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1 Evolution of recycled crust within the mantle: Constraints  
2 from the garnet pyroxenites of the External Ligurian  
3 ophiolites (northern Apennines, Italy)

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10 **ABSTRACT**

11 The pyroxenite-peridotite sequence from the External Ligurian (northern  
12 Apennines, Italy) ophiolites **is evidence of** the evolution of recycled crust within the  
13 mantle. We present new major and trace element **and** Nd-Hf isotopic compositions of  
14 garnet clinopyroxenites and websterites from this mantle section. The garnet  
15 clinopyroxenites display clinopyroxene and bulk-rock rare earth element patterns with  
16 distinct positive Eu anomalies, which argue for the involvement of plagioclase-rich  
17 precursors in their origin. We propose that the garnet clinopyroxenites formed by  
18 crystallization of eclogite-derived melts that **underwent** negligible interaction with the  
19 host peridotites. The garnet websterites are interpreted to have been produced by  
20 reactions between the eclogite-derived melts and peridotites, thereby giving rise to  
21 hybrid, second-stage pyroxenites with a crustal geochemical fingerprint. In our  
22 petrogenetic scenario, a rifting-related event at ca. 220 Ma caused melting of eclogites

23 originating from a mid-oceanic ridge basalt-type gabbroic sequence. These mafic  
24 protoliths underwent a long-lived evolution of recycling in the mantle (1.5–1.0 Ga). We  
25 show that heterogeneity of crustal protoliths, age of recycling, and interaction with the  
26 host peridotites may lead to a significant compositional and isotopic diversity of crust-  
27 derived mantle pyroxenites.

## 28 INTRODUCTION

29 Pyroxenites are the main geochemical heterogeneity in the upper mantle; they are  
30 interpreted to be cumulates crystallized from peridotite-derived melts (Downes, 2007) or  
31 subducted crustal material recycled into the mantle (Allegre and Turcotte, 1986;  
32 Kornprobst et al., 1990). The evolution of recycled crust is accomplished through  
33 multiple stages, so that the original components may be difficult to decipher in the final  
34 products (Blichert-Toft et al., 1999). Experiments (e.g., Yaxley and Green, 1998;  
35 Lambart et al., 2012) and geochemical studies of peridotite-pyroxenite mantle sequences  
36 (e.g., Garrido and Bodinier, 1999; Van Acken et al., 2010) have shown that recycled  
37 pyroxenites record processes such as partial melting, migration of pyroxenite-derived  
38 melts, and variable extents of melt-peridotite reaction.

39 Mantle pyroxenites are believed to play a major role in basalt petrogenesis (e.g.,  
40 Sobolev et al., 2007); they have a wide compositional variability and may produce silica-  
41 oversaturated to undersaturated melts by partial melting (Kogiso et al., 2004). Pyroxenite-  
42 derived melts modified by reactions with mantle peridotites (Mallik and Dasgupta, 2012)  
43 or melts of **second-stage** **[[SU: no quote marks needed]]** pyroxenites formed by these  
44 reactions (Lambart et al., 2012) were proposed to participate in the genesis of ocean  
45 island basalts (OIB). Minor amounts of pyroxenite-derived melts were also suggested

46 (Hirschmann and Stolper, 1996; Sobolev et al., 2007) to explain the origin of mid-oceanic  
47 ridge basalts (MORB). In spite of the robust evidence for the occurrence of recycled  
48 crustal lithologies in the OIB-MORB sources, direct samplings of such mantle  
49 heterogeneities are not available. To provide insights into the composition and the  
50 evolution of crust-derived heterogeneities in the upper mantle, we have carried out a  
51 geochemical investigation of garnet pyroxenites from the mantle sequences of the  
52 External Ligurian ophiolites (northern Apennines, Italy). We show for the first time how  
53 remobilization of melts derived by recycled mafic heterogeneities produces a new  
54 generation of pyroxenites, which are natural analogs of the second-stage pyroxenites  
55 experimentally envisaged as key rocks for OIB formation. This study also reveals that  
56 Hf-Nd isotope compositions of mantle pyroxenites provide timing constraints to the  
57 process of crustal recycling.

## 58 **GEOLOGICAL AND PETROLOGICAL BACKGROUND AND SAMPLES**

59 The External Ligurian ophiolites from the northern Apennines are slices of  
60 oceanic lithosphere formed at a fossil ocean-continent transition (Marroni et al., 1998).  
61 The External Ligurian mantle sequences are interpreted as deep subcontinental  
62 lithosphere exhumed to the ocean floor in response to Mesozoic lithospheric thinning and  
63 opening of the Jurassic Ligurian-Piedmontese basin (Montanini et al., 2006). The  
64 sequences consist of spinel-plagioclase lherzolites with prevailing MORB-type isotopic  
65 signature and diffuse pyroxenite layers (Montanini et al., 2012; Borghini et al., 2013).  
66 The mantle bodies considered in this study include frequent websterite layers (up to 10  
67 cm thick) and rare garnet clinopyroxenite layers (up to 2 m thick) recording an early  
68 stage of equilibration at ca. 2.8 GPa. The websterites in places occur along the garnet

69 clinopyroxenite–herzolite boundaries. Previously recognized garnet clinopyroxenites are  
70  $\text{Al}_2\text{O}_3$  and CaO rich, and have low Mg# values [ $100 \times \text{Mg}/(\text{Mg} + \text{Fe}) = 71\text{--}66$ ; Montanini  
71 et al., 2012]. On the basis of **rare earth element** (REE) compositions, they were  
72 subdivided into two types (**A and B**). These pyroxenites were related to recycling of  
73 crustal mafic protoliths that underwent **a** high degree of partial melting in the garnet  
74 stability field. Lu-Hf isotope correlations suggest **a** Triassic age for the melting event.

75 In this work we report the presence of another thick layer (~1.5 m) of garnet  
76 clinopyroxenites, referred to **here as** type C, displaying distinctive geochemical  
77 characteristics (see **following**). These rocks have a retrogressed assemblage mainly  
78 composed of clinopyroxene, orthopyroxene, and minor plagioclase (5–10 vol%) and  
79 green spinel. Former garnet is replaced by orthopyroxene including vermicular spinel and  
80 secondary clinopyroxene. Our study also includes **the** geochemical characterization of the  
81 websterites, which are spinel-bearing rocks including a substantial amount of  
82 orthopyroxene (40–50 vol%). Evidence for an early garnet-bearing assemblage in the  
83 websterites is given by rounded symplectites consisting of orthopyroxene, clinopyroxene,  
84 and spinel, locally preserving tiny garnet relics (Montanini et al., 2006). Both type C  
85 pyroxenites and websterites contain clinopyroxene porphyroclasts commonly displaying  
86 exsolved lamellae of plagioclase.

## 87 **ELEMENTAL AND Nd-Hf ISOTOPE COMPOSITIONS**

88 Type C garnet clinopyroxenites have relatively high contents of  $\text{Al}_2\text{O}_3$ , CaO, and  
89  $\text{Na}_2\text{O}$ , and Mg# **ranges** from 84 to 78 (Table DR1 in the GSA Data Repository<sup>1</sup>). These  
90 pyroxenites show slightly depleted to weakly enriched light (L) REEs, positive Eu and Sr  
91 anomalies, **and** nearly flat to slightly enriched heavy (H) REEs (Fig. 1A). The websterites

92 have lower contents of  $\text{Al}_2\text{O}_3$  and CaO, and higher Mg# (Table DR1). They are  
93 characterized by moderate LREE depletion, no Eu anomaly, variable Sr enrichment, and  
94 nearly flat to slightly enriched HREEs (Fig. 1A).

95 In the pseudoternary system forsterite (Fo)–Ca-Tschermak (CaTs)–quartz (Qtz)  
96 projected from diopside (Di), the type C pyroxenites plot near the enstatite (En)–CaTs  
97 join (Fig. 2), i.e., close to the garnet-pyroxene plane. This is consistent with a primary  
98 assemblage dominated by garnet + aluminous clinopyroxene. The websterites have low  
99 CaTs contents and plot at the left of the En–CaTs join, in the field of silica-deficient  
100 pyroxenites. Therefore, these rocks most likely contained olivine as an additional phase  
101 beside garnet and pyroxenes (Garrido and Bodinier, 1999; Herzberg, 2011).

102 The porphyroclastic clinopyroxenes from type C garnet clinopyroxenites ( $\text{Al}_2\text{O}_3$   
103 8–11 wt%) have heterogeneous incompatible element compositions (Fig. 1B; Table DR2;  
104 Figs. DR1 and DR2). Some of these clinopyroxenes display low concentrations of  
105 HREEs, Y, and Sc, and chondrite-normalized patterns characterized by depletion of  
106 HREEs with respect to middle (M) REEs. These chemical characteristics are commonly  
107 attributed to geochemical equilibrium with garnet. Therefore, they most likely represent  
108 relics of primary clinopyroxene that were not homogenized during redistribution of trace  
109 elements after garnet breakdown. We also found clinopyroxene porphyroclasts with  
110 higher concentrations of HREEs, Y, Sc, and nearly flat MREE to HREE patterns. These  
111 compositions are interpreted to reflect release of these trace elements from garnet. A  
112 remarkable feature of the clinopyroxene REE patterns is the positive Eu anomaly, which  
113 mirrors that of the whole rocks.

114 The clinopyroxene porphyroclasts from the websterites display REE patterns with  
115 variable MREE-HREE fractionation (Fig. 1B; Fig. DR2). In the websterite BA3-W, relict  
116 Al<sub>2</sub>O<sub>3</sub>-rich (~11 wt%) clinopyroxene with plagioclase exsolution shows HREE depletion  
117 and a positive Eu anomaly (Fig. 1B), whereas the recrystallized clinopyroxene (Fig.  
118 DR1) has lower Al<sub>2</sub>O<sub>3</sub> (~8 wt %), nearly flat HREE, and slight LREE and MREE  
119 enrichment (Fig. DR2). The clinopyroxene from one of the websterites (AM397) has an  
120 REE pattern characterized by a steady increase from LREE to HREE that may originate  
121 from replacement of garnet (Fig. DR2).

122 Some clinopyroxenes from both pyroxenite types have convex-upward REE  
123 patterns and relatively high LREE and MREE contents (type C samples AM288a and  
124 AM404, and websterites AM353 and AM393), which may be associated with slight Nb,  
125 Th, and U enrichment (Table DR2). These observations argue for interaction with  
126 migrating melts during exhumation; therefore, only the few samples lacking evidence for  
127 late metasomatism (type C pyroxenite AM403 and websterite BA3-W) have been taken  
128 into account for the trace element modeling reported in the following.

129 Nd-Hf isotope compositions of the least retrogressed type C garnet  
130 clinopyroxenites and websterites (Table DR3) were corrected for radiogenic ingrowth at  
131 220 Ma. This age is inferred for a fractionation event in the mantle related to continental  
132 rifting and asthenospheric ascent (Montanini et al., 2012). The type C garnet  
133 clinopyroxenite (AM403) shows substantial departure from the OIB-MORB field and  
134 Nd-Hf decoupling due to the highly radiogenic Hf composition (Fig. 3). The websterite  
135 BA3-W has Nd-Hf isotope composition comparable to type A and B garnet  
136 clinopyroxenites.

137 **DISCUSSION**

138 **Origin of Type C Garnet Clinopyroxenites**

139       The presence of positive Eu-Sr anomalies in whole rocks and relict  
140 clinopyroxenes document that plagioclase-rich protoliths, crystallized under relatively  
141 low-pressure (*P*) conditions, played a major role in the petrogenesis of type C  
142 clinopyroxenites. Type C clinopyroxenites are chemically distinct from gabbroic rocks,  
143 which plot on the silica-rich side of the Fo-CaTs-Qtz-Di projection (Fig. 2). This  
144 therefore argues against an origin by subsolidus metamorphic transformations of gabbroic  
145 protoliths. Following the procedure of Wood and Blundy (1997), the parental melts of  
146 type C clinopyroxenites are inferred to have relatively low Mg# of 54–49. Such values  
147 are consistent with those observed for melts produced experimentally by olivine-free  
148 mafic rocks under high-*P* conditions (Yaxley and Sobolev, 2007). We propose that type  
149 C clinopyroxenites represent garnet-clinopyroxene cumulates from eclogite and/or  
150 pyroxenite melts. The low HREE contents and the pronounced positive Eu-Sr anomalies  
151 suggest low-*P* precursors with high plagioclase + olivine modes (see Kornprobst et al.,  
152 1990; Aulbach et al., 2007). Conversely, protoliths displaying a high proportion of  
153 clinopyroxene are expected to produce melts (and derivative cumulates) with higher  
154 HREE contents and less prominent (or absent) Eu anomalies, similar to the type A and B  
155 garnet clinopyroxenites (Fig. 1A).

156       We tested our hypothesis by incompatible trace element (REE, Y, Zr, Hf, Sr)  
157 modeling on the basis of whole-rock and clinopyroxene compositions (Fig. 1C).  
158 Calculated parental melt compositions display slight LREE enrichment, positive Eu and  
159 Sr anomalies, and marked HREE depletion. They show remarkable similarity with

160 computed melts formed by partial melting of eclogites derived from plagioclase- and  
161 olivine-rich protoliths (i.e., troctolites). On the basis of incompatible trace element  
162 compositions, an origin as residua after partial melting of mafic precursors cannot be  
163 ruled out (see also Montanini et al., 2012). This refractory origin is, however, at variance  
164 with the lack of the marked depletion in highly siderophile elements (Montanini et al.,  
165 2013), as expected for melting residua of recycled mafic crust (Van Acken et al., 2010).

166 The highly radiogenic Hf composition of the type C garnet clinopyroxenite  
167 AM403 (Fig. 3) records a time-integrated growth related to high Lu/Hf ratios.  
168 Conversely, type A and B garnet clinopyroxenites plot below the mantle array. These  
169 differences in Nd-Hf isotopic compositions are explained considering that MORB-  
170 derived gabbros are characterized by Lu/Hf ratios lower than their mantle source  
171 (depleted mantle [[SU: correct?]]) and thus evolve with time below the mantle array  
172 (Stracke et al., 2003), whereas MORB-derived troctolites may have significantly higher  
173 Lu/Hf ratios (Borghini et al., 2007; Godard et al., 2009) leading to radiogenic Hf isotope  
174 compositions. In the  $\epsilon_{\text{Nd}}-\epsilon_{\text{Hf}}$  diagram, type A, B, and C garnet clinopyroxenites  
175 approximate the curves of ancient, recycled MORB-derived gabbros and troctolites,  
176 respectively. We propose that no melt extraction or other substantial fractionation of  
177 Sm/Nd and Lu/Hf ratios occurred during the recycling evolution of the garnet  
178 clinopyroxenite protoliths. According to Nd-Hf isotope systematics, old ages may be  
179 postulated for the mafic precursors of all the garnet clinopyroxenites, which are close to  
180 1.5–1.0 Ga oceanic crust (Fig. 3). The Nd-Hf isotopic compositions of type C pyroxenite  
181 document that recycled oceanic crust includes rock types that diverge from the HIMU  
182 (high  $\mu$ ;  $\mu = {}^{238}\text{U}/{}^{204}\text{Pb}$ ) component (and the OIB field). If these rock types were involved

183 in the mantle plumes, their Nd-Hf isotopic contribution to basalt petrogenesis is likely  
184 diluted by mixing during melt formation, extraction, and ascent (Stracke et al., 2003).

### 185 **Origin of the Websterites**

186 The websterites have major and trace element characteristics requiring a distinct  
187 origin with respect to the associated garnet clinopyroxenites. The hypothesis that the  
188 websterites are recycled gabbroic rocks or related to melts formed by eclogites is  
189 **precluded** by their high MgO and low Al<sub>2</sub>O<sub>3</sub> contents. To assess the nature of the parental  
190 melts of the websterites, we calculated the incompatible trace element compositions of  
191 the liquid in equilibrium with the primary Al-rich clinopyroxene. The computed melt is  
192 characterized by a steep LREE-HREE fractionation and positive Sr and Eu anomalies  
193 (Fig. 1D). The websterites therefore preserve the geochemical fingerprint of a **crustal**  
194 **[[SU: no quote marks]]** melt precursor (Sobolev et al., 2000), similar to type A garnet  
195 clinopyroxenites occurring in the same mantle sequence (Montanini et al., 2012; Fig.  
196 1D).

197 Because the eclogite-derived melts have typically low MgO contents (<7 wt%;  
198 Yaxley and Green, 1998; Yaxley and Sobolev, 2007), the formation of the websterites  
199 requires the involvement of an MgO-rich component. We propose that they have a hybrid  
200 origin through peritectic melt-consuming reaction between eclogite-derived melts and  
201 host peridotites, in agreement with the local occurrence of websterites as margins of the  
202 garnet pyroxenite layers (e.g., sample AM393). In the Fo-CaTs-Qtz-Di system,  
203 experimental and thermodynamic simulations (Lambart et al., 2012; Mallik and  
204 Dasgupta, 2012) predict that, at increasing melt/rock ratios, the reaction products of  
205 peridotite and eclogite-derived melts originated at  $P \geq 2$  GPa move away from the

206 peridotite field (located close to the Fo apex) and become progressively enriched in En  
207 and CaTs components. This implies that the eclogite-derived melts produce olivine  
208 dissolution and crystallization of variable proportions of pyroxenes and garnet. The rocks  
209 may thus evolve toward olivine-bearing and, eventually, olivine-free garnet websterites,  
210 according to the melt/rock ratio and the silica activity of the reacting melt. The External  
211 Ligurian websterites have chemical compositions consistent with the products of the  
212 melt/rock reaction experiments of Mallik and Dasgupta (2012) (Fig. 2). Such reactions  
213 are expected to create Mg-rich second-stage pyroxenites, which partially preserve  
214 geochemical and isotopic signature of recycled mafic crust and display melting  
215 relationships similar to peridotites (Lambart et al., 2012). Quantitative trace element  
216 modeling of a replacive origin is hampered by subsolidus reequilibration and late-stage  
217 metasomatism affecting most websterite samples. However, the variations of trace  
218 elements not affected by secondary enrichments (e.g., Zr, Y, HREE) may conceivably be  
219 attributed to the compositional variability of eclogite-derived melts involved in the  
220 websterite formation (Figs. 1C, 1D), and to variable proportions of eclogite-derived and  
221 peridotite components. A hybrid origin would be consistent with the Nd-Hf isotope  
222 characteristics of the analyzed websterite (Fig. 3).

## 223 **CONCLUSIONS**

224 Different types of garnet pyroxenites were recognized in the mantle section of the  
225 External Ligurian ophiolites. The following recycling scenario is envisaged for their  
226 origin: (1) a MORB-derived, heterogeneous gabbroic sequence, developed at 1.5–1.0 Ga,  
227 was recycled into the mantle; (2) a rifting-related event triggered partial melting of the  
228 eclogitized gabbros at ca. 220 Ma, thereby producing melt redistribution within the

229 mantle. The crystallization of these melts gave rise to garnet clinopyroxenites, if no  
230 interaction with host peridotites occurred, and MgO-rich websterites preserving a crustal  
231 geochemical signature by reaction with the peridotites. These hybrid rocks resemble the  
232 second-stage pyroxenites envisioned as a key factor for generating OIB with a pyroxenite  
233 fingerprint. The elemental and isotopic variability of the garnet-pyroxenite  
234 heterogeneities is therefore generated by (1) the compositional heterogeneity of the  
235 crustal protoliths; (2) the time of **of [[SU: meaning amount of time elapsed, or age (as in**  
236 **text)?]]** residence within the mantle; and (3) the interaction with the mantle peridotites.  
237 We conclude that the oceanic recycled crustal component proposed for the OIB sources  
238 may include a variety of isotopic signatures, which do not identify a univocal end  
239 member.

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### 335 **FIGURE CAPTIONS**

336 Figure 1. A: Incompatible trace element composition of type C garnet pyroxenites (grt p.)  
337 and websterites normalized to chondrites (Anders and Grevesse, 1989). Shaded fields:  
338 compositions of type A (light gray) and type B (medium gray) garnet clinopyroxenites  
339 (Montanini et al., 2012). B: Chondrite-normalized rare earth element (REE) patterns for  
340 heavy REE–depleted clinopyroxenes (cpx) in equilibrium with garnet. C: Parental liquids

341 of type C garnet clinopyroxenites computed from Al-rich primary cpx (type C melt, cpx)  
342 and whole rock (type C melt, wr) compared with partial melts of eclogite derived from a  
343 plagioclase-olivine cumulate (eclogite melt I). D: Melt in equilibrium with Al-rich  
344 clinopyroxene from websterite sample BA3-W is compared with liquids produced from  
345 melting of gabbro-derived eclogite (eclogite melt II; Sobolev et al., 2000). Parental melts  
346 of type A clinopyroxenites (Montanini et al., 2012) are also reported. Numbers on the  
347 patterns indicate melting degree. **[[SU: should units be given (e.g., %, °C, other)?]]** See  
348 the Data Repository (see footnote 1) for details of trace element modeling.

349

350 Figure 2. Projection of type C garnet clinopyroxenites, websterites (this work), and  
351 External Ligurian (Italy) peridotites (our data) in the pseudoternary system forsterite–Ca-  
352 Tschermak (CaTs)–quartz (Qtz) from diopside (Di; Ol—olivine; En—enstatite).  
353 Projection according to Herzberg (2011). Sources of data for External Ligurian type A–  
354 type B, Ronda, and Beni Bousera garnet pyroxenite, gabbroic rocks from modern oceanic  
355 crust and ophiolites after Montanini et al. (2012, and references therein). Gray arrow is  
356 compositional trend of eclogite melt-peridotite reactions products (Mallik and Dasgupta,  
357 2012; mixed experiments at 3.0 GPa, 1375 °C).

358

359 Figure 3.  $\epsilon_{\text{HF}}-\epsilon_{\text{Nd}}$  diagram for the pyroxenites of this study. Data for type A–type B garnet  
360 (grt) clinopyroxenites and the enclosing peridotites are after Montanini et al. (2012). All  
361 the plotted data are calculated at 220 Ma. HIMU—high  $\mu$  ( $\mu = {}^{238}\text{U}/{}^{204}\text{Pb}$ ). Black lines  
362 show isotopic composition of ancient recycled oceanic crust for variable recycling ages  
363 (2.0–0.5 Ga). 1—oceanic olivine gabbro, 2—average normal mid-oceanic ridge basalt

364 (MORB), 3—oceanic troctolite. See the Data Repository (see footnote 1) for details of  
365 the calculations. MORB—ocean island basalt (OIB) fields are from Salters et al. (2011).

366

367 <sup>1</sup>GSA Data Repository item 2015xxx, [\[\[SU: Need item name\(s\), brief descriptions](#)  
368 [here.\]\]](#), is available online at [www.geosociety.org/pubs/ft2015.htm](http://www.geosociety.org/pubs/ft2015.htm), or on request from  
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