

1 **Origin of ultramafic–mafic bodies on the Isles of Lewis and Harris (Scotland,**
2 **UK): constraints on the Archean–Paleoproterozoic evolution of the Lewisian**
3 **Gneiss Complex, North Atlantic Craton**

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21 Dykes; Archean tectonics.

22 ABSTRACT

23 The Lewisian Gneiss Complex (LGC) is a tonalite-trondhjemite-granodiorite (TTG)-dominated fragment
24 of the North Atlantic Craton (NAC) in northwest Scotland. End-member models describe the LGC as
25 representing either a continuous piece of Archean crust or up to 12 geologically distinct Archean
26 terranes, with interpretations sitting on a spectrum between these end-members. There is particular
27 uncertainty over the correlations between the Archean–Paleoproterozoic magmatic and
28 metamorphic events recorded by mainland part of the LGC and the part exposed on the Outer
29 Hebridean islands of Lewis and Harris. In this paper, we present the results of field mapping,
30 petrography, and major, trace and platinum-group element (PGE) bulk-rock geochemistry for four
31 ultramafic–mafic bodies in Lewis and Harris, namely: Maaruig, Loch Mhorgail, Coltraiseal Mor and
32 Beinn a’ Chuailean. We consider the effects of metamorphism and element mobility, their
33 petrogenesis, and potential correlations with ultramafic–mafic rocks elsewhere in the LGC. Our data
34 indicate that the studied ultramafic–mafic rocks can be subdivided into two petrologically distinct
35 groups. Metaperidotites and metapyroxenites from Maaruig and Loch Mhorgail are interpreted as
36 Archean (> 2.8 Ga) cumulates distinct from anything currently identified in the mainland LGC, with this
37 interpretation based on distinctive modal layering, a discordance with surrounding TTG gneiss,
38 fractionated PGE patterns ($[Pd/Ir]_N = 1.3–6.6$) and negative HFSE anomalies ($[Nb/La]_N = 0.2–0.8$).
39 Metagabbronorites from Coltraiseal Mor and Beinn a’ Chuailean, which also exhibit negative high field
40 strength-element (HFSE) anomalies ($[Nb/La]_N = 0.2–0.7$) and show mildly fractionated ($[Pd/Ir]_N = 1.2–$
41 2.8) PGE patterns, most likely represent deformed Paleoproterozoic dykes. These occurrences could
42 be correlatives of a suite of ca. 2.4 Ga mafic dykes exposed throughout the mainland LGC (the Scourie
43 Dykes), with the Outer Hebridean occurrences having experienced more intense Paleoproterozoic
44 (Laxfordian) deformation/reworking. These interpretations suggest that the LGC lithologies of Lewis
45 and Harris were proximal to the mainland LGC’s Central Region by the early Paleoproterozoic but
46 raises the possibility that they were distinct crustal blocks in the Mesoarchean.

47 **1.0 INTRODUCTION**

48 The Lewisian Gneiss Complex (LGC) in northwest Scotland (Fig. 1a) is a fragment of tonalite-
49 trondhjemite-granodiorite (TTG) Archean crust variably reworked during the Proterozoic (e.g.,
50 Wheeler et al., 2010). The Archean–Paleoproterozoic evolution of the LGC is described by competing
51 models, including end-member interpretations whereby it is interpreted to represent: (a) a section of
52 broadly continuous Archean crust that has been subsequently faulted to expose different crustal levels
53 (Park and Tarney, 1987); or (b) up to 12 geologically unique terranes that assembled later, during the
54 Proterozoic (Kinny et al., 2005 and references therein).

55 This regional debate, whereby interpretations sit on a spectrum between the two end-member
56 models outlined above, reflects a broader discussion about the nature of Archean geodynamics and
57 the onset of plate tectonics (e.g., Kamber, 2015). While many envisage plate tectonics to have
58 commenced by the late Archean (ca. 2.8 Ga; de Wit et al., 1987; Furnes et al., 2007; Dhuime et al.,
59 2015; Brown and Johnson, 2018; Cawood et al., 2018), others argue that key plate tectonic indicators
60 (e.g., blueschists and ophiolites) are absent in the Archean and rare in the Proterozoic, with this
61 geodynamic regime therefore not predominating on Earth until the Neoproterozoic (ca. 1.0–0.85 Ga;
62 Hamilton, 2003; Stern, 2005, 2008, 2020). Fundamentally, was Archean continental crust — such as
63 that preserved in the LGC — formed and assembled by processes akin to modern-style plate tectonics,
64 or was an alternative geodynamic regime, such as stagnant-lid tectonics, responsible (Bédard, 2013;
65 Debaille et al., 2013; Stern, 2016)?

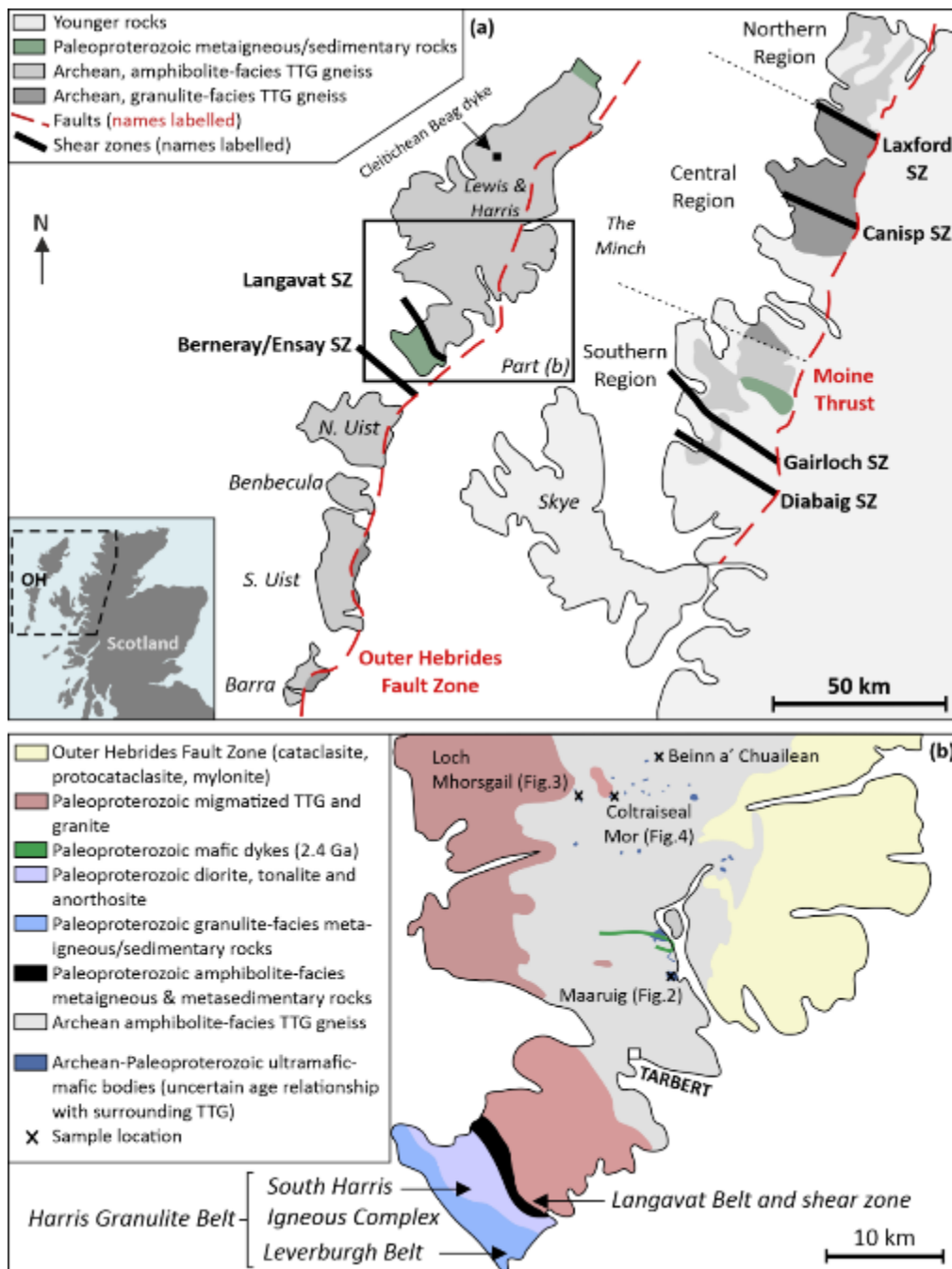
66 Much of the previous research on this topic in the LGC has focused on felsic lithologies (Kinny and
67 Friend, 1997; Friend and Kinny, 2001; Love et al., 2010; Whitehouse and Kemp, 2010; MacDonald et
68 al., 2013), with a smaller body of work considering the mafic rocks (e.g., Sills et al., 1982; Johnson et
69 al., 2012; Guice et al., 2020; Fischer et al., 2021). In this paper, we focus on ultramafic–mafic
70 lithologies, whose origin(s) can potentially be attributed to a wide-range of magmatic settings that
71 have varied potential implications for broad-scale geodynamic processes predominant on Earth. For

72 example, the preservation of fragments of oceanic lithosphere — as suggested for a plethora of
73 Archean–Proterozoic ultramafic complexes globally (Kusky et al., 2001, 2007; Anhaeusser, 2006;
74 Furnes et al., 2007; Dilek and Polat, 2008; Ordóñez-calderón et al., 2009; Kisters and Szilas, 2012; Szilas
75 et al., 2013; Grosch and Slama, 2017) — requires a different suite of magmatic and tectonic processes
76 to that of komatiites or associated layered intrusions. In the latter case, the ultramafic rocks would
77 have crystallized from a magma derived from high degrees of partial melting and became juxtaposed
78 with the TTG gneiss, possibly via. “sagduction” (e.g., Johnson et al., 2016).

79 The temporal and petrogenetic relationship between ultramafic–mafic magmatism exposed in the
80 Scottish mainland (see Guice et al., 2020 and references therein) and Outer Hebridean portions of the
81 LGC also remains unresolved (Fettes and Mendum, 1987; Mason and Brewer, 2004). Some authors
82 have correlated the Archean–Paleoproterozoic rocks in the Outer Hebrides with those in mainland
83 Scotland (Fettes and Mendum, 1987), while others argue — in-line with the terrane model described
84 above — that they comprise several distinctive crustal blocks that amalgamated during the late
85 Paleoproterozoic (Kinny et al., 2005). In the latter scenario, the Outer Hebrides Fault Zone (OHFZ),
86 which is a north-northeast/south-southwest-trending structure that can be traced for 170 km along
87 the east coast of the Outer Hebridean islands of Barra, South Uist, Benbecula, North Uist and Lewis
88 and Harris (Fig. 1a; Jehu and Craig, 1924, 1927, 1934; Cheadle et al., 1987; Imber et al., 1997), could
89 be a late Paleoproterozoic (ca. 1.6 Ga) suture (Friend and Kinny, 2001; Mason and Brewer, 2004; Kinny
90 et al., 2005; Love et al., 2010; Mason, 2016).

91 In this paper, we focus on four Outer Hebridean ultramafic–mafic bodies whose age(s) and origin(s)
92 are enigmatic, namely Maaruib, Loch Mhorgail, Coltraiseal Mor and Beinn a’ Chuailean (Fig. 1b). For
93 these localities, we present the results of field mapping and observations, petrography, and major,
94 trace and platinum-group element (PGE) bulk-rock geochemistry. Using these data, we aim to: (a)
95 establish age relations with the surrounding TTG gneiss; (b) consider the effects of metamorphism and
96 element mobility; (c) discuss the likely origin(s) of the ultramafic–mafic bodies; (d) test the possible

97 correlations with ultramafic–mafic magmatism exposed in the mainland LGC; and (e) consider any
 98 implications for the Archean–Paleoproterozoic evolution of the LGC.



99
 100 **Figure 1: (a)** Simplified geological map of the Lewisian Gneiss Complex, NW Scotland (redrawn after: Wheeler et al., 2010;
 101 MacDonal and Goodenough, 2013). Abbreviations: SZ = shear zone; TTG = tonalite-trondhjemite-granodiorite. **(b)**
 102 Geological map of the island of south Lewis and Harris, which is the focus of this study (redrawn after: Fettes et al., 1992).
 103 The area represented is highlighted in part (a).

104 **2.0 GEOLOGICAL FRAMEWORK**

105 The Archean–Paleoproterozoic LGC is a small fragment of the NAC that crops out on the Outer
106 Hebrides and the northwest Scottish mainland (Fig. 1a). The LGC predominantly comprises Archean,
107 amphibolite- to granulite-facies TTG gneiss, with minor ultramafic, mafic and metasedimentary
108 lithologies (Peach et al., 1907; Sutton and Watson, 1951; Fettes and Mendum, 1987; Wheeler et al.,
109 2010). These lithologies, which have been subject to multiple phases of amphibolite–granulite-facies
110 metamorphism, are cross-cut by Paleoproterozoic mafic dykes (ca. 2.4–2.0 Ga; Teall 1885, Mason and
111 Brewer 2004, Davies and Heaman 2014) and Paleo- to Meso-proterozoic granitic–pegmatitic
112 intrusions (Dearnley, 1962; Davies et al., 1975; Fettes and Mendum, 1987; Park et al., 2002; Shaw et
113 al., 2016). Paleoproterozoic (ca. 2.2–1.9 Ga) belts comprising metasedimentary and meta-igneous
114 lithologies are also a minor component of the LGC; both in the mainland (e.g., Loch Maree Group;
115 Floyd et al. 1989) and Outer Hebrides (e.g., Harris Granulite Belt; Langavat Belt; Whitehouse and
116 Bridgwater 2001, Mason et al. 2004a, 2004b, Hollis et al. 2006, Mason 2016).

117 **2.1 The Lewisian Gneiss Complex of mainland Scotland**

118 Cropping out as a 125 km long, 20 km wide coastal strip (Fig. 1a), the mainland LGC is traditionally
119 subdivided into a granulite-facies Central Region and amphibolite-facies Northern and Southern
120 Regions (e.g., Wheeler et al. 2010 and references therein). The pyroxene-bearing gneisses of the
121 Central Region have been previously interpreted as representing deeper crustal levels than the
122 hornblende-bearing gneisses of the Northern and Southern Regions (Park and Tarney, 1987).
123 Geochronological studies — generally involving U-Pb dating of zircon from the TTG gneisses — have
124 revealed a geographically diverse suite of protolith and metamorphic ages for these lithologies (Kinny
125 and Friend, 1997; Friend and Kinny, 2001; Love et al., 2004, 2010; Kinny et al., 2005). One
126 interpretation of these data, which represents the alternate end-member to the traditional
127 subdivision described above, is that the mainland LGC comprises six distinct “terranes” that have
128 unique Archean histories and were tectonically juxtaposed during the Proterozoic (e.g., Kinny et al.

129 2005). Although the number, extent and significance of individual terranes remains a matter for
130 discussion (e.g., Fischer et al., 2021), the Laxford and Gairloch Shear Zones (Fig. 1a) are generally
131 accepted as major crustal boundaries (Park, 2005; Goodenough et al., 2010, 2013).

132 A broad magmatic and metamorphic chronology is relatively well constrained for the Central Region
133 of the mainland LGC (Fig. 2). The igneous protoliths of the TTG gneisses crystallized between 3.1 and
134 2.7 Ga (Kinny and Friend, 1997; Friend and Kinny, 2001; Love et al., 2004; Whitehouse and Kemp,
135 2010; MacDonald et al., 2013, 2015). This was followed by a granulite–amphibolite-facies
136 tectonothermal episode between 2.7 and 2.5 Ga (Taylor et al., 2020), whereby the lower crust could
137 have been at high temperature and melt-bearing for more than 200 m.y.. This protracted
138 tectonothermal episode has also been interpreted as two discrete metamorphic events (e.g., Fischer
139 et al. 2021 and references therein): known locally as the granulite-facies Badcallian (ca. 2.7 Ga; Evans
140 and Lambert, 1974; Barnicoat, 1983; Cartwright et al., 1985; Corfu et al., 1994; Andersen et al., 1997;
141 Corfu, 1998; Barooah and Bowes, 2009; Crowley et al., 2015; Feisel et al., 2018) and granulite- to
142 amphibolite-facies Inverian (ca. 2.5 Ga Beach, 1973, 1974; Corfu et al., 1994; Whitehouse and Kemp,
143 2010). The Badcallian–Inverian structures are cross-cut by a suite of mafic dykes, largely emplaced
144 2.42–2.38 Ga, which are known locally as the Scourie Dykes (Teall, 1885; Peach et al., 1907; Sutton
145 and Watson, 1951; Davies and Heaman, 2014; Hughes et al., 2014). Dyke emplacement was followed
146 by multiple greenschist- to amphibolite-facies metamorphic events in the late Paleoproterozoic (ca.
147 1.9–1.6 Ga) — collectively referred to as the Laxfordian (Beach, 1974; Beach et al., 1974; Goodenough
148 et al., 2010, 2013).

149 *2.1.1 Archean ultramafic, mafic and metasedimentary rocks in the mainland LGC*

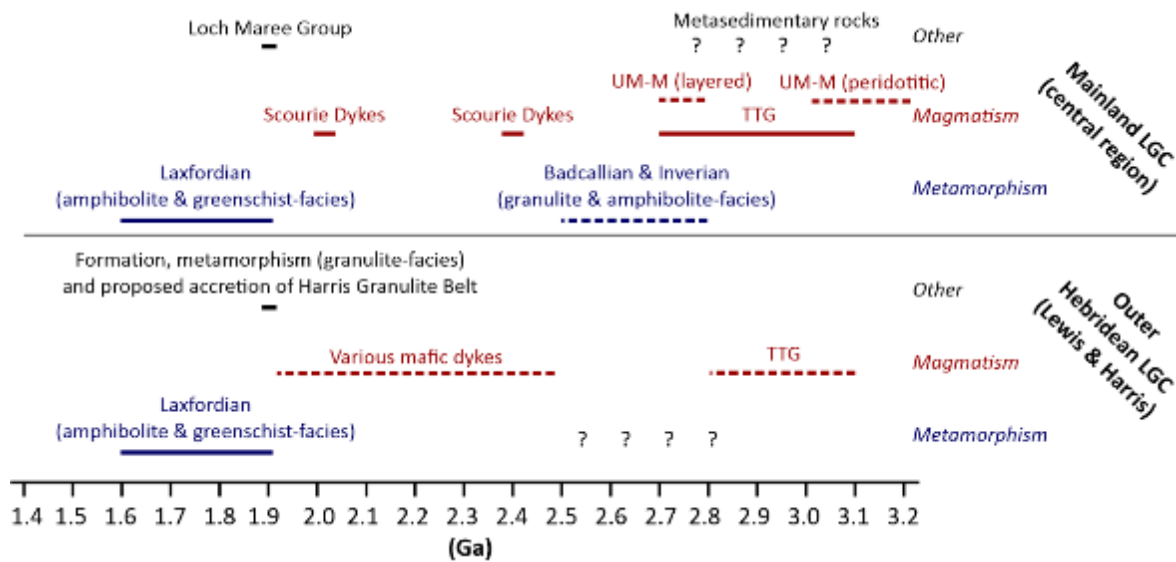
150 Ultramafic–mafic complexes occur throughout the granulite-facies Central Region, ranging from cm-
151 scale pods to km-scale complexes (O’Hara, 1961; Bowes et al., 1964, 1966; Davies, 1974; Rollinson and
152 Windley, 1980; Sills et al., 1982; Rollinson and Gravestock, 2012; Johnson et al., 2016; Guice et al.,
153 2018a, 2018b, 2020). The relative proportions of the ultramafic and mafic rocks vary dramatically
154 between individual complexes, with some containing no ultramafic rocks and others comprising

155 almost entirely these lithologies (Guice, 2019). With some exceptions, the ultramafic portions are
156 generally metapyroxenite-dominated, form the stratigraphic base of individual occurrences and
157 display primary magmatic layering (Sills, 1981; Sills et al., 1982; Guice et al., 2018a, 2020). The mafic
158 portions of these complexes are predominantly mesocratic- to melanocratic (rarely leucocratic),
159 comprise variable portions of garnet, plagioclase, orthopyroxene, clinopyroxene and amphibole
160 (Johnson et al., 2012, 2016), and sometimes preserve relict magmatic layering (Sills, 1981; Sills et al.,
161 1982; Guice et al., 2018a, 2020). Some of these occurrences — notably in the Laxford Shear Zone (Fig.
162 1a), are spatially associated with quartz-feldspar-biotite-(garnet) gneisses that could be interpreted as
163 metamorphosed sedimentary or volcanogenic lithologies (Beach et al., 1974; Cartwright et al., 1985;
164 Goodenough et al., 2010, 2013; Johnson et al., 2016).

165 These ultramafic–mafic complexes have previously been the subject of wide-ranging interpretations,
166 including their formation as: (1) the remnants of an early (possibly oceanic) crust that pre-dates the
167 TTG protoliths (Sills, 1981); (2) fragments of one or more layered intrusions (Bowes et al., 1964; Guice
168 et al., 2018a, 2020); (3) accreted oceanic crust (Park and Tarney, 1987); (4) fragments of Archean
169 mantle (Guice et al., 2020); or (5) sagducted remnants of one or more greenstone belt(s) (Johnson et
170 al., 2016). Recent studies have suggested that these lithologies may reflect more than one origin
171 (Rollinson and Gravestock, 2012; Johnson et al., 2016; Guice et al., 2018a, 2020), and possibly record
172 multiple phases of temporally distinct ultramafic–mafic magmatism (Guice et al., 2020).

173 Based on field mapping, petrographic characteristics, and bulk-rock major, trace and platinum group
174 element geochemistry, Guice et al. (2020) subdivided these mainland Archean ultramafic–mafic
175 complexes into two groups: (1) a large group of distinctly layered bodies, interpreted as representing
176 ca. 2.8 Ga layered intrusions; and (2) a smaller group of peridotite-rich bodies that are generally
177 massive (weakly layered in places), likely pre-date the TTG and record a more enigmatic, possibly
178 mantle origin. Unlike the layered ultramafic–mafic bodies, which display consistent concordance
179 between TTG gneissosity, lithological contacts and layering in the ultramafic–mafic rocks, the second

180 group of ultramafic bodies occur as discordant, elliptical-shaped pods dominated by metaperidotite
 181 (Faithfull et al., 2018; Guice et al., 2020). Note that this “layered” vs. generally massive “peridotitic”
 182 distinction is made for the Archean ultramafic–mafic complexes in the mainland LGC throughout this
 183 paper (Fig. 2).



184
 185 **Figure 2:** Timeline detailing the major magmatic and metamorphic events currently recognized the mainland (Central Region)
 186 and Outer Hebridean (Lewis and Harris; often referred to as the Tarbert Terrane) portions of the LGC. Dashed lines represent
 187 moderate uncertainty. Question marks represent high uncertainty. See text for references.

188 *2.1.2 Paleoproterozoic Scourie Dykes in the mainland LGC*

189 The Scourie Dykes are a suite of steeply-dipping northwest/southeast to east/west-trending mafic
 190 dykes that mostly intruded between 2.42 and 2.38 Ga (Davies and Heaman, 2014), with a smaller
 191 group of ca. 2.0 Ga occurrences also identified (Heaman and Tarney, 1989). Individual dykes are up to
 192 100 m wide, display sharp contacts with the surrounding TTG and, although they are most widespread
 193 in the Central Region, exist in all 3 regions of the mainland LGC (Sutton and Watson, 1951; Weaver
 194 and Tarney, 1981). Based on the primary mineral assemblages, the Scourie Dykes of the Central Region
 195 (Fig. 1) can be subdivided into several groups (Tarney, 1963; Weaver and Tarney, 1981; Tarney and
 196 Weaver, 1987; Hughes et al., 2014): (1) a texturally and mineralogically homogenous dolerite suite —
 197 comprising 90–95 % of the total dykes exposed in the mainland LGC — that is composed of
 198 amphibolitized clinopyroxene, plagioclase, hornblende, quartz and biotite, with accessory magnetite,

199 ilmenite, pyrite, pyrrhotite and apatite; (2) a picrite suite, containing olivine, orthopyroxene and
200 clinopyroxene phenocrysts, alongside interstitial plagioclase, minor phlogopite, and accessory
201 chromite, magnetite, ilmenite and pyrite; and (3) an olivine gabbro suite, comprising orthopyroxene,
202 clinopyroxene, olivine, plagioclase, hornblende and minor phlogopite, alongside accessory magnetite,
203 ilmenite, pyrite and pyrrhotite.

204 The restriction of the most Mg-rich dykes (the picrite suite) to the Central Region has been used to
205 suggest that these magmas did not ascend to the mid-crustal levels represented by the Northern and
206 Southern Regions (Hughes et al., 2014). As has been documented for multiple extension-related
207 Paleoproterozoic dykes swarms globally (e.g., Sandeman and Ryan, 2008; Stepanova and Stepanov,
208 2010), the Scourie Dykes display negative high field strength element (HFSE) anomalies, alongside
209 enrichments in the large ion lithophile elements (LILE: e.g., Th and the light rare earth elements;
210 Hughes et al., 2014). Modelling by Hughes et al. (2014) suggested that this geochemical signature
211 reflects partial melting of sub-continental lithospheric mantle (SCLM), which had been metasomatized
212 either during shallow-angle subduction in the NAC or by carbonatite-induced metasomatism (Yaxley
213 et al., 1991).

214 **2.2 The Lewisian Gneiss Complex of the Outer Hebrides**

215 The OHFZ separates the LGC of the Outer Hebrides into “eastern gneisses” and “western gneisses”
216 (Dearnley, 1962; Fig. 1). The eastern gneisses, which have been most intensely deformed in north
217 Lewis, are often correlated with the mainland LGC, with rocks in southeast Barra preserving evidence
218 for granulite-facies “Badcallian” (see Section 2.1) metamorphism at 2.73 Ga (Fettes and Mendum,
219 1987; Kinny et al., 2005; MacDonald and Goodenough, 2013). Proposed correlations between the
220 Archean–Paleoproterozoic geology of the western gneisses and the mainland LGC (e.g., Dearnley
221 1962, Fettes and Mendum 1987) are more controversial, with some authors interpreting the OHFZ as
222 representing a key suture in the Paleoproterozoic amalgamation of the LGC (e.g., Imber et al. 2002,
223 Kinny et al. 2005).

224 West of the OHFZ, the LGC of Lewis and Harris (often referred to as the “Tarbert Terrane”)
225 predominantly comprises ca. 3.1–2.8 Ga, amphibolite-facies TTG gneiss (Jehu and Craig 1927, 1934,
226 Dearnley 1962, Soldin 1978, Fettes and Mendum 1987, Friend and Kinny 2001, Mason et al. 2004a),
227 with these rocks cross-cut by mafic (Paleoproterozoic) dykes (Mason and Brewer 2004, Davies and
228 Heaman 2014). These lithologies are pervasively affected by a late Paleoproterozoic (1.9–1.6 Ga)
229 amphibolite-facies metamorphic event correlated with the Laxfordian of the mainland LGC (see
230 Section 2.1; Fig. 2) that resulted in local migmatization of the TTG gneiss and the intrusion of granites
231 and pegmatites (Dearnley, 1962; Davies et al., 1975; Shaw et al., 2016). Also present are late Archean
232 to mid-Paleoproterozoic metasedimentary and metaigneous lithologies. This includes the Harris
233 Granulite Belt and Langavat Belt in south Harris (Dearnley 1963, Cliff et al. 1983, Baba 1998, 1999,
234 Whitehouse and Bridgwater 2001, Mason et al. 2004a, Hollis et al. 2006, Kelly et al. 2008, Mason
235 2016), and the Ness Assemblage in northernmost Lewis (Coward et al., 1969; Watson, 1969;
236 Whitehouse, 1990).

237 The Harris Granulite Belt (Fig. 1b) is composed of two broadly defined components: (1) the Leverburgh
238 Belt, which predominantly comprises psammitic and pelitic metasedimentary rocks, alongside minor
239 mafic rocks, ultramafic rocks, chert and marble; and (2) the South Harris Igneous Complex, which is
240 dominated by tonalite, metagabbro and anorthosite, with minor ultramafic rocks and trondhjemite
241 pegmatites (Dearnley, 1963). The Harris Granulite Belt experienced 1.9 Ga, granulite-facies
242 metamorphism ($T \geq 900^\circ$, $P \leq 12.5$ kbar; Baba 1998, 1999, Hollis et al. 2006) not experienced by the
243 TTG gneiss of Lewis and Harris, and is interpreted as an accreted island arc terrane (Whitehouse and
244 Bridgwater 2001, Mason et al. 2004b).

245 The Langavat Belt, which separates the South Harris Igneous Complex from the TTG gneisses (Fig. 1b),
246 contains highly deformed felsic orthogneiss and metasedimentary rocks, alongside minor mafic and
247 ultramafic rocks, and pelitic metasedimentary rocks. Although its precise origin is enigmatic (Mason
248 et al. 2004a), the Langavat Belt is related to the emplacement of the Harris Granulite Belt (Kelly et al.

249 2008; Mason 2012). It has not experienced granulite-facies metamorphism, with Archean zircons from
250 the metasedimentary rocks suggesting that they could have been derived from the TTG gneisses to
251 the north (Mason et al. 2004a).

252 *2.2.1 Ultramafic–mafic rocks in Lewis and north Harris*

253 The ultramafic–mafic rocks in Lewis and Harris appear as < 0.5 km², variably deformed/dismembered
254 lenses and remnants within both the TTG gneiss and Langavat Belt (Dearnley, 1962, 1963; Dearnley
255 and Dunning, 1967; Myers and Lisle, 1971; Cliff et al., 1998; Mason and Brewer, 2004). Some
256 occurrences cross-cut the gneissose foliation of the surrounding TTG, while others display
257 concordance with this fabric and have more uncertain age relationships (Dearnley and Dunning, 1967;
258 Mason and Brewer, 2004). Based on the preserved mineral assemblages, textures and grain sizes,
259 these ultramafic–mafic bodies have been subdivided into two groups (Dearnley, 1962; Myers and
260 Lisle, 1971): (1) mafic rocks — comprising < 0.5 cm diameter clinopyroxene and hornblende “clots”
261 surrounded by a plagioclase-dominated groundmass — that show relatively uniform textures and no
262 evidence for layering or chilled margins; and (2) coarse-grained, commonly layered ultramafic–mafic
263 rocks, comprising olivine, orthopyroxene, clinopyroxene, plagioclase and hornblende in variable
264 proportions, alongside minor chromite and phlogopite. The latter, relatively coarse-grained and more
265 ultramafic group are less common, but are distinctive in the field, typically displaying light brown
266 weathered surfaces (Dearnley, 1962; Myers and Lisle, 1971).

267 Several authors have considered all of the ultramafic–mafic rocks to represent deformed mafic dykes
268 that can likely be correlated with the Scourie Dykes of the mainland LGC (Dearnley, 1962; Myers and
269 Lisle, 1971; Davies et al., 1975; Fettes and Mendum, 1987; Cliff and Rex, 1989). This hypothesis is
270 supported by comparable relict igneous textures and by the age of one ultramafic–mafic body on the
271 island of Lewis — the Cleitichean Beag Dyke — which has been dated at 2.41 Ga using U-Pb zircon
272 geochronology (Davies and Heaman, 2014). In this scenario, the general preservation of ultramafic–
273 mafic rocks as small, < 0.5 km² remnants would reflect the variable deformational effects of the

274 Laxfordian metamorphic event. Moreover, the ultramafic occurrences could represent co-magmatic
275 plutonic complexes that intruded deeper crustal levels than the predominantly mafic Scourie Dykes
276 (Dearnley, 1962).

277 More recent studies have shown that the ultramafic–mafic bodies may record more than one
278 magmatic event. Based on U-Pb zircon geochronology, some of the ultramafic–mafic bodies in Harris
279 (north of the Langavat shear zone and south of Tarbert; Fig. 1b) have been suggested to be a suite of
280 mid-Paleoproterozoic mafic dykes (Mason and Brewer, 2004). It has been speculated that this
281 represents a period of extension — prior to the accretion of the ca. 1.9 Ga Harris Granulite Belt — that
282 could be correlated with the ca. 2.05 Ga Kangamiut dykes in the Greenlandic portion of the NAC
283 (Mason and Brewer, 2004). It has also been suggested that some of the concordant and more
284 ultramafic bodies may represent another group of ultramafic–mafic bodies that could be older than
285 the TTG gneiss (Mason and Brewer, 2004).

286 **3.0 MAPPING AND FIELD RELATIONSHIPS**

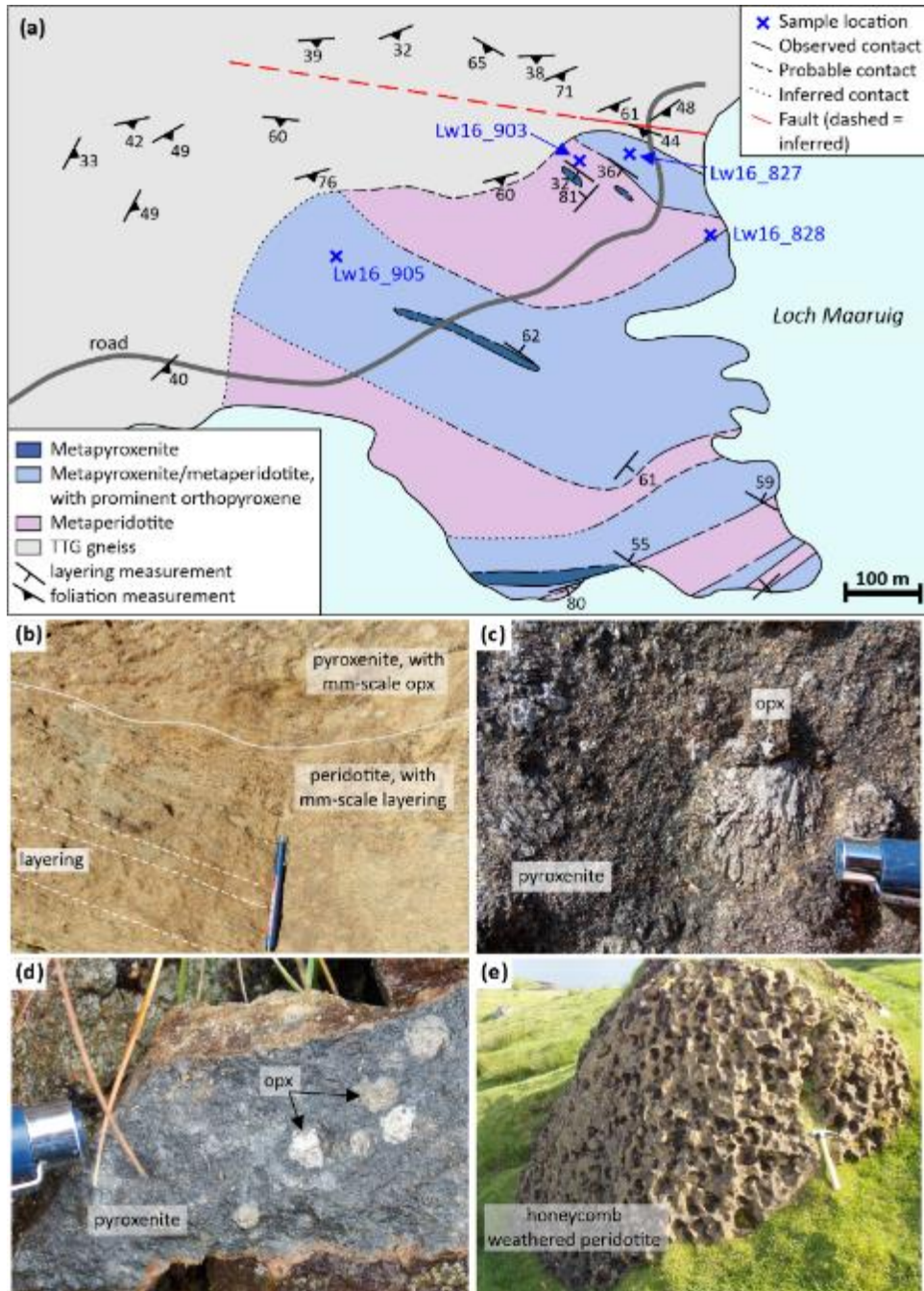
287 Three ultramafic–mafic bodies were mapped and subject to detailed field observations and sampling,
288 namely: Maaruig (Section 3.1); Loch Mhorgail (Section 3.2); and Coltraiseal Mor (Section 3.3). A
289 fourth locality — Beinn a’ Chuailean — was also subject to field observations and sampling (see Section
290 4.0), but was not mapped due to its comparably small size, limited outcrop, and time constraints
291 during fieldwork. It should also be noted that the “meta” prefix is applied to all rock names used
292 throughout this paper, as all four ultramafic–mafic bodies have been subject to amphibolite-facies
293 metamorphism and associated hydrothermal alteration.

294 **3.1 Maaruig**

295 The Maaruig Complex (Myers and Lisle, 1971) forms an elliptical-shaped, 0.6 km x 0.5 km body
296 comprising metapyroxenite and metaperidotite, with localized metre-scale occurrences of
297 metaolivine-gabbro (Fig. 3a). The ultramafic rocks are extremely prominent, forming distinctive brown

298 outcrops that stand proud of the relatively flat and poorly-exposed surrounding TTG gneiss. Our
299 mapping subdivides the Maaruiq Complex into the following units: metapyroxenite (with minor
300 metaperidotite) displaying prominent, mm- to cm-scale orthopyroxene grains; metapyroxenite (with
301 minor metaperidotite) without the prominent orthopyroxene; and honeycomb-weathered
302 metaperidotite that sometimes contains mm- to cm-scale pyroxene oikocrysts (containing olivine and
303 spinel-group mineral chadocrysts). All units commonly contain mm-scale phlogopite. The contacts
304 between metapyroxenite without prominent orthopyroxene and other units are generally sharp,
305 whilst the contacts between metapyroxenite with prominent orthopyroxene and metaperidotite vary
306 from sharp to gradational on the meter-scale. Decimetre-scale, east-west-trending layering can be
307 traced across the mapped area, which is consistent with cm-scale layering observed in individual
308 outcrops (Fig. 3b). The orthopyroxene-bearing metapyroxenite exhibits dark grey to brown weathered
309 surfaces, with 5–40 mm diameter orthopyroxenes which are weathering-resistant relative to the
310 groundmass (Fig. 3c-d). Metaperidotite displays brown weathered surfaces and honeycomb
311 weathering, with individual voids generally 5–10 cm in diameter (Fig. 3e). The orthopyroxene-poor
312 metapyroxenite, which exhibits dark grey weathered surfaces, comprises 1–2 mm diameter olivine
313 and pyroxene grains.

314 The TTG gneiss displays a mm-scale gneissose foliation that exhibits generally moderate dips and is
315 defined by variation in the modal abundance of quartz, feldspar and amphibole (minor mica is also
316 present). The contacts between the ultramafic rocks and surrounding TTG gneiss are obscured by
317 vegetation, with no outcrop-scale, cross-cutting relationships observed. The gneissose foliation in the
318 TTG is consistently parallel to the margins of the ultramafic-mafic complex, but there is some
319 discordance between the map-scale layering, outcrop-scale layering and complex margins, most
320 notably in the northeast of the mapped area (Fig. 3a). Rare plagioclase is noted in the ultramafic rocks
321 close to some contacts with TTG gneiss, with this feature best observed on the foreshore in the
322 northeast of the mapped area (Fig. 3a).



324

325 **Figure 3:** (a) Geological map of the Maaruig Complex. See Fig. 1b for its location in southern Lewis. (b-e) Field photographs

326 detailing the typical field relationships displayed by the ultramafic rocks. Hammer length = 33 cm; pen length = 13.5 cm.

327 3.2 Loch Mhorsgail

328 The ultramafic rocks of the Loch Mhorsgail Complex — exposed over an area of < 0.5 km² and

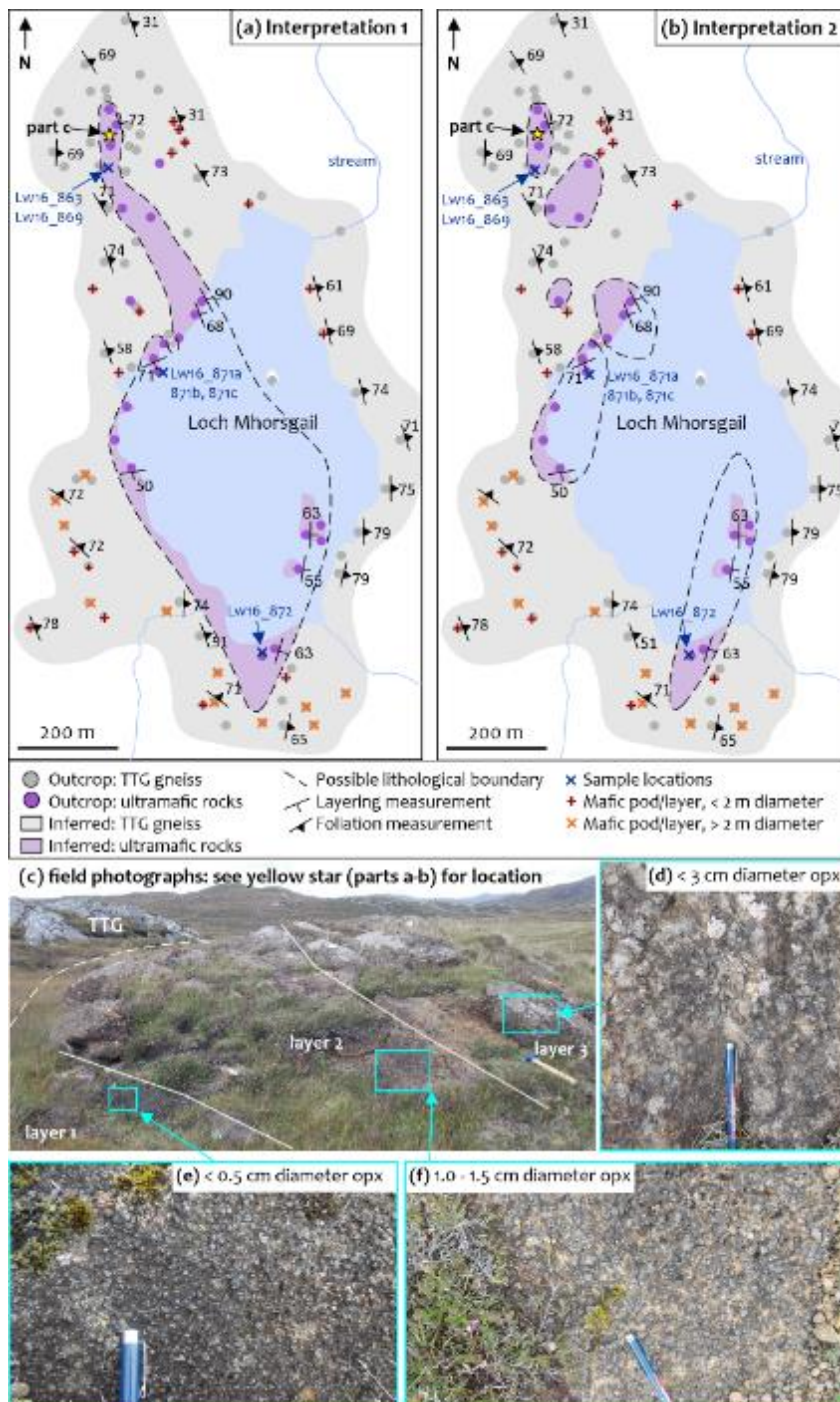
329 described here for the first time — occur as small (generally meter- to decimeter-scale), low-lying

330 outcrops located on the shore and islands of Loch Mhorgail, with minor outcrops inland to the NW
331 of Loch Mhorgail (Fig. 4a–b). Contacts with the surrounding TTG gneiss are not exposed, but the
332 gneissose foliation in the TTG is generally parallel to the layering in the ultramafic rocks where exposed
333 (Fig. 4a–b). Despite this, there exists some map-scale discordance between the layering and gneissose
334 foliation (specifically on the NE shore of the loch; Fig. 4a–b), but the nature of these contacts (i.e.,
335 whether they are tectonic or primary) is unclear. Given both this and the relatively poor exposure, the
336 lateral continuity of the ultramafic rocks is uncertain, with two possible interpretations presented in
337 Figure 4.

338 The Loch Mhorgail Complex contains both metapyroxenite (dark-grey weathered surfaces) and
339 metaperidotite (brown weathered surfaces), with individual outcrops exhibiting modal layering that
340 is also defined by the diameter of orthopyroxene grains (Fig. 4c–f). Boundaries between individual
341 layers range from sharp to gradational (over 5–10 cm), with an outcrop-scale example detailed in
342 Figure 4c. Metapyroxenite, which exhibits dark grey to brown weathered surfaces, contains prominent
343 0.5–4.0 cm diameter orthopyroxene grains alongside rare, mm-scale phlogopite grains.
344 Metaperidotite, which commonly displays honeycomb-weathered textures similar to those at Maaruig
345 (Fig. 3e), also contains prominent, mm-to cm-scale orthopyroxene and rare phlogopite.

346 The TTG gneiss is composed of feldspar, quartz, amphibole and biotite in varied proportions, with
347 amphibole and mica commonly comprising > 30 % of the mineral assemblage observed in hand
348 specimen. The gneissose foliation, which occurs on the mm-scale and reveals cm- to m-scale isoclinal
349 folds in places, generally displays moderate to steep dips towards the east that are broadly parallel
350 with the trend of the ultramafic–mafic body (Fig. 4). TTG gneiss outcrops also commonly exhibit cm-
351 to m-scale mafic layers/pods, with their spatial distribution detailed in Figure 4. Such layers, which are
352 consistently wrapped by the TTG foliation, predominantly comprise amphibole and plagioclase, with
353 garnet preserved in the centre of larger occurrences. Restricted to the TTG, quartz-feldspar
354 pegmatites occur on the cm- to m-scale, and exhibit grain-sizes ranging from 3–10 cm diameter. The

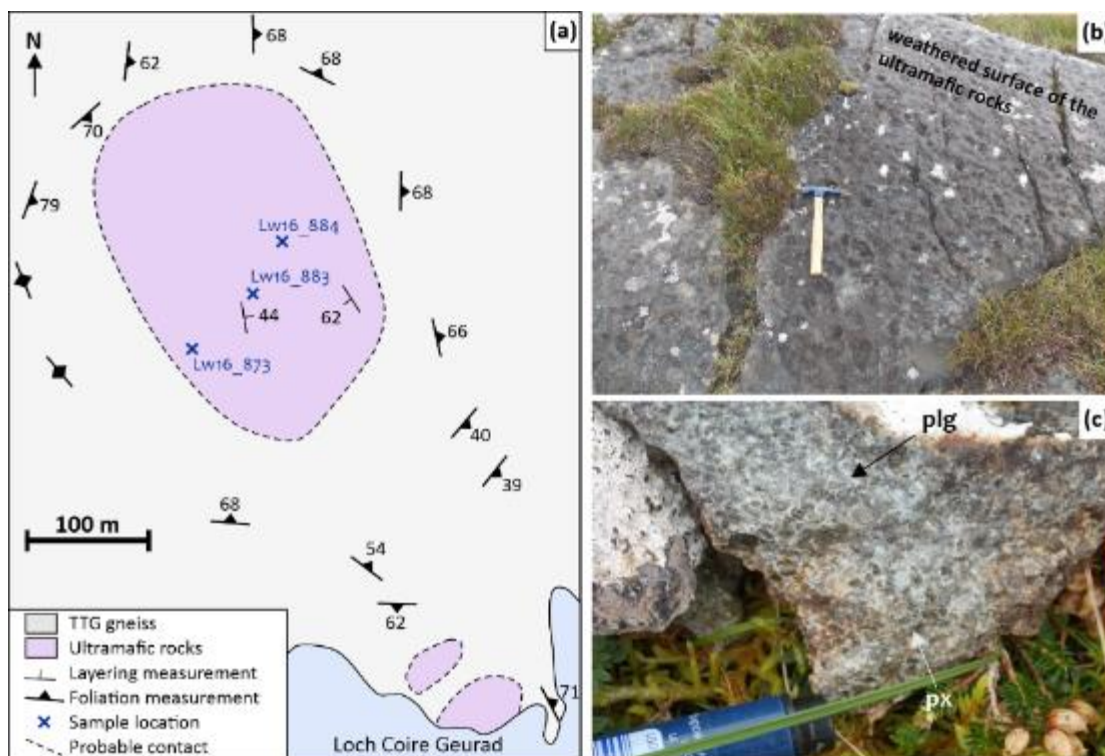
355 pegmatites occur with increasing frequency/size with increasing proximity to the ultramafic–TTG
 356 contact and contain cm scale biotite mica within ~ 10m of the contact. Moreover, these pegmatites
 357 are generally concordant with the TTG foliation, but locally cross-cut it.



358
 359 **Figure 4:** (a–b) Exposure maps of the Loch Mhorgail Complex, with two potential interpretations for the lateral extent of the
 360 ultramafic body. See Fig. 1b for its location in Lewis. (c) Field photograph detailing the outcrop-scale layering shown by the
 361 Loch Mhorgail Complex. (d–f) Smaller-scale field photographs detailing the grain size variation shown by different layers
 362 shown in (c). Note: the different mapping style (relative to Figures 3, 5) used to account for the two possible interpretations.

363 **3.3 Coltraiseal Mor**

364 Ultramafic–mafic rocks at Coltraiseal Mor occur as three elliptical-shaped bodies that stand proud of
365 the generally low-lying and poorly-exposed TTG gneisses (Fig. 5a). The largest ultramafic–mafic body
366 is approximately 220 m x 160 m, while the smaller bodies are < 50 m diameter. The ultramafic–mafic
367 rocks show dark-grey weathered surfaces that often show cm-scale pockmarks (Fig. 5b) and comprise
368 pyroxene (predominantly orthopyroxene), olivine and plagioclase in variable proportions, alongside
369 minor phlogopite (Fig. 5c). Subtle layering is present in rare outcrops, but no systematic variation in
370 modal mineral proportions is identified across the mapped area. The TTG is of similar character to that
371 described at nearby Loch Mhorgsail (Fig. 1b). The contacts between the ultramafic–mafic bodies and
372 the surrounding TTG gneiss are not exposed, with the TTG foliation consistently parallel to the margins
373 of the largest ultramafic–mafic body. Decimetre- to meter-scale pods/layers of mafic material rarely
374 occur in the TTG gneiss, alongside common quartz-feldspar-biotite pegmatites.



375
376 **Figure 5:** (a) Geological map of the Coltraiseal Mor ultramafic body. See Fig. 1b for its location in Lewis. (b-c) Field
377 photographs detailing the weathered and fresh surfaces of the ultramafic rocks.

378 **4.0 SAMPLES AND ANALYTICAL METHODOLOGY**

379 A total of 15 samples were collected for this study, with 13 samples from the mapped ultramafic–
380 mafic bodies at Maaruig ($n=4$), Loch Mhorgail ($n=6$) and Coltraiseal Mor ($n=3$). An additional 2
381 samples were collected from the ultramafic–mafic body at Beinn a' Chuailean (see Fig. 1b for location
382 in Lewis). Brief field observations of Beinn a' Chuailean qualitatively suggests it is similar to the
383 exposures at Coltraiseal Mor (Section 3.3). Sample locations are provided in Table 1 and Figures 3–5.
384 Further to thin sectioning, all samples were crushed and their major- and trace-element bulk-rock
385 geochemistry analyzed. Thirteen of the samples also underwent PGE and Au bulk-rock geochemical
386 analysis, with the methodologies employed described below.

387 **4.1 Bulk-rock geochemistry: Major and trace elements**

388 In preparation for bulk-rock geochemical analysis, weathered surfaces were removed using a
389 diamond-bladed rock-saw, before samples were crushed using a Mn-steel jaw-crusher and ground
390 using an agate ball mill at Cardiff University. Powdered samples were then ignited (at $\sim 900^{\circ}\text{C}$) for 2
391 hours, with loss-on-ignition (LOI) determined gravimetrically.

392 A sample mass of 0.1 g was accurately weighed and mixed with 0.6 g of Li metaborate flux in a Claisse
393 Pt-Rh crucible (see McDonald and Viljoen 2006). Approximately 0.5 mL of a Li iodide solution was
394 added as a non-wetting agent, before the mixture was fused over a propane burner on an automated
395 Claisse FLUXY fusion system. The mixture was dissolved in a Teflon beaker containing 50 ml of 4 %
396 HNO_3 , before the solution was spiked with 1 mL of a 100 ppm Rh spike solution (for use as an internal
397 standard) and made up to 100 mL with 18.2 M Ω de-ionised water. Samples were then analyzed for
398 major and trace elements using a Thermo iCAP 7000 series ICP-OES and Thermo X Series 2 ICP-MS (for
399 solution analyses) respectively. Standard reference materials (SDO-1 and MRG1) and blanks were
400 prepared and analyzed using the same method and instrumentation, with the sample material
401 omitted for the blanks. Accuracy was constrained by analysis of international standard reference
402 materials MRG1 and SDO-1, with precision (reported as the 2se – standard error) constrained by

403 conducting duplicate analyses of selected unknown samples (LW16_900 and LW16_905; see
404 supplementary material).

405 **4.2 Bulk-rock geochemistry: platinum-group elements and gold**

406 All samples were prepared by Ni sulphide fire assay followed by Te co-precipitation, with the full
407 method described by Huber et al. (2001) and McDonald and Viljoen (2006). Typically, 7.5 g of
408 powdered sample was mixed with 7.5 g of silica, 6 g of Na₂CO₃, 12 g of sodium tetraborate decahydrate
409 (borax), 0.9 g of sulfur and 1.1 g of carbonyl-purified Ni. After thoroughly mixing the reagents with the
410 unknown material, the samples were transferred to fire clay crucibles and fired at 1050°C for 90
411 minutes. The resulting sulphide buttons were dissolved using concentrated HCl, with co-precipitation
412 achieved using Te and SnCl₂. The filtered residues were digested using 4 ml of concentrated HCl and 3
413 ml of concentrated HNO₃ in 15 ml Saville screw-top Teflon vials. After the residue had dissolved, liquid
414 contents were transferred to 50 ml volumetric flasks spiked with a 2.5 ppm Tl spike, for use as an
415 internal standard. Samples were made up to 50 ml with 18.2 MΩ deionized water. Solutions were
416 analyzed for PGE and Au using a Thermo X Series 2 ICP-MS (for solution analyses) at Cardiff University.
417 Standard reference material (TDB1) and blanks were prepared and analyzed using the methodology
418 described above, with accuracy and precision (reported as the 2se – standard error) constrained by
419 the analysis of the international standard reference TDB1 (see supplementary material).

420 **5.0 RESULTS**

421 **5.1 Petrography**

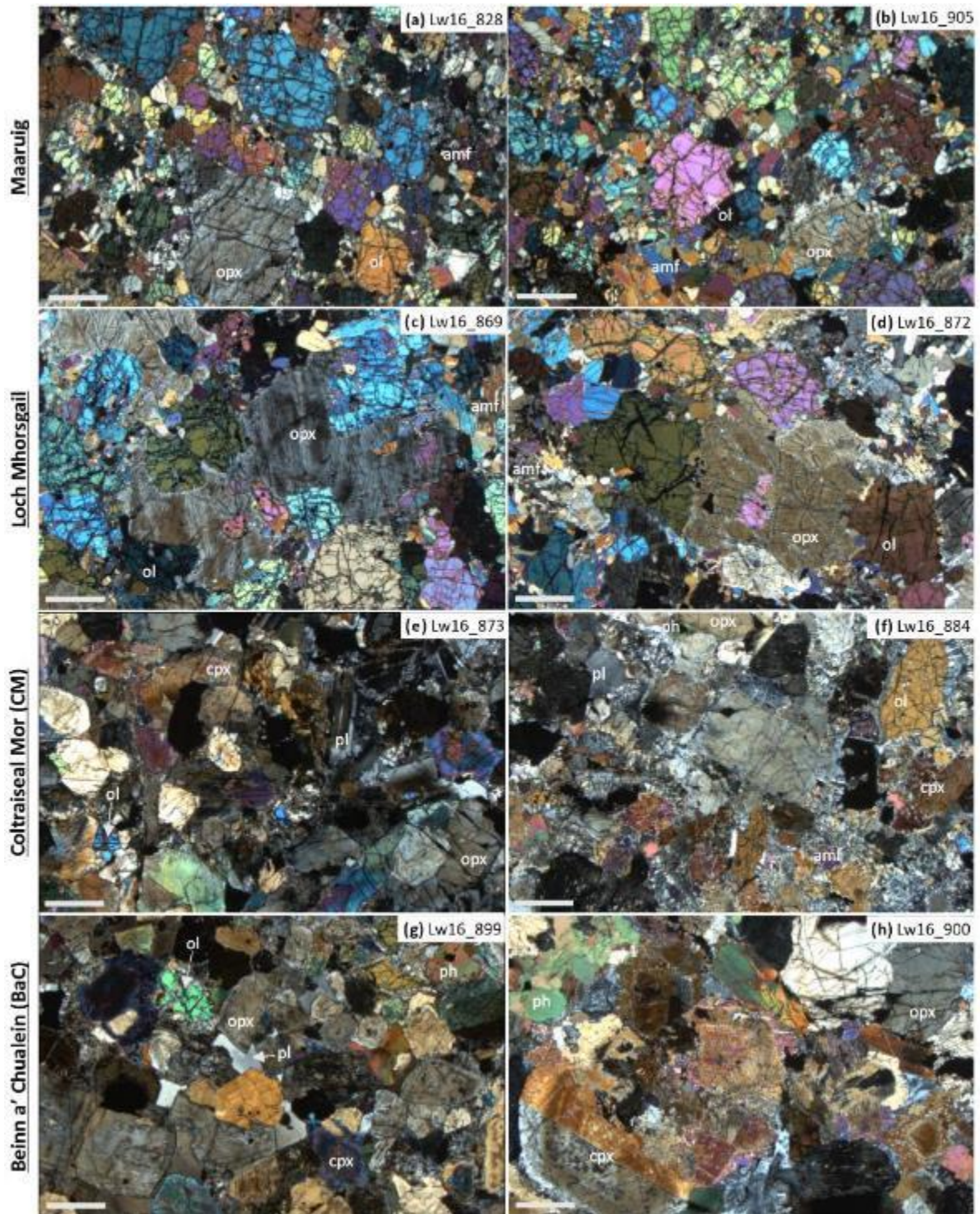
422 *5.1.1 Maaruig*

423 The four Maaruig samples (Fig. 6a–b), which classify as metaperidotite and metapyroxenite (based on
424 modal mineral proportions; see Table 1), comprise olivine, orthopyroxene, clinopyroxene, amphibole
425 and spinel-group minerals, with accessory phlogopite. Olivine occurs as 0.3–3.5 mm diameter,
426 subhedral to anhedral grains that show limited alteration. Congruent with field observations (Section

427 3), orthopyroxene occurs as large, 3–14 mm diameter anhedral to subhedral grains. These grains
428 commonly contain inclusions of anhedral, 0.2 mm diameter oxide phases and < 0.5 mm diameter
429 silicate minerals (predominantly olivine, with rarer amphibole). Clinopyroxene is rare, occurring as
430 0.3–1.5 mm diameter subhedral grains that are amphibolitized, most notably along cleavage planes.
431 Amphibole occurs as subhedral to euhedral, 0.1–0.8 mm in diameter grains that commonly show 120°
432 triple junctions and appear to replace pyroxene. Spinel group minerals — classified as Al-chromite and
433 picotite, with rare Fe-chromite and chromite — are 40–300 µm diameter and euhedral to subhedral.

434 *5.1.2 Mhorskail*

435 The six Loch Mhorskail rocks (Fig. 6c–d), which classify as metaperidotite and metapyroxenite and
436 show slightly more visible alteration (to fine-grained amphibole) than the Maaruig rocks, comprise
437 olivine, orthopyroxene, clinopyroxene, amphibole, and spinel group minerals in varied proportions
438 (see Table 1), with accessory phlogopite, plagioclase, ilmenite, apatite and dolomite. Olivine forms
439 0.4–3.5 mm diameter grains that are subhedral to anhedral. Anhedral orthopyroxene grains are 3.5–
440 15.0 mm in diameter and, like those at Maaruig, contain µm-scale spinel inclusions and mm-scale
441 silicate inclusions (comprising olivine and amphibole). Clinopyroxene, which occurs rarely as cores
442 rimmed by clusters of finer-grained amphibole, is subhedral and 1.0–1.5 mm in diameter. Amphibole
443 is generally 0.1–1.0 mm in diameter and subhedral, with 120° triple junctions common, particularly
444 between smaller grains. Plagioclase occurs rarely, forming anhedral grains 1.0–1.5 mm in diameter.
445 Phlogopite occurs rarely as an accessory phase in some thin sections, forming elongate, pleochroic
446 grains that are euhedral to subhedral and up to 1.5 mm in diameter. Spinel group minerals — classified
447 as Al-chromite and picotite, with rare Fe-chromite and chromite — are 40–300 µm diameter and
448 euhedral to subhedral.



449

450 **Figure 6:** Crossed-polarised light (XPL) photomicrographs for the Maaruig (a-b), Loch Mhorskail (c-d), Coltraiseal Mor (e-f)
 451 and Beinn a' Chualein (g-h) complexes. Abbreviations: amf = amphibole; cpx = clinopyroxene; ol = olivine; opx =
 452 orthopyroxene; ph = phlogopite; pl = plagioclase. White scale bar = 1 mm. See main text for full petrographic descriptions
 453 (Section 5.1).

454 5.1.3 Coltraiseal Mor

455 The three Coltraiseal Mor rocks (Fig. 6e–f), which classify as metagabbronorite, comprise olivine,
456 orthopyroxene, clinopyroxene, amphibole, plagioclase and phlogopite in varied proportions, with
457 accessory pyrrhotite, apatite, pyrite and Cr-spinel. Olivine is subhedral to anhedral and 0.3–2.5 mm in
458 diameter. Orthopyroxene is subhedral to anhedral, 1.0–3.2 mm in diameter and occasionally displays
459 weak pleochroism. Where present, clinopyroxene is subhedral to anhedral and < 3.2 mm in diameter,
460 with common alteration to fine-grained (μm -scale) amphibole. Amphibole also occurs as 0.5–2.5 mm
461 diameter, subhedral grains that show common 120° triple junctions. Highly pleochroic phlogopite is
462 0.1–2.0 mm diameter and anhedral, with subhedral plagioclase generally 0.8–2.5 mm in diameter.

463 5.1.4 Beinn a' Chuailean

464 The two Beinn a' Chuailean rocks (Fig. 6g–h), which classify as meta-olivine gabbronorite and
465 metagabbronorite, comprise olivine, orthopyroxene, clinopyroxene, amphibole, plagioclase and
466 phlogopite, with accessory ilmenite, apatite, Cr-spinel and pyrite. Olivine, which is 0.3–2.5 mm in
467 diameter and anhedral to subhedral, appears to cluster in specific parts of thin sections.
468 Orthopyroxene is 0.8–6.5 mm in diameter and subhedral, with some fine-grained alteration to
469 amphibole restricted to cleavage planes. Clinopyroxene occasionally preserves relic twinning and
470 zoning, with grains typically 0.8–3.0 mm in diameter and subhedral. Plagioclase is anhedral to
471 subhedral and 0.5–2.5 mm in diameter, while phlogopite is 0.2–3.5 mm in diameter and anhedral.

472 5.2 Bulk-rock geochemistry

473 Table 2 presents the major and trace element geochemistry of all samples analyzed in this study,
474 alongside key normalized trace element ratios. To assess potential correlations, our data are
475 compared to comparatively well-defined phases of ultramafic–mafic magmatism recorded in the
476 mainland LGC's Central Region (see Section 2.1). Specifically, we compare these data to: (i) a suite of
477 Paleoproterozoic mafic dykes, known as the “Scourie Dykes” (Hughes et al., 2014); (ii) a large group of
478 distinctly layered Archean ultramafic–mafic rocks (Guice et al. 2018b, 2020); and (iii) a smaller, more

479 enigmatic group of generally massive, peridotitic Archean ultramafic–mafic rocks (Guice et al. 2020).
480 For descriptions of these phases of ultramafic–mafic magmatism, see Section 2.1. All LGC comparison
481 data were collected using the same sample preparation procedures at Cardiff University (see Section
482 4) as for the present study. Also included for comparison — as an end-member — are global residual
483 mantle rocks sampled from ophiolites and as oceanic peridotites (Godard et al., 2000, 2008; Paulick
484 et al., 2006).

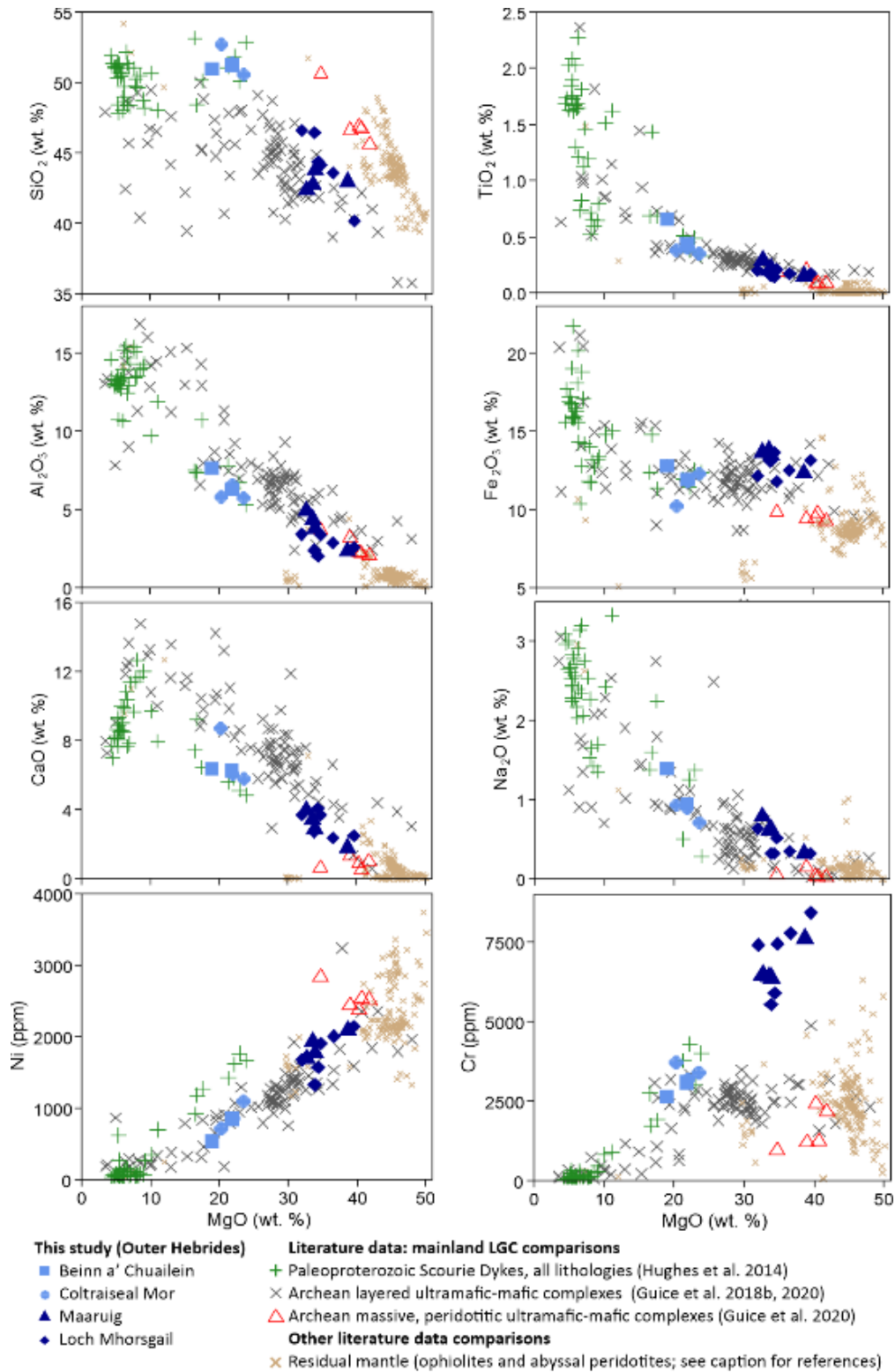
485 *5.2.1 Major elements*

486 The analyzed Outer Hebrides samples form two groups based on their MgO and SiO₂ contents, with
487 this distinction correlating with sample location. The Maaruig and Loch Mhorsgail samples are more
488 MgO-rich, containing 32–40 wt. % MgO and 40–47 wt. % SiO₂, while the Beinn a' Chuailean and
489 Coltraiseal Mor samples are less MgO-rich, containing 19–24 wt. % MgO and 51–53 wt. % SiO₂. This
490 geochemical distinction between the four localities can also be seen for most other major and minor
491 elements (Fig. 7). Collectively, the Outer Hebridean samples form broadly linear trends on bivariate
492 plots (Fig. 7), with MgO showing a strong negative correlation ($R^2 = \geq 0.7$) with SiO₂, TiO₂, Al₂O₃, CaO
493 and Na₂O, a moderate negative correlation ($R^2 = 0.4–0.7$) with K₂O, a strong positive correlation with
494 Ni and Cr, and a weak positive correlation ($R^2 = 0.1–0.4$) with Fe₂O₃. A notable feature is the strong Cr
495 enrichment shown by the Maaruig and Loch Mhorsgail samples relative to other analyzed samples
496 and comparison data, which correlates with the higher modal percentages of spinel-group minerals
497 (see Section 5.1; Table 1).

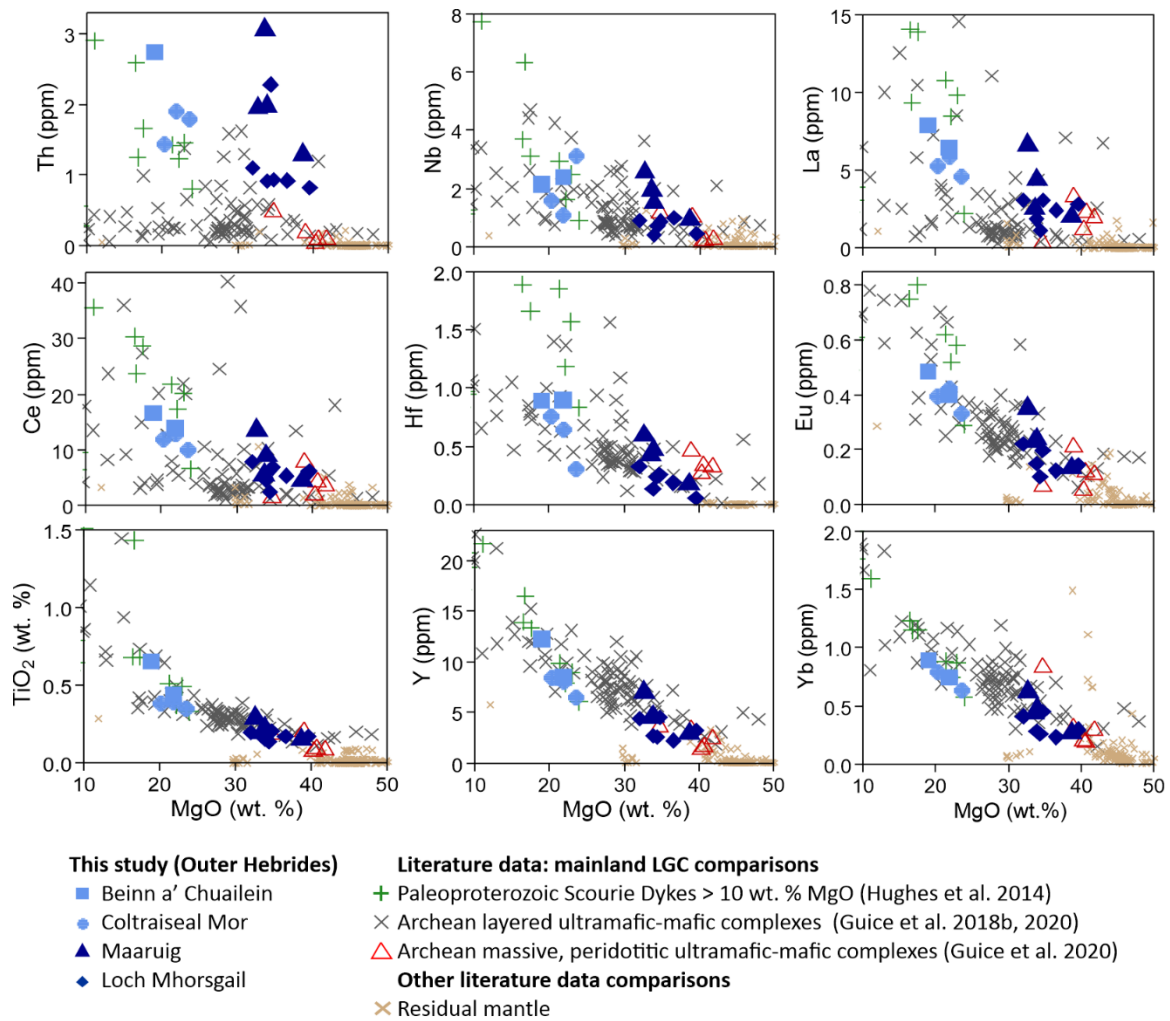
498 *5.2.2 Trace elements*

499 Figure 8 details the trace element compositions of the studied samples according to their MgO
500 contents. The relatively MgO-poor samples from Coltraiseal Mor and Beinn a' Chuailean are generally
501 enriched in trace elements relative to the MgO-rich samples from Maaruig and Loch Mhorsgail, but
502 there is overlap in the absolute concentrations of all trace elements (Fig. 8). When considered

503 collectively, the two groups form broadly linear trends for most trace elements ($R^2 > 0.6$), with the
 504 exception of Th and Nb, which exhibit R^2 values of 0.2 and 0.3 respectively (Fig. 8).



505 **Figure 7:** Bulk-rock major and minor element bivariate plots for the rocks analyzed as part of this study. These data are
 506 compared to the Paleoproterozoic Scourie Dykes and Archean ultramafic–mafic bodies exposed in the mainland LGC (see
 507 figure for references), and to residual mantle rocks (data from: Paulick et al. 2006, Godard et al. 2000, 2008).
 508



509

510 **Figure 8:** Bivariate plots detailing the trace element compositions of the studied ultramafic–mafic rocks, plotted against MgO.

511 For data visualization purposes, the x axis is clipped to include the 10–50 wt. % MgO range.

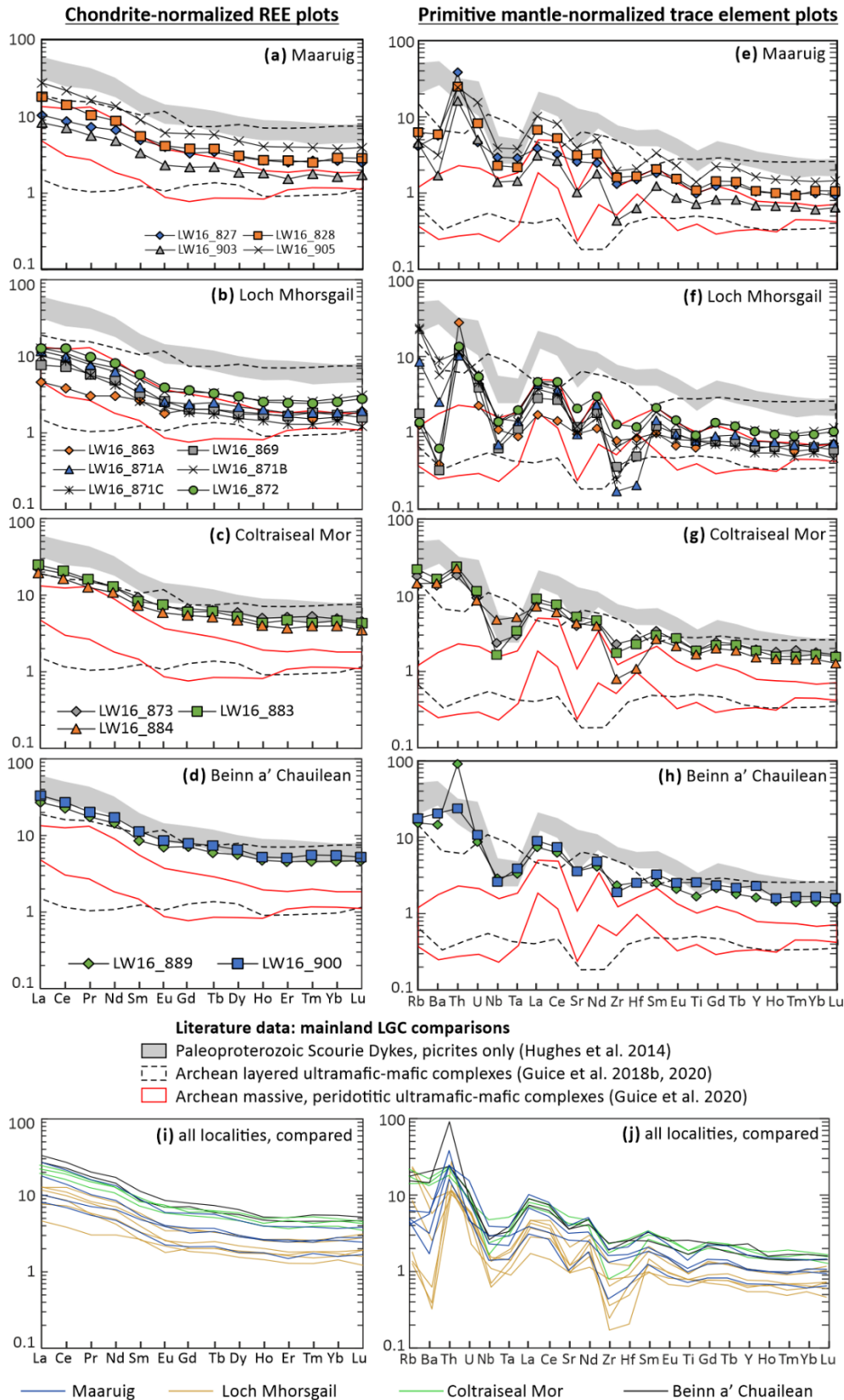
512 On chondrite-normalized rare earth-element (REE) plots (Fig. 9a–d), all of the analyzed rocks from all
 513 four localities display relatively flat heavy-REE (HREE) patterns and negatively sloping light-REE (LREE)
 514 ($[La/Lu]_N = 2.4–8.3$; $[La/Sm]_N = 1.7–3.9$; Fig. 9a–d). The Maaruig and Loch Mhorgail samples are
 515 comparatively depleted in all of the REE, displaying chondrite-normalized values of 1.2–27.9 (Fig. 9a–
 516 b), while the Coltraiseal Mor and Beinn a' Chuailean samples show chondrite-normalized values of
 517 3.5–33.0 (Fig. 9c–d).

518 On primitive mantle-normalized trace element plots (Fig. 9e–h), the analyzed rocks from the Maaruig
 519 and Loch Mhorgail Complexes show negatively sloping patterns ($[Th/Yb]_N = 11.5–48.8$), prominent
 520 negative Nb-Ta anomalies ($[Nb/La]_N = 0.2–0.8$), weak negative Zr-Hf-Ti anomalies and normalized

521 abundances ranging from 0.2 to 38.5 (Fig. 9e–f). The analyzed samples from the Beinn a' Chuailean
522 and Coltraiseal Mor localities also show negatively sloping trace element patterns ($[\text{Th}/\text{Yb}] = 10.1\text{--}$
523 85.4), prominent negative Nb-Ta anomalies ($[\text{Nb}/\text{La}]_N = 0.2\text{--}0.7$) and weak negative Zr-Hf-Ti anomalies,
524 but are relatively enriched in all trace elements, with primitive mantle-normalized trace element
525 abundances range from 0.8–144.6 (Fig. 9g–h). These data are also visualized on several bivariate plots
526 (Fig. 10), which is a simpler visualization of the trace-element variation described in full by the
527 normalized plots.

528 *5.2.3 Platinum-group elements (PGE) and gold (Au)*

529 On chondrite-normalized PGE (+Au) diagrams, the Maarug Complex samples ($n=2$) display
530 fractionated patterns ($[\text{Pd}/\text{Ir}]_N = 5.8\text{--}6.6$), with flat Pd-group PGE (Pt, Pd, Rh; PPGE) and fractionated,
531 positively sloping Ir-group PGE (Os, Ir, Ru; IPGE; Fig. 11a). The majority of Loch Mhorsgail samples
532 ($n=6$) display flat to mildly fractionated chondrite-normalized PGE patterns ($[\text{Pd}/\text{Ir}]_N = 1.3\text{--}4.1$), with
533 one sample having an overall negative slope ($[\text{Pd}/\text{Ir}]_N = 0.2$). They also define positive IPGE patterns
534 and flat to negatively sloping PPGE (Fig. 11b). The Coltraiseal Mor samples ($n=3$) have flat to mildly
535 fractionated chondrite-normalized PGE patterns ($[\text{Pd}/\text{Ir}]_N = 1.2\text{--}2.8$), with flat to positively sloping IPGE
536 and negatively to positively sloping PPGE (Fig. 11c). The Beinn a' Chuailean samples ($n=2$) define mildly
537 fractionated patterns ($[\text{Pd}/\text{Ir}]_N = 1.8\text{--}2.8$), with positively sloping IPGE and flat PPGE (Fig. 11d). These
538 data are further visualized on the Ir versus chondrite-normalized Pd/Ir plot (Fig. 12). The Outer
539 Hebridean samples show moderate scatter, with Ir contents ranging from 0.4 to 2.4 ppb, and Pd/Ir
540 ratios ranging from 0.2 to 8.3.

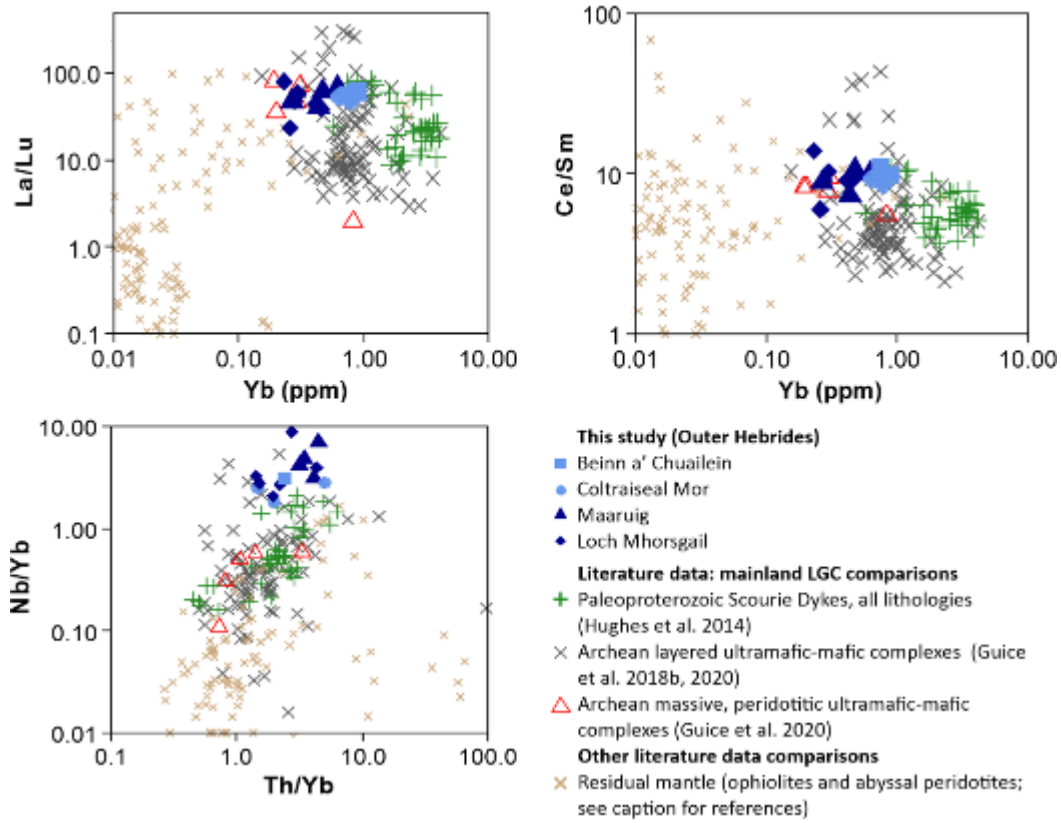


541

542 **Figure 9:** Chondrite-normalized (McDonough and Sun, 1995) REE plots (left-hand column) and primitive mantle-normalized

543 (McDonough and Sun, 1995) trace element plots (right-hand column) for the Maaruig (a–b), Loch Mhorgail (c–d), Coltraiseal

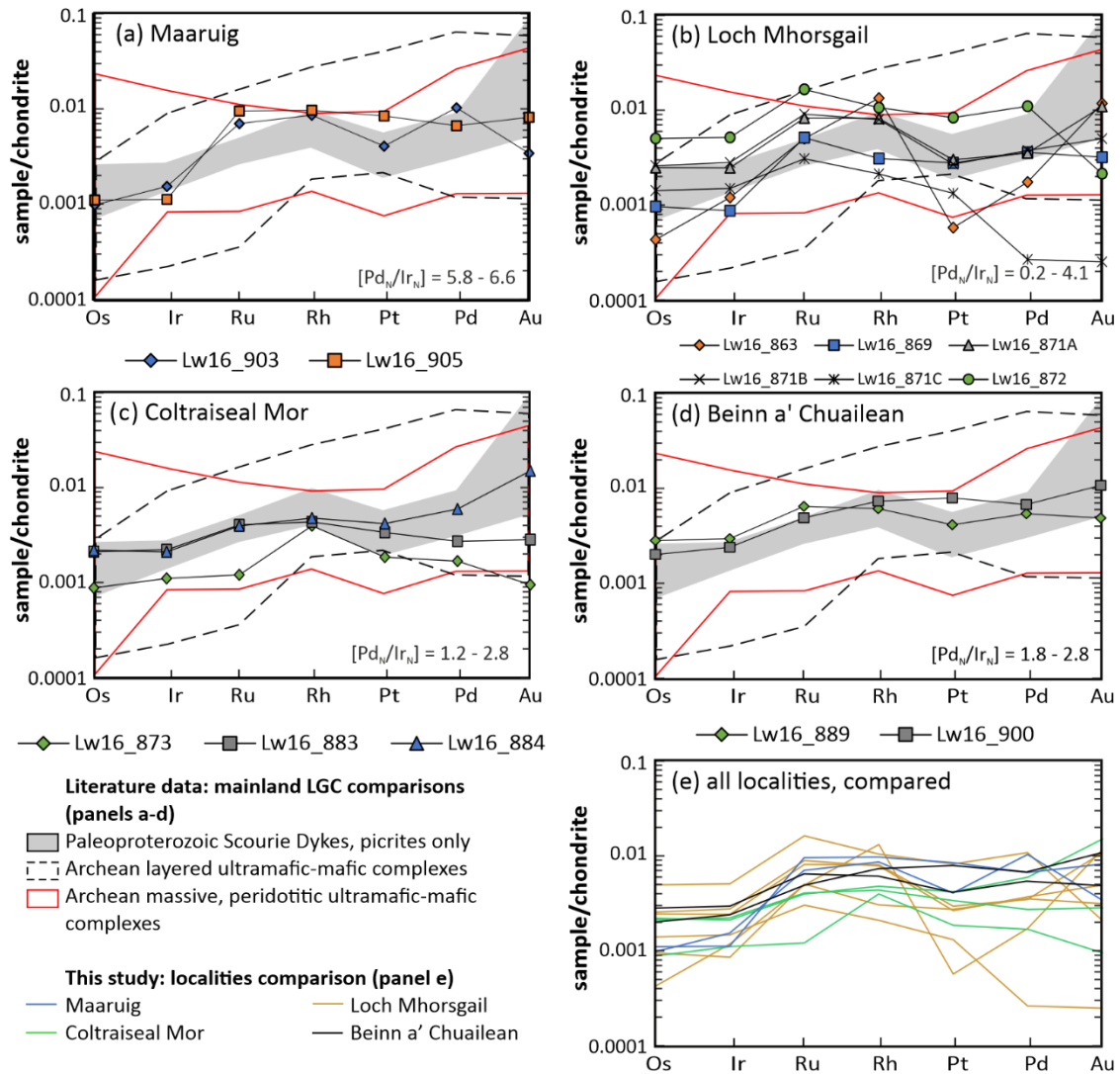
544 Mor (e–f) and Beinn a' Chauilean (g–h) ultramafic–mafic bodies. (i–j) Comparison of all localities.



545

546 **Figure 10:** Yb versus La/Lu (a), Yb versus Ce/Sm (b) and Nb/Yb versus Th/Yb (c) bivariate plots for the rocks analyzed as part

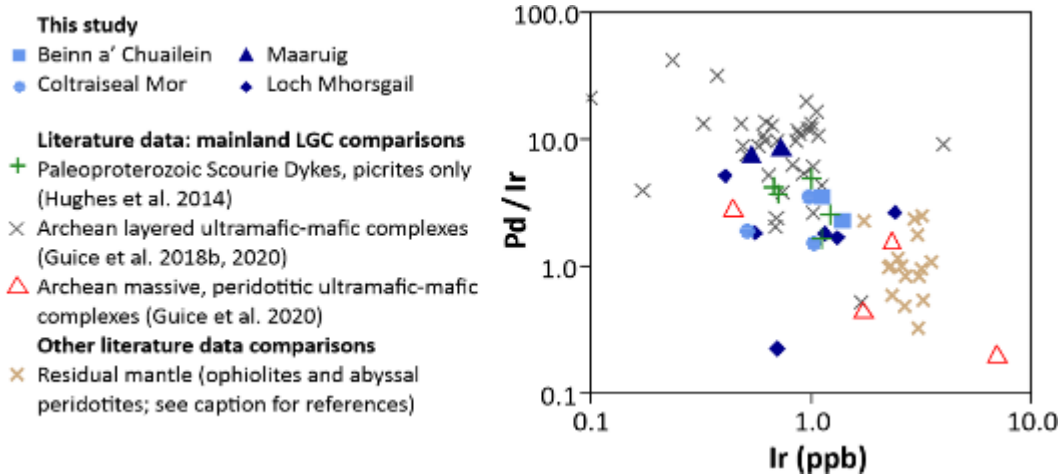
547 of this study. Residual mantle rocks data: Paulick et al. (2006), Godard et al. (2000, 2008).



548

549 **Figure 11:** Chondrite-normalized (Lodders, 2003) platinum-group element (PGE) + Au plots for the analyzed ultramafic rocks.

550 References as in Fig. 9.



551

552 **Figure 12:** Ir versus Pd/Ir for the analyzed ultramafic rocks from the Outer Hebrides.

553 5.3 Comparisons with existing datasets

554 One end-member interpretation for ultramafic remnants in Archean cratons is that they represent
555 fragments of ophiolitic mantle, with ophiolitic hypotheses proposed for a plethora of Archean–
556 Paleoproterozoic ultramafic–mafic complexes globally (Kusky et al., 2001, 2007; Anhaeusser, 2006;
557 Furnes et al., 2007; Polat et al., 2008; Grosch and Slama, 2017). The Maaruig, Loch Mhorskail,
558 Coltraiseal Mor and Beinn a' Chuailean complexes exhibit geochemical characteristics distinct from
559 this field. The major and minor-element compositions show no overlap with the ophiolitic mantle and
560 abyssal peridotites field on bivariate plots (Fig. 7); trace element compositions are enriched by several
561 orders of magnitude relative to those of residual mantle rocks (Figs. 8–9); the bulk-rock Th/Yb and
562 La/Lu ratios are high relative to residual mantle rocks (Fig. 10); and the PGE compositions exhibit
563 relatively low Ir abundances and Pt/Ir ratios (Figs. 11–12).

564 Another possibility is that the studied Outer Hebridean ultramafic–mafic rocks represent direct
565 equivalents of the large group of distinctly layered Archean ultramafic–mafic complexes exposed in
566 the mainland LGC (see Section 2.1.1 for a full description; Fig. 2; Sills et al., 1982; Guice et al., 2018a,
567 2020). The analyzed samples show some overlap with the field for Archean layered complexes on
568 major element bivariate plots, but exhibit relative enrichment in SiO₂ and Cr₂O₃, and relative depletion
569 in Al₂O₃ and CaO (Fig. 7). There are also some similarities in the trace element bulk-rock geochemistry,
570 with samples showing significant overlap with the range of normalized abundances with this field.
571 However, the normalized patterns are distinctive from these Archean layered complexes on both the
572 REE and trace element plots (Fig. 9). Whereas the layered complexes from the mainland LGC exhibit
573 distinctly flat normalized REE and trace element patterns (Fig. 9; Guice et al. 2018b), the Outer
574 Hebridean ultramafic rocks from all localities display negatively sloping LREE (Fig 9a–d) and consistent
575 negative HFSE anomalies (Fig. 9e–h). This chemical distinction is further illustrated by the Yb versus
576 Ce/Sm, Yb versus La/Lu and Nb/Yb versus Th/Yb plots (Fig. 10).

577 Alternatively, the assessed ultramafic–mafic rocks could be correlatives of the generally massive,
578 peridotitic (Archean) complexes in the mainland LGC (see: Guice et al., 2020; Section 2.1.1). On major
579 element bivariate plots, the analyzed rocks are distinct from the small suite of samples for generally
580 massive, peridotitic complexes, displaying relative depletion in SiO₂, MgO and Ni, and relative
581 enrichment in TiO₂, Fe₂O₃, CaO, Na₂O and Cr₂O₃. There are some similarities in the normalized REE and
582 trace element patterns shown by the Maaruig and Loch Mhorskail samples (Fig. 9), with negatively
583 sloping LREE patterns, negative HFSE anomalies and partial overlap with the field for generally
584 massive, peridotitic complexes. However, these samples also show some differences, including
585 relative enrichments in the HREE, some HFSE (e.g., Nb and Ta) and most incompatible elements (e.g.,
586 Rb, Th). Moreover, the Coltraiseal Mor and Beinn a’ Chuailean samples show little or no overlap with
587 this field on normalized REE and trace element plots (Fig. 9c–d, g–h). Samples also show limited
588 overlap with this group of Archean ultramafic-mafic complexes on most trace element bivariate plots
589 (Fig. 8), and on the Nb/Yb versus Th/Yb plot (Fig. 10c).

590 Finally, the studied ultramafic–mafic rocks in Lewis and Harris could be direct equivalents of the
591 Paleoproterozoic (ca. 2.4 Ga) Scourie Dykes exposed in the mainland LGC (see Section 2.1.2), as
592 proposed by several previous authors (e.g., Myers and Lisle, 1971; Davies et al., 1975). The most MgO-
593 poor samples analyzed as part of this study — from the Coltraiseal Mor and Beinn a’ Chuailean
594 complexes — share some geochemical characteristics with the most magnesian mainland Scourie
595 Dykes (classified as picrites). This geochemical similarity can be seen in terms of: major element
596 compositions, with the exception of Ni (Fig. 7); chondrite-normalized REE patterns and normalized REE
597 abundances (Fig. 9c–d); primitive mantle-normalized trace element patterns, which includes negative
598 HFSE anomalies (Fig. 9g–h); and mildly fractionated ($[Pd/Ir]_N = 1.2–2.8$) PGE patterns (Fig. 11c–d; Fig.
599 12). However, while the patterns are parallel to the field for MgO-rich mainland Scourie Dykes, the
600 Coltraiseal Mor and Beinn a’ Chuailean complexes show a small depletion (relative to the MgO-rich
601 mainland Scourie Dykes) in almost all elements on normalized REE and trace element plots (Fig. 9).

602 In contrast, the samples from Maaruig and Loch Mhorgail are geochemically distinct from the
603 mainland Scourie Dykes. They are considerably more magnesian (> 30 wt. % MgO) than the most MgO-
604 rich Scourie Dykes (< 24 wt. % MgO) and show differences in the abundance of most other major
605 elements (Fig. 7). Moreover, samples from these two localities exhibit significant depletion in all trace
606 elements relative to the Scourie Dyke picrites, as illustrated on the trace element bivariate plots (Figs.
607 8, 10), chondrite-normalized REE plot (Fig. 9a–b) and primitive mantle-normalized trace element plots
608 (Fig. 9e–f).

609 **6.0 DISCUSSION**

610 **6.1 Mapping, field observations and relative age constraints**

611 Myers and Lisle (1971) described the ultramafic–mafic bodies in Lewis and Harris as small (generally <
612 0.5 km²) lenses and remnants that can be broadly divided into the following two groups based on
613 lithological characteristics: (1) mafic rocks — comprising < 0.5 cm diameter clinopyroxene and
614 hornblende “clots”, alongside a plagioclase-dominated groundmass — that show relatively
615 homogenous textures and no evidence for layering or chilled margins; and (2) coarse-grained, layered
616 ultramafic–mafic rocks, comprising olivine, orthopyroxene, clinopyroxene, plagioclase and
617 hornblende in variable proportions, alongside minor chromite and phlogopite. Other research
618 conducted by Mason and Brewer (2004) describes a suite of mafic dykes concentrated in Harris that
619 comprise hornblende, plagioclase and quartz, with these occurrences reportedly cross-cutting the TTG
620 gneiss. Most of these ultramafic–mafic rocks in Lewis and Harris are considered younger than the TTG
621 (e.g., Myers and Lisle, 1971), but selected occurrences have been suggested to pre-date the TTG gneiss
622 based on their chaotic distribution within the TTG gneiss (e.g., Mason and Brewer 2004). Fettes and
623 Mendum (1987) describe “Younger Basics” and “Older Basics”, which are interpreted as correlatives
624 of the mainland Scourie Dykes and layered bodies that pre-date the TTG protoliths respectively. In this
625 section, we place the Maaruig, Loch Mhorgail, Coltraiseal Mor and Beinn a’ Chuailean bodies within

626 the context of these previous studies, and consider the relative age relations with the surrounding
627 lithologies.

628 *6.1.1 Maaruig and Loch Mhorgail*

629 The metaperidotite and metapyroxenite samples from Maaruig and Loch Mhorgail have a mineralogy
630 (Table 1; Section 5.1) comparable to the Group 2 occurrences described by Myers and Lisle (1971; see
631 above). This correlation is supported by other aspects of the field characteristics, such as the
632 prominent brown weathered surfaces that stand proud of the surrounding TTG gneiss, and presence
633 of layering on mm-, cm-, dcm-, and m-scales (Section 3; Figs. 3 and 4). These group 2 ultramafic–mafic
634 bodies have previously been considered to represent deformed Paleoproterozoic dykes that are
635 possibly correlatives of the mainland Scourie Dykes (Fig. 2; Myers and Lisle 1971), with this broad
636 hypothesis potentially supported by a ca. 2.1 Ga Sm-Nd (bulk-rock) isochron age yielded from a mafic
637 rock collected from the Maaruig area (Cliff et al. 1998). However, the sample location reported does
638 not correlate with the area mapped here. Rather, our field mapping and observations suggest that the
639 Maaruig and Loch Mhorgail bodies could be older than or broadly coeval with the local TTG gneiss.
640 This interpretation is supported by the map-scale discordance between the ultramafic layering and
641 lithological contacts at both localities (Figs. 3 and 4); and by the presence of meter-scale mafic pods
642 in the TTG immediately adjacent to the ultramafic–mafic bodies at Loch Mhorgail (Fig. 4), which could
643 reflect intrusion by the TTG protoliths or sagduction of ultramafic–mafic material into partially molten
644 TTG (see Johnson et al. 2016 and references therein).

645 Although these observations are not irrefutable evidence of a pre-TTG origin for Maaruig and Loch
646 Mhorgail, an important comparison with the Archean layered bodies of the LGC's mainland can be
647 made, with these occurrences likely post-dating the local TTG gneiss protoliths (Burton et al., 2000;
648 Guice et al., 2018a). This group of mainland ultramafic–mafic complexes exhibit consistent
649 concordance between layering, lithological contacts and the TTG foliation, despite being considerably
650 more deformed than the Maaruig and Loch Mhorgail bodies studied here (Guice et al., 2020).

651 Similarly, there is no spatial relationship between the mainland ultramafic–mafic bodies and meter-
652 scale mafic pods in the surrounding TTG, which is a notable feature of the Loch Mhorgail occurrence.
653 A pre-TTG hypothesis is therefore considered to be more likely than an interpretation whereby these
654 occurrences represent deformed Paleoproterozoic dykes.

655 *6.1.2 Coltraiseal Mor and Beinn a' Chuailean*

656 The metagabbro and meta-olivine gabbro samples from Coltraiseal Mor and Beinn a'
657 Chuailean also contain olivine, orthopyroxene, clinopyroxene amphibole and plagioclase, alongside
658 minor oxide minerals and phlogopite, suggesting that they may also correlate with the Group 2
659 occurrences of Myers and Lisle (1971). However, there are some differences in the field characteristics
660 shown by the Coltraiseal Mor and Beinn a' Chuailean bodies. This includes: relatively homogenous
661 modal mineral proportions between samples (Table 1); presence of plagioclase and phlogopite as
662 major mineral phases (Fig. 6; Table 1); absence of distinctive mineralogical layering on any scale; and
663 consistent parallelism between TTG foliation and lithological boundaries. This raises the possibility
664 that the Coltraiseal Mor and Beinn a' Chuailean bodies represent a different age relationship with the
665 TTG gneiss (and thus record a distinct petrogenesis) to that favoured for Maaruig and Loch Mhorgail
666 (Section 6.1.1).

667 **6.2 Metamorphism and element mobility**

668 Irrespective of the age relations with the TTG gneiss, the studied ultramafic–mafic bodies have all been
669 subject to at least one period of metamorphism: the 1.9–1.6 Ga greenschist–amphibolite-facies
670 Laxfordian tectonothermal event (Fig. 2). This event resulted in local migmatization of the TTG gneiss
671 and the intrusion of associated granites and pegmatites (Dearnley, 1962; Davies et al., 1975; Fettes
672 and Mendum, 1987; Shaw et al., 2016), with such effects most pervasive in the northern and western
673 parts of Lewis (Fig. 1).

674 In the studied thin sections, metamorphic re-crystallization is evidenced by 120° triple junctions shown
675 by 0.5–2.0 mm diameter amphibole grains from all localities (Fig. 6). These rocks show limited

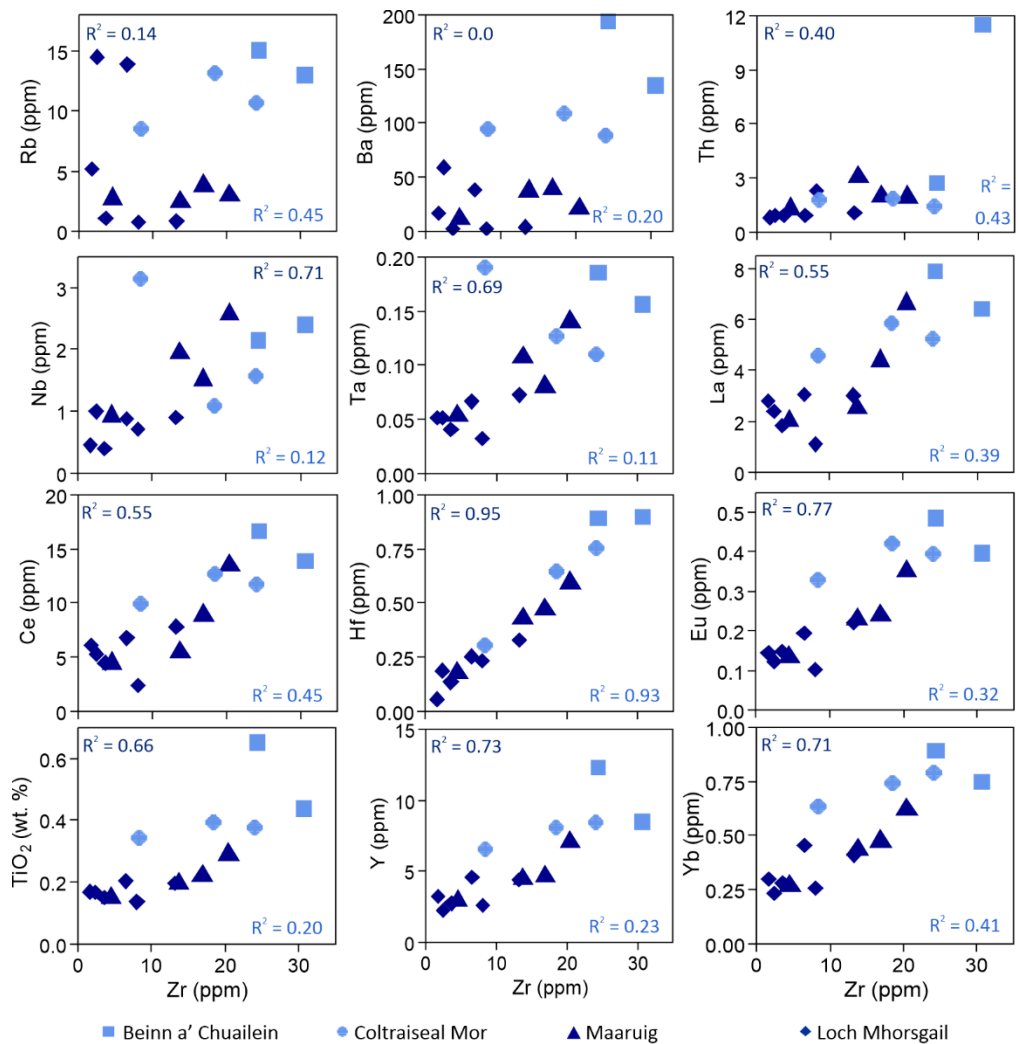
676 petrographic evidence for alteration, with olivine extremely fresh (Fig. 6a–d) and clinopyroxene zoning
677 sometimes preserved (Fig. 6e, g–h). Some samples show slightly more petrographic evidence of
678 alteration (e.g., Lw16_884 from Coltraiseal Mor), which manifests as amphibolitization of
679 clinopyroxene and sericitization of plagioclase (Fig. 6f). However, fresh olivine, pyroxene and
680 plagioclase are all preserved even in the most visibly altered samples.

681 This petrographic evidence, which suggests that the samples studied here have been subject to
682 relatively limited hydrothermal alteration, is supported by the bulk-rock trace element data. Within
683 suites of co-genetic samples, the relative mobility of trace elements can be tested by plotting
684 individual elements against those considered to be most immobile (e.g., Zr, Y and Yb) and calculating
685 the R^2 value (Fig. 13; e.g., Cann, 1970; Guice, 2019). Given the possibility that the Maaruig and Loch
686 Mhorskail bodies were derived from a phase of magmatism distinct from Coltraiseal Mor and Beinn a'
687 Chuailean (see Section 6.1), R^2 values are given (and element mobility tested) for these groups
688 separately (Fig. 13). We here use the samples from Maaruig and Loch Mhorskail to assess element
689 mobility, as they comprise 10 of the 15 samples assessed as part of this study and are therefore more
690 statistically robust.

691 When plotted against Zr on bivariate plots (Fig. 13), a selection of elements, including Yb, Y, Eu and
692 Hf, show high R^2 values ($R^2 \geq 0.7$) for the Maaruig and Loch Mhorskail samples. This suggests that these
693 elements have experienced limited bulk-rock element mobility due to metamorphism and
694 hydrothermal alteration. A second group of elements — including TiO_2 , La, Ce, Nb, Ta and Th — show
695 moderate correlations ($R^2 = 0.4–0.7$) with Zr that likely reflect small amounts of element mobility. The
696 interpretation of these elements as having experienced limited element mobility during
697 metamorphism is supported by the lack of correlation between trace element composition and
698 amphibole modal abundance, which is a phase derived from the alteration of clinopyroxene (see
699 Section 5.1). Samples LW16_873 and LW16_889 contain 45 modal % and 15 modal % amphibole

700 respectively, but exhibit near-identical normalized trace-element patterns for the elements
 701 considered immobile or as having experienced limited mobility (Fig. 9).

702 A final group of elements — Rb ($R^2 = 0.11$), Ba ($R^2=0.0$) and Cu ($R^2 = 0.01$) — display poor correlations
 703 with Zr (Fig. 13). Given their poor correlation with Zr, these elements likely experienced significant
 704 element mobility, with abundances in the studied rocks not reflective of primary magmatic
 705 compositions. As such, genetic interpretations are not based on these elements. Taken together, the
 706 studied ultramafic–mafic samples have experienced relatively limited bulk-rock element mobility as a
 707 consequence of metamorphism and associated hydrothermal alteration.



709 **Fig. 13:** Bivariate plots detailing the correlation between Zr and various trace elements. Separate R^2 values are provided for
 710 Maaruig and Loch Mhorgail (dark blue), and for Coltraiseal Mor and Beinn a' Chuailean (light blue).

711 **6.3 Petrogenesis of the studied ultramafic–mafic bodies**

712 Ultramafic rocks can be produced in a variety of geological and tectonic environments, with several of
713 these hypotheses proposed for ultramafic–mafic rocks exposed in the mainland LGC. This includes,
714 but is not limited to: the lower portions of ophiolites/ oceanic lithosphere, including the mantle
715 portion (Park and Tarney, 1987; Guice et al., 2020); komatiites that once formed part of a greenstone
716 belt and were sagducted into the TTG (Johnson et al., 2016); cumulates, whose parental magmas could
717 potentially be derived from several tectonic environments (Bowes et al., 1964; Guice et al., 2018a); or
718 extension-related dykes (Hughes et al., 2014 and references therein).

719 In this section, we discuss the origin of the Maaruig, Loch Mhorskail, Coltraiseal Mor and Beinn a'
720 Chuailean bodies, and consider potential correlations with ultramafic–mafic rocks exposed in the
721 mainland LGC's Central Region (see Section 2.1 for descriptions of these rocks). A comparison between
722 the data presented here and various global/regional-scale datasets is provided in Section 5.3. Based
723 on the chemical distinction between all analyzed ultramafic–mafic rocks studied here and residual
724 mantle rocks, we consider it improbable that any of the analyzed samples are fragments of ophiolitic
725 mantle. This hypothesis is therefore not considered in the succeeding sections.

726 *6.3.1 Maaruig and Loch Mhorskail*

727 As discussed in Section 6.1, we consider it most likely that the Maaruig and Loch Mhorskail bodies are
728 older than or coeval with the local TTG gneiss. This Archean-age interpretation contradicts the broad
729 hypothesis that these rocks represent deformed Paleoproterozoic dykes and, by extension, questions
730 any direct correlation with the mainland Scourie Dykes (Dearnley, 1962; Myers and Lisle, 1971; Davies
731 et al., 1975; Fettes and Mendum, 1987; Cliff and Rex, 1989). Moreover, other field and geochemical
732 evidence contradict a direct equivalence between the Scourie Dykes and the Maaruig/Loch Mhorskail
733 bodies. While subtle layering is present in some Scourie Dykes, the scale and prominence of the
734 layering presented in the Maaruig and Loch Mhorskail bodies (Section 3.1–3.2) is not observed in the
735 Scourie Dykes. The Maaruig and Loch Mhorskail samples are also extremely MgO-rich (> 30 wt. %

736 MgO). For comparison, most previously analyzed Scourie Dykes generally contain < 10 wt. % MgO,
737 with a small group of picrites containing 15–25 wt. % MgO (Fig. 7).

738 Instead, both Maaruig and Loch Mhorgsail exhibit several features characteristics of cumulates, with
739 evidence for *in-situ* fractionation demonstrated by the distinctive modal layering observed on mm-,
740 cm-, dcm- and m-scales. *In-situ* fractionation is also evidenced by the chemical data, with most major
741 elements correlated with MgO (see Fig. 7), alongside fractionated PGE patterns (Fig. 11a–b). Given
742 this interpretation, it is unlikely that these ultramafic bodies can be correlated with the “generally
743 massive, peridotitic” (Archean) complexes exposed in the mainland LGC (see Section 2.1), with this
744 distinction supported by the chemical data (see Section 5.3). It is, however, potentially suggestive of
745 a correlation with the Archean “layered” complexes from the mainland LGC (see Fig. 2). Like this group
746 of Archean ultramafic–mafic rocks, the Maaruig and Loch Mhorgsail occurrences display distinctive
747 mineralogical layering on a variety of scales (Sills et al., 1982; Guice et al., 2018a). There are also some
748 chemical similarities, with significant overlap with the field for Archean layered complexes on
749 normalized REE and trace element plots (Fig. 9).

750 Despite this overlap, the flat patterns shown by the “layered” mainland ultramafic–mafic complexes
751 are distinct from the patterns shown by the Maaruig and Loch Mhorgsail samples, which exhibit
752 distinctive negative HFSE anomalies and LREE enrichment. Moreover, other aspects of the field
753 relationships are distinct from this group. Rather than forming relatively thin (on a scale of tens of
754 metres) units that can be traced for several hundred meters along strike (Guice et al., 2018a), the
755 Maaruig and Loch Mhorgsail rocks occur as elliptical-shaped pods that exhibit discordance between
756 layering and lithological contacts (see Section 6.1). As such, we consider it unlikely that the studied
757 Outer Hebridean ultramafic–mafic bodies are direct equivalents of the layered complexes exposed in
758 the mainland LGC, and Maaruig and Loch Mhorgsail may thus represent a distinct type of ultramafic–
759 mafic magmatism in the LGC that is currently recognized only in the Outer Hebridean islands west of
760 the OHFZ.

761 An alternative hypothesis is that the Maaruig and Loch Mhorgsail bodies represent cumulates that are
762 genetically related to the Scourie Dykes. However, as well as being unlikely based on the favoured age
763 relationships (Section 6.1), this interpretation is inconsistent with normalized trace element patterns
764 shown by the Maaruig and Loch Mhorgsail samples. These samples exhibit negative HFSE anomalies
765 and are parallel with those of the Scourie Dykes, but are depleted in all trace elements relative to this
766 field (see Fig. 9). If they were cumulate parents of the melts that formed the Scourie Dykes,
767 complimentary geochemical signatures would be expected, rather than the parallel patterns observed
768 (Charlier et al., 2005; Ma et al., 2016; Schaen et al., 2017). For example, ultramafic cumulates from
769 the Nincheng complex (North China Craton) are highly depleted in Sr and LREE, with the derivative
770 melts (preserved as adakites) exhibiting relatively high Sr/Y and La/Yb ratios (Ma et al., 2016).

771 The distinctive negative HFSE anomalies — alongside the associated LREE enrichment shown by all of
772 the analyzed samples from Maaruig and Loch Mhorgsail (as well as Coltraiseal Mor and Beinn a'
773 Chuailean) — are a bulk-rock geochemical signature that could reflect one or more of the following
774 potential origins: (a) parental magmas derived from subduction-related processes (Klemme et al.,
775 2005); (b) crustal contamination during magma ascent (Arndt, 1999); (c) secondary processes, such as
776 interaction with hydrothermal fluids (Lahaye et al., 1995); (d) cumulate effects, whereby LREE-rich
777 minerals are concentrated in these specific layers/units whilst the HFSE have been retained in the
778 residual magma; and (e) parental magmas partially derived from a previously metasomatized sub-
779 continental lithospheric mantle (Hughes et al., 2014). While it is not possible to assess these
780 hypotheses robustly using the data presented here, several comments can be made. First, as the
781 Maaruig and Loch Mhorgsail rocks are considered to have experienced limited element mobility (see
782 Section 6.2), hypothesis (c) is considered unlikely, but is not completely ruled out given the small
783 sample set and potentially cryptic effects of hydrothermal alteration in Archean cratons (Guice et al.,
784 2018b). Second, no distinctly LREE-rich mineral phases are identified in the Maaruig and Loch
785 Mhorgsail samples. Given the evidence for limited hydrothermal alteration, which is a process capable
786 of generating LREE enrichment (Rollinson and Gravestock, 2012; Guice et al., 2018b), hypothesis (d) is

787 also considered improbable, but not ruled out. It is therefore likely that the distinctive geochemical
788 signature is derived from the parental magma, but the specific process(es) responsible remains an
789 open question.

790 *6.3.2 Coltraiseal Mor and Beinn a' Chuailean*

791 One hypothesis is that these two ultramafic–mafic bodies are genetically related to the ultramafic
792 rocks at Maaruig and Loch Mhorgail, with all occurrences representing variably fractionated products
793 of the same parental magma. Despite the aforementioned differences in field characteristics (see
794 above; Section 6.1), this hypothesis is potentially supported by some of the petrographic and chemical
795 characteristics. The mineral assemblage is broadly comparable to Maaruig and Loch Mhorgail, with
796 the main differences being the prevalence of clinopyroxene, plagioclase and phlogopite in the
797 Coltraiseal Mor and Beinn a' Chuailean samples, which could potentially be attributed to magmatic
798 differentiation. Moreover, the chondrite-normalized REE and primitive mantle-normalized patterns
799 shown by all four localities studied here are parallel to one another.

800 Alternatively, there are several geochemical similarities between the Coltraiseal Mor and Beinn a'
801 Chuailean samples and the mainland Scourie Dykes (Section 5.3), raising the alternate possibility that
802 these occurrences are Outer Hebridean equivalents of this well-documented suite of ca. 2.4 Ga dykes
803 exposed in the mainland LGC (Fig. 2; Section 2.1.2). This hypothesis is potentially supported by the
804 favoured age relationship with the TTG gneiss (see Section 6.1), and by several field characteristics.
805 Unlike Maaruig and Loch Mhorgail, Coltraiseal Mor and Beinn a' Chuailean show relatively consistent
806 modal mineral proportions between samples, alongside an absence of modal mineralogical layering.
807 On balance, we consider this latter hypothesis more likely, whereby the Coltraiseal Mor and Beinn a'
808 Chuailean are Paleoproterozoic dyke fragments that could be direct equivalents of the Scourie Dykes
809 exposed on the mainland.

810 **6.4 Implications for the Lewisian Gneiss Complex, North Atlantic Craton**

811 This study identifies a new phase of Archean ultramafic–mafic magmatism in the LGC: a suite of
812 Archean cumulates in the Outer Hebrides of Lewis and Harris, as represented by the Maaruig and Loch
813 Mhorsgail bodies. In terms of their field, petrographic and geochemical characteristics, these
814 occurrences are distinct from any of the Archean ultramafic–mafic magmatism currently described in
815 the mainland LGC’s well-studied Central Region (see Section 2.1). This raises the possibility that Lewis
816 and Harris (west of the OHFZ) formed a distinct crustal block that was separate from the mainland
817 LGC’s Central Region during the Mesoarchean, although the implications of this finding are open to
818 interpretation. These two (or more) crustal blocks could have been separate terranes during the
819 Mesoarchean, with their unique magmatic evolutions during this Era reflective of their origin as
820 separate (micro)continents (e.g., Mason and Brewer, 2004; Kinny et al., 2005; Love et al., 2010).
821 Alternatively, it is possible that the distinctive Mesoarchean evolution reflects heterogeneity within a
822 once continuous piece of Archean crust.

823 In either case, the interpretation of the Coltraiseal Mor and Beinn a’ Chuailean bodies as deformed
824 Paleoproterozoic dykes that could be correlatives of the mainland LGC Scourie Dykes — as is
825 considered most likely (see Section 6.3.2) — suggests that the Outer Hebrides (west of the OHFZ) and
826 mainland portions of the LGC were proximal to one another by ca. 2.4 Ga. This hypothesis is consistent
827 with recent U-Pb zircon geochronology conducted by Davies and Heaman (2014), who assigned a 2.41
828 Ga crystallization age to the Cleitichean Beag Dyke, which is located in central Lewis and is considered
829 to be an Outer Hebridean “Scourie Dyke” (Fig. 1b). When combined with the identification of a distinct
830 phase of Archean ultramafic–mafic magmatism in Lewis and Harris (represented by Maaruig and Loch
831 Mhorsgail), this finding suggests that a major tectonic juxtaposition — whether via subduction-
832 accretion or large-scale crustal faulting — likely occurred in the LGC in the Neoproterozoic or early
833 Paleoproterozoic prior to 2.42 Ga.

834 **6.5 Ultramafic–mafic rocks: a crucial component of the early Earth puzzle**

835 As described in the introduction to this paper, the Archean–Paleoproterozoic evolution of the LGC is
836 described by competing models, including end-member models whereby it is interpreted to represent:
837 (a) a section of broadly continuous Archean crust (Park and Tarney, 1987); or (b) multiple geologically
838 unique terranes assembled via plate tectonic processes (Kinny et al., 2005 and references therein).
839 This craton-scale debate reflects a broader discussion about the nature and evolution of Earth’s
840 geodynamic regime(s) during the Archean and Proterozoic Eons, which remains a topic of significant
841 debate (e.g., Arndt, 2013; Kamber, 2015; Bédard, 2018).

842 In the last 10 years, several important contributions to this research topic have assessed the temporal
843 evolution of several geochemical and geological proxies, including the thermobarometric ratio
844 recorded by metamorphic rocks (e.g., Brown, 2008; Brown and Johnson, 2018; Holder et al., 2019;
845 Brown et al., 2020) and the bulk-rock composition of mafic and/or granitoid rocks (Dhuime et al., 2015;
846 Condie, 2018; Moyen and Laurent, 2018; Smithies et al., 2018; Johnson et al., 2019). These records
847 highlight evidence for a significant evolution on Earth between 3.5 and 2.2 Ga, which is often
848 interpreted as reflecting the onset of plate tectonics (Cawood et al., 2018; Tang et al., 2018). Other
849 authors point out that direct geological evidence for plate tectonics — such as the presence of
850 ophiolites and blueschists — are almost exclusively concentrated in rocks younger than 1 Ga (Stern,
851 2008, 2020). Alternatively, zircon grains from Jack Hills record an emergence of peraluminous granitic
852 rocks at 3.6 Ga, which could be the product of widespread subduction and the onset of plate tectonics
853 (Ackerson et al., 2021).

854 By combining these important observations with more spatially focused projects aiming to constrain
855 the petrogenesis of units within their geological context, it may be possible to establish with greater
856 confidence the specific geodynamic processes that operated during the Archean and Proterozoic Eons,
857 and how these processes evolved through time. Ultramafic–mafic rocks have the potential to provide
858 particularly important insights due to their formation in a broad-range of geological environments. In

859 the LGC, these potential interpretations include: layered intrusions remnants (Bowes et al., 1964;
860 Guice et al., 2018a, 2020); accreted oceanic crust (Park and Tarney, 1987); fragments of Archean
861 mantle (Guice et al., 2020); sagducted remnants of one or more greenstone belt(s) (Johnson et al.,
862 2016); and mafic dyke fragments (Mason and Brewer 2004). The historic limitation — when
863 attempting to constrain which of these hypothesis is most likely for individual, or suites of, Archean–
864 Paleoproterozoic ultramafic–mafic bodies — is that the primary stratigraphy is disrupted by
865 deformation and the primary geochemistry affected by metamorphism (e.g., Guice et al., 2018a,
866 2018b).

867 In this study, we have combined detailed field observations with widely available petrographic and
868 geochemical techniques to demonstrate that four apparently similar ultramafic–mafic bodies in the
869 Lewisian Gneiss Complex of the Outer Hebrides likely represent multiple magmatic events. We have
870 also deciphered broad geological environments for their formation and considered how they relate,
871 petrogenetically and temporally, to other ultramafic–mafic rocks in the LGC and broader NAC (Section
872 6.4). This adds to a number of recent papers — focusing on ultramafic–mafic rocks — that have
873 successfully utilized similar approaches to extract crucial information about the evolution of various
874 Archean cratons (Szilas et al., 2014, 2015; Anhaeusser, 2015; Grosch and Slama, 2017; Guice et al.,
875 2018a, 2018b, 2019; Pinheiro et al., 2021). Such first-order geological constraints are vitally important
876 if we are to successfully apply modern, high-precision isotopic constraints (e.g., Sm-Nd; Re-Os) to
877 ultramafic–mafic bodies and, ultimately, utilize their potential to provide crucial insights into the
878 geodynamic processes responsible for the creation and destruction of Earth’s lithosphere during the
879 Archean and Proterozoic Eons.

880 **7.0 CONCLUSIONS**

881 1. This study identifies a previously unrecognized phase of ultramafic–mafic magmatism in the LGC: a
882 suite of Archean cumulates represented by the Maaruig and Loch Mhorskail bodies in the Outer
883 Hebrides of Lewis and Harris. These occurrences are likely older than, or broadly coeval with, the

884 protoliths to the TTG gneiss that comprise the majority of the LGC (> 2.8 Ga). They are also
885 geochemically distinct from anything currently identified in the mainland LGC's Central Region,
886 potentially suggesting that the Outer Hebrides of Lewis and Harris (west of the OHFZ) was a distinct
887 crustal block to that of the mainland LGC's Central Region in the late Mesoproterozoic.

888 2. A second group of ultramafic–mafic bodies — represented by the Coltraiseal Mor and Beinn a'
889 Chuailean bodies — are likely dismembered Paleoproterozoic dykes that could be correlatives of the
890 mainland Scourie dykes (extension-related, ca. 2.4 Ga ultramafic–mafic dykes). This supports recent
891 U-Pb zircon geochronology (Davies and Heaman 2014), which assigned a 2.41 Ga crystallization age to
892 a proposed Scourie Dykes in the Outer Hebrides (the Cleitichean Beag Dyke). It also suggests that the
893 Outer Hebridean and mainland portions of the LGC were proximal to one another by the early
894 Paleoproterozoic, placing an important constraint on the tectonic evolution of the LGC.

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903 **TABLES**

904 **Table 1:** Modal mineralogy for each sample assessed as part of this study. Abbreviations: Col. Mor =

905 Coltraiseal Mor; BaC = Beinn a' Chuailean; ol = olivine; opx = orthopyroxene; cpx = clinopyroxene;

906 amph = amphibole; ox = oxide phases; plag = plagioclase; phlo = phlogopite; Acc. = accessory phase.

Locality	Sample	Grid ref (NB)	ol	opx	cpx	amph	ox	plag	phlo	Rock name
Maaruig	Lw16_827	20398/06292	60	6	2.5	30	1.5			Metaperidotite
Maaruig	Lw16_828	20502/06187	40	22.5	1	35	1.5			Metapyroxenite
Maaruig	Lw16_903	20332/06278	79	11	1	7	2			Metaperidotite
Maaruig	Lw16_905	20023/06159	35	17	2	45	1			Metapyroxenite
Mhorsgail	Lw16_863	13415/22692	27	22	3	46	2			Metapyroxenite
Mhorsgail	Lw16_869	13409/22624	34	40		25	1		Acc.	Metapyroxenite
Mhorsgail	Lw16_871a	13516/22167	35	15		47	2.5		0.5	Metapyroxenite
Mhorsgail	Lw16_871b	13516/22167	68			30	2	Acc.	Acc.	Metaperidotite
Mhorsgail	Lw16_871c	13516/22167	50	5		43	2			Metaperidotite
Mhorsgail	Lw16_872	13726/21610	34	22		43	1		Acc.	Metapyroxenite
Col. Mor	Lw16_873	16498/22005	5	38	4	45	Acc.	6	2	Metagabbronorite
Col. Mor	Lw16_883	16546/22050	4	39	10	31	Acc	15	1	Metagabbronorite
Col. Mor	Lw16_884	16570/22091	5	38	12	30	1	12.5	1.5	Metagabbronorite
BaC	Lw16_899	19791/24719	15	40	20	15		8	2	Meta-ol gabbronorite
BaC	Lw16_900	19741/24717	3	25	25	19		8	20	Metagabbronorite

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917 **Table 2:** Major and trace element analyses for each of the samples analyzed as part of this study.

Locality Sample (Lw16_)	Maaruig				Loch Mhorskail					
	827	828	903	905	863	869	871A	871B	871C	872
Major elements (wt. %)										
SiO ₂	42.76	43.77	42.95	42.34	44.33	46.41	40.14	44.10	43.55	46.57
TiO ₂	0.19	0.22	0.15	0.29	0.14	0.15	0.17	0.20	0.17	0.20
Al ₂ O ₃	4.35	3.79	2.31	4.91	1.99	2.39	2.54	3.38	2.84	3.40
Fe ₂ O ₃	13.79	13.45	12.33	13.65	13.63	13.06	13.15	11.79	12.48	12.11
MnO	0.18	0.18	0.17	0.18	0.20	0.20	0.18	0.16	0.17	0.18
MgO	33.60	33.92	38.68	32.69	34.37	33.85	39.58	34.74	36.59	32.01
CaO	3.39	2.86	1.78	3.98	4.06	2.69	2.49	3.67	2.35	3.70
Na ₂ O	0.61	0.64	0.32	0.78	0.32	0.31	0.32	0.51	0.34	0.63
K ₂ O	0.16	0.19	0.12	0.22	0.06	0.05	0.14	0.29	0.30	0.08
P ₂ O ₅	0.02	0.03	0.02	0.05	0.01	0.01	0.03	0.02	0.02	0.02
LOI	2.46	2.03	4.76	1.71	2.10	1.13	3.77	1.69	2.56	1.14
trace elements (ppm)										
V	93.1	83.5	62.6	104.1	95.4	88.7	76.4	90.1	80.3	96.8
Cr	6415	6362	7607	6461	5888	5536	8408	7437	7765	7404
Co	100.7	98.1	97.7	206.4	93.6	93.0	103.2	88.0	95.3	88.4
Ni	1927	1768	2092	1692	1570	1325	2141	1899	2005	1674
Cu	16.63	26.94	10.52	132.94	8.65	10.77	12.18	11.30	14.56	16.35
Zn	129.9	124.6	107.6	152.5	112.3	100.4	111.8	98.6	119.6	91.4
Ga	4.94	4.41	3.02	5.96	2.57	3.11	3.45	4.68	3.80	5.62
Rb	2.46	3.76	2.72	2.94	0.74	1.07	5.16	13.84	14.48	0.81
Sr	51.00	62.93	20.37	78.66	18.73	23.44	18.56	23.67	19.27	41.22
Y	4.46	4.61	2.94	7.04	2.57	2.74	3.21	4.57	2.26	4.38
Zr	13.74	16.91	4.56	20.45	8.08	3.61	1.71	6.56	2.46	13.24
Nb	1.94	1.51	0.92	2.57	0.71	0.40	0.45	0.88	1.00	0.90
Cs	n.d	0.07	0.04	0.03	n.d	0.03	0.15	0.40	0.30	n.d
Ba	37.17	39.03	11.22	20.70	2.46	2.05	16.58	37.96	59.12	4.00
La	2.51	4.38	2.00	6.61	1.10	1.83	2.79	3.05	2.40	3.02
Ce	5.43	8.84	4.42	13.50	2.35	4.48	6.06	6.76	5.31	7.81
Pr	0.69	0.98	0.53	1.54	0.28	0.54	0.72	0.77	0.53	0.91
Nd	3.10	4.08	2.24	6.35	1.40	2.24	2.89	3.26	1.93	3.72
Sm	0.74	0.84	0.50	1.36	0.39	0.49	0.59	0.75	0.38	0.86
Eu	0.23	0.24	0.13	0.35	0.10	0.15	0.14	0.19	0.12	0.22
Gd	0.67	0.78	0.44	1.20	0.41	0.40	0.48	0.69	0.37	0.72
Tb	0.13	0.14	0.08	0.21	0.07	0.07	0.09	0.12	0.06	0.12
Dy	0.74	0.77	0.47	1.20	0.47	0.43	0.54	0.71	0.38	0.74
Ho	0.15	0.15	0.10	0.23	0.10	0.09	0.11	0.15	0.08	0.14
Er	0.41	0.44	0.25	0.65	0.26	0.27	0.29	0.43	0.21	0.40
Tm	0.07	0.06	0.04	0.10	0.04	0.05	0.05	0.06	0.03	0.06
Yb	0.43	0.47	0.27	0.62	0.26	0.28	0.30	0.45	0.23	0.41
Lu	0.06	0.07	0.04	0.10	0.05	0.04	0.05	0.08	0.03	0.07
Hf	0.43	0.47	0.18	0.59	0.23	0.13	0.06	0.25	0.19	0.33
Ta	0.11	0.08	0.05	0.14	0.03	0.04	0.05	0.07	0.05	0.07
Pb	0.59	0.93	0.51	0.80	1.80	0.30	2.15	4.23	50.54	0.66
Th	3.06	1.98	1.29	1.95	2.28	0.91	0.82	0.94	0.92	1.09
U	0.09	0.17	0.10	0.31	0.05	0.08	0.12	0.12	0.09	0.11
Normalized trace element ratios										
[La/Lu] _{ch} N	4.13	6.33	4.78	6.98	2.39	4.83	6.06	4.10	8.27	4.56
[La/Sm] _{ch} N	2.11	3.27	2.49	3.04	1.74	2.33	2.98	2.54	3.90	2.20
[Nb/La] _{pm} N	0.76	0.34	0.45	0.38	0.63	0.21	0.16	0.28	0.41	0.29
[Th/Yb] _{pm} N	39.10	23.12	26.70	17.37	48.80	18.16	15.28	11.46	21.93	14.80
[Ta/Yb] _{pm} N	2.94	2.02	2.38	2.69	1.49	1.73	2.06	1.75	2.63	2.11

Locality Sample (Lw16_)	Coltraiseal Mor			Beinn a' Chuailein	
	873	883	884	889	900
Major elements (wt. %)					
SiO ₂	52.70	51.37	50.54	51.22	50.96
TiO ₂	0.38	0.39	0.35	0.44	0.65
Al ₂ O ₃	5.78	6.56	5.72	6.26	7.62
Fe ₂ O ₃	10.20	11.76	12.29	11.91	12.76
MnO	0.18	0.19	0.19	0.19	0.19
MgO	20.29	21.94	23.55	21.85	18.96
CaO	8.67	6.00	5.78	6.25	6.35
Na ₂ O	0.93	0.88	0.71	0.95	1.39
K ₂ O	0.31	0.41	0.32	0.44	0.67
P ₂ O ₅	0.02	0.04	0.03	0.05	0.08
LOI	0.00	1.12	0.00	0.00	0.00
trace elements (ppm)					
V	180.0	145.5	148.3	146.1	178.3
Cr	3725	3172	3373	3089	2641
Co	55.6	66.8	587.6	66.2	60.9
Ni	709	862	1087	850	543
Cu	28.83	26.79	648.88	39.91	41.48
Zn	77.6	95.0	94.4	100.0	94.2
Ga	9.04	7.57	7.49	8.99	9.47
Rb	10.61	13.11	8.51	12.97	15.02
Sr	78.52	104.23	84.72	91.12	91.03
Y	8.40	8.07	6.54	8.49	12.27
Zr	24.03	18.43	8.36	30.65	24.38
Nb	1.56	1.09	3.13	2.40	2.14
Cs	0.18	0.15	0.12	0.18	0.21
Ba	88.00	108.76	94.35	134.67	194.10
La	5.23	5.86	4.58	6.41	7.86
Ce	11.73	12.68	9.96	13.87	16.65
Pr	1.40	1.49	1.17	1.63	1.88
Nd	5.87	5.89	4.93	6.61	7.88
Sm	1.39	1.23	1.07	1.27	1.68
Eu	0.39	0.42	0.33	0.40	0.49
Gd	1.32	1.20	1.08	1.43	1.58
Tb	0.23	0.22	0.19	0.22	0.27
Dy	1.45	1.28	1.15	1.39	1.62
Ho	0.27	0.24	0.22	0.26	0.29
Er	0.82	0.75	0.59	0.73	0.82
Tm	0.13	0.11	0.10	0.11	0.14
Yb	0.79	0.74	0.63	0.75	0.89
Lu	0.11	0.11	0.09	0.12	0.13
Hf	0.75	0.64	0.31	0.90	0.89
Ta	0.11	0.13	0.19	0.16	0.19
Pb	2.01	2.31	1.39	4.04	3.63
Th	1.44	1.89	1.79	11.49	2.74
U	0.19	0.23	0.17	0.24	0.30
Normalized trace element ratios					
[La/Lu] _{ch} N	4.86	5.77	5.53	5.75	6.29
[La/Sm] _{ch} N	2.35	2.98	2.66	3.15	2.93
[Nb/La] _{pm} N	0.29	0.18	0.67	0.37	0.27
[Th/Yb] _{pm} N	10.13	14.16	15.66	85.42	17.02
[Ta/Yb] _{pm} N	1.66	2.03	3.58	2.49	2.48

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