of Fe-rich non-metallic precipitates (FeO, FeS, Fe₃Si) 937 at the top of the core, then undergo fractional crystallization by precipitating solid metal 938 grains and leaving behind the residual metallic liquid. The residual liquid is then incorporated 939 into the base of the mantle, where it subsequently may be entrained by mantle plumes.

Based on the discovery of negative μ^{182} W values in some modern ocean island basalts, 940 941 Mundl et al. (2017), Rizo et al. (2019), and Mundl-Petermeier et al. (2019, 2020) have also 942 argued for some form of core-mantle exchange/equilibration in order to transfer the inferred negative μ^{182} W value of the core of -220 to plumes rising from the CMB. Although the 943 ^{186,187}Os and ¹⁸²W data obtained on the same samples are still limited, all Hawaiian lavas with 944 negative μ^{182} W (ranging between -7.8 and -20.2) are also characterized by positive μ^{186} Os 945 (ranging between +18.6 and +62.8) and positive γ^{187} Os (ranging between +2.0 and +7.2). 946 Furthermore, Puchtel et al. (2020) obtained a negative μ^{182} W value of -10 for the Lapland 947 komatiites, which is within the range of the μ^{182} W values obtained for Hawaii, and proposed 948 949 that Lapland komatiites could be early Proterozoic equivalents of Hawaiian picrites. A core-950 mantle interaction process, as put forward by Puchtel et al. (2020), would be expected to have 951 decreased the ¹⁸²W value in the Lapland komatiite source relative to the ambient mantle, and 952 also have increased the HSE abundances in this source over ambient mantle levels, which is 953 consistent with the observations (Fig. 6). That study further proposed that if the residual metal 954 in the model of Humayun (2011) had a composition similar to that of fractionated iron 955 meteorites, its entrainment into the mantle plume that gave rise to the Lapland komatiites could explain the decoupled ^{186,187}Os systematics of these komatiites. 956

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6.4. Crustal recycling processes

958 Yet another important mechanism for creating mantle chemical heterogeneities is 959 recycling of Earth's mantle lithosphere, oceanic crust, and sedimentary layers back into the 960 mantle (Hofmann and White, 1982; Shirey and Hanson, 1986; Chase and Patchett, 1988; 961 Campbell and Griffiths, 1992; Hart et al., 1992; Carlson, 1994; Roy-Barman and Allègre, 962 1995; Widom, 1997; Lassiter and Hauri, 1998; Blichert-Toft et al., 1999; Lassiter, 2006). This 963 recycling may occur via subduction – a process by which one tectonic plate moves under 964 another and sinks into the mantle as the plates converge; subduction is the driving force 965 behind modern plate tectonics. The subducted material may age in the mantle for hundreds of millions of years until entrained in mantle plumes. Although there is little agreement on the 966 967 timing of the onset or even existence of modern-style plate tectonics in the Hadean and 968 Archean (e.g., Brown, 2007; Shirey et al., 2008; Gerya, 2014; Sizova et al., 2015; Johnson et 969 al., 2017; Bédard, 2018), several lines of geochemical evidence put forward by Shirey et al. 970 (2008) require that some version of modern-style plate tectonics already operated as far back 971 in Earth history as 3.5 billion years ago, followed by a transition from the stagnant-lid to 972 mobile-lid plate tectonic regimes sometime in the late Archean (O'Neill and Debaille, 2014).

973 Crustal recycling, e.g., in the form of subduction of pelagic sediments into the mantle, 974 may also cause decoupling of Nd and Hf isotope systematics of lavas from the Nd-Hf mantle 975 array, as has been argued to be the case for a suite of drill core Komati komatiite samples 976 (Blichert-Toft et al., 2015). This is due to the fact that the Lu/Hf ratio is strongly fractionated 977 relative to Sm/Nd in the Earth's sedimentary system (Patchett et al., 1984). This is caused by 978 high resistance to chemical weathering of the mineral zircon, which contains most of the Hf 979 budget of continental crustal rocks. During weathering of continental crustal rocks, they break 980 down into zircon-bearing sandy sediments with very low Lu/Hf and fine-grained clay material 981 with up to three times the chondritic Lu/Hf ratio. During sediment transport via turbidity 982 currents, sandy sediments stay on or near the continent, while the pelagic material is carried 983 out onto the ocean floor, where it may eventually be subducted into the mantle.

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7. The Nd-Hf-Os isotope paradox and its implications for early Earth history

As noted above, there is evidence for decoupling between the radiogenic lithophile ¹⁴³Nd 985 and ¹⁷⁶Hf systems in certain komatiltes mantle sources, but there is also apparent decoupling 986 between these lithophile isotope systems and the highly siderophile ¹⁸⁷Os system in some 987 988 komatiite mantle sources. The majority of the Archean and Proterozoic komatiite systems are 989 characterized by variable, and generally strong, long-term depletions in more incompatible 990 lithophile trace elements relative to less incompatible elements, e.g., Nd versus Sm and Hf 991 versus Lu (Fig. 1), yet they retain chondritic time-integrated Re/Os (Fig. 2). These komatiite 992 mantle sources must have experienced relatively early melt depletion events that fractionated 993 the Sm/Nd and Lu/Hf ratios to suprachondritic values high enough to bring the ¹⁴³Nd and ¹⁷⁶Hf compositions to the variably positive ε^{143} Nd and ε^{176} Hf values. Since Re is generally 994 995 moderately to highly incompatible during mantle melting, whereas Os is moderately to highly 996 compatible (e.g., Barnes et al., 1985; Mallmann and O'Neill, 2007), these melt extraction 997 events should have also decreased the Re/Os ratio in the komatiite-basalt mantle sources. This would have resulted in retardation of ¹⁸⁷Os ingrowth and, thus, evolution of the sources to 998 variably negative γ^{187} Os values. However, such isotopic relationships are not observed in the 999 1000 global komatiite record.

Puchtel et al. (2022) named this phenomenon the Nd-Hf-Os isotope paradox, to which Puchtel et al. (2020, 2022) offered a solution by developing a model whereby the komatiitebasalt mantle sources underwent early (~100 Ma into Solar System history), low-degree (F =1004 1.5-2.0%) partial melting and melt extraction events that would have sufficiently fractionated the Sm/Nd and Lu/Hf ratios in the mantle sources, but would have had little effect on the 1006 Re/Os ratio due to the significantly lower incompatibility of Re compared to Nd and Hf1007 during low-degrees of partial mantle melting.

Because this depleted reservoir is apparently persistent throughout the Archean and is 1008 1009 globally distributed, it requires early formation and subsequent complete isolation of a 1010 complementary basaltic crust enriched in incompatible lithophile trace elements. Calculations 1011 by Puchtel et al. (2022) indicate that such crust formed as a result of 1.5-2.0% batch melting 1012 of a BSE-like mantle would contain 31-27 ppm Nd, 7.1-6.5 ppm Sm, 0.62-0.59 ppm Lu, and 5.2-4.7 ppm Hf, and would have 147 Sm/ 144 Nd = 0.140-0.145 and 176 Lu/ 177 Hf = 0.0166-0.0174 1013 (compared to 0.1967 and 0.0336 in the BSE, respectively). Based on these estimates of Sm 1014 and Nd concentrations in the early crust, the mass of this reservoir must have represented 1015 1016 \leq 2.0% of the mass of the BSE, which is consistent with this reservoir being the size of the D" 1017 layer. The existence of such crust has been previously advocated for by Galer and Goldstein 1018 (1991). Alternatively, this earliest mafic crust could have served as the source of the oldest 1019 tonalite-trondhjemite-granodiorite (TTG) complexes (Carlson et al., 2019), such as those from 1020 the Nuvvuagittuq Greenstone Belt in Québec (O'Neil et al., 2008, 2016) or the Vodla Block 1021 TTG complex in the Fennoscandian Shield (Puchtel et al., 2016b).

1022 The residence time of this early mafic crust in the mantle is difficult to constrain. The 1023 available komatiite-basalt record would require its isolation through at least the late Archean, 1024 *i.e.*, when the ¹⁴²Nd anomalies were still present in the mantle (Debaille et al., 2013). 1025 However, there is no evidence that this crust was stored in the mantle until the present day, 1026 based on the generally chondritic ¹³⁸Ce, ^{142,143}Nd, and ¹⁷⁶Hf composition estimates for the 1027 BSE (Jackson and Carlson, 2012; Horan et al., 2018; Willig and Stracke, 2019; Willig et al., 1028 2020; Hyung and Jacobsen, 2020).

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8. Komatiite constraints on early mixing rates of the terrestrial mantle

1030 It has been proposed that the mixing rates of the mantle, or the average time of mantle 1031 homogenization, can be evaluated through studies of short-lived radiogenic isotope 1032 systematics and HSE abundances in mantle-derived rocks (e.g., Jacobsen and Yu, 2015; 1033 Carlson et al., 2019; Hyung and Jacobsen, 2020; Tusch et al., 2021; Puchtel et al., 2022). Isotopic variations due to the decay of the now extinct ¹⁴⁶Sm and ¹⁸²Hf nuclides recorded 1034 1035 processes in the Earth that fractionated Sm/Nd and Hf/W ratios between different terrestrial 1036 reservoirs at the times when these parent nuclides were still alive. When these events occurred 1037 during the first ~600 and ~60 Ma of Earth history, respectively, the differentiation processes resulted in formation of ¹⁴²Nd and ¹⁸²W anomalies in these terrestrial reservoirs. Due to the 1038

1039 protracted, violent accretion history of the Earth accompanied by vigorous convective mixing 1040 during the Hadean and Archean eons, these reservoirs and, thus, the isotopic anomalies they 1041 hosted, were gradually erased from the geological record. Hence, tracing the variations in the 1042 magnitude of these isotopic anomalies over geological time can be used to determine the 1043 mixing rates of the terrestrial mantle (*e.g.*, Jacobsen and Yu, 2015; Hyung and Jacobsen, 1044 2020).

1045 Early mantle convection models (e.g., Blichert-Toft and Albarède, 1994; Coltice and 1046 Schmalzl, 2006) concluded that the mixing times in the hot Hadean and Archean mantle were 1047 10 times faster than today, and that any large-scale primordial chemical and isotopic heterogeneities in the mantle would have been erased within ~100 Ma of their creation. 1048 However, during the past two decades, numerous ¹⁴²Nd and ¹⁸²W anomalies have been 1049 discovered in the terrestrial mantle-derived rock record (Fig. 7). The largest ¹⁴²Nd anomalies 1050 $(\mu^{142}Nd up to +20 ppm and down to -20 ppm)$ have been reported for Eo- to Paleoarchean 1051 1052 supracrustal rocks from the Isua Greenstone Belt, Greenland (Boyet et al., 2003; Caro et al., 1053 2006; Bennett et al., 2007; Rizo et al., 2011; 2012; 2013; O'Neil et al., 2016), the 1054 Nuvvuagittuq Greenstone Belt, Québec (O'Neil et al., 2008; 2012; Roth et al., 2013), the 1055 Acasta Gneiss Complex, Canada (Roth et al., 2014; Reimink et al., 2018), and the Barberton 1056 Greenstone Belt, Kaapvaal Craton, South Africa (Puchtel et al., 2016a; Boyet et al., 2021). 1057 These isotopic anomalies must have been created before ~4.0 Ga by silicate-silicate 1058 differentiation and, thus, have survived for >1.0 Ga, implying much slower mixing times of the mantle for ¹⁴²Nd than was predicted by the early mantle convection models. Following 1059 1060 these discoveries, more recent mantle convection models argue for complete homogenization 1061 of the mantle by the end of the Archean (e.g., Rosas and Korenaga, 2018; Korenaga, 2021). Only few terrestrial samples with ages between 3.0 and 2.7 Ga have μ^{142} Nd values deviating 1062 from the terrestrial standard by more than ± 5 ppm, the only known exception being 2.72 Ga 1063 tholeiites from the Abitibi Greenstone Belt, Canada (Debaille et al., 2013). Although there is a 1064 significant gap in the mafic-ultramafic rock record for ¹⁴²Nd data between ~2.7 Ga and the 1065 present day, it indeed appears that ¹⁴²Nd anomalies largely disappeared by the end of the 1066 Archean (Fig. 7). As a result, on the basis of the now much larger 142 Nd record and using a 1067 mathematical mantle mixing box model, Hyung and Jacobsen (2020) concluded that the 1068 1069 relationships in Fig. 7 are consistent with mantle mixing rates of ~ 400 Ma that operated since 1070 the early Hadean. These authors further argued that such fast mixing rates are consistent with 1071 Earth's thermal and chemical evolution having been largely regulated by plate tectonics for 1072 most of its history.

1073 Nonetheless, recent studies have shown that some modern plume-derived OIB (**Fig. 7**) 1074 have 142 Nd compositions resolved, albeit marginally, from the terrestrial standard (*e.g.*, Horan 1075 et al., 2018; Peters et al., 2018), indicating that some 142 Nd isotopic signatures dating back to 1076 Earth's earliest differentiation events may still be preserved in deepest parts of the modern 1077 mantle.

1078 In contrast to the earliest ¹⁴²Nd record, mostly positive ¹⁸²W anomalies, of up to $\sim +20$ 1079 ppm, have been reported for mafic-ultramafic rocks ranging in age from 4.0 to 2.7 Ga (Fig. 7b). The only exceptions are komatilites from the Schapenburg system (Puchtel et al., 2016a), 1080 which have a negative 182 W anomaly of -8 ppm. On the other hand, some recent and modern 1081 1082 OIB, such as Hawaii, Samoa, Iceland, Galapagos, Heard, Pitcairn (Mundl et al., 2017; Mundl-Petermeier et al., 2019, 2020), and Kerguelen and Reunion (Rizo et al., 2019) are 1083 characterized by negative 182 W anomalies down to -25 ppm, although the volumetrically 1084 dominant signature in OIB is most likely ${}^{182}W = 0$. The very limited data available (Puchtel et 1085 al., 2016b, 2020) provide a hint that there may be a transition at ~2.5 Ga from mostly positive 1086 μ^{182} W-dominated to no-anomaly μ^{182} W-dominated mantle (Fig. 7b). With the relatively 1087 voung 2.05 Ga Lapland komatiites already registering a negative 182 W anomaly of -10 ppm 1088 (Puchtel et al., 2020), and some OIB sources registering negative ¹⁸²W anomalies down to -1089 1090 25 ppm, this transition may indicate the combined effects of the onset of modern-style plate 1091 tectonics and core-mantle interaction taking over as the main driving forces controlling the W 1092 isoltopic composition of the deep plume mantle sources (e.g., Rizo et al., 2019; Carlson et al., 2019). 1093

The different behavior of the two short-lived isotopic systems in the mantle may be 1094 possible to reconcile when the mechanisms behind the origin of ¹⁴²Nd and ¹⁸²W anomalies are 1095 considered. Because both Sm and Nd are uniformly strongly lithophile elements under the 1096 redox conditions applicable to Earth's formation, ¹⁴²Nd anomalies are solely the result of 1097 1098 primordial silicate-silicate planetary differentiation (e.g., Boyet and Carlson, 2005). No new ¹⁴²Nd anomalies could have been created in the BSE after ~4.0 Ga, when ¹⁴⁶Sm became 1099 extinct. In contrast to the ¹⁴⁶Sm-¹⁴²Nd system, Hf is strongly lithophile, while W is 1100 1101 moderately siderophile. Hafnium, therefore, can be fractionated from W during metal-silicate 1102 differentiation, such as that which occurred during planetary core segregation. During silicate-1103 silicate differentiation, W is more incompatible than Hf, which can result in fractionation of 1104 Hf from W in purely silicate systems. Both processes are considered to be capable of creating ¹⁸²W anomalies within the first 60 Ma of Earth's history, while ¹⁸²Hf was still extant. The 1105 additional processes that are thought to be capable of introducing ¹⁸²W heterogeneities to the 1106

1107 mantle and to which the ¹⁴²Nd system was insensitive, were late accretion and core-mantle 1108 interaction, as discussed in **Section 6**. As a result of the combination of all these factors in 1109 variable, and generally unconstrained, proportions, the use of the ¹⁸²Hf-¹⁸²W isotopic system 1110 for tracing mixing times of the mantle appears to be of lesser value than the ¹⁴⁶Sm-¹⁴²Nd 1111 systematics.

1112 Mantle mixing rates in terms of HSE have been studied *via* tracing the changes in HSE 1113 abundances in the mantle over the span of geological time (e.g., Maier et al., 2009; Puchtel et al., 2018, 2020, 2022). Because the lunar highland crust, which was formed at ~4.4 Ga (e.g., 1114 1115 Borg et al., 2014), is only negligibly contaminated by meteoritic material (e.g., Day et al., 1116 2010), late accretion must have been largely complete prior to 4.4 Ga, *i.e.*, within the first 1117 ~150 Ma of the Solar System history. Puchtel et al. (2022) used the estimated HSE 1118 abundances in the sources of the Archean and Proterozoic komatiite systems plotted in Fig. 3 1119 as a function of their ages and *Isoplot* regression analysis to calculate the average time by which the late accreted materials would have been completely homogenized within the 1120 1121 mantle, and found 2.5±0.2 Ga. These data require that survival times of the late accreted 1122 planetesimals within the mantle, before complete homogenization, were on average 1.9±0.2 1123 Ga (*i.e.*, the time interval between 4.4 and 2.5 Ga), thus constraining the average mixing rates 1124 of the terrestrial mantle in terms of siderophile element abundances in the Hadean and the 1125 Archean. This period in time coincides with the timing of onset of modern-style plate tectonics on Earth, as argued by some (Brown, 2007; Brown and Johnson, 2018), and with the 1126 near-complete disappearance of all ¹⁴²Nd and positive ¹⁸²W anomalies in the mantle (Carlson 1127 1128 et al., 2019; Reimink et al., 2020, Tusch et al., 2021).

It is possible that some inefficient mixing could be due to early Earth tectonic regimes differing from those of modern-style plate tectonics (O'Neill and Debaille, 2014). A stagnantlid, or episodic, subduction regime in the Hadean is consistent with the persistence of shortlived isotopic anomalies and compositional heterogeneities, although more recent modeling indicates that such mantle heterogeneities can survive for billions of years, even under a mobile-lid tectonic regime (Foley and Rizo, 2017).

1135 9. The diverse nature of komatiite sources reflects on the complexity of the terrestrial1136 mantle

As is evident from the chemical and isotopic data summarized in **Tables 1** and **2** and discussed throughout this review, komatiite mantle sources were characterized by extreme diversity throughout the Archean and early Proterozoic in terms of both trace- and HSE abundances and radiogenic isotope systematics. Early Archean komatiites were likely derived from lower mantle domains that were created very early in Earth history as a result of deepseated magma ocean crystallization processes. These mantle domains were characterized by highly variable, and often strong, long-term depletions in the highly incompatible lithophile trace elements and generally large deficits in the late accreted component, as attested to by their HSE abundance and ¹⁸²W systematics.

The late Archean and Proterozoic komatiites show evidence in their mantle sources for limited variability of long-term depletions in highly incompatible lithophile trace elements as a result of prior melt extractions. With the exception of the Kostomuksha and possibly Lapland systems, these komatiites were derived from mantle plumes that originated in the upper mantle, most likely in the MTZ. These komatiite sources were characterized by smaller deficits to excesses in the late accreted component, as testified to by their HSE abundance and ¹⁸²W systematics.

Finally, our compliation of the geochemical data relevant to the mantle sources of Archean and early Proterozoic komatiites reveals that none of the projected sources were similar in composition to estimates for the BSE. The komatiite systems considered here, which span wide geographical and temporal distributions, further demonstrate that no single petrogenetic model can account for all the diverse chemical compositions of komatiites.

1158 **10. Conclusions**

Komatiites, due to their unique properties and, with a few exceptions, very old ages, are among the best samples to constrain the evolution of the chemical composition of the early Earth's mantle. Isotopic and elemental signatures created in early-formed silicate reservoirs *via* radioactive decay of short- and long-lived nuclides, including ^{146,147}Sm, ¹⁷⁶Lu, ¹⁸²Hf, ¹⁸⁷Re, and ¹⁹⁰Pt, have been sampled by komatiitic magmas and preserved in the geological rock record. These signatures have been used here to constrain the nature and timing of formation of these, by now largely vanished, mantle reservoirs.

The komatiites considered in the present review are interpreted to have been derived from anhydrous melting in mantle plumes that were initiated in both the lower and upper mantle, although, in contrast to modern plumes, all evidence for the depths of ancient mantle plume initiation is circumstantial in nature.

1170 The data for the existing 3.6-2.0 Ga komatiite record provide robust evidence for the 1171 presence of remarkably ancient isotopic and chemical heterogeneities in the mantle at that 1172 time interval in Earth history. These heterogeneities have been shown here to reflect the

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1173 combined effects of (i) the co-existence of diverse post-magma ocean silicate domains 1174 characterized by variably-fractionated lithophile and siderophile element abundances; (ii) the 1175 presence of distinct reservoirs representing mantles and cores of large, differentiated 1176 planetesimals delivered to Earth during late accretion; and (iii) isotopic exchange across the 1177 core-mantle boundary.

1178 The near-complete disappearance of resolvable ¹⁴²Nd anomalies and of decoupled ¹⁴³Nd-1179 ¹⁷⁶Hf isotopic signatures in the mafic-ultramafic rock record by ~2.5 Ga indicates that the earliest silicate reservoirs, formed as a result of primordial magma ocean crystallization, had 1180 been largely destroyed by that time through vigorous, convective mantle mixing, implying 1181 1182 mixing rates of the mantle on the order of ~ 1.5 Ga. The shift from mostly positive ¹⁸²W offsets in the pre-2.5 Ga mantle sources of mafic-ultramafic lavas to no ¹⁸²W offsets at ~2.5 1183 Ga, as well as disapperance of HSE abundance anomalies at ~2.5 Ga, may indicate the 1184 1185 combined effects of the onset of modern-style plate tectonics and core-mantle interaction that took over as the main driving force of creating ¹⁸²W isotopic heterogeneities in the mantle. 1186

1187 This review further emphasizes the great compositional complexity of early Archean to 1188 early Proterozoic komatiite mantle sources in terms of both lithophile and siderophile element 1189 abundances and isotope systematics; apparently, none of these sources were similar in 1190 composition to that of the BSE. Obviously, no single petrogenetic model can fully account for 1191 the chemical and isotopic diversity of komatiites.

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Figure captions

2074 Fig. 1. Variations of model-specific (see the references below for the specific models used), time-integrated Sm/Nd and Lu/Hf ratios in the mantle sources of Archean and 2075 Proterozoic komatiite-basalt systems studied to date. The individual models assume a 2076 2077 minimum-degree fractionation of Sm/Nd and Lu/Hf in the particular mantle domains from either the chondritic values or values defined by the combined ^{142,143}Nd/¹⁴⁴Nd systematics 2078 (where available) to those required to bring the ϵ^{143} Nd and ϵ^{176} Hf in the mantle sources to the 2079 initial ϵ^{143} Nd and ϵ^{176} Hf by the times of the respective komatiite formation. The solid line 2080 connects Sm/Nd and Lu/Hf ratios inferred for the chondritic uniform reservoir (CHUR) and 2081 2082 modern depleted mantle (DMM). Data are from: Ottawa – Blichert-Toft and Arndt (1999); 2083 2.05 Ga Lapland – Puchtel et al. (2020); 2.41 Ga Vetreny – Puchtel et al. (2016b); 2.72 Ga 2084 Pyke Hill and Alexo – Dupre et al. (1984), Walker et al. (1988), Blichert-Toft and Arndt (1999); 2.72 Ga Boston Creek – Puchtel et al. (2018); 2.82 Ga Kostomuksha – Puchtel et al. 2085 (1998), Blichert-Toft and Puchtel (2010); 3.48 Ga Komati and 3.26 Ga Weltevreden -2086 2087 Puchtel et al. (2013); 3.32 Ga Commondale - Wilson and Carlson (1989), Hoffmann and 2088 Wilson (2017); 3.55 Ga Schapenburg – Puchtel et al. (2009a, 2016a); 3.53 Ga Coonterunah, 2089 3.34 Ga Kelly, and 3.18 Ga Ruth Well and Regal – Puchtel et al. (2022). The CHUR and 2090 DMM parameters are from Jacobsen and Wasserburg (1980), Hamilton et al. (1983), Goldstein et al. (1984), Vervoort and Blichert-Toft (1999), and Bouvier et al. (2008). All 2091 uncertainties are 2SD of the mean. 2092

Fig. 2. (a) Initial ¹⁸⁷Os/¹⁸⁸Os expressed as γ^{187} Os and (b) initial ¹⁸⁶Os/¹⁸⁸Os expressed as 2093 μ^{186} Os of Archean komatiite systems studied to date, plotted as a function of their age. The 2094 blue bars for the modern BSE estimates represent the 2SD of the mean from Meisel et al. 2095 2096 (2001) and Brandon et al. (2006) for (a) and (b), respectively. Data are from: 2.05 Ga Lapland 2097 - Puchtel et al. (2020); 2.41 Ga Vetreny - Puchtel et al. (2016b); 2.69 Ga Belingwe and 2.72 2098 Ga Pyke Hill – Puchtel et al. (2009a); 2.72 Ga Boston Creek – Puchtel et al. (2018); 2.82 Ga Kostomuksha - Puchtel et al. (2005); 2.88 Ga Volotsk - Puchtel et al. (2007); 3.48 Ga 2099 Komati and 3.26 Ga Weltevreden - Puchtel et al. (2014); 3.55 Ga Schapenburg - Puchtel et 2100 2101 al. (2009b, 2016a); 3.34 Ga Kelly and 3.18 Ga Ruth Well and Regal – Puchtel et al. (2022). The data for chondritic meteorites are compiled from Walker et al. (2002), Horan et al. 2102 (2003), Brandon et al. (2005; 2006), and Fischer-Gödde et al. (2010) and are plotted as an 2103 envelope enclosed between the slanting purple lines and corresponding to the entire range of 2104 calculated modern γ^{187} Os and μ^{186} Os values projected back to the Solar System initial 2105 ¹⁸⁷Os/¹⁸⁸Os and ¹⁸⁶Os/¹⁸⁸Os ratios. All uncertainties are 2SD of the mean. 2106

Fig. 3. Calculated total HSE abundances in the sources of Archean and Paleoproterozoic komatiite systems plotted as *percent* of those in estimates of the modern BSE of Becker et al. (2006). The blue dashed lines correspond to the range for the BSE estimates from Becker et al. (2006). The dark-red arrow represents the regression line through the data. The dark-red vertical arrows represent the projections to the x-axis from the intersections of the regression line with the range for the BSE estimate, thus, illustrating the uncertainties on the average time of homogenization of late accreted materials within the mantle (2.5 ± 0.2 Ga). Data are 2114 from: 2.05 Ga Lapland – Puchtel et al. (2020); 2.41 Ga Vetreny Belt – Puchtel et al. (2016b); 2.69 Ga Belingwe – Puchtel et al. (2009a); 2.72 Ga Pyke Hill and Alexo – Puchtel et al. 2115 (2004b, 2009a); 2.72 Ga Boston Creek - Puchtel et al. (2018); 2.82 Ga Kostomuksha -2116 2117 Puchtel and Humayun (2005); 2.88 Ga Volotsk – Puchtel et al. (2007); 3.26 Ga Weltevreden 2118 and 3.48 Ga Komati – Puchtel et al. (2014); 3.55 Ga Schapenburg – Puchtel et al. (2009b, 2016a); 3.53 Ga Coonterunah, 3.34 Ga Kelly, and 3.18 Ga Ruth Well and Regal – Puchtel et 2119 2120 al. (2022). Uncertainties are 2SD. The data reveal a broad trend of increasing HSE 2121 abundances in komatiite mantle sources over geological time. All uncertainties are 2SD of the 2122 mean. See text for additional details and the *Electronic Supplement* for the algorithm used to 2123 estimate the HSE abundances in the sources of the komatiite systems.

Fig. 4. Evolution of the time-integrated ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁷⁶Lu/¹⁷⁷Hf in the calculated 2124 sources of the Komati and Weltevreden komatiite systems formed during crystallization of a 2125 2126 primordial magma ocean. The differentiation trends (shown by tick marks in percentage) 2127 depend on the relative proportions of the fractionating lower mantle mineral phases, *i.e.*, 2128 bridgmanite and Ca-Pv. The composition of the magma ocean prior to onset of crystallization 2129 is represented by that of the CHUR. The upper part of the panel represents aggregate 2130 compositions of magma ocean cumulates, while the lower part of the panel represents 2131 compositions of residual liquids after removal of the respective amounts of cumulate 2132 bridgmanite and Ca-Pv. All uncertainties are 2SD of the mean. See text for additional details

2133 of the model and the *Electronic Supplement* for the algorithm used in the calculations.

Fig. 5. Values of μ^{182} W versus (A) μ^{186} Os and (B) γ^{187} Os in the Kostomuksha komatiites 2134 illustrating mixing between the modern BSE and a mantle reservoir preserved from an early 2135 magma ocean crystallization event. The Re/Os, Pt/Os, and Hf/W ratios were established by 2136 high-pressure and -temperature metal-silicate equilibrium, resulting in ^{187,186}Os and ¹⁸²W 2137 enrichments at 2.82 Ga (Touboul et al., 2012). The coupled ^{186,187}Os and ¹⁸²W excesses in the 2138 2139 source of the Kostomuksha komatiites (yellow circle) are explained by a ~50% contribution 2140 from the isotopically enriched reservoir to the modern BSE. All uncertainties are 2SD of the 2141 mean. See text for additional details.

Fig. 6. μ^{182} W (ppm) versus calculated total HSE abundances in the sources of Archean 2142 and Paleoproterozoic komatiite-basalt systems studied to date relative to those in the estimates 2143 2144 of the present-day BSE of Becker et al. (2006). This proportion corresponds to the fraction of 2145 the total HSE budget of the BSE added during late accretion assuming an HSE-free terrestrial mantle prior to late accretion. The average μ^{182} W value for the Moon of +25±5 is from 2146 Kruijer et al. (2015) and Touboul et al. (2015). The μ^{182} W of the BSE prior to late accretion is 2147 constrained via Isoplot regression analysis of the $\mu^{182}W$ and HSE compositions of all 2148 komatiite-basalt systems except for those of Kostomuksha and Schapenburg and the present-2149 2150 day BSE to be $+17\pm7$. The W isotopic data and estimates of the HSE contents for the 2151 komatiite systems are from Puchtel and Humayun (2005), Touboul et al. (2012), and Puchtel et al. (2014; 2009b, 2016a,b; 2018; 2020; 2022). All uncertainties are 2SD of the mean. See 2152 2153 text for additional details.

Fig. 7. ¹⁸²W/¹⁸⁴W and ¹⁴²Nd/¹⁴⁴Nd data for the terrestrial mafic-ultramafic rock record 2154 obtained to date. The colored bands represent the 2SD uncertainty on the mean $\mu^{182}W$ and 2155 μ^{142} Nd values for each locality. The red arrows illustrate the change in direction and 2156 magnitude of the ¹⁸²W and ¹⁴²Nd anomalies. Note the relative scarcity of data for the period 2157 between ~2.7 Ga and present day. Data are from: 3.96 Ga Acasta - Roth et al. (2014), 2158 2159 Willbold et al. (2015), Reimink et al. (2018); 3.85 Ga Isua – Willbold et al. (2011), Rizo et al. (2013, 2016), Dale et al. (2017), Tusch et al. (2019), Caro et al. (2006), Bennett et al. (2007), 2160 2161 Saji et al. (2018); 3.80 Ga Nuvvuagittuq – O'Neil et al. (2012), Touboul et al. (2014); 3.78 Ga 2162 Saglek – Liu et al. (2016), Morino et al. (2017); 3.75 Ga Ukalig – Caro et al. (2017); 3.72 Ga Isua - Rizo et al. (2011, 2016), Tusch et al. (2019), O'Neil et al. (2016); 3.50 Ga Ameralik -2163 Saji et al. (2018); 3.37 Ga Ameralik - Rizo et al. (2012, 2016), Tusch et al. (2019); 3.55 Ga 2164 Schapenburg – Puchtel et al. (2016a); 3.53 Ga Coonterunah – Tusch et al. (2021), Puchtel et 2165 2166 al. (2021); 3.48 Ga Komati and 3.26 Weltevreden – Touboul et al. (2012), Caro et al. (2006), Puchtel et al. (2013), Schneider et al. (2018); 3.45 Ga Warrawoona – Archer et al. (2019), 2167 2168 Rizo et al. (2019), Murphy et al. (2021); 3.35 Ga Kelly – Puchtel et al. (2022); 3.18 Ga Ruth 2169 Well and Regal – Archer et al. (2019), Tusch et al. (2021), Puchtel et al. (2022); 3.14 Ga Banasandra – Maya et al. (2017); 2.82 Ga Kostomuksha and 2.69 Ga Belingwe – Touboul et 2170 2171 al. (2012), Boyet and Carlson (2006); 2.72 Ga Boston Creek - Puchtel et al. (2018); 2.72 Ga Theo's Flow – Debaille et al. (2013); 2.41 Ga Vetreny Belt – Puchtel et al. (2016b); 2.05 Ga 2172 Lapland - Puchtel et al. (2020); modern OIB - Caro et al. (2006), Andreasen et al. (2008), 2173 2174 Murphy et al. (2010), Touboul et al. (2012), Mundl et al. (2017), Mundl-Petermeier et al. 2175 (2019, 2020), Rizo et al. (2019), Horan et al. (2018), Saji et al. (2018), Peters et al. (2018), 2176 Hyung and Jacobsen (2020); modern MORB – Caro et al. (2006), Mundl et al. (2017), Hyung 2177 and Jacobsen (2020).

Komatiite-basalt system	Age, Ga	MgO _{liq}	T _{liq} °C	T _p ° C	Dmelt init, km	Dplume init, km	Al ₂ O ₃ /TiO ₂	(La/Sm) _N	(Gd/Yb) _N	Nb/Nb*	W/Th
Kaapvaal Craton, South Afr	ica									I I	
Schapenburg	3.55	29.1±2.8	1582	1811	443	LM	10.0±0.8	0.93±0.04	1.57±0.10	1.2±0.1	9.7±5.5
Komati	3.48	29.9±0.5	1591	1822	494	LM	10.1±0.7	0.97±0.24	1.39±0.05	1.1±0.2	0.25±0.1
Weltevreden	3.26	31.4±0.9	1606	1841	605	LM	29.2±1.1	0.68 ± 0.06	0.84±0.03	1.4±0.1	1.5±1.4
Pilbara Craton, Western Au	stralia	1		1			•		1	1	
Coonterunah	3.53	23.0±0.3	1498	1705	203	MTZ	23.8±1.7	0.80 ± 0.04	0.96±0.04	1.08±0.02	0.91±0.6
Kelly	3.34	27.4±1.0	1561	1786	351	MTZ	22.1±1.7	0.65±0.02	0.91±0.04	1.03±0.04	0.81±0.6
Ruth Well	3.18	29.3±0.7	1584	1814	455	MTZ	11.0±0.5	0.55±0.09†	1.31±0.04†	1.0±0.1*	3.4±2.3
Ruth Weh	5.10							0.77±0.09	1.32±0.04	0.51±0.05	
Regal	3.18	29.3±0.7	1584	1814	455	MTZ	11.9±0.8	0.61±0.11*	1.28±0.12†	1.0±0.1*	3.5±2.8
Regal	5.10							0.83±0.11	1.31±0.12	0.55±0.09	
Fennoscandian Shield, nort	hern Europe					•			•		
Kostomuksha	2.82	27.6±1.0	1564	1789	361	LM	17.2±1.2	0.48±0.13	1.16±0.02	1.2±0.1	0.84±0.3
Vetreny	2.41	27.0±1.0	1556	1779	333	MTZ	19.7±0.5	0.41±0.10†	1.2±0.1†	1.0±0.1†	0.05±0.02
veneny								2.2±0.1	1.3±0.1	0.29±0.03	
Loulond	2.05	25.2±0.5	1532	1748	264	LM	13.9±0.4	0.14±0.02†	1.48±0.02†	1.0±0.1†	0.16±0.06
Lapland	2.03							0.26±0.01	1.49±0.02	0.58±0.01	
Superior Craton, Canada		1		1			•	•	•	1	
Boston Creek	2.72					MTZ	5.1±0.8	1.9±0.3	2.0±0.2	1.2±0.2	0.9±1.4
Pyke Hill-Alexo	2.72	28.2±0.5	1571	1789	392	MTZ	20.4±0.6	0.53±0.16	0.98±0.03	1.0±0.1	0.15±0.0
Rhodesian Craton, Zimbabw	ve	1		1			1		1	·I	
Belingwe	2.69	25.6±0.5	1537	1755	277	MTZ	19.8±0.6	0.68±0.02	1.02 ± 0.04	0.97±0.05	0.20±0.0

Table 1. Summary of ages, thermodynamic parameters, and chemical features of the komatiite systems considered in this review.

2180 †Values corrected for AFC. The italicized values are those measured in the emplaced lavas, before correction for AFC. All uncertainties are 2SD of the mean. MgO_{liq} – the
 2181 MgO content of the original emplaced komatiite magma for each komatiite system compiled from Nicklas et al. (2018, 2019) and Puchtel et al. (1998, 2016a, 2020, 2022).

2182 $\mathbf{T}_{liq}^{\circ}\mathbf{C}, \mathbf{T}_{p}^{\circ}\mathbf{C}$ – liquidus temperatures of the emplaced komatiite magmas and mantle potential temperatures for the studied komatiite systems. $\mathbf{D}_{melt init}$ – depths of melting

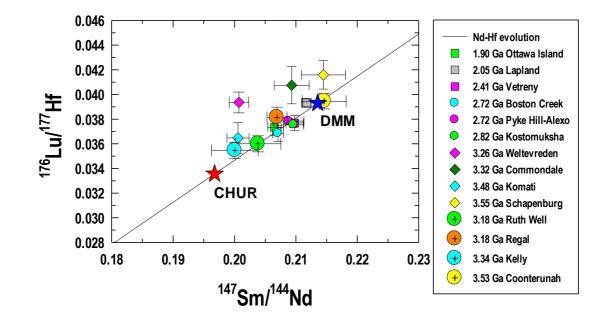
2183 initiation. D_{plume init} – inferred depths of plume initiation for the studied komatiite systems. Nb/Nb* = Nb_N/ $\sqrt{(Th_N \times La_N)}$, where N are the BSE normalizing values from

Hofmann (1988). For sources of the data see Table 2. See text and the *Electronic Supplement* for details.

Komatiite-basalt system	$\epsilon^{143}Nd(T)$	$\epsilon^{176} Hf(T)$	μ^{142} Nd	$\mu^{182}W$	$\gamma^{187}Os(T)$	μ ¹⁸⁶ Os(T)	∑HSE	Data sourc
Kaapvaal Craton, South Africa					I	I I		I
Schapenburg	+2.4±0.5	+5.8±0.8	-5.0±2.8	-8.4±4.1	+3.7±0.3		29±9	[1-2]
Komati	+0.46±0.39	$+1.9{\pm}1.0$	-0.9 ± 2.7	$+2.7\pm4.5$	+0.3±0.3	-12±8	60±8	[3-5]
Weltevreden	$+0.54\pm0.40$	$+4.7\pm2.1$	$+2.5\pm3.8$		-0.1±0.2	+22±7	65±6	[3-4]
Pilbara Craton, Western Australi	a	•			•	1		
Coonterunah	$+2.4\pm0.5$	+4.4±0.3		+11.4±4.6			29±6	[6]
Kelly	+0.5±0.6	+1.7±0.6		+8.2±3.3	$+0.8\pm0.4$		38±7	[6]
Ruth Well	+1.3±0.7†	+2.4±0.4†	-1.8±3.8	+7.7±5.0	-0.4±0.4†		55±4	[6]
Ruth Weh	+0.6±0.7	+1.7±0.4			-0.4 ± 0.4			
Decel	+1.9±0.3*	+4.6±0.8†		+7.7±5.0	+0.9±0.3†		70±5	[6]
Regal	+1.0±0.3	+4.1±0.8			$+0.9\pm0.3$			
Fennoscandian Shield, northern	Europe					1		
Kostomuksha	+3.0±0.4	+4.9±0.3	$+0.4\pm0.9$	+15.0±4.8	$+2.5\pm0.6$	+22±6	79±6	[7 – 11]
Vetreny	+3.7±0.4†	+6.3±0.9†	+0.5±2.1	-0.5±5.2†	+1.3±0.2†	+3.2±1.7	66±10	[12]
Veueny	-0.9 ± 0.4	$+0.4\pm0.9$	+0.5±2.1	$+7.5\pm5.2$	+1.7±0.2			
Lonlord	+4.9±0.3†	+10.2±0.7†		-10.0±5.0†	-0.2±0.3†	+29±2	120±5	[13]
Lapland	+3.7±0.3	+8.7±0.7		+1.5±3.3	-0.2 ± 0.3			
Superior Craton, Canada	I					1		
Boston Creek	+2.5±0.2	+4.3±0.9	-3.8 ± 2.8	+11.7±4.5	+0.1±0.3		35±10	[14]
Pyke Hill-Alexo	+3.0±0.5	+5.5±0.6	$+6.8\pm2.5$	l l	+0.4±0.1	-0.1±4.2	85±5	[15 - 19]
Rhodesian Craton, Zimbabwe	I				·			I
Belingwe	+2.9±0.2		$+3.7\pm7.0$		+0.1±0.2	$+0.6\pm2.5$	57±7	[20 - 21]

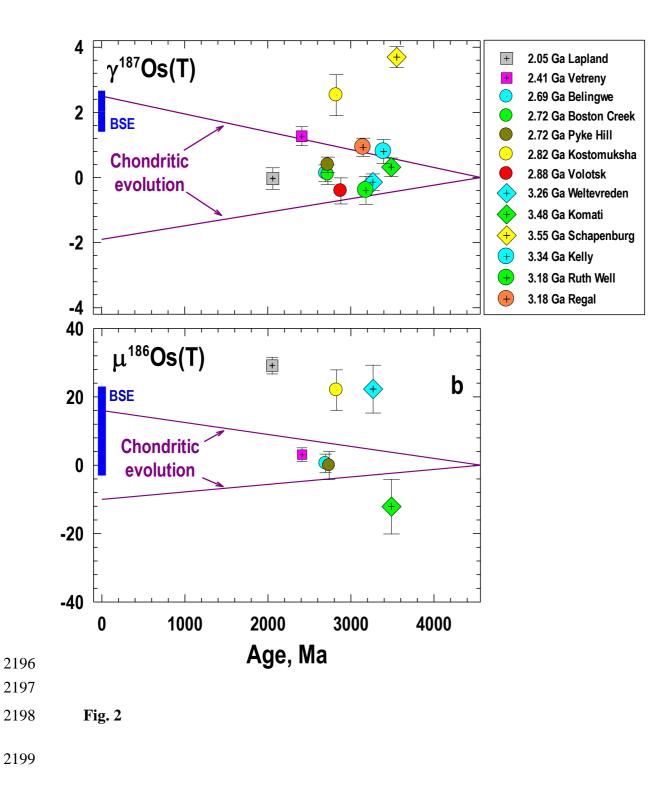
Table 2. Summary of isotopic features and HSE abundances of the komatiite systems considered in this review

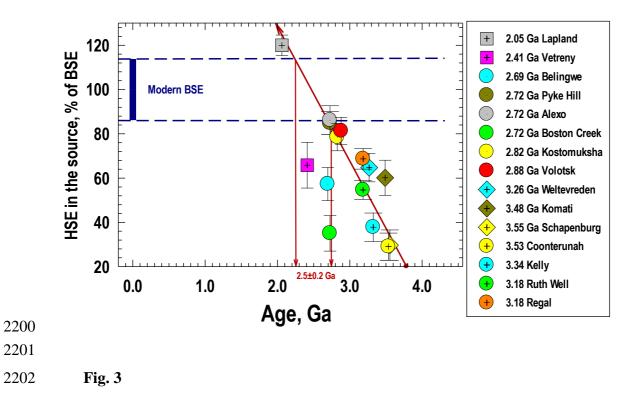
[†]Values corrected for AFC. The italicized values are those measured in the emplaced lavas, before correction for AFC. All uncertainties are 2SD of the mean. Data sources are as follows: [1] - Puchtel et al. (2009a); [2] - Puchtel et al. (2016a); [3] - Puchtel et al. (2013); [4] - Puchtel et al. (2014); [5] - Touboul et al. (2012); [6] - Puchtel et al. (2022); [7] - Puchtel et al. (1998); [8] - Boyet and Carlson (2006); [9] - Touboul et al. (2012); [10] - Puchtel et al. (2005); [11] - Puchtel and Humayun (2005); [12] - Puchtel et al. (2016b); [13] - Puchtel et al. (2020); [14] - Puchtel et al. (2018); [15] - Blichert-Toft and Arndt (1999); [16] - Puchtel et al. (2004a); [17] - Puchtel et al. (2004b); [18] - Puchtel et al. (2009b); [19] - Debaille et al. (2013); [20] - Puchtel et al. (2009b); [21] - Boyet and Carlson (2006).

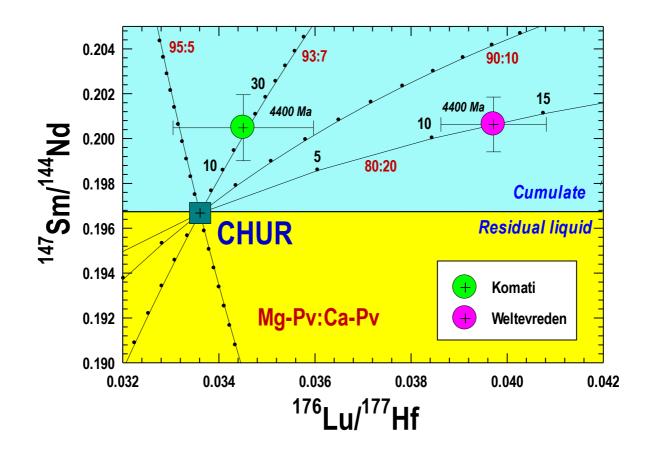




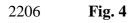


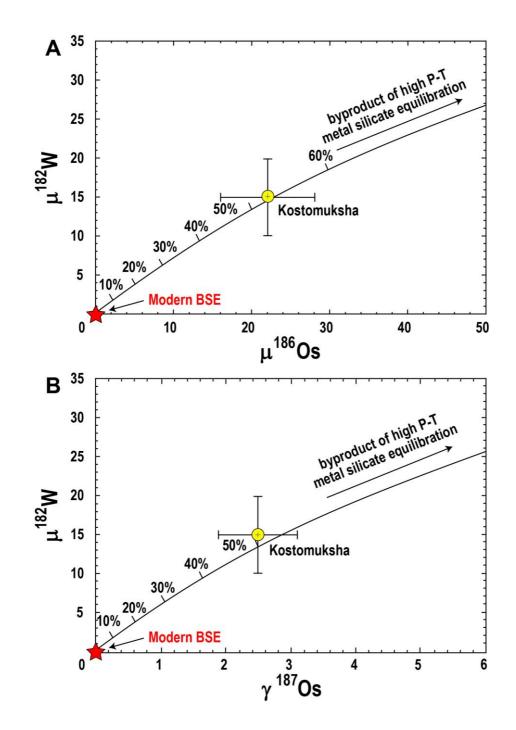






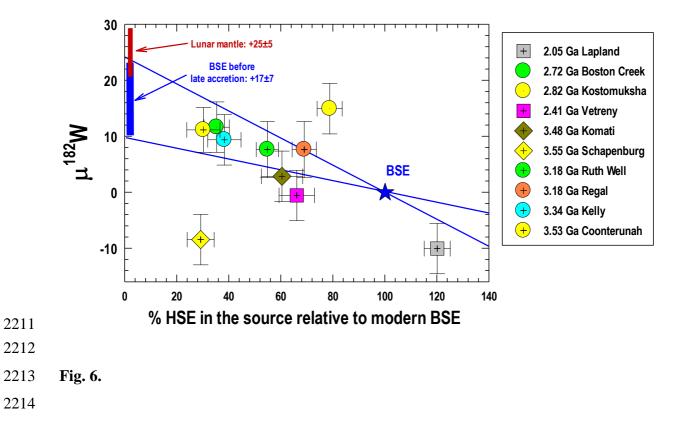








2209 Fig. 5.



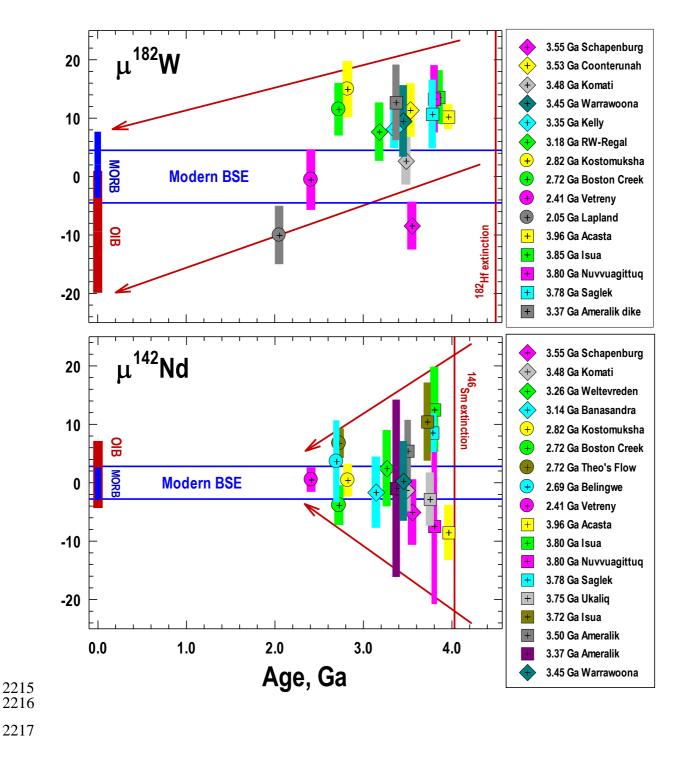


Fig. 7