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**Geochemistry of serpentized and multiphase altered
Atlantis Massif peridotites (IODP Expedition 357):
Petrogenesis and discrimination of melt-rock vs.
fluid-rock processes**

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41

42

Abstract

43 International Ocean Discovery Program (IODP) Expedition 357 drilled 17
44 shallow sites distributed ~ 10 km in the spreading direction (from west to east) across the
45 Atlantis Massif oceanic core complex (Mid-Atlantic Ridge, 30 °N). Mantle exposed in
46 the footwall of the Atlantis Massif oceanic core complex is predominantly nearly wholly
47 serpentinized harzburgite with subordinate dunite. Altered peridotites are subdivided into
48 three types: (I) serpentinites, (II) melt-impregnated serpentinites, and (III) metasomatic
49 serpentinites. Type I serpentinites show no evidence of melt-impregnation or
50 metasomatism apart from serpentinization and local oxidation. Type II serpentinites have
51 been intruded by gabbroic melts and are distinguishable in some cases on the basis of
52 macroscopic and microscopic observations, e.g., mm-cm scale mafic-melt veinlets, rare
53 plagioclase (<0.5 modal % in one sample) or by the local presence of secondary
54 (replacive) olivine after orthopyroxene; in other cases, ‘cryptic’ melt-impregnation is
55 inferred on the basis of incompatible element enrichments. Type III serpentinites are
56 characterized by silica metasomatism manifested by alteration of orthopyroxene to talc
57 and amphibole, and by anomalously high anhydrous SiO₂ concentrations (59-61 wt.%)
58 and low MgO/SiO₂ values (0.48-0.52). Although many chondrite-normalized rare earth
59 element (REE) and primitive mantle-normalized incompatible trace element anomalies,
60 e.g., negative Ce-anomalies, are attributable to serpentinization, other compositional
61 heterogeneities are due to melt-impregnation. On the basis of whole rock incompatible

62 trace elements, a dominant mechanism of melt-impregnation is distinguished in the
63 central and eastern serpentinites from fluid-rock alteration (mostly serpentinization) in
64 the western serpentinites, with increasing melt-impregnation manifest as a west to east
65 increase in enrichment in high-field strength elements and light REE. High degrees of
66 melt extraction are evident in low whole-rock $\text{Al}_2\text{O}_3/\text{SiO}_2$ values and low concentrations
67 of Al_2O_3 , CaO and incompatible elements. Estimates of the degree of melt extraction
68 based on whole rock REE patterns suggest a maximum of $\sim 20\%$ non-modal fractional
69 melting, with little variation between sites. As some serpentinite samples are *ex situ*
70 rubble, the magmatic histories observed at each site are consistent with a local source
71 (from the fault zone) rather than rafted rubble that would be expected to show more
72 heterogeneity and no spatial pattern. In this case, the studied sites may provide a record
73 of enhanced melt-rock interactions with time, consistent with proposed geological
74 models. Alternatively, sites may signify heterogeneities in these processes at spatial scales
75 of a few km.

76

77 *Keywords:* IODP Expedition 357; Atlantis Massif; mantle peridotite; fluid-rock interaction;
78 melt-rock interaction

79 **1. Introduction**

80 Spreading rate exerts a profound influence on the geometry and architecture of
81 the oceanic crust. Slow-spreading ridges (≤ 5 cm/year), as exemplified by the Mid-
82 Atlantic ridge (MAR), are characterized by wide (up to 30 km) and deep (1–2 km) axial
83 valleys bounded by uplifted shoulders and transient magma reserves. In contrast, fast-
84 spreading ridges (≥ 9 cm/year), as characterized by the East Pacific Rise, exhibit much
85 narrower to absent axial troughs (a few hundred meters wide) and a more continuous
86 magma supply (e.g., Macdonald, 1982; Karson, 2002; Stewart et al., 2005). Analogous
87 with ‘metamorphic core complexes’ found in extended continental regions (e.g., John,
88 1987), ‘oceanic core complexes’ (OCCs), such as the Atlantis Massif (30 °N, Fig. 1a)
89 along the MAR, represent segments of a slow spreading ridge comprised of elevated
90 seafloor massifs that display flat or gently curved upper surfaces with prominent
91 corrugations or ‘megamullions’ (Escartin et al., 2017; Tucholke et al., 1998). OCCs
92 comprise segments of lower crustal and upper mantle rocks exposed at the seafloor (Cann
93 et al., 1997; Blackman et al., 1998; Tucholke et al., 1998) and represent uplifted footwalls
94 of large-offset low-angle normal faults, commonly referred to as detachment faults.
95 Mantle rocks of OCCs characteristically comprise olivine-rich peridotite (i.e., harzburgite,
96 dunite) that interacted with seawater to produce serpentinite over a range of temperatures
97 (Andreani et al., 2007; Blackman et al., 1998; Boschi et al., 2006a; Boschi et al., 2006b;
98 Cann et al., 1997; Cannat, 1993; Früh-Green et al., 2004; Karson et al., 2006; Kelemen et
99 al., 2007; Rouméjon et al., 2015; Rouméjon et al., 2018a; Schroeder et al., 2002).

100 Four OCCs have been drilled by the Ocean Drilling Program (ODP), the
101 Integrated Ocean Drilling Program (IODP) and the International Ocean Discovery
102 Program (IODP). The Atlantis Massif is one of the best studied and well-known OCC as
103 it hosts the off-axis, peridotite-hosted Lost City hydrothermal field (Kelley et al., 2001)

104 on its southern wall (Fig. 1b). Other drilled OCCs include the Atlantis Bank, SW Indian
105 Ridge (ODP Hole 735B, Dick et al., 2000), the MARK (Mid-Atlantic Ridge Kane fracture
106 zone) at 23°32' N (ODP Leg 153, Sites 921–924, Cannat et al., 1995), and an OCC on the
107 MAR at 15°44' N (ODP Leg 209, Site 1275, Kelemen et al., 2004). The Atlantis Massif
108 (30 °N, Fig. 1a) was drilled during IODP Expeditions 304/305 (Blackman et al., 2006),
109 at Site U1309 and Expedition 357 (Früh-Green et al., 2017, 2018; Roumejon et al., 2018a,
110 2018b; Akizawa et al., 2020).

111 Research on detachment faulting at OCCs implies that oceanic spreading is closely
112 linked to the development of hydrothermal circulation patterns and encompasses a wide
113 variety of fluid flow and hydrothermal regimes (McCaig et al., 2007; Escartin et al.,
114 2008). High-temperature fluid circulation both within the footwall and along the fault
115 zone is well documented on the basis of mineralogy and geochemistry (Schroeder and
116 John, 2004; Boschi et al., 2006a; McCaig et al., 2010, Picazo et al., 2012, Verlaguet et
117 al., 2021). Importantly, uplift along detachment faults appears to promote circulation and
118 alteration within the footwall. Several studies propose a temporal evolution in the style of
119 hydrothermal circulation associated with OCC formation. High-temperature systems are
120 hosted in the basaltic hanging wall within the rift valley, whereas high-temperature
121 ultramafic-hosted systems occur within the OCC footwall. Ultimately, hydrothermal
122 circulation transitions to off-axis ultramafic-hosted systems within the footwall (Andreani
123 et al., 2007; McCaig et al., 2007; Fouquet et al., 2010). In ultramafic and mafic systems,
124 metasomatic assemblages of talc-tremolite-chlorite or quartz form at > 350 °C (Boschi et
125 al., 2006a, 2008; McCaig et al., 2010; Verlaguet et al., 2021), typical of black smoker
126 discharge zones, whereas serpentine-prehnite-hydrogarnet assemblages form at lower
127 temperatures (Frost et al., 2008; Bach and Klein, 2009).

128 Cores recovered during IODP Expedition 357 have highly heterogeneous rock
129 types, and are variably altered and deformed. Ultramafic rocks are dominated by
130 serpentized harzburgite with intervals of serpentized dunite and minor pyroxenite
131 veins; gabbroic layers occur as melt impregnations and veins. Dolerite dikes and basalts
132 are the latest phase of magmatism. Overall, the peridotites show a high degree of
133 serpentization (> 80 %) and are locally oxidized. In cores with gabbroic intrusions,
134 hydrothermal alteration of mixed peridotite and gabbroic lithologies forms serpentine-
135 talc-amphibole-chlorite assemblages (Boschi et al., 2006b; Picazo et al., 2012).

136 The petrogenesis of mantle-derived peridotites recovered during IODP Expedition
137 357 is the subject of this communication. In this paper we use whole rock major, trace
138 and rare earth element (REE) chemistry of Expedition 357 peridotites to discriminate
139 dominant melt-impregnation vs. fluid-dominated processes (i.e., serpentization, Si
140 metasomatism) and constrain the degrees of partial melting recorded in the peridotites.
141 The altered peridotites are subdivided into Type I serpentinites, Type II melt-impregnated
142 serpentinites and Type III metasomatic serpentinites. Type I serpentinites show no
143 evidence of melt-impregnation or metasomatism apart from serpentization and local
144 oxidation. Type II serpentinites are distinguishable in some cases on the basis of
145 macroscopic or microscopic evidence and in other cases, are inferred on the basis of
146 incompatible element enrichment in the case of ‘cryptically’ melt-impregnated
147 serpentinites. Type III serpentinites are characterized by silica metasomatism and the
148 formation of talc-rich alteration assemblages and anomalously high anhydrous SiO₂
149 concentrations and low MgO/SiO₂ values. In so doing, we geochemically characterize the
150 evolution of mantle lithosphere at a slow-spreading ridge associated with OCC formation,
151 and document spatial compositional variations.

152

153 2. Geological setting

154 The dome-shaped Atlantis Massif OCC is located at 30 °N on the western edge of
155 the MAR axial valley where it intersects the Atlantis Fracture Zone (Fig. 1b). The OCC
156 stretches 15-20 km N–S parallel to the ridge and is 8-12 km wide. Exhumation occurred
157 via low-angle detachment faulting (Cann et al., 1997; Blackman et al., 2002; Schroeder
158 and John, 2004; Karson et al., 2006; Ildefonse et al., 2007). On the basis of its distance
159 from the spreading axis and a calculated spreading half-rate of 12 mm/yr (Zervas et al.,
160 1995), the lithosphere of the massif is considered to be 2.0 to 0.5 Ma (Blackman et al.,
161 2006), a span which encompasses eighteen SHRIMP U/Pb zircon ages of oxide gabbro
162 and felsic dike melt intrusions recovered from Hole U1309D, which range from $1.28 \pm$
163 0.05 to 1.08 ± 0.07 Ma (Grimes et al., 2008). Dredging and submersible dives at the
164 massif revealed the dominance of serpentinized peridotite (Blackman et al., 2002; Boschi
165 et al., 2006a; Karson et al., 2006) concentrated primarily along the southern and most
166 elevated portion of the detachment. Samples recovered during IODP Expeditions
167 304/305, with a drill hole reaching ~ 1500 m depth, consisted almost entirely of olivine-
168 rich mafic intrusive rocks of gabbro and troctolite (Drouin et al., 2009; Ferrando et al.,
169 2018; Godard et al., 2009; Suhr et al., 2008). These geological observations suggest a
170 compositional change of the footwall, with peridotites decreasing in abundance from the
171 segment end towards the north, and also reflecting variations in time of the emplacement
172 of magma in the footwall (Ildefonse et al., 2007).

173 IODP Expedition 357 cored seventeen shallow holes at nine sites (Fig. 1b) along
174 the detachment fault surface of the Atlantis Massif (Früh-Green et al., 2016). The two
175 eastern sites (M0075, M0068) and one western site (M0071) recovered fault scarp
176 deposits whereas the central sites yielded *in situ* sequences. Most of the sites are aligned
177 along the southern edge of the detachment fault by the Atlantis Fracture Zone wall, with

178 the northernmost hole (M0074) located ~ 6 km north of the detachment's southern edge,
179 and ~ 1 km to the southwest of U1309D. In terms of igneous lithologies and their
180 metamorphosed equivalents, IODP Expedition 357 recovered primarily serpentized
181 peridotite comprising harzburgite with subordinate dunite (Fig. 1c) and wehrlite with
182 lesser amounts of variably altered mafic inclusions of basalt, gabbro and dolerite (Früh-
183 Green et al., 2018; Rouméjon et al., 2018a; Akizawa et al., 2020). Core lengths range
184 from approximately 1.3 to 16.5 m. Recovery was ~ 75 %.

185

186 **3. Samples and petrography**

187 Except for one talc schist, the protolith of which was mafic, all rocks chosen for
188 study are serpentized peridotites, and from holes immediately adjacent to the transform
189 fault scarp and drilled at the southern edge of the detachment fault surface (Figs. 1b, c).
190 All peridotites were subjected to high degrees of serpentization on the order of > 80 %.
191 On the basis of collective macroscopic, microscopic and whole rock chemistry evidence,
192 the studied samples are divided into three types: Type I are serpentinites, Type II are melt-
193 impregnated serpentinites, and Type III are metasomatic serpentinites. Type I
194 serpentinites are ones primarily subjected to serpentization only, and Types II and III
195 serpentinites have been subjected to mm- to cm-scale mafic melt intrusions and
196 subsequent silica metasomatism, respectively. In the case of melt-impregnation, this is
197 evident in Type II serpentinites by the presence of small veinlets of dolerite (as reported
198 in Expedition 357 petrology logs), and in one case by the presence of rare plagioclase and
199 in another case, by the presence of recrystallized olivine along the peripheries of partially
200 dissolved orthopyroxene (see Fig. 2). Further details regarding evidence for classification
201 of Types II and III are provided in Table 1.

202 All samples were studied by polarization microscopy. Representative thin section
203 images are given in Figure 2. Sample numbers are taken directly from Früh-Green et al.
204 (2017, Supplementary material). Below, we describe the petrography of each sample type.
205 The mineralogy and mode of the serpentinites are summarized in Table 2.

206

207 *3.1. Type I serpentinites*

208 Type I serpentinites (Fig. 2a-d) consist of dominant harzburgite (14 of 20 samples)
209 and lesser dunite. Outlines of olivine are generally clear under plane-polarized light (PPL)
210 as serpentinization is most intense around grain boundaries and fractures cutting olivine
211 (e.g., Fig. 2a). Classic mesh-textures are commonly observed. In the harzburgite,
212 orthopyroxene is always altered to bastite, but is nonetheless recognizable under crossed
213 polars by remnant and deformed (curved) cleavage and parallel extinction.
214 Orthopyroxene is typically < 2 mm and olivine is 2-4 mm. Reddish-orange to brown
215 spinel is easily identifiable under PPL and is typically less than 0.5 mm but in some cases
216 up to 4 mm. Spinel occurs both as isolated, angular grains and as subangular and rounded
217 grains commonly arranged in masses or trails of multiple grains. Clinopyroxene was not
218 observed in any samples, reflecting the high degrees of partial melting and melt extraction
219 that many of these peridotites have undergone (see Fig. 3). Modal percentages of the
220 volumetrically dominant harzburgite lie in the range of 75-90 % for olivine and 10-25 %
221 for orthopyroxene; spinel ranges from 0-3 % (Table 2). In the dunite, orthopyroxene is
222 absent and spinel is less common (0 to <1 %) than in the harzburgite.

223 Four Type I serpentinites exhibit anomalously high CaO contents ranging from 6-
224 13 wt.% (see Table 3). Microscopic inspection reveals that the Ca-rich peridotites exhibit
225 an extensive and densely concentrated, anastomosing network of ~ 0.1 mm thick
226 carbonate veinlets (Fig. 2d).

227

228 *3.2. Type II Melt-impregnated serpentinites*

229 Melt-impregnated serpentinites consist of harzburgite only and comprise six of
230 the 29 studied samples (Table 1). Type II serpentinites in most cases are observed to have
231 mm to cm-scale mafic veins at the scale of drill core (Table 2) although these are not
232 obvious in thin section. Orthopyroxene ranges from ~ 1-5 mm. Modal percentages of
233 olivine and orthopyroxene are 70-90 % and 10-30 %, respectively, with spinel typically
234 absent but reaching 2-3 % in one sample (Table 2).

235 In some Type II serpentinites, other petrographic evidence is present to confirm
236 melt-impregnation. For example, one central harzburgite (76B-7R1-81-83) is the only one
237 in which rare 0.5-0.7 mm plagioclase (< 0.5 % of the mode) is present (Fig. 2e).
238 Plagioclase is highly altered (epidote, sericite) but preserves relict twinning. In another
239 harzburgite from eastern Site M0068B, fresh, 0.5-1 mm olivine constitutes ~ 5 % of the
240 mode, and is observed mantling orthopyroxene (Fig. 2f), which we interpret as replacive
241 following dissolution of orthopyroxene accompanying melt injection.

242

243 *3.3. Type III Metasomatic serpentinites*

244 Type III serpentinites from this study include two harzburgites that are restricted
245 to the eastern sites (Fig. 1c), and are distinguishable on the basis of talc-amphibole
246 alteration of orthopyroxene (Fig. 2e) and whole rock chemistry (see Section 5.1).
247 Orthopyroxene ranges from 1-6 mm. Brown spinel ranges from 0.5-2 mm and are always
248 embedded in orthopyroxene. Modal percentages of olivine and orthopyroxene are ~ 70 %
249 and 30 % respectively, with spinel occupying around 2 % (Table 2).

250

251 **4. Analytical methods and data compilation**

252 *4.1. Whole rock geochemistry*

253 Major and trace element contents of the serpentinites were determined by
254 inductively coupled plasma mass spectrometry (ICP-MS) and atomic emission
255 spectroscopy (ICP-AES) at ALS Geochemistry, North Vancouver, Canada. Prior to
256 shipping to ALS, samples for whole-rock analysis were trimmed to remove weathered
257 surfaces. Samples sent to ALS were crushed to > 70 % passing through a 2 mm mesh,
258 and a 250-g split was pulverized to > 85 % of the material being < 75 µm in diameter.
259 Powders were then analyzed for whole rock major and trace element determinations. For
260 major element oxides, 100 mg of powdered sample was added to lithium
261 metaborate/lithium tetraborate flux, mixed well and fused in a furnace at 1000 °C. The
262 resulting melt was dissolved in 100 mL of 4 % HNO₃/2 % HCl. This solution was then
263 analyzed by ICP-AES (ALS Geochemistry method ME-ICP06) and the results were
264 corrected for spectral inter-element interferences. Major element detection limits are
265 0.01% for all oxides except Cr₂O₃, which is 0.002 % (Supplementary Table S1). For loss
266 on ignition (LOI), 1 g of sample powder was heated at 1000° C for one hour, cooled and
267 then reweighed, with percent LOI calculated by the difference in weight.

268 For trace and rare earth elements, the ME-MS61L Super Trace and MS61L-REE
269 methods (<https://www.alsglobal.com/>) were employed, which combines a four-acid
270 digestion with ICP-MS instrumentation utilizing collision/reaction cell technologies to
271 provide the lowest detection limits available. In the first method, analyzed elements
272 include Ba, Cd, Ce, Co, Cr, Cs, Cu, Ga, Hf, La, Li, Mo, Nb, Ni, Pb, Rb, Sc, Sn, Sr, Ta,
273 Te, Th, U, V, W, Zn and Zr; in the second method, analyzed elements include Dy, Er, Eu,
274 Gd, Ho, Lu, Nd, Pr, Sm, Tb, Tm and Yb. This trace method has been optimized for long-
275 term robust ICP-MS signal stability, in particular for samples with high Ca content. The
276 prepared samples (nominal weight 0.25 g) were digested with 1.5 mL concentrated HNO₃

277 and HClO₄, followed by concentrated HF. Subsequently, the mixture was heated at 185
278 °C until incipient dryness, leached with 50 % HCl and diluted with weak HCl. The final
279 solution was then analyzed by ICP-MS with results corrected for spectral inter-element
280 interferences. Apart from Ba, Cr, Sn, V, W and Zr with detection limits that range from
281 0.3-5 µg/g, detection limits of the remaining trace elements ranged from 0.001-0.2 µg/g
282 (Supplementary Table S1).

283 Results of duplicate analyses and analytical standards used are provided in
284 Supplementary Table S2. Reproducibility of these reference samples is generally better
285 than 5 % for concentrations more than 10 x above the detection limit, and typically 5-20
286 % closer to the detection limit.

287

288 *4.2. Data compilation*

289 The stratigraphic location of samples analyzed for whole rock chemistry in this
290 study are shown in Fig. 1c. The new whole rock geochemical data of 29 whole rock ICP-
291 MS and ICP-AES geochemical analyses from this study (Table 3) are compared with the
292 ten Expedition 357 peridotite samples of Früh-Green et al. (2018) and the four Atlantis
293 Massif peridotite samples of Godard et al. (2009) from Site 1309 in Figs. 3-10. We also
294 compare these data with whole rock geochemical data compiled from global abyssal
295 peridotites of Niu (2004), Paulick et al. (2006), and Godard et al. (2008).

296

297 **5. Results**

298

299 *5.1. Whole rock chemistry major elements and Ni and Cr abundances*

300 In MgO/SiO₂ vs. Al₂O₃/SiO₂ space (Fig. 3) the ‘terrestrial array’ represents the
301 successive magmatic depletion (or melt extraction) trend of primitive mantle. Highly
302 depleted compositions are characterized by low Al₂O₃/SiO₂ of < 0.01 (Jagoutz et al.,

1979; Hart and Zindler, 1986). Verification of this melt extraction trend is reflected by a global data set of MOR peridotites (compilation of Niu, 2004; Paulick et al., 2006; Godard et al., 2008). Atlantis Massif serpentinitized harzburgites record $\text{Al}_2\text{O}_3/\text{SiO}_2$ that range from ~ 0.01 - 0.04 indicating moderate to high degrees of magmatic depletion. Two Type I serpentinitized dunites from the central sites (M0076B-3R1-78-82, M0076B-5R1-59-61, Table 1), and one dunite (1309D-31R-25-28) from Site U1309 reported by Godard et al. (2009) exhibit $\text{Al}_2\text{O}_3/\text{SiO}_2 < 0.01$. Very low $\text{Al}_2\text{O}_3/\text{SiO}_2$ values can alternatively be caused by replacive melt-rock interaction. Vertical deviations from the terrestrial array are indicative of Mg-Si mass transfer; peridotites that exhibit MgO/SiO_2 below the terrestrial array may reflect magnesium loss or silica addition during serpentinitization or magnesium loss during seafloor weathering (Snow and Dick, 1995; Niu, 2004). A trend defined by increasing $\text{Al}_2\text{O}_3/\text{SiO}_2$ with most samples offset to moderately low MgO/SiO_2 , as seen in the Atlantis Massif serpentinites (Fig. 3), can also be partially attributed to melt-impregnation processes (Niu, 2004). Half of the eight Type II melt-impregnated serpentinites of the eastern and central sites from this study and the study of Früh-Green et al. (2018) exhibit MgO/SiO_2 of < 0.9 , consistent with this process; nonetheless, all melt-impregnated serpentinites plot within the range of the Type I serpentinites. Two central serpentinites yield MgO/SiO_2 values of ~ 0.76 - 0.86 , within the lower range of the melt-impregnated serpentinites, which suggests that these two samples may also have been subjected to processes associated with melt-impregnation. Silica-metasomatism is characterized by low MgO/SiO_2 (< 0.55) and low $\text{Al}_2\text{O}_3/\text{SiO}_2$ (< 0.025). One exception is the talc schist, which exhibits low MgO/SiO_2 (~ 0.4) but high $\text{Al}_2\text{O}_3/\text{SiO}_2$ of 0.11 , which suggests the addition of silicic melt (Paulick et al., 2006).

In Fig. 4 we plot MgO vs. selected major element oxides and loss on ignition (LOI) to illustrate the effects of serpentinitization and multiphase alteration processes on

328 major element chemistry. In terms of MgO and other major element oxides, the Type I
329 serpentinites generally plot similarly to global MOR peridotites. Apart from two
330 serpentinitized harzburgites, which exhibit anomalously high Al₂O₃ and CaO, the
331 remaining serpentinites have MgO contents that ranges from 41.25 to 46.06 wt.% and
332 Mg# (Mg/Mg+Fe²⁺) of 87.6-91.8 (Table 3). In SiO₂ vs. MgO space (Fig. 4a) the
333 serpentinites and melt-impregnated serpentinites fall almost completely within the range
334 of global abyssal peridotite and plot similarly to, but with slightly lower SiO₂ contents
335 than the bulk composition of serpentine. In contrast, the Type III talc-altered serpentinites
336 and talc schists plot near the bulk composition of talc and ODP Leg 209 talc-altered
337 peridotites, but with slightly lower SiO₂ and MgO. The Ca-rich Type I serpentinites (6.66-
338 13.30 wt.% CaO) plot off the linear array of the serpentinites and melt-impregnated
339 peridotites in SiO₂ vs. MgO space with significantly lower SiO₂ at similar MgO. Fe₂O₃,
340 Al₂O₃ and CaO contents (Figs. 4b-d) of the talc-altered serpentinites and talc schists
341 generally fall within the range of the Type I and II serpentinites. Apart from two Type II
342 melt-impregnated serpentinitized harzburgites, which exhibit slightly higher Al₂O₃
343 contents than the remaining serpentinites, other major element oxide concentrations of
344 the melt-impregnated serpentinites are similar to those of the serpentinites. CaO contents
345 in the western site Type I serpentinites (Fig. 4d) record lower concentrations (0.05-0.30
346 wt.%) than those of the central and eastern sites. Similar to the SiO₂ vs. MgO plot,
347 significant differences exist in LOI values between the Ca-rich serpentinites and all other
348 serpentinites (Fig. 4e). Whereas the bulk of the peridotites exhibit LOI of ~ 12-14 wt.%,
349 similar to those of global MOR peridotites and the talc-altered harzburgites and schists
350 have low LOI contents that range from ~ 4-6 wt.%, similar to ODP Leg 209 talc-altered
351 peridotites, the Ca-rich serpentinites have high LOI of ~ 16-20 wt.%. The Type II melt-

352 impregnated serpentinites have LOI values that are up to ~ 9 wt.%, intermediate between
353 those of the Type I and serpentinites and Type III serpentinites and schists.

354 The majority of the Expedition 357 serpentinites have Ni and Cr concentrations
355 of ~ 1000-3000 $\mu\text{g/g}$ (Table 3), similar to that of primitive mantle (PM), and fall within
356 the range of global MOR peridotites (Supplementary Fig. S1). Exceptions are two central
357 and two western serpentinite samples from Früh-Green et al. (2018), which range to Ni >
358 10000 $\mu\text{g/g}$ and Cr > 30000 $\mu\text{g/g}$, and two central serpentinitized dunites and two eastern
359 melt-impregnated harzburgites from our study, which exhibit low Cr (< 400 $\mu\text{g/g}$).

360

361 *5.2. Whole rock trace element concentrations*

362 Significant differences in the relative degree of depletion, pattern morphology,
363 and the existence of prominent element anomalies, e.g., conspicuous positive or Ce-
364 anomalies depending on serpentinite type, are apparent on the basis of chondrite-
365 normalized REE and PM-normalized incompatible trace element patterns (Fig. 5).
366 Patterns range from those similar to and significantly more depleted than depleted MORB
367 mantle (DMM) and mean abyssal peridotite in western Site M0071, central Sites M0069
368 and M0076, and Atlantis Massif IODP Site U1309 (Godard et al., 2009) to patterns
369 enriched relative to DMM and mean abyssal peridotite as shown by central Site M0072
370 and most of eastern Site M0068. The serpentinites from eastern Site M0068 are atypical
371 to other Expedition 357 serpentinites in that they exhibit LREE enrichments but HREE
372 concentrations intermediate to that of DMM and mean abyssal peridotite. Central Site
373 M0072 and eastern Site M0068 serpentinites typically exhibit REE concentrations that
374 are more enriched than DMM and range from ~ 1-10 x chondrite; two M0068 talc schists
375 are exhibit high REE concentrations of ~ 20 x chondrite.

376 Type I serpentinites sampled at western Site M0071, and central Sites M0069 and
377 M0076 exhibit low Yb as well as low abundances of other incompatible elements, for
378 example the high-field strength elements (HFSE) Nb and Zr (Table 3). Apart from one
379 central Site M0072 serpentinitized dunite with similarly low Yb, Nb and Zr, the remaining
380 central Site M0072 and eastern site serpentinites, many of which are Type II melt-
381 impregnated or Type III metasomatic serpentinites, exhibit comparatively higher
382 concentrations of these incompatible elements (Table 1). As we show in Section 6.2.3,
383 the melt-impregnated serpentinites, in particular in central Site M0072 and eastern Site
384 M0068, show high concentrations of most incompatible elements relative to other sites.

385 Although the highly depleted Type I serpentinites from the western and central
386 sites exhibit mostly concave upwards REE patterns with a steady decrease in REE
387 abundances from the HREE to LREE, central Site M0072 Type I and Type II serpentinites
388 exhibit flatter patterns; the eastern Types I, II and III serpentinites are atypical relative to
389 the serpentinites to the west (i.e., at the central and western sites) in that they exhibit
390 LREE enrichment. Excluding the lone, Ca-rich (12.46 wt.% CaO) serpentinitized dunite
391 with La/Sm_{CN} (CN denotes chondrite-normalized) of 4.70 and La/Yb_{CN} of 2.39, the
392 western Type I serpentinites from Site M0071 exhibit mostly moderate but variable
393 LREE/MREE fractionation (La/Sm_{CN} of 0.40-3.03) with La/Yb_{CN} ranging from 0.11-
394 0.68. In contrast, the two eastern Type I serpentinites exhibit relatively minor LREE
395 enrichment with La/Sm_{CN} and La/Yb_{CN} ranging from 1.08-1.17 and 1.13-1.30,
396 respectively; the eastern Type II melt-impregnated serpentinites have higher LREE
397 enrichments with $La/Sm_{CN} = 1.72-2.15$ and $La/Yb_{CN} = 1.43-1.68$ and the Type III
398 metasomatic serpentinites have $LaSm_{CN} = 0.41-1.41$ and $La/Yb_{CN} = 0.48-1.36$.

399

400 **6. Discussion**

401 On the basis of trace and rare earth element chemistry, we address differences in
402 the compositions of the different serpentinite types to discriminate dominant fluid-rock
403 interaction processes associated with serpentinization, from processes associated with
404 mafic melt-rock interaction. We then provide estimates of degrees of partial melting (melt
405 extraction) recorded in serpentinites sampled across the Atlantis Massif (IODP Exp. 357),
406 before linking the geochemistry to processes associated with oceanic core complex
407 formation.

408

409 *6.1. Evidence of fluid-dominated serpentinization processes*

410 Significant observations from the chondrite-normalized REE and PM-normalized
411 patterns are: (1) prominent negative Ce-anomalies (i.e., Ce/Ce^* where $Ce^* =$
412 $\sqrt{(La_{CN} * Pr_{CN})}$ and $Ce = Ce_{CN}$) in the western Type I serpentinites and two central Type I
413 serpentinites, and positive Ce-anomalies in almost all eastern serpentinites (the vast
414 majority of other central serpentinites show $Ce/Ce^* \sim 1$); (2) extreme U-enrichment in
415 many serpentinites with values that range to nearly 300 x PM; and (3) extreme Sr-
416 anomalies with enrichments of up 100 x PM in most Ca-rich Type I serpentinites and
417 depletion to below 0.1 x PM in the serpentinites of Sites M0072 and M0068.

418 With respect to the negative Ce-anomalies exhibited mostly by the western Type
419 I serpentinites (site M0071), such anomalies in peridotites have been attributed to
420 seawater-peridotite interaction (e.g., Frisby et al., 2016a, b) and indeed, (oxygenated)
421 seawater itself exhibits a negative Ce-anomaly (e.g., Elderfield and Greaves, 1982).

422 The western site Type I serpentinites range to the highest U concentrations (5.05
423 $\mu\text{g/g}$), whereas the Type II melt-impregnated serpentinites of central site M0072 and
424 eastern site M0068 range to the lowest U concentrations (0.01 $\mu\text{g/g}$) (Fig. 6a, Table 3),
425 which suggests that U enrichment is associated with serpentinization and oxidation, and

426 not melt-impregnation. Uranium enrichment in serpentinites is common as U is hosted by
427 serpentine phases (e.g., Deschamps et al., 2010). Frisby et al. (2016a, b) have shown that
428 U enrichments correlate with Ce anomalies and the amount of Nd derived from seawater,
429 which is a proxy for water/rock values. Thus, U enrichments have been shown explicitly
430 to reflect fluid-rock interaction in abyssal peridotites.

431 In the case of Sr enrichment (Fig. 6b), evidence is not as clear. However, we note
432 that the Type II melt-impregnated central site M0072 and eastern site M0068 serpentinites
433 range to the lowest Sr concentrations, with the former exhibiting the lowest
434 concentrations of Sr (1.02-4.01 $\mu\text{g/g}$) of all sites, whereas the other sites locally show
435 distinct Sr enrichments. This suggests that Sr enrichment is not associated with melt-
436 impregnation.

437

438 *6.2. Evidence for melt-impregnation processes: Enrichment in incompatible elements,* 439 *LREE, LREE fractionations and ΣREE*

440 As explained above, we infer melt-impregnation in central site M0072
441 serpentinites on the basis of compositional similarities with melt-impregnated
442 serpentinites constrained on the basis of macroscopic and microscopic evidence, even
443 though site M0072 serpentinites lack observable evidence of melt-impregnation at the
444 scale of core and thin section. Hence, due to the absence of such mineralogical evidence,
445 we term these M0072 serpentinites ‘cryptically melt-impregnated’.

446 The HFSE, e.g., Th, Hf, Zr (Figs. 7a-c), Nb, and Ti are highest in Type II melt-
447 impregnated serpentinites at central Site M0072 and eastern Site M0068 and lowest in
448 western sites Type I serpentinites. In addition, Figures 7d and 7e show an increase in
449 LREE and ΣREE , and possibly LREE fractionation from Type I serpentinites to Types II
450 and III serpentinites, and from west to central to east. For example, apart from a lone Ca-

451 rich Type I serpentinized dunite, which exhibits anomalously high La and anomalously
452 low Ce and La/Yb_{CN} of 2.39, the remaining western Type I serpentinites yield La/Yb_{CN}
453 of 0.11-0.47 and mean ΣREE of 0.39 μg/g (including the Ca-rich dunite). The central
454 Type I and II serpentinites yield a range of La/Yb_{CN} of 0.15-1.31 and mean ΣREE of 2.40
455 μg/g, and the eastern Types I, II and III serpentinites yield a range of La/Yb_{CN} of 0.61-
456 1.68 and mean ΣREE of 12.66 μg/g. Thus, overall, melt impregnation produces less LREE
457 fractionation and more elevated ΣREE compared to processes dominated by fluid-rock
458 interaction.

459

460 *6.3. Evidence of silica metasomatism*

461 Talc- and amphibole-rich fault rocks form as the result of silica metasomatism and
462 have been recovered from detachment fault surfaces along slow and ultra-slow spreading
463 mid-ocean ridges including oceanic core complexes (e.g., Escartín et al., 2003; Schroeder
464 and John, 2004; Boschi et al., 2006b; McCaig et al., 2007; Picazo et al., 2021). These
465 rocks record heterogeneous deformation under greenschist-facies conditions and are
466 commonly restricted to localized shear zones (< 200 m), which are associated with intense
467 talc-amphibole metasomatism (Escartín et al., 2003; Boschi et al., 2006b). Talc is
468 mechanically weak and thus may be critical to the development of such fault zones and
469 may enhance unroofing of upper mantle peridotites and lower crustal gabbroic rocks
470 during seafloor spreading. In the case of the Atlantis Massif, talc metasomatism is
471 associated with serpentinite dehydration (Boschi et al., 2008). Strontium isotope
472 compositions of talc-rich fault rocks indicate that talc metasomatism along detachment
473 faults occurs at low water/rock values (0.2–0.7) and reflect interaction with Si-rich,
474 evolved fluids with a mafic component derived from interaction with gabbro lenses within
475 a peridotite-dominated ridge segment (Boschi et al., 2008).

476 In our study, silica metasomatism is reflected by plots of MgO/SiO₂ vs.
477 Al₂O₃/SiO₂ (Fig. 3) and MgO vs. SiO₂ (Fig. 4a). These figures demonstrate the distinct
478 whole rock geochemistry of the talc-altered Type III metasomatic serpentinites and talc
479 schists from the eastern site M0068. In MgO vs. SiO₂ space (Fig. 4a), the talc-bearing
480 serpentinites and talc schists are clearly distinct from all other Exp. 357 serpentinites by
481 much lower MgO and much higher SiO₂ contents. These low MgO/SiO₂ and Al₂O₃/SiO₂
482 values of < 0.02 are indicative of silica metasomatism and are close to compositions of
483 talc-bearing harzburgites recovered during ODP Leg 209 (Paulick et al., 2006).

484

485 *6.4. Discrimination of fluid-rock vs. melt-dominated processes*

486 *6.4.1. Comparison with global abyssal peridotites*

487 Figure 8a highlights trace element variations of mean global abyssal peridotites
488 with compositions dominated by melt-rock interaction vs. those dominated by fluid-rock
489 interaction associated with serpentinization (as interpreted by Paulick et al., 2006). We
490 note that (in Fig. 8) we are comparing patterns, as absolute concentrations are subject to
491 other processes (e.g., melt addition). Serpentinites subjected to mafic melt-impregnation
492 exhibit enrichments in incompatible elements relative to those that have not been affected
493 by melt-impregnation (Fig. 8a). In Figure 8b, we plot the mean serpentinite composition
494 of each Expedition 357 location (i.e., west, central, east) vs. the mean compositions of
495 melt-dominated and fluid-dominated global abyssal peridotites as shown in Fig. 8a. In
496 Fig. 8c, we parse the mean peridotite compositions according to borehole of the central
497 sites. There are clear differences in incompatible element concentrations between Exp.
498 357 Type II melt-impregnated serpentinites and Type III metasomatic serpentinites
499 relative to Type I serpentinites, with Types II and III exhibiting greater abundances of all
500 incompatible elements (Fig. 8b). In particular, the Type I serpentinites from the western

501 site have PM-normalized incompatible element abundances that are most similar to fluid-
502 dominated peridotites, whereas central sites serpentinites exhibit concentrations most
503 similar to melt-dominated peridotites; eastern site serpentinites have incompatible
504 abundances more enriched than melt-rock dominated peridotites (Fig. 8b). Apart from U
505 and Sr, the Exp. 357 serpentinites show a clear enrichment in all other incompatible
506 elements from the western to central to eastern sites (Fig. 8b).

507

508 6.4.2. HFSE vs. LREE

509 On the basis of selected HFSE vs. LREE and HFSE vs. MREE/HREE variations,
510 Paulick et al. (2006) showed that the global abyssal peridotite dataset of Niu (2004)
511 exhibit relatively steep positive trends, which was interpreted as indicative of dominant
512 melt-rock interaction processes. The reason for this trend is related to differences in the
513 behavior of LREE and HFSE; LREE are more hydrophilic than HFSE, and so are more
514 readily transported in solution. Although melt-rock reaction mobilizes both LREE and
515 HFSE, fluid-dominated reactions preferentially mobilize LREE, resulting in distinctly
516 different patterns (Niu, 2004). A trend defined by only minor increase in HFSE
517 concentrations (i.e., a sub-horizontal as opposed to steep positive trend) on the other hand,
518 is indicative of dominantly hydrothermal alteration processes (i.e., fluid-rock interaction),
519 which affect the more immobile HFSE to a lesser extent than the REE (Paulick et al.,
520 2006). In the dataset of ODP Leg 209 peridotites, Paulick et al. (2006) showed that those
521 at Sites 1270 and 1271 exhibit steep trends similar to the dataset of Niu (2004), whereas
522 those of Sites 1268, 1272 and 1274 show less steep, subhorizontal trends, which they
523 interpreted as more consistent with fluid-rock interaction.

524 Fig. 9 shows Expedition 357 serpentinites vs. the trends of global abyssal
525 peridotites, with compositions dominated by (i) melt-rock interaction, and (ii) fluid-rock

526 interaction in Nb vs. La and Ti vs. Dy space. These plots produce two distinct linear
527 trends, which mimic the two described above, in agreement with the observations
528 presented in Fig. 8. The serpentinites of the central and eastern sites, which show ample
529 evidence of melt-impregnation, exhibit steep trends indicative of compositions
530 dominantly controlled by melt-rock interaction similar to those of the global MOR
531 peridotite dataset of Niu (2004) and ODP Leg 209 Sites 1270 and 1271 of Paulick et al.
532 (2006). In contrast, Type I serpentinites from the western site, which show minimal
533 evidence of melt-impregnation, instead show flatter, subhorizontal trends suggestive of
534 compositions mostly controlled by fluid-rock reactions, similar to trends of peridotites of
535 ODP Leg 209 Sites 1268, 1272 and 1274. We note also that similar relations and trends
536 occur on other plots of HFSE vs. LREE and MREE (e.g., Th vs. Ce, Ti v. Ce, Hf vs. Pr,
537 Nb vs. $[Gd/Lu]_{CN}$ and Ti vs. $[Gd/Lu]_{CN}$, Supplementary Fig. S2). In the case of Expedition
538 357 serpentinites, in all instances, the slope of the trends of the central and eastern site
539 serpentinites are steeper than the slope of the trend of the western site serpentinites,
540 consistent with compositions of the former being the result of dominant melt-rock
541 reaction and compositions of the latter being the result of dominantly fluid-rock
542 interaction. When the central location is parsed into discrete sites, compositions of
543 serpentinites from central Site M0069 are consistent with being dominated by fluid-rock
544 interaction, whereas those of central Sites M0072 and M0076 are consistent with being
545 dominated by melt-rock interaction.

546

547 *6.5. Petrogenesis: Estimates of degrees of melt extraction*

548 Chondrite-normalized REE and PM-normalized patterns (Fig. 5) are used to
549 explain differences in the relative depletion of peridotites of each site investigated and
550 provide evidence of fluid-dominated processes and melt-impregnation processes

551 (Sections 6.1.-6.4.), based on the compositional variation of Expedition 357 serpentinites.
552 Below, we model estimates of partial melting undergone by the peridotites based on trace
553 and REE chemistry.

554 Various highly incompatible and immobile trace elements (Yb, Nb, Zr) in
555 serpentinites from western Site M0071 and central Sites M0069 and M0076 exhibit
556 extreme depletion relative to DMM, in contrast to the central Site M0072 serpentinites
557 that exhibit enriched chondrite-normalized patterns relative to DMM apart from a lone
558 moderately depleted Type I serpentinized dunite. The eastern Site M0068 serpentinites
559 exhibit LREE concentrations more enriched than DMM but HREE slightly to moderately
560 more depleted than DMM. Evidence in support of high degrees of melt depletion includes
561 the high depletion in Al_2O_3 and CaO (Fig. 4), and the aforementioned other incompatible
562 trace elements (Fig. 7, Table 3). Below, we provide estimates of the maximum degrees of
563 melt extraction based on modelling of whole rock REE concentrations and note that REE
564 patterns of many samples are strongly affected by refertilization, which thus needs to be
565 taken into account to derive accurate estimates of melt depletion.

566 The REE patterns of the most melt-depleted sample for each of the five sites are
567 shown in Fig. 10 and compared with curves predicted by modeling of melt depletion
568 followed by subsequent metasomatism. As LREE are most strongly depleted during
569 partial melting, and therefore sensitive to refertilization, this part of the REE pattern is
570 essentially that of the metasomatizing agent. As a result, different amounts of
571 metasomatism will shift the LREE pattern up or down, but the slope of the LREE pattern
572 (La/Sm) is barely affected. This slope is strongly variable in our dataset, ranging from
573 LREE-depleted ($[\text{La}/\text{Sm}]_{\text{CN}} = 0.3\text{-}0.9$) to strongly enriched ($[\text{La}/\text{Sm}]_{\text{CN}} = 2\text{-}5$). The REE
574 patterns in Figs. 5a-e show at least two distinct melts that have infiltrated the peridotites,
575 an enriched (*E*) and a depleted type (*D*), of which the most REE-rich examples are from

576 eastern Site M0068 (samples 68A-1R1-34-35 and 68A-1R1-1-6). In these samples, the
577 original peridotite REE patterns have been completely overprinted by the infiltrating
578 melts. Therefore, we chose these peridotites to estimate the composition of the two
579 endmember contaminants during modeling (*E* and *D*, respectively). Both types can be
580 observed at each site, although central Sites M0072 and M0076 are dominated by a *D*-
581 type contaminant. Some serpentinites show evidence of infiltration by an even more
582 enriched contaminant (*SE*), the composition of which is more difficult to estimate. As this
583 contaminant is strongly LREE-enriched and therefore relatively HREE poor, its exact
584 composition will have little effect on our melt fraction estimates, which are based on the
585 HREE part of the patterns.

586 The calculated volume of fractional non-modal melt extraction (see Fig. 10
587 caption for model parameters) of the most depleted sample from each site ranges from 19
588 to 21 % (Fig. 10), which indicates little difference in the maximum amount of melt
589 extraction between the sites. The difference in the REE patterns between the sites are
590 primarily due to small but variable amounts of infiltrating melts (<1 % for each of the
591 five most depleted samples, Fig. 10).

592 These modeling results show little difference in the maximum amount of melt
593 extraction between the different sites across the footwall. Unfortunately, significant melt
594 infiltration experienced by many of the serpentinites makes it difficult to evaluate the
595 average amount of melt extraction experienced by serpentinites of the different sites. For
596 example, nearly all whole rock compositions of eastern Site M0068 and central Site
597 M0072 are more enriched than DMM, which could be interpreted as very limited melt
598 extraction, but refertilization of these samples will have obliterated any previous melt
599 extraction history. Our modeling results suggest that differences in REE patterns observed
600 between sites are probably due to differences in refertilization, not partial melting.

601

602 *6.6. Spatial variations in geochemical processes and peridotite composition*

603 It was noted by Fröh-Green et al. (2018) that there exists a weakly defined
604 enrichment in peridotites from west to east. This pattern is actually strongly defined in
605 terms of enrichment in LREE, LREE fractionation, Σ REE, and HFSE (Figs. 7-9 and
606 Supplementary Fig. S2). For example, Fig. 8 shows a distinct increase in all incompatible
607 elements except U, Pb, and Sr in the mean serpentinite compositions moving from west
608 to central to east. Specifically, the western Type I serpentinites exhibit the lowest
609 concentrations of HFSE of Nb, Zr, and Th whereas the eastern serpentinites exhibit the
610 highest concentrations of these elements (Figs. 7a-c); the central serpentinites have
611 concentrations intermediate to the western and eastern serpentinites. However, as we have
612 also noted, there are distinctions in the central location when parsed into individual sites
613 with central Site M0069 being depleted to the same degree as the western serpentinites
614 and with the two other central sites (M0072 and M0076) being comparatively more
615 enriched.

616 Moreover, this pattern of west to east enrichment is complementary to the relative
617 volume of melt-impregnation and silica metasomatism recorded in sampled peridotite at
618 each site. The eastern site comprises the largest percentage of melt-impregnated and
619 silica-metasomatized peridotites (70%), relative to the central sites (35%), and the
620 western site (a single sample out of nine, or 11%, is melt-impregnated, with no Type III
621 silica-metasomatized serpentinites). However, the calculation for the central sites is
622 underestimated as we do not include here the four cryptically melt-impregnated
623 serpentinites of central Site M0072 from our dataset, which show no macroscopic or
624 microscopic evidence of melt-impregnation. If we recalculate with the inclusion of these

625 four M0072 serpentinites, the percentage of melt-impregnated and silica-metasomatized
626 peridotites from the central sites increases to 52 %.

627

628 *6.7. Links to OCC processes*

629 The observed spatial variations in serpentinite composition needs to be interpreted
630 in the context of both the limitations of our observations and taking into account the
631 complex history of detachment fault formation and exhumation. We have shown that the
632 western site Type I serpentinites record compositions consistent with dominantly fluid-
633 rock interaction processes, whereas the central site serpentinites, considered collectively,
634 and those from the eastern site record compositions consistent with having undergone
635 dominant melt-rock interaction. Furthermore, there is a clear progression in the volume
636 of melt-impregnations from west to central and east and a corresponding increase in
637 incompatible element enrichment, a record of these increasing volumes of melt-
638 enrichment.

639 Serpentinites from the western sites originated from shallower borehole depths of
640 ~ 0-10 m relative to those of the central sites (a maximum of ~ 16 m in the cases of central
641 Sites M00069A and M0076A, Fig. 1c). The present-day position of these samples cannot
642 be directly related to that of the history of each sample, from lithospheric emplacement,
643 subsequent interaction with melt, alteration, and exhumation. First, the detachment fault
644 zone is likely a complex, anastomosing structure, with a thickness of ~100 m to ~ a few
645 100s of m (e.g., Karson et al., 2006; Escartín et al., 2017; Parnell-Turner et al., 2018),
646 where fault blocks are transposed in the across-axis direction. In this case, variations in
647 composition vs. borehole depth may not correspond to actual structural depths relative to
648 the fault surface exposed at the seafloor, and now adjacent materials may have originated
649 from different positions in the deformed fault zone. Second, the boreholes also include

650 talus material that is therefore not *in situ*. The present-day location of these samples
651 cannot be directly interpreted as an indicator of spatial and temporal relationships with
652 samples *in situ*, and therefore have an ambiguous structural position. If these debris fields
653 correspond to local mass-wasting deposits, visible at small scales over the detachment
654 fault, the present-day position may be close to the original *in situ* position. Oceanic
655 detachments also display a cover of rubble and sediment that is sourced from the hanging
656 wall, and rafted during extension (e.g., Dick et al., 2008; Escartín et al., 2017). Thus, if
657 the talus material corresponds to rubble cover, its source may be the hanging wall rather
658 than the fault zone or footwall instead, and its present-day location does not necessarily
659 reflect its position at the time of formation, emplacement, impregnation, or alteration.

660 The consistent grouping of samples with a similar melt-impregnation (or lack
661 thereof) and alteration history at the eastern, central, and western sites may suggest that
662 *in situ* samples have not witnessed significant (km-scale), lateral transposition along the
663 fault zone, or at least at spatial scales that are smaller than any local spatial variation
664 associated with melt-rock interactions. Similarly, the samples that are not *in situ*, with
665 similar melt impregnation and alteration histories, suggest a local source; sampling of
666 hanging wall rubble would likely result in an extremely heterogeneous and variable set
667 of ultramafic samples, as observed from the rubble cover from other oceanic detachments
668 (e.g., Escartín et al., 2007). The homogeneity is therefore consistent with a local origin,
669 with material sourced from the mass-wasted fault zone, eventually mixed with basaltic
670 rubble that is present in the boreholes (e.g., Früh-Green et al., 2018).

671 If the samples were either *in situ*, or sourced locally where *ex situ*, and if the lateral
672 transposition of material within an anastomosing fault zone occurs at a smaller scale (< 1
673 km parallel to extension) than the distance between sites (3-5 km), then spatial patterns
674 observed may be interpreted in the context of the internal structure and evolution of the

675 OCC and its detachment fault. Ildefonse et al. (2007) suggested a temporal variation in
676 melt supply to the ridge axis, with an initial phase of accretion that has limited melt
677 supply, and a late phase of accretion with increased melt supply and the emplacement of
678 gabbros in the footwall. This interpretation is consistent with early peridotites just
679 showing melt extraction (western sites), and melt-rock reactions in more recent time
680 associated with this recent magmatic phase (eastern sites).

681 Alternatively, the sites may simply record an inherently heterogeneous system,
682 where melt percolation, and hence melt-rock reactions, are inherently inhomogeneous. If
683 this is the case, the east-west patterns may not reflect a temporal evolution but instead
684 indicate that these heterogeneities occur over distances of a few km, corresponding to the
685 separation among the different sites. This interpretation is consistent with geological
686 observations and drilling that document a change in footwall composition along-axis,
687 with peridotites near the transform fault wall, and gabbros ~ 10 km to the north, hence at
688 spatial scales similar to those shown by the western, central, and eastern sites.

689

690 **7. Conclusions**

691 Peridotites sampled during IODP Expedition 357 comprise predominantly Type I
692 serpentinites, with lesser Type II melt-impregnated and Type III metasomatic
693 serpentinites. A principal variation in the nature and composition of Expedition 357
694 serpentinites is an increase in the volume of melt-impregnation products and a
695 concomitant increase in HFSE and LREE enrichments from the western to central to
696 eastern sites. Whole rock chemical compositions of western site Type I serpentinites are
697 consistent with dominantly fluid-rock interaction associated with serpentinization and
698 oxidation, whereas compositions of eastern site Type II melt-impregnated and Type III
699 metasomatic serpentinites are dominated by melt-rock interaction processes. High to

700 moderate degrees of melt extraction in the peridotites are evident in low concentrations
701 of Al₂O₃, CaO and incompatible elements. Degrees of melt extraction based on whole
702 rock REE suggest a high range of 19-21 % with no apparent variation between sites. Some
703 serpentinite samples are rubble and therefore *ex situ*. Thus, the coherent magmatic history
704 observed at each site may be consistent with a local source originating in the fault zone
705 rather than rafted rubble that would be expected to show more heterogeneity and no
706 spatial pattern. If this is accurate, our sites may provide a time-record of enhanced melt-
707 rock interaction with time, consistent with proposed geological models. Otherwise, the
708 magmatic history may represent heterogeneities in these processes at spatial scales of a
709 few km.

710

711

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727

728 **Figure captions**

729 Figure 1. (a) Location of the Atlantis Massif at 30°N within the context of the greater
730 MAR spreading system (modified from Kelley et al., 2001). (b) Bathymetric map
731 centered on the southern wall of the Atlantis Massif (modified after Rouméjon et al.,
732 2018a). Inset shows the location of the Atlantis Massif on the western flank of the Mid-
733 Atlantic Ridge axial valley, bordered to the south by the Atlantis Fracture Zone. Circles
734 with numbers in italics and beginning with M00 represent shallow drill hole locations
735 from IODP Exp. 357 (Früh-Green et al., 2017, 2018). White circles indicate drill holes
736 with no recovered peridotite; blue, red and yellow circles represent western (M0071),
737 central (M0069, M0072, M0076) and eastern (M0068) sites, with recovered peridotite
738 samples investigated in this study. White squares represent sites (U1309, U1310, U1311)
739 drilled during Exp. 304/305 (Blackman et al., 2006); white star shows the location of the
740 Lost City Hydrothermal Field (LCHF) (Kelley et al., 2001). (c) Simplified downhole plots
741 showing dominant rock types recovered in the seven drill holes hosting serpentized
742 peridotites (modified from Roumejon et al., 2018a; see Fruh-Green et al., 2018 for
743 complete expedition drill hole vertical sections). The sections are arranged from right to
744 left according to longitude (see Fig. 1b), and from hole top to bottom. The central holes
745 embody *in situ* segments of the detachment footwall, whereas the western and eastern site
746 holes which yield rubbly intervals and sedimentary structures are interpreted as artefacts
747 of mass wasting and local faulting. Symbols: circles, harzburgite; squares, dunite;
748 triangles, Ca-rich harzburgite; triangles with slash, Ca-rich dunite; crosses, melt-
749 impregnated harzburgite; x's, plagioclase-impregnated harzburgite; diamonds, talc-

750 amphibole altered harzburgite; diamonds with slash; talc-amphibole-chlorite schist. Blue,
751 red and yellow symbols represent samples of western, central and eastern serpentinitized
752 harzburgites and dunites, respectively, analyzed for whole rock chemical analysis. Note
753 ‘metamafic’ includes metabasalt, metadolerite and metagabbro composition samples.

754

755 Figure 2. Representative thin section images of Expedition 357 Type I serpentinites (a-
756 d), Type II (mafic) melt-impregnated (mm-i) serpentinite (e, f) and Type III metasomatic
757 serpentinites (msp) (g, h). ‘Typical’ completely serpentinitized peridotite samples from (a,
758 b) 71A-1R2-85-89, harzburgite and (c) 71C-5R1-6-8, harzburgite. (d) Ca-rich
759 serpentinitized harzburgite 69A-9R2-8-12 showing extensive network of carbonate
760 veinlets. (e, f) Mafic melt-impregnated (mm-i) harzburgites 76B-7R1-81-83 and 68B-
761 4R1-23-29 with (e) the presence of rare plagioclase feldspar (f) and secondary (replacive)
762 olivine mantling partially dissolved orthopyroxene porphyroclast. (e) Talc-amphibole
763 altered (t-aa) harzburgite 68B-1R1-10-13, showing replacement of orthopyroxene by talc
764 (Tlc) and tremolite (Tr), and (f) talc-schist 68B-1R1-1-6. Image (a) is under plane
765 polarized light; images in (b-h) are under cross-nicols.

766

767 Figure 3. Plot of MgO/SiO_2 vs. Al_2O_3/SiO_2 for all studied serpentinites (top panel, Type
768 I serpentinites, bottom panel, Type II melt-impregnated and Type III metasomatic
769 serpentinites). The lighter shaded samples (light yellow, light blue and pink) of the same
770 shape here and in all other figures are Expedition 357 Atlantis Massif peridotites from
771 Früh-Green et al. (2016); light orange circles and squares harzburgites and dunites,
772 respectively, from IODP Site U1309 Atlantis Massif peridotites (Godard et al., 2009).
773 Global abyssal peridotite data from Niu (2004), Paulick et al. (2006) and Godard et al.
774 (2008), unless stated otherwise. Fields of serpentinitized peridotite and talc-altered

775 peridotite are from Paulick et al. (2006) and melt-impregnated peridotites (MIP) are from
776 Paulick et al. (2006), Godard et al. (2008) and as referenced in Whattam et al. (2011).
777 DMM (depleted MORB mantle) and PM (primitive mantle) from Workman and Hart
778 (2005) and Palme and O'Neill (1983) respectively. The terrestrial array is from Jagoutz
779 (1979).

780

781 Figure 4. Variation diagrams of MgO vs. (a) SiO₂, (b) Fe₂O₃, (c) Al₂O₃, (d) CaO and (e)
782 LOI of Atlantic Massif serpentinites from this study, and peridotites of Godard et al.
783 (2009) and Früh-Green et al. (2018) compared with global MOR peridotite and bulk
784 compositions of serpentine and talc (Blanco-Quintero et al. 2011, mean of ten and eight
785 serpentine and talc analyzes). Symbols as in Fig. 3.

786

787 Figure 5. (a-e) Chondrite-normalized REE and (f-j) primitive mantle-normalized
788 incompatible element plots of Atlantis Massif serpentinites from this study and peridotites
789 of the study of Früh-Green et al. (2018). Chondrite and primitive mantle values are from
790 McDonough and Sun (1995). Shown for comparison is mean abyssal peridotite
791 composition determined on the compilations of Niu (2004) and Bodinier and Godard
792 (2003) by Godard et al. (2008), and depleted MORB mantle (DMM) (Workman and Hart,
793 2005).

794

795 Figure 6. IODP Expedition 357 serpentinite samples from this study, and peridotites of
796 the study of Früh-Green et al. (2018) on plots of (a) U and (b) Sr vs. site. Note that three
797 of the four high-CaO Type I serpentinites (with 5.59-10.50 wt.% CaO) also exhibit the
798 highest concentrations of Sr (> 1000 µg/g). Symbols as in Fig. 3.

799

800 Figure 7. Expedition 357 Atlantis Massif serpentinites from this study and peridotites of
801 the study of Früh-Green et al. (2018) in plots of (a) Th, (b) Hf, (c) Zr, (d) Ce/Yb and (e)
802 total REE vs. site. Plotted for comparison are the ranges of fluid-rock dominated and melt-
803 rock dominated global abyssal peridotite (GAP, references as in the caption for Fig. 5).
804 Depleted mantle (DM) and primitive mantle (PM) compositions are from Workman and
805 Hart (2005) and Lyubetskaya and Korenaga (2007), respectively. Symbols as in Fig. 3.

806

807 Figure 8. Primitive mantle-normalized plots of the mean compositions of (a) global
808 abyssal peridotites interpreted as being dominated by fluid-rock and melt-rock reactions,
809 (b) Atlantis Massif serpentinites from this study and (c) central serpentinites parsed into
810 site. The fluid-rock dominated peridotite is from Paulick et al. (2006) and represent
811 peridotites collected during ODP Leg 209, Sites 1268, 1772, 1274. The melt-rock
812 dominated peridotite is from Niu (2004) and Paulick et al. (2006) and in the case of
813 Paulick et al. (2006), represent peridotites collected from ODP Leg 209, Sites, 1270,
814 1271. Symbols as in Fig. 3

815

816 Figure 9. Expedition 357 Atlantis Massif serpentinites from this study and peridotites of
817 the studies of Godard et al. (2009) and Früh-Green et al. (2018) in plots of (a, b) Nb vs.
818 La, and (c, d) Ti vs. Dy. Element abundances listed as '0' were assumed to be below
819 detection limit and not used in calculation of the regression. Colors of the regressed lines
820 and text for the Expedition 357 serpentinites include: blue, western; red, central; and
821 black, eastern. In (b) and (d), the light blue, pink and light grey regressed lines represent
822 mean of the western, central and eastern sites (i.e., the blue, red and black lines in (a) and
823 (c)). R^2 values shown for Exp. 357 serpentinites represent ones which include the
824 peridotite samples of Früh-Green et al. (2018) in addition to the serpentinites from this

825 study and the numbers in italics in brackets beside R^2 values represent number of samples
826 used in regression calculation. The orange line represents the dataset of Atlantis Massif
827 peridotites from Site U1309 (Godard et al., 2009). Linearly regressed vectors (grey)
828 labelled *A*, *B*, and *C* in (a) and (c) represent data from global abyssal peridotite (GAP)
829 and for visual clarity, samples are omitted from the plot. The *A* trend represents the dataset
830 of Niu (2004) which was interpreted by Paulick et al. (2006) as being representative of
831 dominant melt-rock interaction (i.e., melt-impregnation); *B* and *C* are from Paulick et al.
832 (2006) and represent peridotites interpreted as having compositions dominantly
833 associated with melt-rock reaction (*B*, ODP Leg 209, MAR, Sites, 1270, 1271) and fluid-
834 rock reaction (*C*, Sites 1268, 1772, 1274). Depleted mantle (DM) and primitive mantle
835 (PM) compositions from Workman and Hart (2005) and Lyubetskaya and Korenaga
836 (2007), respectively. Symbols as in Fig. 3.

837

838 Figure 10. (a) REE compositions of the most melt-depleted sample from each site
839 compared with model estimates of 10, 15, and 20 % partial melting of DMM as well as
840 three ‘best fit’ models of melting and subsequent refertilization to estimate the extent of
841 melting experienced by these samples. Modeling is of non-modal fractional melting
842 (dashed curves encompassing light green shade) of partial melting in % (10, 15, 20) of a
843 four phase DMM spinel lherzolite source comprising ol: opx: cpx: sp in modal
844 proportions of 0.53: 0.27: 0.17: 0.03 and a melt mode of -0.06: 0.28: 0.67: 0.11
845 (Hellebrand et al. 2002). Initial source composition, DMM from Workman and Hart
846 (2005). Partition coefficients from Suhr et al., 1998; missing REE interpolated. Also
847 indicated are compositions of rocks after 20 % non-modal fractional melting and
848 subsequent 1 % or 10 % addition of a mafic melt (mm, uppermost grey shaded area) with
849 a REE composition of the most enriched sample (eastern site M0068A-1R1-34-35 talc-

850 schist, Früh-Green et al., 2018, see Fig. 4e) as a proxy for a slightly LREE-enriched melt
851 (darker green shade). Symbols as in Fig. 3.

852

853 **Table captions**

854 Table 1 Evidence and means for classification of Expedition 357 serpentinites subjected
855 to melt-impregnation and (silica) metasomatism.

856

857 Table 2 Mineralogy and modality of Expedition 357 serpentinites.

858

859 Table 3 Whole rock ICP-AES and ICP-MS analyses of Expedition 357 Atlantis Massif
860 serpentinites from this study.

861

862 **Supplementary material**

863 See Supplementary Document

864

865 **References**

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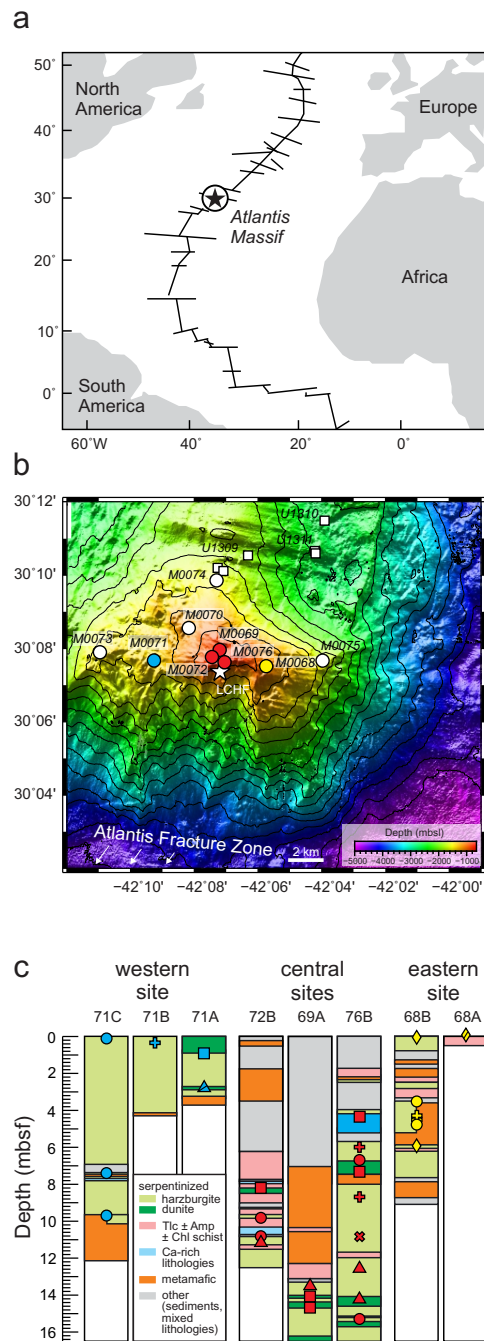


Figure 1 PR1 Whattam et al. (2021)

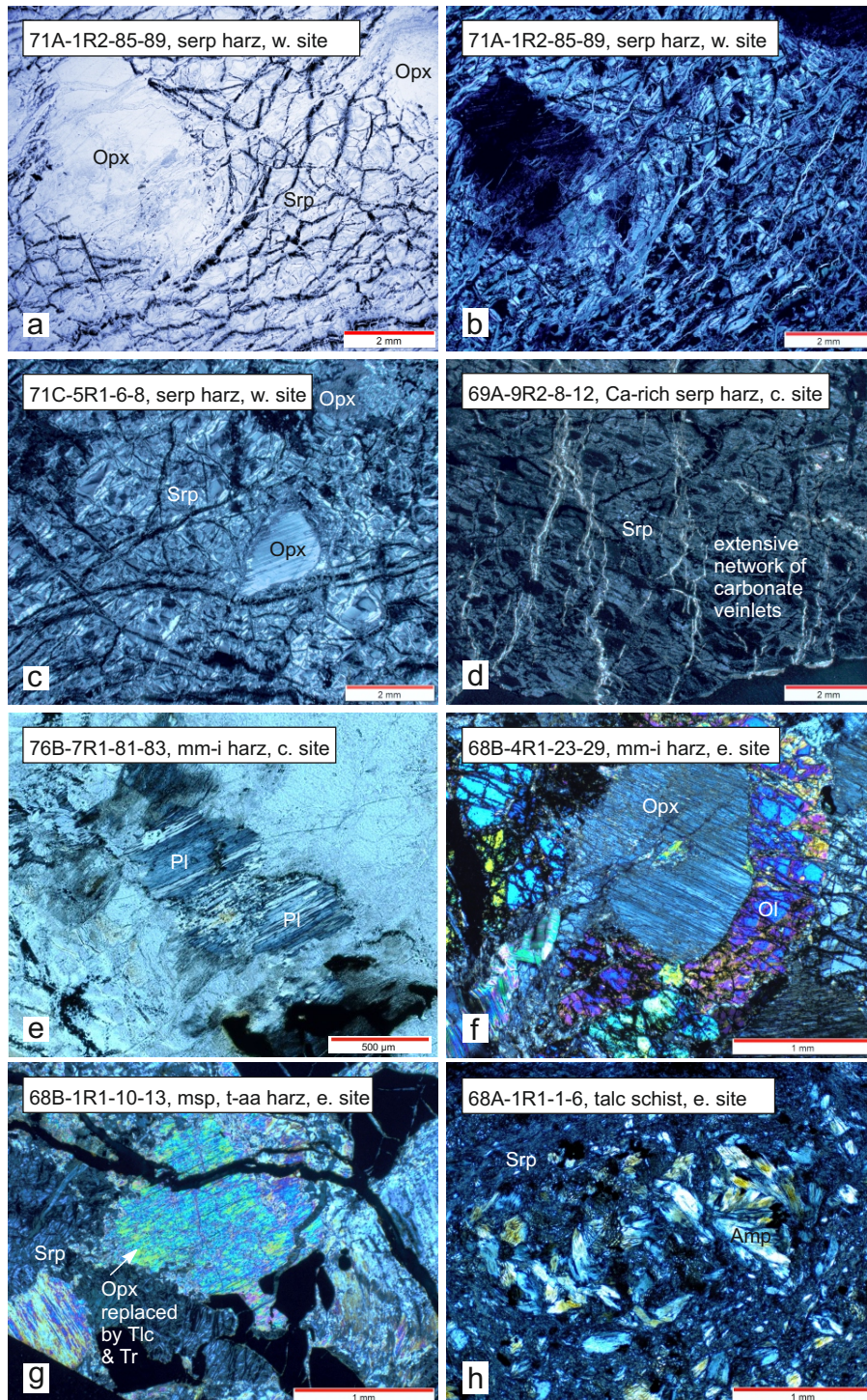


Figure 2 P-S 1A Whattam et al. (2021)

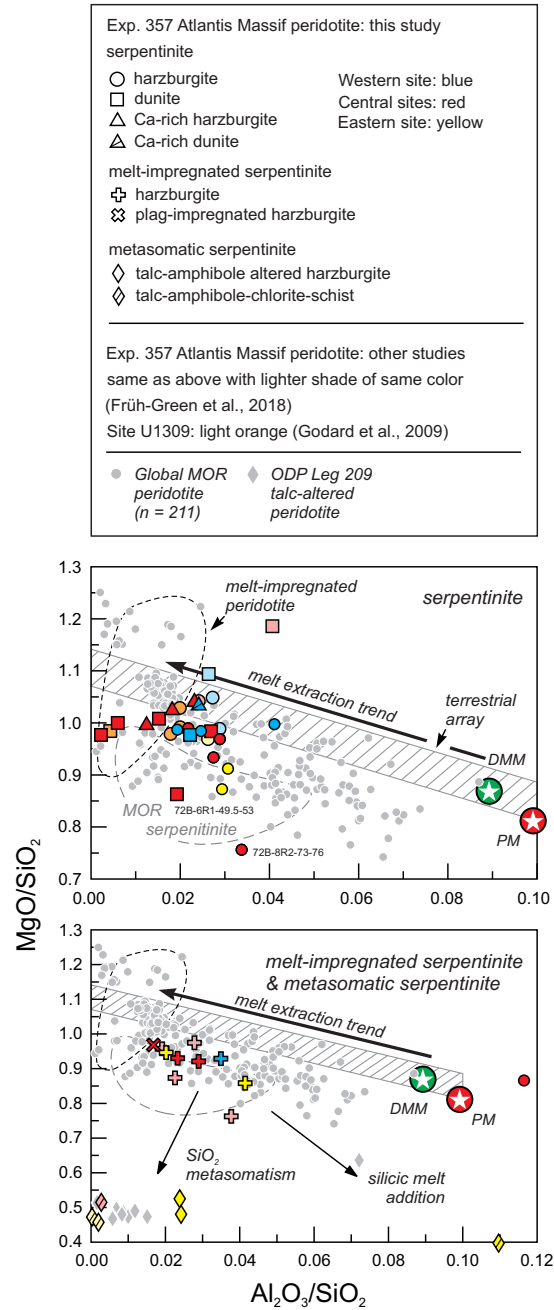


Figure 3 PR1 Whattam et al. (2021)

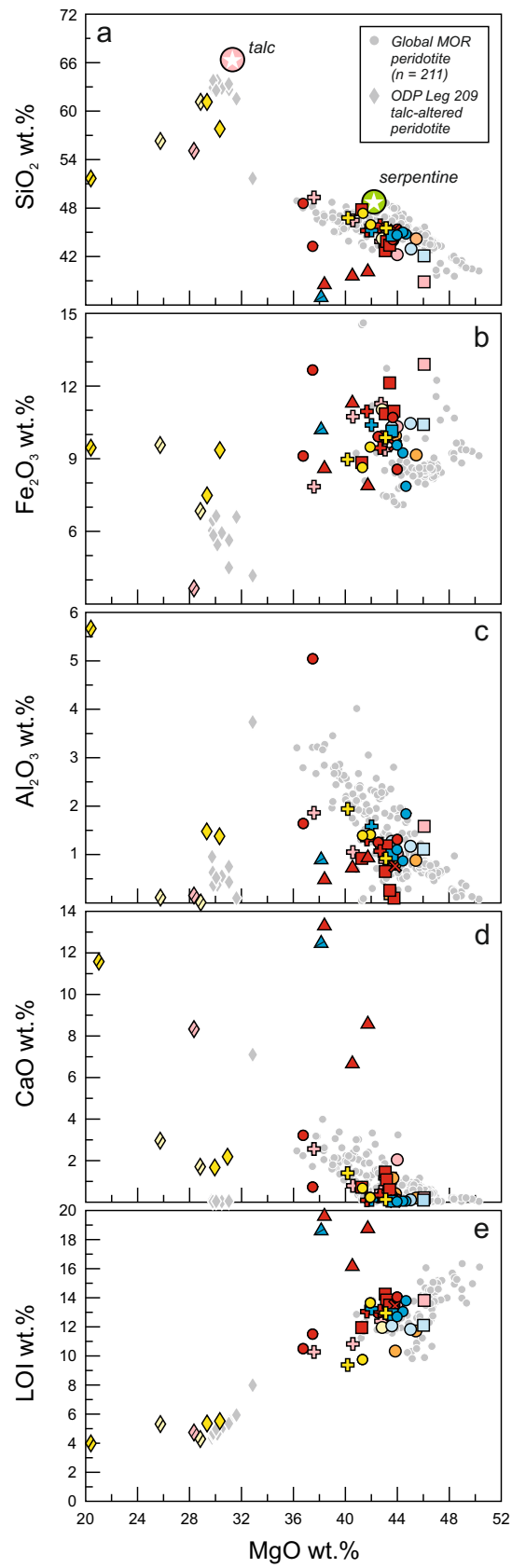


Figure 4 P-S 1A Whattam et al. (2021)

Figure 5

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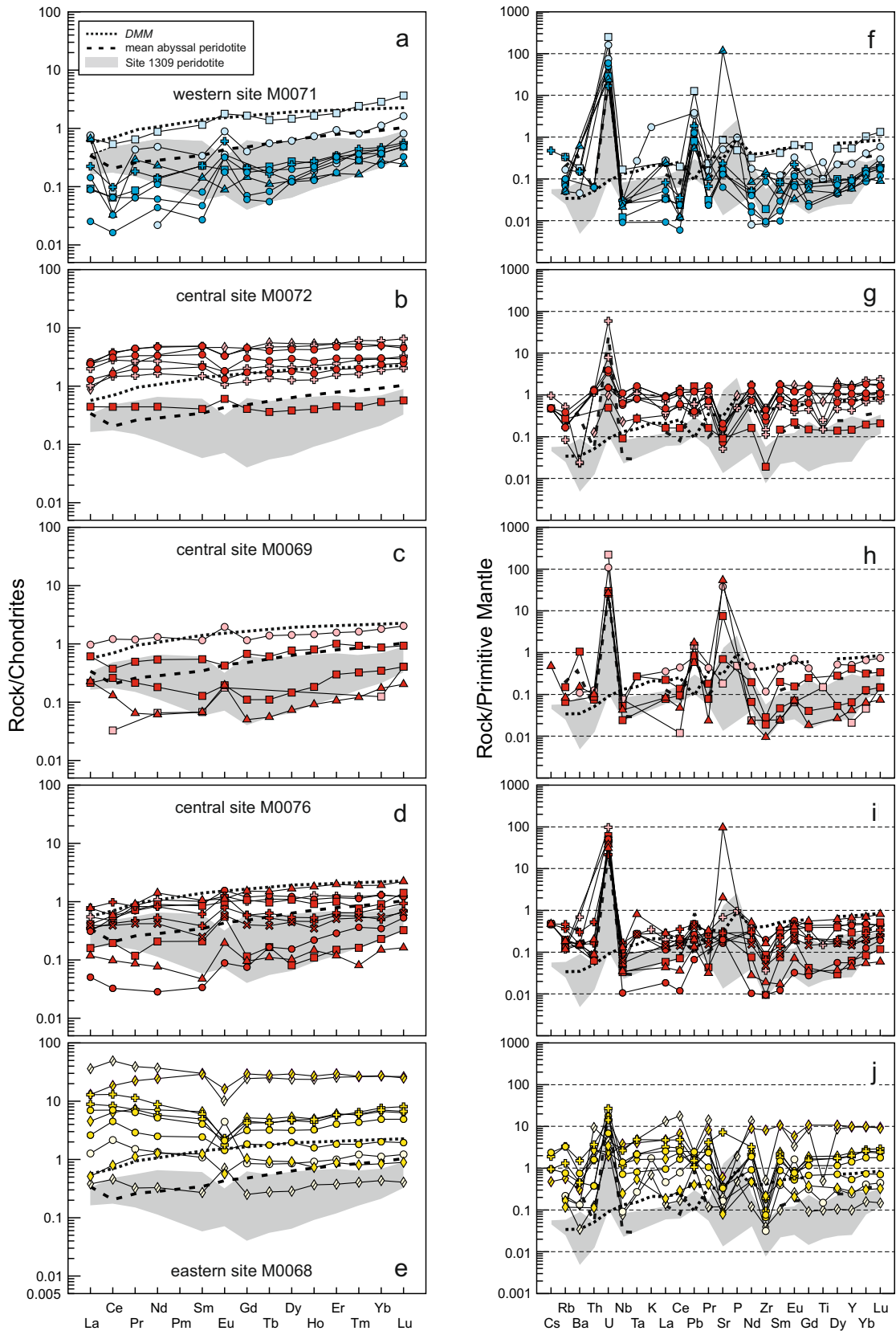


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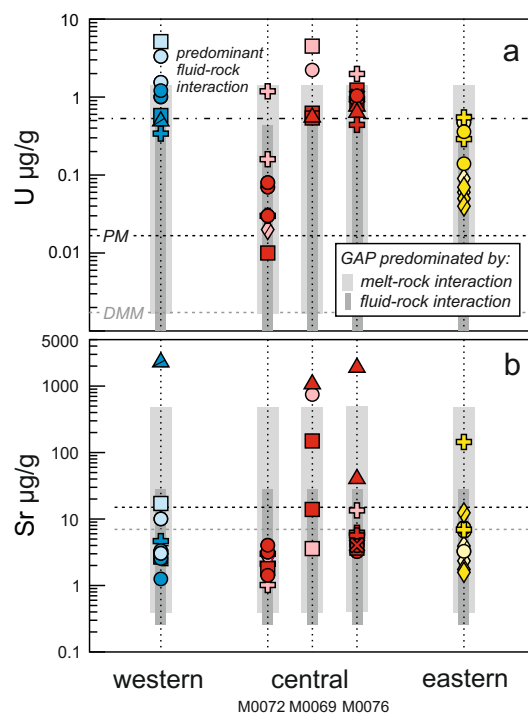


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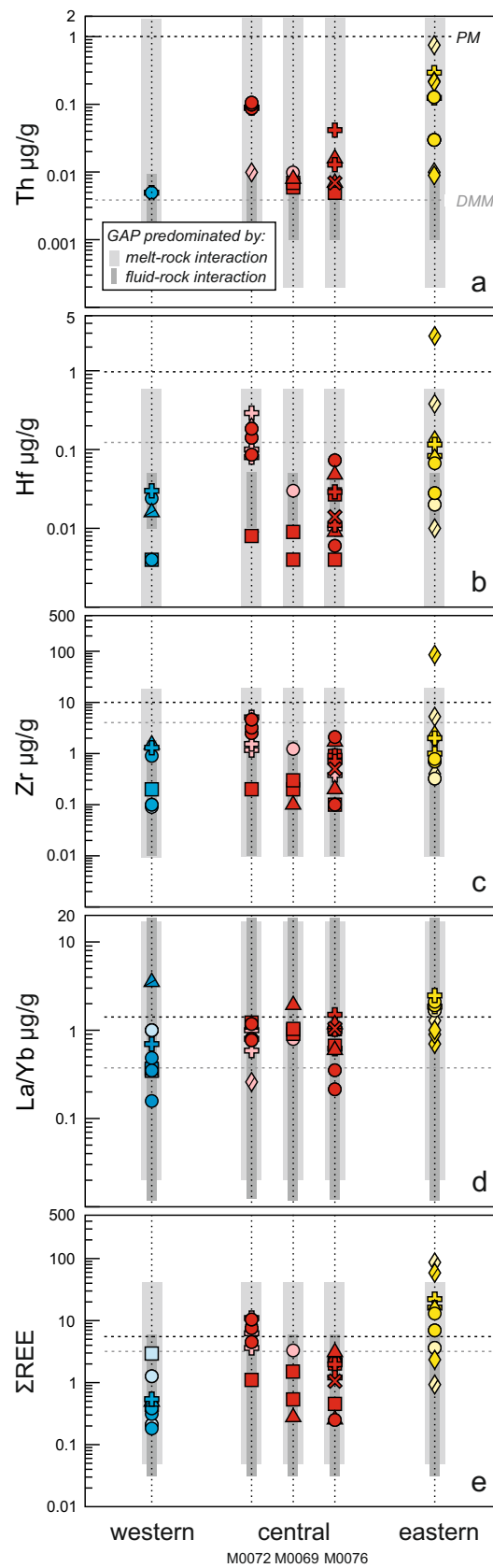


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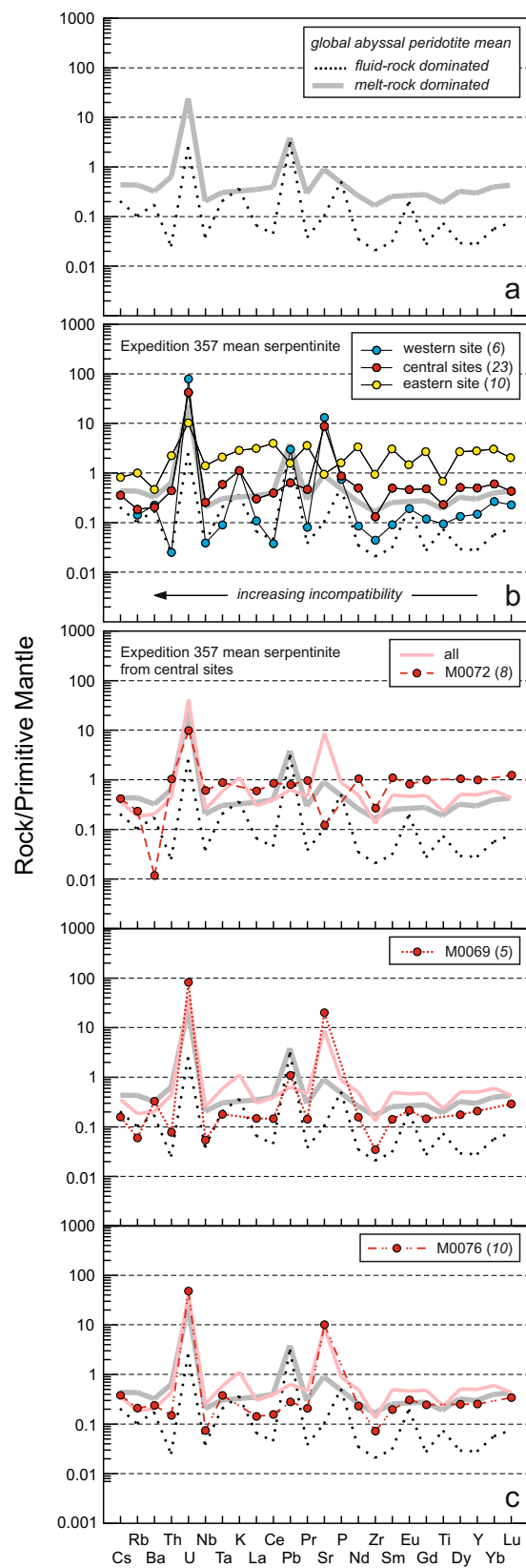


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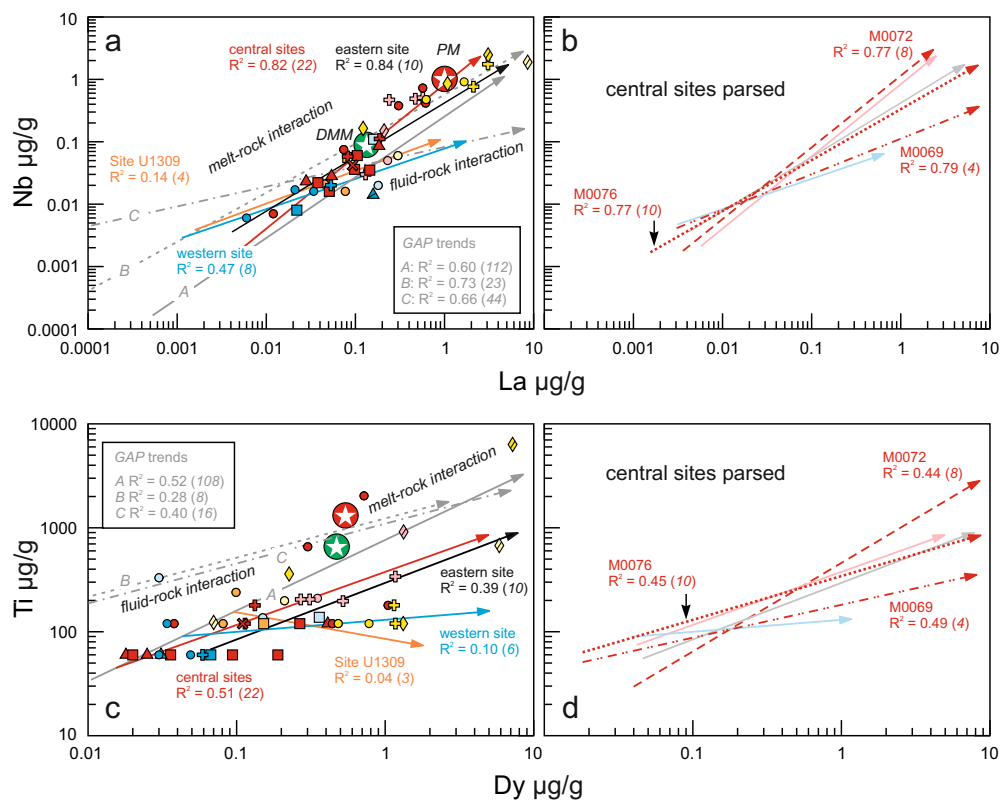


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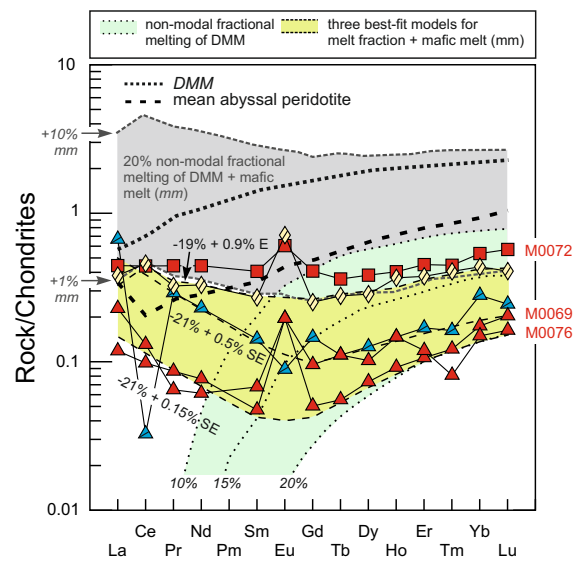


Figure 10 PR1 Whattam et al. (2021)

Table 1 Evidence and means for classification of Expedition 357 Atlantis Massif serpentinites subjected to melt-impregnation and (silica) metasomatism.

Site	Hole	Sample	Petrologic Type	Lithology	Evidence
western	M0071B	71B-1RCC-1-3	II	mm-i harzburgite	**macroscopic: presence of gabbro veins
	M0072B	72B-7R1-76.5-79.5	II	*cryptically mm-i harzburgite	whole rock chemistry: incompatible element enrichment
		72B-8R2-23-26	II	cryptically mm-i harzburgite	whole rock chemistry: incompatible element enrichment
central	72B-8R2-73-76		II	cryptically mm-i harzburgite	whole rock chemistry: incompatible element enrichment
		M0076B	76B-4R1-73-78	II	mm-i harzburgite
		76B-6R1-65-68	II	mm-i harzburgite	macroscopic: presence of mafic melt intrusions
		76B-7R1-81-83	II	plagioclase-impregnated	petrographic: presence of rare plagioclase feldspar
	M0068A	68A-1R1-1-6	III	talc-schist	petrographic: prevalence of secondary talc-amphibole whole rock chemistry: high SiO ₂ (51.7 wt.%), low MgO/SiO ₂ (0.39), high Al ₂ O ₃ /SiO ₂ (0.11)
eastern	M0068B	68B-1R1-10-13	III	t-aa harzburgite	petrographic: talc-amphibole replacement of orthopyroxene whole rock chemistry: high SiO ₂ (57.8 wt.%), low MgO/SiO ₂ (0.52)
		68B-4R1-23-29	II	mm-i harzburgite	petrographic: fresh olivine mantling orthopyroxene; olivine interpreted as recrystallized from mafic melt
		68B-4R1-45-49	II	mm-i harzburgite	macroscopic: presence of gabbro intrusions
		68B-6R1-8-10	III	t-aa harzburgite	petrographic: talc-amphibole replacement of orthopyroxene whole rock chemistry: high SiO ₂ (61.1 wt.%), low MgO/SiO ₂ (0.48)

Notes:

Type I serpentinites ($n = 20$ of 29 total samples, see Table 3) are not listed as the only evidence for their classification is lack of melt-impregnation products and metasomatism. Type I includes the Ca-rich serpentinites ($n = 4$).

Abbreviations: mm-i, mafic melt-impregnated; t-aa, talc-amphibole altered.

Type II mm-i peridotite include plagioclase-impregnated peridotite.

*cryptically melt-impregnated refers to those peridotite which bear no evidence of impregnation at the scale of core or thin section (see text for further details).

**macroscopic evidence of mafic melt-impregnation determined by Expedition 357 Scientific Party at Onshore Scientific Party, University of Bremen, 2016 (see also Frth-Green et al., 2017, 2018).

Table 2 Mineralogy and modality of Expedition 357 serpentinites.

Informal sample no.	Hole	Core	Interval (cm)	Top depth (m)	Bottom depth (m)	Unit	Site	Petrologic Type*	Rock type	OI	Modal proportions (%)****	Spl****	Notes
											Opx	Spl****	
19-QU	M0071C	1R1	6-10	0	2.68	1	W	I	serpentinized harzburgite	85	15	1-2	Spl ranges up to 4 mm
5-GT	M0071C	5R1	6-8	7.38	9.63	1	W	I	serpentinized harzburgite	80	20	2-3	Spl usually embedded in Opx
10-GT	M0072B	8R2	23-26	10.71	12.43	1	C	I	serpentinized harzburgite	75	25	3	Spl embedded in Opx
7-GT	M0069A	9R2	8-12	13.04	14.72	1	C	I	Ca-rich harzburgite	90	10	0	secondary carbonate veins (sub-mm) comprises ~5% by volume
2-QU	M0069A	9R2	103-106	13.04	14.72	4	C	I	serpentinized dunite	100	0	0	
8-QU	M0076B	3R1	78-82	3.44	5.16	3	C	I	serpentinized dunite	100	0	<1	
13-QU	M0076B	5R1	59-61	6.73	7.98	1	C	I	serpentinized dunite	100	0	0	
13-QU	M0076B	6R1	65-68	7.98	9.72	1	C	II	mm-1 harzburgite	70	30	0	
11-GT	M0076B	7R1	81-83	9.72	11.15	1	C	II	plg-impreg. harzburgite	85	15	0	pl present only as two ~500 µm discrete grains
14-GT	M0076B	9R1	96-100	12.87	14.99	1	C	III	Ca-rich harzburgite	85	15	3	secondary carbonate veins (sub-mm) comprises ~10% by volume
6-GT	M0068B	1R1	10-13	0	1.72	1	E	III	1-aa harzburgite	70	30	2	Spl embedded in Opx
19-GT	M0068B	4R1	23-29	4	5.72	2	E	II	mm-1 harzburgite	75	25	2-3	Spl embedded in Opx
12-QU	M0068B	4R1	45-49	4	5.72	3	E	II	mm-1 harzburgite	90	10	0	secondary (replacive) OI comprises ~5% by volume
18-GT	M0068A	1R1	1-6	0	1.97	1	E	III	talc schists**	-	-	0	

Notes:

Abbreviations: mm-1, mafic melt-impregnated; 1-aa, talc-amphibole altered; GT-Cent; QU, Queen's University.

*Petrologic Types: I, serpentinites; II, melt-impregnated serpentinites; III, mafic serpentinites.

Type I includes Ca-rich serpentinites; Type II includes plagioclase-impregnated (plg-impreg) serpentinites.

**The high abundances of Hf,Se (e.g., Ti, Nb, Zr; see Table 3) suggests the protolith of the talc schist was mafic.

***Modal proportions of the protolith are visually estimated.

****Spl is usually sub-mm.



Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: