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1           Metasomatism and deformation of block-in-matrix  
2           structures in Syros: the role of inheritance and fluid-rock  
3           interactions along the subduction interface

4  
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13

14 **Keywords**

15 Syros Island; Cycladic Blueschist unit; Mélange; Block-in-matrix structure; Metasomatism;  
16 Subduction interface

17

18

## Abstract

19 *The behavior of fluids at the subduction plate interface and their chemical and rheological*  
20 *impacts remain poorly constrained. Based on detailed fieldwork, petrographic and geochemical*  
21 *analyses and thermobarometry, the present study documents an example of a block-in-matrix*  
22 *'mélange' metasomatized along the subduction interface (Kampos-Lia mélange zone, Cycladic*  
23 *Blueschists, Syros island). We show that this particular mélange zone is a preserved fragment of*  
24 *discontinuous oceanic domain associated with the Pindos basin, which underwent dominant*  
25 *exhumation-related deformation with top to the east shearing. A large part of the 'mélange'*  
26 *structure is inherited from the initial intraoceanic setting and should not be considered as a*  
27 *tectonic 'mélange'. Through exhaustive, meter-scale mapping of the nature of blocks and matrix*  
28 *we show that metasomatism dominantly occurs at the contact between metavolcanic layers and*  
29 *serpentine, with diffusion of Ca from the metavolcanics to the matrix and diffusion of Mg in the*  
30 *opposite direction. Most of the metavolcanic layers and blocks (mafic and carbonate) are only*  
31 *partly digested contrary to the ultramafic matrix which has been largely metasomatized and*  
32 *forms a tremolite-chlorite-talc schist, i.e. a 'hybrid' rock with intermediate chemical composition.*  
33 *Geochemical data demonstrate that metasomatic element mobilization (e.g. Li, B, U, LILE and*  
34 *REE) varies at the scale of the unit, suggesting a complex evolution of fluid composition at the*  
35 *kilometer scale. We suggest that the fluid which intruded the Kampos subunit was derived from*  
36 *the subducting slab and caused an enrichment in Li, B, U, LILE (Rb, K, Na, Ba, Eu), MREE and*  
37 *HREE. Mineralogical and P-T constraints indicate that the dominant part of metasomatism and*  
38 *rock hybridization occurred during exhumation, at ~1 GPa, well after peak burial and*  
39 *detachment from the slab (~2 GPa). Due to the absence of major tectonic mixing and the scarce*

40 *preservation of prograde and peak metasomatic reactions, this metasomatism may be*  
41 *representative of fore-arc hybridization processes at depths of ~35 km. Deeper hybridization*  
42 *processes as hypothesized to produce a potential source for arc magmas are not well preserved*  
43 *in the studied examples. However, by changing the mineralogy of the matrix, metasomatism*  
44 *changes the rheological properties of the mélangé and could thus manipulate the rheology of the*  
45 *subduction interface and influence exhumation processes.*

## 46 **1 Introduction**

47 The interface between the downgoing slab and the mantle wedge exerts a fundamental control  
48 on element recycling, arc volcanism and seismicity in subduction zones (e.g., Stern 2002; Audet  
49 et al. 2009; Wada and Wang 2009; Marschall and Schumacher 2012; Spandler and Pirard 2013).  
50 However, chemical (Bebout, 2007) and physical processes (Agard et al., 2009, 2018) operating  
51 along the interface, and even its exact nature (i.e., lithologies, thickness, strain patterns, ...)  
52 remain ill-constrained. In particular, the net rheological and chemical impact of fluids at the  
53 subduction interface constitutes a major unknown which can only be addressed, at the m- to  
54 km-scale, by studying the fossil subduction rock record (Angiboust et al., 2015; Bebout and  
55 Penniston-Dorland, 2016).

56 Around the world, exhumed high pressure low temperature (HP-LT) units appear as either  
57 continuous slices or block-in-matrix structures, and have been used as a proxy to describe the  
58 plate interface (Guillot et al., 2015; Agard et al., 2018). Block-in-matrix structures are broadly  
59 coined in the literature as 'mélangé' zones (Hsü, 1968; Raymond, 1984; Grigull et al., 2012;

60 Bebout and Penniston-Dorland, 2016; Festa et al., 2019). Those mélange terranes have a wide  
61 range in size (m- to km-scale), structure (block to matrix ratio, deformation features...), variety  
62 of lithologies (e.g. pelitic, ultramafic or gneissic matrix), metamorphic grades (between 0.1 and 3  
63 GPa and 150 to 800°C) and degree of metasomatism. Significant metasomatism is frequently  
64 described and interpreted to progressively homogenize the various protoliths, forming 'hybrid'  
65 rocks (Spandler et al., 2008; Marschall and Schumacher, 2012; Angiboust et al., 2014; Penniston-  
66 Dorland et al., 2014; Bebout and Penniston-Dorland, 2016). Low-density hybrid rocks could form  
67 'diapir' structures from the slab interface into the mantle wedge and directly contribute to arc  
68 volcanism (Marschall and Schumacher, 2012; Nielsen and Marschall, 2017; Bayet et al., 2018;  
69 Codillo et al., 2018).

70 The formation of block-in-matrix structures and metasomatism can occur before burial (e.g.,  
71 hydrothermal circulation at the ridge, sedimentary mixing, discontinuity of the oceanic  
72 lithosphere, core complexes formation), during the prograde or retrograde path (e.g.,  
73 metasomatism through slab derived fluids, deformation and tectonic mixing) and after  
74 exhumation (e.g., weathering, erosion). The behavior of the resulting block-in-matrix structure  
75 (Stöckhert, 2002; Grigull et al., 2012) and the metasomatized rock rheology (Penniston-Dorland  
76 et al., 2018) may have significant implications on the mechanical properties of the slab interface.  
77 However, the significance and scale at which these processes occur, both in time and space, are  
78 still mostly unconstrained.

79 Syros island (Fig. 1), in the Cycladic Archipelago, is renowned for its well-preserved eclogites  
80 and blueschists and allows studying the tectonometamorphic evolution of a subduction  
81 interface. In contact with metasediment-dominated units, km-scale mélange zones comprise

82 meta-igneous or metasedimentary lenses and blocks within an ultramafic serpentinite and/or  
83 chlorite-schist matrix (already described by Dixon, 1968). This ophiolitic unit is thought to belong  
84 to a restricted oceanic domain in the Pindos 'ocean' (Bonneau, 1984; Cooperdock et al., 2018;  
85 Schmid et al., 2020). Reaction zones at the contact between those different lithologies have  
86 been described for decades (Ktenas, 1907; Dixon, 1968; Ridley, 1984; Okrusch and Bröcker,  
87 1990; Seck et al., 1996; Bröcker and Enders, 2001; Marschall et al., 2006). In particular, several  
88 studies have been performed on a m-scale continuous reaction zone (called the Lia beach  
89 transect in this study; Breeding et al. 2004; Miller et al. 2009; Pogge von Strandmann et al. 2015;  
90 Gorman et al. 2019). However, an understanding of the metasomatism at the scale of the  
91 *mélange* unit is still missing (Ague, 2007).

92 This study therefore aims to provide an image of a reacted plate interface '*mélange*',  
93 through the example of Syros island, by studying both chemical and mechanical processes on a  
94 km-scale, trying to answer the following questions:

95 (1) What is the detailed organization (distribution, size and deformation of blocks and layers  
96 inside the matrix) of such a *mélange*, block-in-matrix structure? How does it relate to the  
97 original structure?

98 (2) Under which depth/temperature/deformation conditions did this metasomatism occur ?  
99 When did it happen in the overall subduction history ?

100 (3) What is the nature of the reaction zones and to what extent/scale do they depend on  
101 initial lithology, block size and/or matrix type? At the  $\mu\text{m}$  to m scale, what are the detailed  
102 transformations affecting rocks, minerals, and major and trace elements?

103 (4) Does the observed metasomatism produce chemically hybrid rocks, and to what extent ?  
104 What are the consequences of this mélange on the subduction interface rheology? Is this a  
105 relevant example to estimate arc magma sources ?

106 To approach these aims, we herein present a combined tectonometamorphic and  
107 geochemical approach (rock and mineral major and trace element compositions) with detailed  
108 field mapping between Kampos and Lia beach on the northern tip of the island, together with  
109 complementary investigations in Kini and Galissas (Fig. 1).

## 110 **2 Geological setting**

### 111 **2.1 The Aegean domain**

112 The Aegean domain corresponds to a collapsed segment of the Hellenic belt formed from the  
113 Late Cretaceous to the Early Eocene following subduction and collision of Africa with Eurasia (Le  
114 Pichon and Angelier, 1981; Bonneau and Kienast, 1982; Jolivet et al., 1994; Ring et al., 2010).  
115 During convergence, subducted HP-LT metamorphic units of the Cycladic Blueschist Unit (CBU)  
116 were exhumed beneath a top-to-the east/northeast thrust at the top and a top-to-the south  
117 detachment at the base (Augier et al., 2015; Huet et al., 2009; Trotet et al., 2001). From 35 to 30  
118 Ma, the acceleration of African slab retreat led to the collapse of the Hellenic belt in the back-  
119 arc domain (Le Pichon and Angelier, 1981; Lister et al., 1984; Jolivet et al., 2004, 2013) and the  
120 exhumation of HP-LT units was completed via large-scale detachments, forming metamorphic  
121 core complexes (Lister et al., 1984; Urai et al., 1990; Gautier et al., 1993; Jolivet and Patriat,  
122 1999), in a low-pressure and high-temperature (LP-HT) environment.

123 It has been proposed that the Cycladic Blueschist Unit may represent subducted fragments  
124 of the Pindos oceanic domain (Bonneau and Kienast, 1982; Bonneau, 1984), but the oceanic  
125 nature of this domain is somewhat controversial. Apart from thin ophiolitic slices, the CBU is  
126 mostly associated with continental crust (Keay and Lister, 2002; Zlatkin et al., 2018) and no  
127 ophiolite is found in the Pindos unit, i.e. in the non-metamorphosed equivalent of the CBU in the  
128 mainland (Schmid et al., 2020). In the ophiolitic slices, metagabbros and metaplagiogranites  
129 formed across a restricted time period only (mostly 76 to 81 Ma; Broecker and Enders, 1999;  
130 Tomaschek et al., 2003; Bröcker and Keasling, 2006; Bulle et al., 2010; Bröcker et al., 2014;  
131 Cooperdock et al., 2018); isotopic and trace element compositions of serpentinites from Syros  
132 hint to a hyper-extended margin or refertilized mid-ocean ridge setting. If oceanic at all, the  
133 Pindos basin was probably narrow and characterized by extended and thinned continental crust,  
134 mantle exhumation, and incipient oceanic crust formation.

135 The Cycladic Archipelago (Fig. 1) is located in the center of the Aegean domain and  
136 corresponds to the deepest exhumed portions of the Hellenides-Taurides belt. The CBU reached  
137 peak-pressure conditions at 53-42 Ma (Trotet et al., 2001; Tomaschek et al., 2003; Putlitz et al.,  
138 2005; Lagos et al., 2007; Lister and Forster, 2016; Laurent et al., 2017; Uunk et al., 2018).  
139 Eclogites and blueschists of the CBU are spectacularly preserved on some of the islands. Syros  
140 island, in particular, is world-famous for being the type locality of glaucophane (Hausmann,  
141 1845), and has been the focus of numerous studies in geology, petrology, geochronology and  
142 structural geology.

## 143 **2.2 Tectonic units on Syros island**

144 Most of the surface of Syros exposes the CBU except to the southeast, where the Vari Unit crops  
145 out (Fig. 1; Keiter et al. 2011; Laurent et al. 2017). The Vari Unit lies above and is separated from  
146 the CBU by a large-scale detachment (Trotet et al., 2001; Keiter et al., 2004, 2011; Soukis and  
147 Stockli, 2013). It is made of a greenschist-facies mylonitic unit overlain by orthogneiss intruded  
148 by amphibolite-facies metabasites. No HP imprint was recognized (Soukis and Stockli, 2013;  
149 Laurent et al., 2016).

150 Laurent et al. (2016) subdivided the Syros CBU nappe stacks into three subunits, based on  
151 their predominant lithology and metamorphic facies, bounded by top-to-the-east shear zones,  
152 which are, from bottom to top:

153 1) Posidonia Subunit, composed of gneiss overlain by albitic micaschists with intercalated  
154 boudins of metabasites and thin marble layers. Most of this subunit is overprinted under  
155 greenschist-facies conditions and only a few blueschist- and eclogite-facies parageneses are  
156 observed within the core of metabasite boudins.

157 2) Chroussa Subunit, composed of alternating micaschists, thick marble layers and  
158 metabasites. In this subunit, both eclogites and blueschist- and greenschist-overprinted  
159 eclogites are observed.

160 3) Kampos Subunit, mainly composed of a mélange of metabasites in an ultramafic matrix  
161 with minor metasediments and marbles. Eclogite- and blueschist-facies parageneses are  
162 particularly well preserved and mostly escaped significant retrogression. The Kampos and  
163 Chroussa subunits are bounded by the Kastri zone. The northern part of the island is composed

164 of a subunit structurally above the Kamos subunit, interpreted as a klippe of Chroussa Subunit  
165 and bounded by the Lia shear zone (Laurent et al., 2016).

166 The estimated age of maximum burial lies between 42 to 53 Ma (Tomaschek et al., 2003;  
167 Putlitz et al., 2005; Lagos et al., 2007; Lister and Forster, 2016; Laurent et al., 2017; Kotowski et  
168 al., 2020), whereas the age of syn-kinematic retrogression under greenschist-facies conditions  
169 has been estimated from 25 to 21 Ma (Bröcker et al., 2013) and 43 to 20 Ma (Kotowski et al.,  
170 2020).

171 The Kamos, Chroussa and Posidonia subunits were stacked by thrusting (Laurent et al.,  
172 2016), probably during the prograde evolution and at the pressure peak with a top-to-the S/ SW  
173 sense of shear (Philippon et al., 2011) and associated large-scale open folds (Keiter et al., 2011).  
174 Thrusting accounts for the structurally upper position of the Kamos ophiolitic slice on top of  
175 metasedimentary-dominated subunits (Keiter et al., 2004, 2011). During retrograde evolution  
176 and exhumation, deformation is accommodated along the major contacts or distributed within  
177 the whole Posidonia Subunit with a top-to-the-east sense of shear (Trotet et al., 2001; Laurent  
178 et al., 2016).

179 Metamorphic peak conditions were estimated in the range 450-530°C and in excess of 1.3  
180 GPa up to 2.3 GPa by various authors, depending on the rocks and methods used (Okrusch and  
181 Bröcker 1990; Trotet et al. 2001; Rosenbaum et al. 2002; Keiter et al. 2004; Schumacher et al.  
182 2008; Philippon et al. 2013; Lister and Forster 2016; Laurent et al. 2018; Kotowski et al. 2020).  
183 Laurent et al. (2018) obtained similar peak metamorphic P-T conditions from all three CBU  
184 subunits using independent thermobarometric methods at  $2.2 \pm 0.2$  GPa and  $530 \pm 30$ °C. They  
185 proposed a two-step retrograde P-T path: (1) first, exhumation under HP-LT conditions from 2.2

186 GPa to 1.0 GPa for all units and (2) second, isobaric heating at 1.0-1.2 GPa and 500-570°C for the  
187 two lowermost subunits (Chroussa and Posidonia). This heating is not observed in the upper  
188 Kampos subunit where peak parageneses are better preserved and the exhumation occurred  
189 under HP-LT conditions.

190 This heating phase was also identified on Tinos and Andros islands (Parra et al., 2002; Huet  
191 et al., 2015) and dated between 37 and 30 Ma (Bröcker et al., 1993, 2004; Huet et al., 2015). It  
192 coincides with a change in subduction dynamics around 30 Ma, with an increase in the rate of  
193 slab retreat, inducing major back-arc extension and marking the transition between a cold syn-  
194 orogenic regime in the subduction zone to a warmer post-orogenic regime in the back-arc  
195 domain (Parra et al., 2002; Jolivet and Brun, 2010; Laurent et al., 2018).

### 196 **2.3 Mélange zone and metasomatism**

197 Around the world, mélanges have been described as formations containing either similar or  
198 distinct blocks scattered within a weaker matrix (e.g., Franciscan mélange, Catalina Schists, New  
199 Caledonia, Syros, see Bebout and Penniston-Dorland 2016 for a review). On Syros Island, the  
200 Kampos subunit is characterized by the presence of block-in-matrix structures consisting of  
201 metamorphosed lenses and blocks of metasedimentary and meta-igneous rocks within a  
202 serpentinite and talc-chlorite-tremolite schist matrix. Several of those mélange zones are  
203 observed around the villages of Kampos, Kini, Galissas and near Syros airport. The contacts  
204 between these different lithologies are characterized by reaction zones, also called blackwalls or  
205 rinds (Ridley, 1984; Okrusch and Bröcker, 1990; Seck et al., 1996; Bröcker and Enders, 2001).  
206 Marschall et al. (2006) showed, by modelling fluid compositions, that the high  $\delta^{11}\text{B}$  of

207 tourmalines from reaction zones (from +18 to +28‰) can be explained by the interaction  
208 between slab-derived fluids and the blocks in the exhumation channel. Marschall et al. (2009)  
209 documented high Li concentrations (up to 88  $\mu\text{g/g}$ ) and highly elevated Li/Be ratios (up to 80) in  
210 metasomatic rinds, suggesting specific fluid enrichment in Li. They proposed that B/Be ratios are  
211 dominantly controlled by the presence or absence of tourmaline and Cl/Be ratio variations could  
212 be linked to fluid inclusions.

213 A meter-scale reaction zone between a metavolcanic and serpentinite, hereafter mentioned  
214 as the Lia beach transect, has been studied in detail. Firstly, Breeding et al. (2004) noted a  
215 preferential leaching of Large Ion Lithophile Elements (LILE), U and Pb by fluids in this reaction  
216 zone. Miller et al. (2009) described five different zones: (I) glaucophane + garnet + phengite +  
217 epidote, (II) glaucophane + epidote + chlorite, (III) chlorite + epidote + omphacite  $\pm$  albite (IV)  
218 chlorite  $\pm$  titanite  $\pm$  rutile  $\pm$  apatite and (V) serpentine + chromite. They suggested a diffusion of  
219 elements (such as Mg and Ca) through the reaction zone, and advective leaching of LILE and  
220 enrichment in B and LREE by the fluid. Pogge von Strandmann et al. (2015) documented an  
221 important decrease of the Mg isotopic composition (by  $\sim 1\text{‰}$ ) through the reaction zone and  
222 proposed a duration of the metasomatism of 100 kyr based on diffusion modelling. Finally,  
223 Gorman et al. (2019) noticed a strong fluid mobilization of Os, Ir, Ru and Pt in this transect with  
224 no influence of mechanical mixing and a minor advective enrichment in Re and Pd.

225 Contrasting P-T conditions were estimated for the formation of those reaction zones during  
226 exhumation: at 1.17-1.23 GPa, 500-550°C (Breeding et al., 2004), at 0.62-0.72 GPa, 400-430°C  
227 (Marschall et al., 2006) and at 1.2 GPa, 430°C Miller et al. (2009). This spread of estimated

228 pressures and temperatures could indicate that metasomatism occurred over a significant  
229 period of time with punctually short pulses of fluid influx (Pogge von Strandmann et al., 2015).

### 230 **3 Kampos-Lia mélange zone mapping**

231 This study focused on the Kampos subunit between Lia beach on the west coast of Syros and  
232 Kampos, and along the coast in Kini and Galissas (Fig. 1). The Kampos-Lia mélange zone is  
233 delimited by the Kastri and Lia shear zones (Laurent et al., 2016): the Kastri shear zone is the  
234 basal contact with the Chroussa Subunit, while the Lia shear zones is the roof contact with a  
235 klippe of similar lithology to the Chroussa Subunit. Those contacts are identified in the field  
236 thanks to shearing structures and lithological changes from the metavolcanic and/or ultramafic  
237 matrix of the Kampos subunit, to marbles and/or metapelites.

#### 238 **3.1 Major lithologies**

239 Six main lithologies were recognized in the field:

240 (1) Metavolcanics are mainly composed of variable amounts of glaucophane, omphacite,  
241 garnet, epidote and lawsonite pseudomorphs. They form layers several meters thick. Felsic  
242 layers or dikes composed of jadeite and quartz are also found. More thinly foliated horizons are  
243 interpreted as reflecting a larger tuffitic component.

244 (2) Metagabbros are made of coarse-grained omphacite and clinozoisite with minor  
245 glaucophane replacing omphacite. This lithology is encountered in close association or within  
246 metavolcanic layers.

247 (3) Marbles form a meter-scale discontinuous, boudinaged marble horizon observed under  
248 the northern contact of the mélange zone. It resembles the laminated to massive, medium- to  
249 coarse-grained light gray marble, with frequent impurities, of the Chroussa subunit (visible at  
250 the contact south of the Kampos-Lia mélange zone).

251 (4) Chlorite  $\pm$  tremolite  $\pm$  talc schists matrix associated with blocks. Those blocks are  
252 centimeter to decameter scale and are mostly metabasites, Fe-Ti or Mg-Al-rich, or intermediate  
253 in composition. Blocks are partly rounded and metasomatized. For clarity, we will call 'rind' the  
254 metasomatized part of the blocks.

255 (5) Serpentinities are partly non-foliated, but are isotropic and are blocks themselves with  
256 preserved peridotite micro-textures (pyroxene pseudomorphs and mesh-textured serpentine-  
257 magnetite intergrowths). These serpentinites trend into sheared/foliated serpentinite and into  
258 talc- and chlorite-rich serpentinites where they transition into the chlorite  $\pm$  talc schist.

259 (6) Micaschists are present at the northern contact of the mélange zone, where they are  
260 particularly deformed and show spectacular garnet-rich, centimeter to decameter scale sheath  
261 folds with mica, quartz and variable amounts of glaucophane, epidote and lawsonite  
262 (pseudomorphs).

### 263 **3.2 Overall structure and deformation features**

264 Metavolcanic layers are folded along W-E striking axes and show intense boudinage on the  
265 kilometer scale. These layers usually show a 30 to 80° dip to the N/NE (Fig. 2a). Lineations  
266 dominantly show a W-E orientation with some top to east indications. Well-preserved lawsonite  
267 pseudomorphs are often observed in layers alongside contacts and near the coastline.

268 GPS locations were taken for each matrix outcrop and block, distinguishing those still in  
269 place and those potentially later displaced by gravity sliding and erosion. Size, orientation of the  
270 long axis were measured for each block and their lithologic group and mineralogical composition  
271 of the rinds were determined. The nature of the matrix has been estimated with a spatial  
272 resolution of ~1 meter (Fig. 2a). In total, 499 matrix spots have been identified, with 45.1% of  
273 serpentinite, 4.6% of serpentinite and talc, 0.4% of talc, 1.6% of chlorite and talc, 13.4% of  
274 chlorite and tremolite with major chlorite, 17.6% of chlorite and tremolite with major tremolite  
275 and 17.2% of tremolite. This indicates that approximately half of the matrix comprises hybrid  
276 rocks (products of mixing or modal metasomatism). The serpentinite is never observed at the  
277 contact with blocks or metavolcanics. Consistently, the further the metavolcanic the more the  
278 matrix is made of serpentinite (e.g., near the northern contact; Fig. 2a).

279 Altogether 328 blocks have been identified (Figs. 2bcd), comprising 49.7% of Fe-Ti blocks,  
280 30.2% of Mg-Al blocks and 15.5% of blocks intermediate in composition, 4% of metavolcanic  
281 blocks and 0.6% of metasedimentary blocks. The majority of blocks are Fe-Ti or have a Fe-Ti  
282 component. Block sizes range from centimeter to decameter sizes, most ranging between 1-2  
283 meters with a length/width ratio usually between 1 and 2 with an E/NE-W/SW elongation and  
284 orientation (Fig. 2e), consistent with an overall EW stretching. The amount of block smaller than  
285 a meter may be underestimated because of the difficulty of observing them in the field. Several  
286 intermediate and Fe-Ti blocks (n=25) show pillow-lavas like structures and are mostly located in  
287 the northern part of the zone, as for the Mg-Al blocks (Fig. 2a).

288 Using m- to hm-scale observations, complemented by panorama and satellite pictures for  
289 careful extrapolation, a structural map has been realized (Fig. 2e). Pervasive top-to-the-east

290 shear sense is observed in the unit (Fig. 3), whether at the mineral scale or kilometer scale. It is  
291 outlined by the boudinage of layers of metavolcanics and marbles. A good example is the sheath  
292 folds affecting garnet-rich layers (Fig. 2f) observable along the Lia shear zone near the coast.

293 Detailed mapping reveals that most blocks lie in the variably metasomatized matrix.  
294 Metavolcanics can be mapped continuously and wrap around the blocks and matrix located in  
295 the core of a broad anticline. Therefore, those blocks form a tectonic horizon structurally  
296 beneath the metavolcanic layers. As a result, simplified cross-sections can be proposed based on  
297 a simple, first-order lithostratigraphic succession (Fig. 4). Our structural map based on m-scale  
298 observations broadly confirms the previous hm-scale mapping of Keiter et al. (2011; Fig. 1). It  
299 differs on the presence of a level of metasedimentary rocks described by Keiter et al. (2011),  
300 that we consider as a metavolcanic tuffitic component with a thinner foliation spacing and a  
301 greater abundance of phyllosilicates.

302 Our maps make it possible to estimate the proportion of the different lithologies in the  
303 Kampos-Lia mélange zone: 30.4% of matrix and blocks, 54.4% of metavolcanics, 1.0% of marble  
304 and 14.5% of quaternary ground cover. The cumulative area of the blocks corresponds to 2677  
305 m<sup>2</sup>, i.e. 2.5% of the matrix.

### 306 **3.3 Block and matrix diversity**

307 Samples were collected across different reaction zones to ensure representativeness (Table 1;  
308 Fig. 1). Mineral abbreviations are after Whitney and Evans (2010) except for tremolite (Trem).  
309 Two transects have been more thoroughly sampled:

310 (1) Transect L (Fig. 5), sampled near Lia beach, comprises 8 samples: n290, n57, n10 and n1  
311 in a metavolcanic layer and p5, p125, p135 and p165 in the matrix. Numbers refer to the  
312 distance to the block/matrix contact in centimeters: n (for negative) refers to the distance into  
313 the block and p (for positive) for the distance away from the block (into the matrix).

314 (2) Transect G, sampled near Galissas, contains 7 samples: n50, n30, n8, n8b and n2 in a Fe-  
315 Ti block and p1 and p30 for the matrix.

316 Three main meta-igneous or metavolcanoclastic lithologies interact with the ultramafic  
317 matrix: Fe-Ti blocks, Mg-Al blocks and layers of metavolcanics. The different types of rinds  
318 developing at the contact with the matrix are displayed in figures 6 and 7. These rinds, identified  
319 by the disappearance and/or appearance of phases, replace the eclogitic or blueschist  
320 paragenesis. This suggests that they are mostly near peak and/or retrograde but the possibility  
321 remains that they witnessed earlier transformations (e.g. during peak burial). As shown by  
322 figures 6 and 7, the nature of the rinds largely depends on the nature of the block/layer (i.e.  
323 which lithologies were juxtaposed).

324 — Variations in the composition of metavolcanics are reflected, for example, by the  
325 presence or absence of quartz veins and lawsonite pseudomorphs (Fig. 6a). A decametric,  
326 weathered chloritic rind is observed on a few metavolcanics blocks (Fig. 6b). However, in almost  
327 all cases, metavolcanics are characterized by a decimetric rind of  $\text{Gln}+\text{Ep}+\text{Ph} \pm \text{Cpx}, \pm \text{Chl}, \pm \text{Ab}$   
328 (Fig. 6c). Whenever present, omphacite shows W-E stretching (Fig. 6d). Lawsonite  
329 pseudomorphs are still visible in the rind in places (Fig. 6e). Given that metavolcanics  
330 predominate in the block/layer lithology, this is the most common type of rind in the mélange.  
331 Finally, the specific well-studied outcrop north of Lia beach shows a meter-scale rind with a

332 Cpx+Chl+Ep association and then dominantly chlorite (Fig. 6f; similar to the Lia beach transect;  
333 Miller et al. 2009). Figure 6f also shows that the reaction limits are oblique with respect to the  
334 foliation.

335 — The Fe-Ti blocks show particularly large variations in block and rind mineral assemblages  
336 (Figs. 6,7). Fe-Ti blocks always contain garnet and rutile associated with glaucophane and/or  
337 omphacite (Figs. 6gh). A less widespread type of rind shows the presence of decimetric  
338 clinopyroxene growing perpendicular to the contact (Fig. 6i). The presence of fresh garnet grains  
339 in some Fe-Ti block rinds (Fig. 6j) seems to indicate early metasomatism at peak conditions or  
340 during early retrogression. Such an early metasomatism, if any, has been either very local or  
341 poorly preserved.

342 — The Mg-Al blocks mostly comprise clinopyroxene and/or glaucophane (Fig. 6k). Their  
343 rinds, in addition to clinopyroxene and/or glaucophane, are generally composed of chlorite,  
344 epidote, phengite, tourmaline and with rare occurrences of decimetric clinopyroxene similar to  
345 the one observed in Fe-Ti rinds.

346 Blocks intermediate in composition (Fig. 6l) often show pillow-like structures or microlithic  
347 dyke cross-cutting blocks and display complex rinds with assemblages similar to those of the Fe-  
348 Ti blocks.

## 349 **4 Analytical methods**

### 350 **4.1 Electron probe microanalysis (EPMA) and Scanning electron microscope** 351 **(SEM) analysis**

352 EPMA and SEM were carried out at CAMPARIS (UPMC-IPGP, Paris, France) using a CAMECA SX-  
353 100 and a CAMECA SX-FIVE microprobes and the data reducing method of Pouchou and Pichoir  
354 (1991). Analytical conditions were 15 kV accelerating voltage and 10 nA specimen current for  
355 spot analyses and 15 kV accelerating voltage and 10 nA specimen current with a dwell time of 50  
356 ms and a step size between 3 and 5  $\mu\text{m}$ .  $\text{Fe}_2\text{O}_3$ ,  $\text{MnTiO}_3$ , diopside,  $\text{Cr}_2\text{O}_3$ , orthoclase, anorthite  
357 and albite were used as standards.

358 Representative mineral compositions are provided in Table 2. The X-ray and backscattered  
359 electron compositional maps were processed using the program XMAPTOOLS 2.2.1 (Lanari et al.,  
360 2014).

### 361 **4.2 Bulk-rock analysis**

362 Eighteen mafic samples were sent for bulk-rock chemical analyses to the SARM (Nancy). Sample  
363 preparation consisted of fusion with  $\text{LiBO}_2$  and dissolution in  $\text{HNO}_3$ . Major elements ( $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  
364  $\text{Fe}_2\text{O}_3$ ,  $\text{MnO}$ ,  $\text{MgO}$ ,  $\text{CaO}$ ,  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$ ,  $\text{TiO}_2$  and  $\text{P}_2\text{O}_5$  with a detection limit less than or equal to  
365 0.1%) and Sc were analyzed by inductively coupled plasma-optical emission spectroscopy (ICP-  
366 OES iCap6500). Trace element concentrations were determined by inductively coupled plasma  
367 mass spectrometry (ICPMS iCapQ). Boron was measured after fusion of the sample with sodium  
368 carbonate and measurement by UV-visible spectrophotometry of boron-carmin complex 40 in

369 concentrated sulfuric acid either directly or after concentration and separation on resin  
370 (detection limit of 0.5 µg/g for Li and 2 µg/g for B). Finally, Li was measured after acid attack of  
371 the sample with HF-HClO<sub>4</sub> and measurement with flame atomic absorption spectrometer.  
372 Whole rock compositions are given in Table 3.

373

### 374 **4.3 LA-ICP-MS analysis**

375 Extensive in-situ mineral analyses were performed using a Thermo Scientific Element 2 sector  
376 field laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) at the Frankfurt  
377 Isotope & Element Research Center (FIERCE) of Goethe Universität Frankfurt. This ICP-MS is  
378 coupled to a Resolution S-155 (Resonetics) 193 nm ArF Excimer laser equipped with a two-  
379 volume ablation cell (Laurin Technic, Australia). Samples were ablated in a helium atmosphere  
380 and mixed in the ablation funnel with argon and nitrogen to minimize oxide production.  
381 Analyses were carried out using an 8Hz laser pulse rate, 40- or 60-µm diameter round spots .

382 Data were acquired in five independent analytical sessions for a total of 1687 analyses  
383 (counting GSD-1G, NIST-SRM612, BCR-2G and BHVO-2G reference material analyses). Each  
384 analysis consists of 21s background acquisition followed by 26 or 23.5s of sample ablation.  
385 During data acquisition, the signals of trace elements were detected by peak jumping in pulse  
386 counting mode, analogue mode or Faraday mode depending of the amount of signal with a total  
387 of 20 mass scans. Prior to analysis, each spot was pre-ablated for 6s to remove surface  
388 contamination.

389 GSD-1G reference glass was used for external sensitivity calibration and mean  
390 concentrations of SiO<sub>2</sub> determined by EPMA were used as internal standards. Data reduction

391 was done using the LADR program (Norris A, 2018), with limits of detection calculated for each  
392 element in every analysis.

393 After treatment, the data show a strong dispersion. This may be due to some important drift  
394 of the noise measurement during a session (probably due to vacuum variations), sometimes  
395 presence of inclusion in the standards and/or simplistic internal calibration. We herein only  
396 consider data whose sum of cations does not vary by more than 5% compared to the  
397 microprobe data. Summarized data are provided in supplementary material.

## 398 **5 Petrography**

399 We describe below mineral occurrences in the various transects, from block or layer to matrix.

400 Transect L (Lia; Figs. 5,8a) is composed of a metavolcanic layer (block: L\_n290, L\_n57; rind:  
401 L\_n10 and L\_n1) and matrix samples (L\_p5, L\_p125, L\_p135 and L\_p165). From L\_n290 to L\_n1,  
402 glaucophane represents more than half of the mineral mode. Garnet and epidote disappear and  
403 albite, phengite and chlorite progressively appear. Garnet grains mainly have quartz, epidote  
404 and glaucophane inclusions and are replaced by phengite in L\_n10 and L\_n1. L\_n290 has the  
405 particularity of being cross-cut by a quartz vein. Variable amounts of tremolite, chlorite and talc  
406 characterize samples L\_p5 to L\_p135, with complete absence of tremolite in L\_p125. Mineral  
407 grains are generally oriented parallel to the contact in L\_n10, L\_n1, L\_p125 and L\_p135, whereas  
408 L\_p5 is chaotically deformed. Chlorite in L\_p135 forms large-scale crystals with lenticular shapes  
409 in a tremolite-dominated matrix. In contrast L\_p165 is mostly made of serpentine with a few  
410 percent of chlorite and talc.

411 Transect G (Galissas; Fig. 8b) is composed of a Fe-Ti rich block (block: G\_n50, G\_n30; rind:  
412 G\_n8, G\_n8b, G\_n2) and matrix samples (G\_p1 and G\_p30). The amounts of omphacite and  
413 epidote vary between G\_n50 and G\_n8, and omphacite is no longer present in G\_n2. The  
414 amount of chlorite varies greatly, from 13% in G\_p1 to 90% in G\_n2. Garnet is increasingly  
415 altered from G\_n50 to G\_n8b and absent in G\_n2. Two single rounded crystals of chloritoid are  
416 observed in G\_n50. Rutile occurs in G\_n50 to G\_n8b, and rimmed by titanite in G\_n2. In samples  
417 G\_n50 to G\_n8b clinopyroxene is present both as large centimetric altered porphyroblasts and  
418 as micrometric prismatic crystals forming localized aggregates. Sample G\_n8b is crosscut by a  
419 chlorite-tremolite vein perpendicular to the vein walls, similar to the matrix assemblage. Sample  
420 G\_n2 contains >80% chlorite with rutile, titanite and apatite. Sample G\_p1 contains tremolite  
421 and chlorite, while G\_p30 contain chlorite oriented parallel to the contact.

422 Transect K (18, Kini) is composed of a Fe-Ti rich block and one matrix sample (K\_18c).  
423 Samples K\_18a to d contain chlorite, clinopyroxene and garnet, together with minor rutile and  
424 apatite. Garnet is partly altered and replaced by chlorite and hosts clinopyroxene inclusions in  
425 both K\_18b and e. Sample K\_18c matrix assemblage comprises tremolite porphyroblasts and  
426 chlorite.

427 Transect 9 (Lia) is composed of a Mg-Al-rich block (9a,b,c and d). Samples 9a, b and c are  
428 mostly comprised of clinopyroxene, chlorite and epidote with variable amounts of  
429 microcrystalline glaucophane and phengite, and minor amounts of albite and apatite. Sample 9d  
430 is composed of chlorite and microcrystalline glaucophane.

431 Transect 19 (Lia) is composed of a Fe-Ti rich block (19a,b and c). Samples 19a and b are  
432 mainly composed of euhedral garnet and glaucophane with quartz fringes around garnet and

433 minor phengite, paragonite, rutile and titanite. Sample 19c is dominantly made of  
434 microcrystalline glaucophane and minor chlorite.

435 Other samples were collected to cover the whole range of existing lithologies (Fig. 7) and/or  
436 to generalize and strengthen our observations.

437 Sample 22 is a metavolcanic rind with elongated aggregates of clinopyroxene surrounded by  
438 glaucophane and phengite, together with minor albite. Sample 24b is a metavolcanic rind mostly  
439 composed of centimetric pseudomorph of lawsonite with epidote, albite, chlorite and tremolite  
440 associated with a glaucophane-dominated matrix. Sample 17 is from a metamorphosed acidic  
441 dyke or vein with >90% quartz and some paragonite, albite and chlorite.

442 Sample 582 is a Fe-Ti block with cogenetic garnet, glaucophane, omphacite and minor rutile.  
443 Sample 4 is a Fe-Ti-rich block with alternating clinopyroxene- and glaucophane-rich layers,  
444 seemingly cogenetic garnet, minor rutile and phengite. Sample 27 is a Fe-Ti-rich rind with  
445 glaucophane, epidote, clinopyroxene, phengite and minor albite, chlorite, altered garnet, rutile  
446 and titanite.

447 Sample 21 is a Mg-Al-rich rind mostly composed of clinopyroxene, with subordinate amounts  
448 of glaucophane, epidote, phengite and some tourmaline phenocrysts with clinopyroxene  
449 inclusions. Sample 23 is a matrix sample composed mostly of chlorite and tremolite with major  
450 rutile and minor apatite and titanite. Sample 581 is a serpentinite block with pervasive  
451 replacement of serpentine by chlorite and minor talc.

## 452 **6 Bulk-rock geochemistry**

453 The bulk chemistry including major (Fig. 8) and minor (Fig. 9) elements allow recognizing four  
454 zones on transects L and G:

455 (1) the block interior with little or no metasomatism (L transect: n290, n57; G transect: n50,  
456 n30, n8), marked by low concentrations of Cr, Ni and Co, relatively low Mg, low enrichment in  
457 LREE.

458 (2) an inner rind (L: n10, n1; G: n2) characterized by marked changes in major element  
459 contents, and in particular a depletion in Ca, with strong variation of the mobile elements.

460 (3) a metasomatized matrix (L: p5, p125, p135; G: p1, p30) with high concentrations of Mg,  
461 Cr, Ni, Co and an enrichment in Ca. Samples are depleted in REE (with also a negative Eu  
462 anomaly), some particularly in LREE (L\_p135 and L\_p1) and one in HREE (L\_p125). The strong  
463 variation of trace element contents is reflected by the strong variation of mineral assemblages.

464 (4) the serpentinite matrix (L\_p165) with very high Mg, Cr, Ni and Co and very little Ca.  
465 Values obtained for L\_p165 are similar to those obtained by Cooperdock et al. (2018), i.e. with  
466 no LREE addition, overall depletion of REE concentrations and lack of major Eu anomaly.

467 Figure 9a shows that the composition of the Fe-Ti block of transect G is similar to N-MORB.  
468 In contrast, the metavolcanic samples of transect L are intermediate between MORB and  
469 sediments and show a large tuffitic and/or sedimentary component (Fig. 9a).

470 Figure 9b compares our two main transects with the Lia beach transect of Miller et al.  
471 (2009). In both transects L and G, the evolution from block to rind is characterized by a decrease  
472 of Ca associated with an increase of Mg. This trend compares with the overall trend of Miller et  
473 al. (2009), yet no increase in Ca content, as reported in zone 3 by Miller et al., (2009), is

474 observed in our transects. Their zone 4 is similar to G\_n2 in transect G, with a rind dominated by  
475 chlorite and associated with titanite and apatite, yet they do not observe a tremolite-rich  
476 metasomatized matrix with high Ca content. Miller et al. (2009) inferred that the original block-  
477 matrix contact lied between their zones 4 and 5 (serpentinite). Some of our hybrid samples  
478 show a higher Ca content than others described in Santa Catalina Island or New Caledonia (King  
479 et al., 2006; Spandler et al., 2008).

480 The minor and major compositions of the metasomatized matrix are compatible with a  
481 serpentinite protolith with high concentration of Mg, Cr and Ni (Fig. 8), negative Eu anomaly  
482 (Fig. 9c), low REE concentration and similar concentration of relatively immobile elements such  
483 as Al, Zr, Ti, V and Y (Figs. 8, 9d-h). The sharp drop of those element concentrations at the rind-  
484 matrix contact suggests that the initial contact was located at the contact between the rind and  
485 the matrix. No linear correlation is observed, however, between these relatively immobile  
486 elements (Figs. 9d-f).

487 Figure 9c shows that the block and rind samples of transect L have similar Eu anomalies  
488 (close to 1), while the block and rind samples of transect G show contrasting Eu anomalies (~1.3  
489 and 0.7-0.9, respectively). Figure 9g and h show important variations (up to hundred times  
490 more) in the concentration in REE and mobile elements such as B, Li and some LILE (Cs, Rb, K,  
491 Na, Pb, Ba, Sr and Eu).

492 Ratios of normalized element concentrations of metasomatized samples with respect to  
493 protolith concentrations are shown in figure 10a for transect L. Inferring the mobility of  
494 elements is not straightforward, especially when comparing lithologies with contrasting  
495 chemistries, i.e. mafic rocks with a variable sedimentary component with (potentially

496 metasomatized) ultramafic rocks. All measured elements may show some mobility with  
497 metasomatism. Since we are interested in relative differences between the various transects,  
498 the strategy followed here is to use only one relatively immobile element for normalization to  
499 compare element gain/loss across all the transects.

500 In order to assess the extent of relative 'immobility', ratios of element concentrations in the  
501 unmetasomatized and metasomatized rock were estimated. Values close to one, in all studied  
502 cases, were mostly obtained for Al and all element concentrations were therefore normalized to  
503  $\text{Al}_2\text{O}_3$ . Samples used to compare metasomatized and unmetasomatized rocks were collected  
504 close to one another to limit the influence of variations in initial composition. The spread of  
505 initial variations is estimated in figure 8 based on our samples and published data (Pogge von  
506 Strandmann et al., 2015; Cooperdock et al., 2018). Another advantage of  $\text{Al}_2\text{O}_3$  is that this major  
507 element can be analyzed to high accuracy and precision and is evenly distributed as a  
508 constituent of the rock-forming minerals, contrary to minor elements which can be affected by  
509 the presence of one extra grain of zircon, rutile or monazite/xenotime.

510 In detail, the concentration in the metasomatized rock is multiplied by the ratio of the  $\text{Al}_2\text{O}_3$   
511 concentration in the unmetasomatized rock and the  $\text{Al}_2\text{O}_3$  concentration in the metasomatized  
512 rock. Comparing ratios on either side of the contact ( $R_b = n1/n57$  for the block;  $R_m = p5/p165$  for  
513 the matrix; Fig. 10a) enables assessing which elements are exchanged between the block and  
514 the matrix (i.e.,  $R_b > 1$  and  $R_m < 1$  for elements lost from the metasomatized matrix and gained by  
515 the rind; or vice-versa:  $R_b < 1$  and  $R_m > 1$ ) and which ones are added ( $R_b$  and  $R_m > 1$  for higher  
516 content alongside the contact than in both protoliths) or removed during fluid infiltration ( $R_b$   
517 and  $R_m < 1$  for lower content alongside the contact than in both protoliths). For the sake of

518 comparison, matrix gains and losses from transect L are evaluated using two different matrix  
519 ratios ( $R_m=p5/p165$  or  $R_m=p125/p165$ ; Figs. 10b,c), while block gains and losses are compared  
520 between transects L and G ( $R_b=n1/n57$  or  $R_b=n2/n50$ ; Figs. 10d,e).

521 The elements most affected by exchange along the contact are Ca and Mg. We see an  
522 exchange of LREE, some LILE (Cs, Pb, Sr) and marginally some other elements such as Co and Ni  
523 (Fig. 10a). The rind and metasomatized matrix are enriched in Li, B, U, some LILE (Rb, K, Na, Ba,  
524 Eu), MREE and HREE. External loss, i.e. to an infiltrating fluid, characterizes Ga, Ta, Zn, Mn, P,  
525 and marginally Cr from both the rind and the metasomatized matrix. The ratio for the block in  
526 transect G (Fig. 10e) shows that, in contrast to the metavolcanic transect L, the rind is enriched  
527 in Cr, Ta and Mn and depleted in Li, B, U, most LILE (Cs, Rb, K, Na, Pb, Ba, Sr) and HREE.

## 528 **7 Mineral chemistry**

### 529 **7.1 Chlorite**

530 Chlorite shows a marked increase of  $X_{Mg}$  ( $Mg/(Mg+Fe_{tot})$ ) from block to matrix (from 0.39 to  
531 0.88; Fig. 11a), associated with a decrease in Al content in the matrix (2Al from 2.40 to 1.46).  
532 This variation in Mg content is mainly explained by the Fe-Mg substitution in all samples, and  
533 the Tschermak substitution in the chlorite of the matrix and that of some of the rinds (19c and  
534 G\_n2), where chlorite is commonly hypersilicic (Si pfu from 2.87 to 3.36). Chlorite  $X_{Mg}$  in blocks  
535 and rinds varies as a function of whole-rock compositions: 0.42-0.62 for transect L, ~0.9 for  
536 metavolcanics (e.g., 24b), 0.39-0.80 for Fe-Ti blocks and 0.6-0.8 for Mg-Al blocks.

## 537 **7.2 Amphibole**

538 Amphibole shows a wide range of compositions with XMg from 0.40 to 0.93 and XNa  
539 (Na/(Na+Ca)) from 0.02 to 0.99 (Fig. 11b). Two groups can be differentiated: (1) Na-rich  
540 amphibole in blocks and rind (XNa from 0.78 to 0.99) with glaucophane and a few  
541 magnesioriebeckite compositions and (2) Ca-rich amphibole in the matrix (e.g., G\_n8b vein or  
542 24b; XNa= 0.02-0.35) with tremolite, actinolite and a few winchite compositions (Leake et al.,  
543 1997). Sample 24b shows two types of amphibole with winchite compositions and intermediate  
544 XNa values (0.52 and 0.67). This sample shows the entire range of amphibole compositions from  
545 Na-rich to Ca-rich (XNa from 0.05 to 0.95) with a high XMg (from 0.82 to 0.90). Ca-rich  
546 amphibole has a higher XMg (from 0.75 to 0.93) compared to Na-rich amphibole (from 0.38 to  
547 0.78, or 0.90 for 24b). In the matrix, amphibole shows a zoning in XMg content (e.g., p135; Fig.  
548 11b, inset), with XMg ~0.83 in the core to ~0.93 in the rim. The XMg of amphibole in blocks and  
549 rinds also depends on whole-rock compositions, and is very close to that of chlorite: from 0.42  
550 to 0.70 (transect L) or 0.90 (24b) for metavolcanics; from 0.38 to 0.68 for Fe-Ti blocks and from  
551 0.61 to 0.78 for Mg-Al blocks.

552

## 553 **7.3 Clinopyroxene**

554 The composition of clinopyroxene (Fig. 12a) shows a wide range of composition  $Jd_{09-60}$   
555  $Ac_{m00-48} Q_{21-76}$  (Q=diopside + hedenbergite + clinoenstatite + clinoferrosilite) and XMg from 0.31  
556 to 0.80.

557 In transect G, measurements in altered clinopyroxene blasts (Jd<sub>12-46</sub> Ac<sub>m12-43</sub> Q<sub>27-76</sub>; XMg  
558 from 0.32 to 0.66) and the small metasomatic crystals (Jd<sub>09-53</sub> Ac<sub>m15-48</sub> Q<sub>21-63</sub>; XMg from 0.31 to  
559 0.68) have similar values. An increase in Ca and Mg content is observed from block (Jd<sub>20-53</sub>  
560 Ac<sub>m17-48</sub> Q<sub>21-43</sub>; XMg from 0.31 to 0.58) to rind (Jd<sub>09-49</sub> Ac<sub>m12-40</sub> Q<sub>27-76</sub>; XMg from 0.44 to 0.68).

561 The same Ca and Mg increase is seen in transect 9, from 9a (Jd<sub>30-44</sub> Ac<sub>m10-22</sub> Q<sub>38-54</sub>; XMg  
562 from 0.53 to 0.72) to 9c (Jd<sub>23-44</sub> Ac<sub>m06-16</sub> Q<sub>44-69</sub>; XMg from 0.65 to 0.80), but not in transect K  
563 (Jd<sub>23-60</sub> Ac<sub>m00-21</sub> Q<sub>36-74</sub>; XMg from 0.51 to 0.79; comprising 18a: Jd<sub>32-49</sub> Ac<sub>m01-19</sub> Q<sub>38-56</sub>; XMg from  
564 0.51 to 0.77). Peak clinopyroxene shows a narrower range of values compared to metasomatic  
565 clinopyroxene (e.g. 4 et 582: Jd<sub>32-48</sub> Ac<sub>m06-23</sub> Q<sub>40-52</sub>; XMg from 0.48 to 0.66).

#### 566 **7.4 Garnet**

567 Zoned garnet is observed in transects G, L, 19 and samples 27 and 4 (Figs. 12b,c), with  
568 spessartine and pyrope content decreasing and increasing, respectively, from core (Alm<sub>51</sub> Prp<sub>3</sub>  
569 Sps<sub>18</sub> Grs<sub>32</sub>) to rim (Alm<sub>71</sub> Prp<sub>13</sub> Sps<sub>0</sub> Grs<sub>20</sub>), and XMg increasing from core (XMg=0.04) to rim  
570 (XMg=0.20).

571 Transect K shows two garnet populations: (1) In samples K\_18a and d, one with similar  
572 composition to other samples, from core (Alm<sub>49</sub> Prp<sub>5</sub> Sps<sub>13</sub> Grs<sub>31</sub>; XMg=0.09) to rim (Alm<sub>64</sub> Prp<sub>15</sub>  
573 Sps<sub>0</sub> Grs<sub>24</sub>; XMg=0.21); (2) In samples K\_18b and e, a higher spessartine and grossular content  
574 from core (Alm<sub>35</sub> Prp<sub>3</sub> Sps<sub>29</sub> Grs<sub>39</sub>; XMg=0.09) to rim (Alm<sub>60</sub> Prp<sub>14</sub> Sps<sub>1</sub> Grs<sub>26</sub>; XMg=0.21).

575 The second population of garnet shows inclusions of metasomatic clinopyroxene and is  
576 therefore interpreted as metasomatic garnet.

## 577 **7.5 White mica**

578 Phengitic compositions are observed in transect 9, with an increase in Mg content from block  
579 (Tri<sub>4</sub> Prl<sub>1</sub> Ms<sub>50</sub> Cel<sub>41</sub> Pg<sub>4</sub>; XMg=0.68, Si pfu=3.41) to rind (Tri<sub>4</sub> Prl<sub>1</sub> Ms<sub>47</sub> Cel<sub>41</sub> Pg<sub>7</sub>; XMg=0.74, Si  
580 pfu=3.42). A somewhat different trend is observed in transect L from block (block: Tri<sub>8</sub> Prl<sub>14</sub> Ms<sub>45</sub>  
581 Cel<sub>28</sub> Pg<sub>4</sub>, XMg=0.74, Si pfu=3.42) to rind (rind: Tri<sub>8</sub> Prl<sub>12</sub> Ms<sub>45</sub> Cel<sub>32</sub> Pg<sub>4</sub>; XMg=0.66, Si pfu= 3.44).  
582 Sample 19a shows a distinct population of mica (Tri<sub>3</sub> Prl<sub>4</sub> Ms<sub>50</sub> Cel<sub>36</sub> Pg<sub>8</sub>, XMg=0.57, Si pfu=3.38)

583 Other samples have a wide range of mica compositions with dominant muscovite  
584 component: #4 (Tri<sub>3</sub> Prl<sub>0</sub> Ms<sub>49</sub> Cel<sub>42</sub> Pg<sub>6</sub>, XMg=0.62, Si pfu=3.41), #21 (Tri<sub>5</sub> Prl<sub>1</sub> Ms<sub>53</sub> Cel<sub>31</sub> Pg<sub>9</sub>,  
585 XMg=0.69, Si pfu= 3.31), #22 (Tri<sub>3</sub> Prl<sub>1</sub> Ms<sub>51</sub> Cel<sub>41</sub> Pg<sub>5</sub>, XMg=0.7, Si pfu=3.40), #27 (Tri<sub>3</sub> Prl<sub>1</sub> Ms<sub>49</sub>  
586 Cel<sub>38</sub> Pg<sub>10</sub>, XMg=0.68, Si pfu=3.37) and #n50 (Tri<sub>7</sub> Prl<sub>1</sub> Ms<sub>59</sub> Cel<sub>25</sub> Pg<sub>8</sub>, XMg=0.53, Si pfu= 3.25).

587 Paragonite is observed in both samples 17 (Tri<sub>1</sub> Prl<sub>2</sub> Ms<sub>4</sub> Cel<sub>0</sub> Pg<sub>93</sub>, XMg=0.27, Si pfu=3.01) and  
588 19a (Tri<sub>2</sub> Prl<sub>3</sub> Ms<sub>4</sub> Cel<sub>0</sub> Pg<sub>91</sub>, XMg=0.26, Si pfu=3.02).

589

## 590 **7.6 Epidote**

591 The composition of epidote varies in each sample, but the range of compositions does not  
592 significantly change between the different samples for a given transect. The range of  
593 compositions is as follows: Transect L (from 0.55 to 0.76 Fe<sup>3+</sup> p.f.u.), transect G (from 0.63 to  
594 0.86 Fe<sup>3+</sup> p.f.u.), transect K (from 0.41 to 0.88 Fe<sup>3+</sup> p.f.u.), 19b (from 0.41 to 0.76 Fe<sup>3+</sup> p.f.u.), 17

595 (from 0.39 to 0.93 Fe<sup>3+</sup> p.f.u.), 21 (from 0.48 to 0.68 Fe<sup>3+</sup> p.f.u.), 24b (from 0.08 to 0.62 Fe<sup>3+</sup>  
596 p.f.u.), 4 (from 0.54 to 0.69 Fe<sup>3+</sup> p.f.u.) and 9b (from 0.64 to 0.82 Fe<sup>3+</sup> p.f.u.).

597

## 598 **7.7 Talc**

599 In transect L, the Mg content of talc decreases closer to the block contact, from L\_p165 (XMg  
600 from 0.94 to 0.96), L\_p135 (XMg from 0.94 to 0.95) to p125 (XMg=0.4). Samples G\_p30 (XMg  
601 from 0.93 to 0.96) and 581 (XMg from 0.94 to 0.96) show a similar range of Mg content.

## 602 **7.8 Serpentine**

603 The serpentine is chemically homogenous in both L\_p165 (XMg from 0.81 to 0.84) and 581 (XMg  
604 from 0.87 to 0.88), with a slightly higher Mg content in 581.

## 605 **7.9 Others**

606 Two rounded chloritoid blasts are observed in sample G\_n50 in transect G, with oscillatory Mg  
607 zoning ((Fe<sub>1.47-1.61</sub> Mg<sub>0.45-0.66</sub> Mn<sub>0-0.02</sub>) Al<sub>3.84-3.91</sub> Si<sub>2.01-2.08</sub> O<sub>10</sub> (OH)<sub>4</sub>).

608 Sample 21 contains Na-Mg rich dravitic tourmaline (from 0.88 to 0.94 Na p.f.u.; from 0.02 to  
609 0.06 Ca p.f.u and XMg from 0.74 to 0.79).

# 610 **8 Trace element in-situ geochemistry**

## 611 **8.1 Chlorite**

612 Chlorite shows fairly flat REE spectra, except for a few analyses likely contaminated by inclusions  
613 (mostly in transect G; Figs. 13ab), and a marked decrease in REE content from block to matrix. In

614 transects L and G, chlorite has a higher Ba content near the contact. In comparison, Cs and Rb  
615 contents increase in the rind of transect L (n1) but decrease at the contact of transect G (n2 and  
616 p1). Opposite trends are observed for Pb and Sr contents, which decrease from block to matrix  
617 in transect L and increase in transect G. The Li content decreases from block to matrix in  
618 transects L, G and K, except near the contact in transect L where L\_p5 is slightly enriched  
619 compared to L\_n1 (Fig. 14a). The Li content of chlorite in transects L and K is overall higher than  
620 in transect G. The Li content of chlorite in the matrix, far away from the contact, is generally very  
621 low (<5µg/g). In all three transects (with the exception of a few values for L\_p135 in transect L),  
622 chlorite composition shows an increase in Ni and Cr from block to matrix, with a strong contrast  
623 at the contact (Fig. 14b). The Co content increases from block to matrix in transect L and G but  
624 decreases in transect K (Fig. 14c).

## 625 **8.2 Amphibole**

626 Na-amphibole (Fig. 13c) shows a wide range of REE spectra, from flat to enriched in LREE,  
627 and no significant nor systematic Eu anomaly. On average, Na-amphibole has higher Cr, Ni and  
628 Co contents in blocks compared to rinds (Figs. 14bc).

629 Ca-amphibole shows an enrichment in HREE and a negative anomaly of Eu. In transect L, REE  
630 contents are lower near the contact. In transect G, tremolite (in vein sample G\_n8b) has a lower  
631 REE content compared to G\_p1, and a less pronounced Eu anomaly. The range of Li, Co and LILE  
632 contents is similar in all samples (Fig. 14). In transect L, L\_p5 and L\_p135 have a similar Ni  
633 content, but L\_p5 has a higher Cr content. Ca-amphibole in G\_n8b is depleted in Cr, Ni, Th, Nb,  
634 Sc and enriched in Zr compared to G\_p1. Ca-amphibole in transect K has a lower Ni and Co  
635 content on average than other transects.

### 636 **8.3 Clinopyroxene**

637 Clinopyroxene shows a trace element composition with HREE enrichment and a negative Eu  
638 anomaly in G\_n8. Omphacite present in G\_n8b (crosscut by a tremolite-chlorite vein) is enriched  
639 in REE, Sc, V and depleted in Ba, Ta, Zr, Hf compared to G\_n8. The contents of Li, Cr, Co and Ni  
640 are similar in G\_n8 and G\_n8b with the exception of a few lower values of Cr in G\_n8 (Fig. 14).  
641 Clinopyroxene in transect K (Fig. 14) shows similar Li, Cr and Ni contents as in transect G.

642

### 643 **8.4 Garnet**

644 Garnet shows classical HREE enrichment (Fig. 13g). Transects L and K show more dispersion  
645 of the data and higher values for LREE, Th, Nb and LILE than transect G. Garnet in transect K has  
646 lower Li and Cr contents and shows a marked Eu positive anomaly in the core (up to 11, Fig.  
647 13h). The different trace element compositions of garnet in transect K further support the  
648 existence of a distinct generation of metasomatic garnet.

649

## 650 **9 Thermobarometry**

651 In order to determine the equilibrium conditions for the unmetasomatized metavolcanic rock of  
652 transect L (sample L\_n57), thermodynamic modelling was performed in the NCKFMASHTO  
653 system using the Perple\_X software (version 6.7.5, December 2016; Connolly 1990, 2005; see  
654 supplementary data). The thermodynamic dataset from Holland and Powell (1998) was used  
655 with the following set of activity models for solid solutions: garnet, epidote (Holland and Powell,

656 1998), chlorite (Holland et al., 1998), omphacite (Green et al., 2007), amphibole (Diener et al.,  
657 2007), feldspar (Holland and Powell, 2003), phengite (White et al., 2014) and talc as an ideal  
658 solution. Pseudosection calculation was performed with excess water, as justified by the  
659 abundance of hydrated phases. In the absence of carbonates in the studied sample, CO<sub>2</sub> was  
660 neglected and a fixed water activity of 1 was used for the fluid. P-T conditions, estimated based  
661 on the Mg content of glaucophane and garnet and on the Fe<sup>3+</sup> content of epidote, are around  
662 530-550°C and 1.8-2 GPa. This value is similar to those estimated by most studies (Okrusch and  
663 Bröcker 1990; Trotet et al. 2001; Rosenbaum et al. 2002; Keiter et al. 2004; Schumacher et al.  
664 2008; Philippon et al. 2013; Lister and Forster 2016; Laurent et al. 2018), indicating that the peak  
665 paragenesis of the block core in transect L is largely preserved.

666 In contrast, the nature of metasomatic minerals replacing the peak paragenesis suggests that  
667 most if not all of the rind formation process occurred during exhumation. For example, in  
668 transect L, garnet is replaced by phengite (Fig. 7b), which is the only K host. The addition of K,  
669 likely related to the metasomatic event, therefore occurred after garnet growth. In transect K,  
670 the composition of the garnet population in the rind differs from that in the block. Garnet from  
671 the rind also contains inclusions of metasomatic clinopyroxene, indicating formation after the  
672 inception of metasomatism (Fig. 7i).

673 In order to further assess the P-T conditions of metasomatism, thermobarometric calculations  
674 were performed for three samples (K\_18e, 24b and L\_p135) using Thermocalc V. 3.21 (Holland  
675 and Powell, 1998, 2011). The H<sub>2</sub>O activity was set at 1.0 based on the lack of significant halogen

676 and carbon contents in hydrous minerals. Mineral activities were determined using the AX  
677 software (Holland and Powell, 1998; updated in 2011).

678 — Matrix sample L\_p135 (L transect): the metasomatic assemblage of Ca-amphibole ( $\text{Na}_{0.438}$   
679  $\text{Ca}_{1.545} \text{Mg}_{4.120} \text{Fe}_{2+0.561} \text{Fe}_{3+0.206} \text{Mn}_{0.016} \text{K}_{0.019} \text{Al}_{0.285} \text{Si}_{7.890} \text{O}_{22} (\text{OH})_2$ ), chlorite ( $\text{Mg}_{4.153} \text{Fe}_{2+0.838}$   
680  $\text{Mn}_{0.009} \text{Al}_{1.912} \text{Si}_{3.063} \text{O}_{10} (\text{OH})_8$ ) and talc ( $\text{Mg}_{2.794} \text{Fe}_{2+0.138} \text{Si}_{4.030} \text{O}_{10} (\text{OH})_2$ ) is unconstrained for  
681 pressure ( $1.90 \pm 1.99$  GPa) but yield temperatures of  $561 \pm 78^\circ\text{C}$ .

682 — Rind sample 24b: the equilibrium assemblage made of Ca-amphibole ( $\text{Na}_{0.507} \text{Ca}_{1.538} \text{Mg}_{3.905}$   
683  $\text{Fe}_{2+0.565} \text{Mn}_{0.015} \text{K}_{0.029} \text{Al}_{0.698} \text{Si}_{7.870} \text{O}_{22} (\text{OH})_2$ ), chlorite ( $\text{Mg}_{3.634} \text{Fe}_{2+0.633} \text{Mn}_{0.010} \text{Al}_{2.214} \text{Si}_{3.198} \text{O}_{10}$   
684  $(\text{OH})_8$ ) and albite ( $\text{Na}_{0.964} \text{Al}_{1.014} \text{Si}_{2.995} \text{O}_8$ ) is poorly constrained for temperature ( $505 \pm 155^\circ\text{C}$ ) but  
685 yield pressures around  $1.02 \pm 0.15$  GPa.

686 — Sample K\_18e: using the garnet-clinopyroxene Fe-Mg exchange thermometer of Ravna (2000)  
687 at a pressure of 1.0 GPa, the garnet core ( $\text{Ca}_{0.897} \text{Mn}_{0.694} \text{Mg}_{0.170} \text{Fe}_{2+1.183} \text{Fe}_{3+0.134} \text{Al}_{1.977} \text{Si}_{2.935}$   
688  $\text{O}_{12}$ ) and clinopyroxene inclusion ( $\text{Ca}_{0.494} \text{Na}_{0.467} \text{Mg}_{0.399} \text{Fe}_{2+0.124} \text{Fe}_{3+0.153} \text{Al}_{0.379} \text{Si}_{1.966} \text{O}_6$ )  
689 predicts a temperature of  $561^\circ\text{C}$ . We estimate an uncertainty of  $\pm 50^\circ$  due to uncertainties in the  
690 AX software models. The assemblage of garnet rim ( $\text{Ca}_{1.006} \text{Mn}_{0.047} \text{Mg}_{0.417} \text{Fe}_{2+1.685} \text{Fe}_{3+0.068}$   
691  $\text{Al}_{1.999} \text{Si}_{2.967} \text{O}_{12}$ ), in direct contact with clinopyroxene ( $\text{Ca}_{0.447} \text{Na}_{0.497} \text{Mg}_{0.399} \text{Fe}_{2+0.112} \text{Fe}_{3+0.088}$   
692  $\text{Al}_{0.425} \text{Si}_{1.992} \text{O}_6$ ), chlorite ( $\text{Mg}_{3.330} \text{Fe}_{2+1.276} \text{Mn}_{0.027} \text{Al}_{2.487} \text{Si}_{2.818} \text{O}_{10} (\text{OH})_8$ ), epidote ( $\text{Ca}_{2.026} \text{Mn}_{0.014}$   
693  $\text{Fe}_{2+0.074} \text{Fe}_{3+0.426} \text{Al}_{2.565} \text{Si}_{2.938} \text{O}_{12} (\text{OH})$ ) and albite ( $\text{Na}_{1.001} \text{Al}_{1.026} \text{Si}_{2.978} \text{O}_8$ ) yield pressures of  
694  $1.03 \pm 0.11$  GPa for temperatures of  $653 \pm 27^\circ\text{C}$ .

695 Estimates for metasomatism therefore lie mostly between 0.90-1.15 GPa and 500-650°C.  
696 Around 1 GPa, the breakdown of serpentine should occur for temperatures above 600°C

697 (Wunder et al., 1997; Guillot et al., 2015). Serpentinite unaffected by metasomatism is still  
698 observed in significant amounts in the mélange unit (Fig. 2). Therefore, it is more likely that the  
699 Kampos-Lia mélange experienced a temperature between 500-600°C during metasomatism, and  
700 that the high temperature from the multi-mineral equilibrium in sample 18e is overestimated.  
701 Our results are consistent with the estimation made by Breeding et al. (2004) using Thermocalc  
702 (1.17-1.23 GPa and 500-550 °C). However, we find higher temperatures than estimated by Miller  
703 et al. (2009; 1.20 GPa and 430°C) using Perple\_X (Connolly, 2005). Finally, the estimation of  
704 Marschall et al. (2006; 0.60-0.75 GPa and 400-430°C) for some tourmaline-chlorite-albite  
705 bearing reaction zones is both lower in temperature and pressure. It could represent a later  
706 stage of metasomatism during further exhumation.

## 707 **10 Discussion**

### 708 **10.1 Mélange structure and deformation**

709 The Kampos-Lia mélange zone, ascribed to the Pindos oceanic domain (Bonneau and Kienast,  
710 1982; Bonneau, 1984), is mainly composed of metavolcanics (54.4% of the surface, Fig. 2)  
711 associated with an ultramafic matrix derived from or composed of serpentinite (34% of the  
712 surface). Blocks in the ultramafic matrix are mainly metagabbros, and some show meta-pillow  
713 like structure. Acidic or basaltic dikes are observed in the metavolcanics and in some blocks.  
714 Metavolcanics have a tuffitic and/or pelitic component (Fig. 9a). Marbles are mostly observed  
715 near the upper contact. The distribution of lithologies, with metavolcanic layers on top of  
716 serpentinite associated with gabbro (Figs. 2,3), and the abundance of serpentinites (variably

717 metasomatized) advocate for a slow-spreading oceanic realm dominated by mantle exhumation,  
718 located close to the continental margin but further outboard than the Posidonia and Chroussa  
719 units. Trace element compositions of Syros metabasites are consistent with an oceanic origin  
720 (Marschall et al., 2009; Seck et al., 1996), with a stable isotope signature of seawater alteration  
721 (Katzir et al., 2007; Putlitz et al., 2000; Seck et al., 1996).

722 Metavolcanic layers are folded with a west-east orientation and are structurally distinct from  
723 the major part of the block-in-matrix structure (Figs. 2,4). This folding indicates north-south  
724 shortening and is consistent with prograde thrusting deformation (Philippon et al., 2011; Keiter  
725 et al., 2011; Laurent et al., 2016). Laurent et al. (2016) already proposed that the shear zones on  
726 either side of the Kampos-Lia mélange zone (the Kastri and Lia shear zones) were prograde  
727 thrusts later reactivated as top-to-the east shear zones during exhumation. At the scale of the  
728 Kampos-Lia mélange zone, large-scale as well as outcrop-scale boudinage is indeed compatible  
729 with top to the east shearing (Fig. 3). A boudinaged marble layer is observed near and parallel  
730 to the north contact of the mélange zone. This shearing is observable all over Syros and  
731 occurred during the syn- to post-blueschist facies exhumation (Laurent et al., 2016).

732 The overall organization and distribution of lithologies in the Kampos-Lia mélange is  
733 therefore consistent with an oceanic or more likely an ocean-continent transitional domain.  
734 These conclusions are consistent with the recent reappraisal of the Pindos oceanic domain as an  
735 extended and thinned continental crust associated with mantle exhumation and incipient  
736 oceanic crust formation during the Cretaceous (Cooperdock et al., 2018; Kotowski et al., 2020;  
737 Schmid et al., 2020). Despite burial and exhumation-related deformation, an inherited internal  
738 structure can still be recognized (Fig. 4), suggesting that tectonic disruption was far from being

739 complete. Unlike other mélanges zones (e.g., Franciscan Complex, Catalina Schist or Northern  
740 Serpentinite Mélange of Cuba), the Kampos-Lia mélange zone is not a chaotic mélange and  
741 remained relatively coherent during burial and exhumation. A significant part of the 'mélange'  
742 structure can therefore be ascribed to the initial oceanic structure rather than to deformation  
743 along the plate interface.

744 This coherent structure and the absence of exotic blocks (with original lithology and/or  
745 metamorphic grade) suggest a relatively low mechanical mixing, incompatible with the  
746 incorporation of mantle wedge derived serpentinite. The geochemical study of Kampos sub-unit  
747 serpentinites (Cooperdock et al., 2018) further support these conclusions. These authors  
748 showed that the geochemical signature of Kampos serpentinites is consistent with derivation  
749 from abyssal peridotites in a hyper-extended margin setting or mid-ocean ridge and fracture  
750 zone environment (even if a certain overlap in trace elements patterns exists between the  
751 different tectonic settings; Deschamps et al., 2013). While the occurrence of serpentinite mesh  
752 texture has been associated with a late stage of serpentinization (Pogge von Strandmann et al.,  
753 2015), the relatively low and heterogeneous deformation in the mélange may have allowed for  
754 the preservation of undeformed serpentinite. In conclusion, the Kampos-Lia block-in-matrix  
755 structures essentially reflect lithological heterogeneities in an ocean-continent transitional  
756 domain and did not undergo major mechanical mixing during burial and exhumation. The  
757 Kampos Unit is therefore not a tectonic mélange as previously thought.

## 758 **10.2 Blocks, rinds and matrix**

759 The diversity of reaction zones seems to depend first and foremost on the nature of the  
760 protolith (Figs. 6,7). Whole-rock compositions, petrography and major/trace element mineral  
761 chemistry (Figs. 8-14) allows differentiating four parts in the reaction zones:

762 (1) blocks or layer, slightly metasomatized at most, retaining the eclogitic or blueschist-  
763 facies paragenesis dictated by the chemical nature of the protolith (e.g., Fe-Ti rich to Mg-Al rich).

764 (2) a rind, where the block/layer is thoroughly metasomatized. It generally comprises  
765 chlorite, amphibole, sodic pyroxene, albite, phengite, apatite and titanite. Large variations exist  
766 in whole-rock major (SiO<sub>2</sub>, MgO, CaO) or incompatible elements (LILE, B, U, Li and some REE)  
767 and in mineral compositions.

768 (3) a metasomatized matrix chiefly composed of tremolite, chlorite and/or talc, with  
769 decreasing Ca and increasing Cr, Ni and Mg content, both in bulk chemistry and mineral  
770 composition, from the matrix/block contact to the serpentinite. Enrichment in incompatible  
771 elements (some REE, U and Li), particularly near the contact, is observed in the metasomatized  
772 matrix compared to the serpentinite.

773 (4) a serpentine matrix with various proportions of talc, with classic high Cr, Ni and Mg  
774 content and insignificant Ca.

775 The whole-rock concentrations of relatively immobile elements (Al, Zr, Ti, V, Y, Cr and Ni) and  
776 their sharp drop from rind to matrix (Figs. 8,9) suggest a serpentinite origin for the  
777 metasomatized matrix, and that the matrix/block 'front' did not move spatially during  
778 metasomatism. The main major elements exchanged across the contact are Mg (derived from

779 serpentinite) and Ca (derived from the blocks, Fig. 8). Metavolcanic layers, since they  
780 predominate across the Kampos-Lia mélange at least, have the largest contribution to  
781 metasomatic exchanges and transformation of matrix.

782 The two transects studied in detail (L,G; Fig. 15) show different trace element patterns.  
783 Transect L (across metavolcanics) points to an enrichment in Li, U, LILE and HREE with a  
784 significant exchange of B and LREE between the block and matrix (Fig. 15a). Transect G (across a  
785 Fe-Ti-rich gabbro) points to an enrichment in LREE and depletion in U and LILE in the rind and  
786 metasomatized matrix, and exchanges of HREE, Li and B between the block and matrix (Fig.  
787 15b). These contrasts reveal the complexity of fluid-assisted exchanges in Kampos at subunit  
788 scale.

789 The strong variation in Mg and Ca (Fig. 15) documented here is similar to that previously  
790 observed in the Lia beach transect (Breeding et al., 2004; Miller et al., 2009). In this  
791 metavolcanic rind, Breeding et al. (2004) observed a depletion in LILE and U without change in  
792 REE concentration and Miller et al. (2009) observed a depletion in LILE but an enrichment in B  
793 and LREE (Fig. 15c). The Lia beach transect, however, shows only very limited metasomatic  
794 overprint of the serpentinite along the contact. The enrichment or depletion in LILE  
795 concentration observed in this study has been documented in various metasomatized rocks, but  
796 more particularly the enrichment for mafic and ultramafic rocks, which are initially poor in these  
797 elements (Bebout, 2007; Bebout and Penniston-Dorland, 2016). Enrichment in LILE, usually  
798 associated with phengite crystallization (such as in the rind of transect L), has been documented  
799 for both prograde and retrograde metamorphism (e.g. in the Catalina Schist and Samana  
800 Complex; Sorensen et al., 1997).

### 801 **10.3 Fluid composition and behavior**

802 Chemical exchanges during metasomatism can occur in several ways: (1) diffusion following  
803 a chemical potential gradient, (2) advection with fluid flow in fractures and intergranular spaces  
804 and (3) mechanical mixing.

805 Contrary to other mélanges (e.g., Catalina schist; Bebout and Barton, 2002; Bebout and  
806 Penniston-Dorland, 2016; Gorman et al. 2019), there is no evidence for mechanical mixing at the  
807 scale of a transect: clearly defined intermediate composition are not found (Figs. 8,9d-f) and  
808 element concentrations throughout the transects do not show systematic, diagnostic parallel  
809 evolutions (in the sense of Gorman et al. 2019, their figure 9b). In addition, no strong correlation  
810 is observed between relatively immobile elements (Figs. 9d-f). The rinds are moreover  
811 commonly foliated parallel to the inner block foliation, and do not show any additional  
812 deformation patterns which could be associated with incorporation and digestion of matrix  
813 clast(s). Nor do we observe any dismemberment of block or rind into the matrix. Mechanical  
814 mixing, if any, is therefore not a dominant process and is mostly hidden by advective and  
815 diffusive processes.

816 Element exchange is more likely controlled by advection at the scale of the unit (such as B, Li,  
817 U and LILE or Re and Pd; Gorman et al. 2019) with high enrichment or depletion along the  
818 contact. Other elements (such as Mg and Ca) are redistributed locally and strongly governed by  
819 local rock composition through diffusion in the intergranular fluid network at the meter scale, as  
820 demonstrated by Pogge von Strandmann et al. (2015). Pogge von Strandmann et al. (2015)  
821 estimated that the formation of the Lia Beach transect reaction zone may have taken ~100 ka,  
822 suggesting that the influx of fluids did not last throughout the entire exhumation process but

823 rather during one or several short events. A full integration of the entire reaction zone is  
824 necessary to tell if REE were advectively moved in or out of the area and it remains to be  
825 quantified how much of the REE are really mobilized over larger distances by fluid advection as  
826 opposed to local redistribution across the growing contacts. Indeed, REE enrichment is generally  
827 associated with the crystallization of accessory minerals such as epidote, apatite, monazite and  
828 titanite (Miller et al., 2009; Sorensen and Grossman, 1993), which can form narrow domains in  
829 the reaction zone. Relatively REE-enriched phases such as Ca-amphibole in the matrix (i.e.,  
830 tremolite; Fig. 13) also participate in REE redistribution during fluid-rock interactions.

831  
832 Significant fluid flow occurs where dynamic permeability is higher, namely at the contact  
833 between sheared metavolcanics and the matrix, where Ca and Mg diffusion takes place. Fluid  
834 flow is also expected in the high-permeability matrix, as suggested by numerical modeling  
835 (Ague, 2007). In the matrix, tremolite is mainly present near blocks and layers but also as  
836 patches with no visible associated block. Pathways and residence time of the fluid probably  
837 dictate the location of tremolite pods in the metasomatized matrix and may explain differences  
838 between transects across common metavolcanics (this study) and the Lia beach transect (Miller  
839 et al., 2009).

840 Fluids have a dominant impact on the composition of rinds and metasomatized matrix in  
841 mobile elements (e.g., Li, B, U, LILE and REE; Fig. 15). Differences between transects L, G and Lia  
842 Beach suggest that fluid composition changes at the m- to hm-scale in the Kampos unit, and  
843 through time, when migrating through the *mélange* zone and interacting with various  
844 lithologies. If representative, transect L, which corresponds to the most common reaction zone

845 in the Kampos-Lia mélange, would indicate that infiltration of external fluid caused an  
846 enrichment in Li, B, U, some LILE (Rb, K, Na, Ba, Eu), MREE and HREE (Figs. 9,10,15) and the fluid  
847 was therefore probably enriched in these elements.

848 The rind of transect L shows very high Li concentrations (up to 45  $\mu\text{g/g}$ ). Marschall et al.  
849 (2009) documented even higher Li concentrations in some metasomatized rocks of Syros (up to  
850 88 $\mu\text{g/g}$ ) and argued for an external fluid origin of Li. Based on high  $\delta^{11}\text{B}$  values in tourmaline and  
851 low silica activity (as a result of interaction with serpentinite), these authors argued for a slab-  
852 derived fluid modified through migration along the subduction interface. This is also compatible  
853 with the sulfur isotopic composition of metasomatic sulfides (Walters et al., 2019). Relatively  
854 high fluid concentration in LILE along with Li, B and Pb has been attributed to interaction with  
855 sedimentary rocks, given their enrichment in those elements compared to mafic and ultramafic  
856 rocks (King et al., 2006; Penniston-Dorland et al., 2012). It is therefore likely that the  
857 metasomatic fluid derived from the deeper dehydration in the downgoing slab was modified by  
858 its infiltration into various lithologies (e.g. sedimentary and ultramafic) before reaching the  
859 Kampos-Lia unit along the slab interface.

860 Finally, considering that metasomatism was probably rather short-lived ( $\sim 100$  ka integrated  
861 duration; Pogge von Strandmann et al., 2015) and that fluid compositions vary largely at the  
862 meter to hectometer scale, with some elements mobilized and advected over large distances or  
863 redistributed locally and strongly governed by local rock composition, caution is needed when  
864 extrapolating fluid composition to be representative of fluid-rock interactions at the scale of the  
865 subduction plate interface.

## 866 **10.4 Metasomatism in a subduction context?**

867

### 868 **10.4.1 Timing and significance of the CBU metasomatism in the subduction history**

869 After entering subduction, the small CBU oceanic domain, typified by mantle exhumation  
870 and incipient oceanic crust formation, was sheared by top-to-the south reverse shear zones  
871 along the prograde path (Philippon et al., 2011; Laurent et al., 2016), and reached maximum  
872 burial between 42 and 53 Ma (Tomaschek et al., 2003; Putlitz et al., 2005; Lagos et al., 2007;  
873 Lister and Forster, 2016; Laurent et al., 2017). Tectonic slices of the CBU were then scrapped off  
874 the downgoing slab and underplated and/or started exhuming along the plate interface (Fig.  
875 16b). Exhumation patterns are consistent with top-to-the east shearing (Trotet et al., 2001;  
876 Laurent et al., 2016).

877 Detachment from the slab may already have triggered the inflow of externally derived fluids  
878 and early metasomatism, at or near peak burial conditions, but supporting evidence is lacking  
879 for now. Instead, metasomatic minerals replacing the peak paragenesis suggest that most if not  
880 all of the rind formation process occurred during exhumation. Our estimates suggest that  
881 metasomatism occurred at 0.90-1.15 GPa and 500-600°C (Fig. 16a), hence much lower pressure  
882 than peak P-T conditions (530-550°C and 1.8-2 GPa; supplementary material). This is comparable  
883 with values documented by Laurent et al. (2018) for the Posidonia and Chroussa units, who  
884 related this slight isobaric heating to underplating at the base of the crust. Contrary to them,  
885 however, we suggest that some heating also affected the Kampos unit and that metasomatism  
886 began or largely developed at that time (Fig. 16c). This episode of heating could have played a  
887 role in the onset of significant fluid migration and metasomatism in the Kampos unit. It was

888 dated between 37 and 30 Ma on Tinos and Andros islands (Bröcker et al., 1993, 2004; Huet et  
889 al., 2015). In support of this interpretation, Brooks et al. (2019) documented a fluid circulation  
890 and entrapment during the late stages of exhumation on Sifnos beginning at about 1.0 GPa and  
891 540°C. Those conditions were estimated using fluid inclusion microthermometry and are  
892 consistent with this heating phase and our own PT estimations for the metasomatism in Syros.

893 Despite the m- to hm-scale block-in-matrix discontinuities/heterogeneities likely inherited  
894 from the slow-spreading environment, data indicate that the Kampos-Lia mélange zone was only  
895 mildly deformed and infiltrated by fluids during prograde and peak conditions or immediately  
896 after the critical step of detachment from the slab (Agard et al., 2018) and incorporation along  
897 the plate interface. Significant fluid ingress and metasomatism only occurred during  
898 exhumation and heating. In this respect the Kampos-Lia mélange differs from a number of  
899 studied mélanges interpreted as the result of prograde disaggregation and mechanical mixing  
900 (e.g., the Santa Catalina schist, Penniston-Dorland et al. 2014, 2018), and/or retrograde mixing  
901 of blocks with variable peak temperatures having witnessed fluid infiltration across a range of  
902 metamorphic grades (e.g., Franciscan complex; Oh et al. 1991; Cooper et al. 2011; Wakabayashi  
903 and Dilek 2011).

#### 904 **10.4.2 Subduction interface metasomatism and rheology**

905 The data presented here show that exhumation-related metasomatism did not fully digest the  
906 metavolcanic layers or competent blocks, at least not the largest ones. In contrast,  
907 approximately half of the ultramafic matrix has been metasomatized (Fig. 2) and was  
908 transformed into a hybrid rock with intermediate chemical composition, probably in part by

909 infiltrating subduction-derived fluids. Such hybridization could resemble the one inferred to  
910 occur deeper down in subduction zones, which may ultimately form hybrid buoyant diapirs  
911 feeding volcanic arcs (Marschall and Schumacher, 2012; Nielsen and Marschall, 2017; Codillo et  
912 al., 2018). The Syros example show, however, that no significant metasomatism and chemical  
913 mixing occurred in the slab during subduction, at least until peak conditions of ~2 GPa and 500-  
914 550°C were reached, despite the fact that it represents relatively deformable slow-spreading  
915 lithosphere (Ruh et al., 2015). This echoes the conclusions of Bayet et al. (2018), who suggested  
916 that metasedimentary diapirs may occur only at depth greater than 80 km and/or at some  
917 distance from the plate interface.

918 In the case of Syros, the low proportion of blocks in the matrix (2.5%) precludes any  
919 significant role of these blocks on the mechanical behavior of the Kampos unit as a whole,  
920 contrary to mélanges studied by Grigull et al. (2012; i.e., Ouaième Complex, New Caledonia; Rio  
921 San Juan Complex, Dominican Republic; Franciscan Complex and Santa Catalina schist, USA). It is  
922 likely that the hectometer to km-scale subcontinuous metavolcanic layers (Figs. 2-4), despite  
923 boudinage and mild digestion by metasomatism, had a much larger impact on the bulk rheology  
924 of the Kampos-Lia mélange zone.

925 The formation of both rheologically stiffer (tremolite) and weaker (chlorite and talc) matrix  
926 minerals at the expense of serpentinite may affect the rheology of the mélange in the context of  
927 exhumation along a plate contact. This is reminiscent of the Santa Catalina schist (Penniston-  
928 Dorland et al., 2018) where metasomatism induces the formation of hard minerals and may

929 increase the strength of the matrix. This may have to be considered to understand the  
930 exhumation rheology along the subduction interface.

## 931 **11 Conclusions**

932 On Syros island, the presence of km-scale 'mélange' provides the opportunity to study  
933 metasomatism and its impact along the subduction plate interface. The combination of detailed  
934 mapping, structural, petrological and geochemical data suggests that:

935 (1) This mélange is, to a first approximation, composed of folded metavolcanics on top of an  
936 ultramafic matrix hosting a limited block fraction (2-3vol%). This 'mélange' is a preserved  
937 fragment of a discontinuous oceanic domain dominated by mantle exhumation, which was only  
938 mildly tectonically disrupted during burial, and even exhumation. Deformation associated with  
939 burial produced north-south folding whereas pervasive top-to-the-east boudinage prevailed  
940 during exhumation, without significant tectonic mixing. This 'mélange' is not a tectonic mélange  
941 as previously thought.

942 (2) Metasomatism occurred mostly, if not entirely, during exhumation, at ~0.90-1.15 GPa  
943 and 500-600°C, probably in a context of underplating and isobaric heating.

944 (3) During metasomatism, Ca diffuses from blocks into the matrix (metasomatizing the  
945 serpentinite) and Mg diffuses from the serpentinite into the blocks (forming a metasomatic rind  
946 at the block margins). In detail, the variety of rinds largely depends on the block/layer lithology.  
947 Significant mobilization of some elements (Li, B, U and LILE) by fluids is detected by whole-rock  
948 and mineral compositions. Large variations nevertheless exist at the scale of the mélange,  
949 suggesting a complex evolution of the fluid composition. Some elements are homogenized and

950 mobilized over larger scales (Li, B, U and LILE) and controlled by the advecting fluid, whereas  
951 other elements (Ca and Mg) are locally controlled by the rock compositions and only  
952 redistributed locally by diffusion across lithological boundaries.

953 (4) About half of the serpentinite matrix was metasomatized into a chlorite-tremolite-talc  
954 schist. This rock is chemically hybrid and most of it formed during exhumation. The Syros  
955 example also shows that no significant metasomatism and chemical mixing occurred within the  
956 slab, at least until the rocks reached peak conditions of ~2 GPa and 500-550°C, despite the fact  
957 that the Syros mélange represents relatively deformable slow-spreading partly oceanic  
958 lithosphere. Together with internal heterogeneities (i.e. metavolcanic layers), distributed  
959 metasomatism of the matrix (particularly tremolite formation) may have influenced the  
960 rheology and mechanical behavior of the mélange along the subduction interface during  
961 exhumation.

962

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1274

1275 **Tables**

1276

| Transect        | Samples      | Location | Lithology    | Mineralogy |     |        |    |     |    |    |    |     |        |     |     |    |     |    |    |     | Lat. N/Lon. E |     |                 |   |  |
|-----------------|--------------|----------|--------------|------------|-----|--------|----|-----|----|----|----|-----|--------|-----|-----|----|-----|----|----|-----|---------------|-----|-----------------|---|--|
|                 |              |          |              | Grt        | Lws | Na-Amp | Ep | Cpx | Ph | Pg | Ab | Chl | Ca-Amp | Tlc | Srp | Rt | Ttn | Qz | Ap | Tur |               | Cld | Ox              |   |  |
| Transect L      | n290         | Lia      | metavolcanic | x          |     | x      | x  |     | x  |    | x  | x   |        |     |     |    | x   |    | x  | x   |               | x   | 37.4924/24.9087 |   |  |
|                 | n57          |          | metavolcanic | x          |     | x      | x  |     | x  |    | x  | x   |        |     |     |    |     | x  | x  |     |               |     | x               |   |  |
|                 | n10          |          | rind         | x          |     | x      | x  |     | x  |    | x  | x   |        |     |     |    |     | x  | x  |     |               |     | x               |   |  |
|                 | n1           |          | rind         |            |     | x      | x  |     | x  |    | x  | x   |        |     |     |    |     | x  | x  |     |               |     | x               |   |  |
|                 | p5           |          | matrix       |            |     |        |    |     |    |    |    | x   |        |     |     |    |     |    | x  |     |               |     |                 |   |  |
|                 | p125         |          | matrix       |            |     |        |    |     |    |    |    | x   |        |     | x   |    |     |    |    |     |               |     |                 | x |  |
|                 | p135         |          | matrix       |            |     |        |    |     |    |    |    | x   |        | x   |     |    |     |    |    |     |               |     |                 | x |  |
| p165            | serpentinite |          |              |            |     |        |    |     |    |    |    |     | x      | x   |     |    |     |    |    |     |               | x   |                 |   |  |
| Transect G      | n50          | Galissas | block Fe-Ti  | x          |     |        | x  | x   | x  |    |    |     |        |     |     |    | x   |    |    |     | x             | x   | 37.4306/24.8818 |   |  |
|                 | n30          |          | block Fe-Ti  | x          |     |        | x  | x   | x  |    |    |     |        |     |     |    |     | x  |    |     |               | x   | x               |   |  |
|                 | n8           |          | rind         | x          |     |        | x  | x   | x  |    |    |     |        |     |     |    |     | x  |    |     |               | x   |                 |   |  |
|                 | n8b          |          | rind         | x          |     |        | x  | x   | x  |    |    |     |        | x   |     |    |     | x  | x  |     |               | x   |                 |   |  |
|                 | n2           |          | rind         |            |     |        |    |     |    |    |    |     |        |     |     |    |     | x  | x  |     |               | x   |                 |   |  |
|                 | p1           |          | matrix       |            |     |        |    |     |    |    |    | x   |        |     |     |    |     |    |    |     |               |     |                 | x |  |
|                 | p30          |          | matrix       |            |     |        |    |     |    |    |    | x   |        |     |     | x  |     |    |    |     |               |     |                 | x |  |
| Transect K (18) | 18a          | Kini     | block Fe-Ti  | x          |     |        | x  | x   |    |    |    | x   | x      |     |     |    |     |    |    |     | x             | x   | 37.4439/24.8890 |   |  |
|                 | 18b          |          | rind         | x          |     |        | x  | x   |    |    |    | x   | x      |     |     |    |     |    |    |     | x             | x   |                 |   |  |
|                 | 18c          |          | matrix       |            |     |        |    |     |    |    |    |     | x      |     |     |    |     |    |    |     |               | x   | x               |   |  |
|                 | 18d          |          | rind         | x          |     |        | x  | x   |    |    |    |     |        |     |     |    |     |    |    |     |               | x   | x               |   |  |
|                 | 18e          |          | rind         | x          |     |        | x  | x   |    |    |    |     |        |     |     |    |     |    |    |     |               | x   | x               |   |  |
| Transect 9      | 9a           | Lia      | block Mg-Al  |            |     | x      | x  | x   | x  |    |    |     |        |     |     |    |     |    |    |     | x             | x   | 37.4924/24.9089 |   |  |
|                 | 9b           |          | block Mg-Al  |            |     | x      | x  | x   | x  |    |    |     |        |     |     |    |     |    |    |     |               | x   | x               |   |  |



PF 1,96 1,92 1,70 2,58 4,97 6,17 4,90 10,33 1,94 1,70 3,51 8,93 3,27 5,29 10,31

1280

1281 *Table 2: Samples whole-rock major (%) and trace element ( $\mu\text{g/g}$ ) concentrations.*

1282

| Sample                         | Amp   |        | Chl                                      |                                       | Grt                                            |                                               |                                                   |                                                 | Cpx                        |                                        | Ep               | Ph                                             |                                              | Srp    | Tlc    | Tur   |
|--------------------------------|-------|--------|------------------------------------------|---------------------------------------|------------------------------------------------|-----------------------------------------------|---------------------------------------------------|-------------------------------------------------|----------------------------|----------------------------------------|------------------|------------------------------------------------|----------------------------------------------|--------|--------|-------|
|                                | L_n10 | L_p135 | L_n10                                    | L_p135                                | L_n57                                          | L_n57                                         | K_18e                                             | K_18e                                           | G_n8                       | 4                                      | L_n10            | L_n10                                          | 17                                           | L_p165 | L_p165 | 21    |
| Location                       | rind  | matrix | rind                                     | matrix                                | core                                           | rim                                           | core                                              | rim                                             | rind                       | peak                                   | rind             | rind                                           | block                                        | matrix | matrix | rind  |
| SiO <sub>2</sub>               | 58,15 | 58,01  | 26,60                                    | 31,50                                 | 37,82                                          | 38,14                                         | 37,88                                             | 36,62                                           | 55,27                      | 56,12                                  | 38,64            | 51,78                                          | 47,31                                        | 41,23  | 60,91  | 37,57 |
| TiO <sub>2</sub>               | 0,03  | 0,02   | 0,06                                     | 0,02                                  | 0,19                                           | 0,10                                          | 0,07                                              | 0,17                                            | 0,12                       | 0,07                                   | 0,07             | 0,15                                           | 0,03                                         | 0,01   | 0,02   | 0,53  |
| Al <sub>2</sub> O <sub>3</sub> | 10,52 | 0,56   | 18,94                                    | 16,68                                 | 20,94                                          | 21,44                                         | 21,42                                             | 20,92                                           | 6,80                       | 10,16                                  | 26,41            | 26,37                                          | 39,78                                        | 2,81   | 0,37   | 31,77 |
| Cr <sub>2</sub> O <sub>3</sub> | 0,01  | 0,01   | 0,00                                     | 0,00                                  | 0,15                                           | 0,15                                          | 0,01                                              | 0,00                                            | 0,00                       | 0,02                                   | 0,00             | 0,00                                           | 0,00                                         | 0,43   | 0,11   | 0,07  |
| FeO                            | 10,81 | 5,97   | 25,09                                    | 10,31                                 | 25,45                                          | 28,63                                         | 26,63                                             | 19,64                                           | 9,41                       | 6,98                                   | 9,91             | 2,94                                           | 0,23                                         | 10,73  | 3,40   | 5,03  |
| MnO                            | 0,14  | 0,23   | 0,77                                     | 0,11                                  | 4,41                                           | 0,90                                          | 1,11                                              | 10,22                                           | 0,20                       | 0,01                                   | 0,22             | 0,03                                           | 0,00                                         | 0,17   | 0,03   | 0,00  |
| MgO                            | 10,74 | 20,66  | 15,84                                    | 28,66                                 | 1,89                                           | 3,08                                          | 2,84                                              | 1,43                                            | 8,60                       | 7,05                                   | 0,00             | 3,65                                           | 0,03                                         | 31,83  | 29,12  | 8,49  |
| CaO                            | 0,93  | 12,43  | 0,03                                     | 0,00                                  | 9,86                                           | 8,66                                          | 10,06                                             | 10,44                                           | 13,65                      | 11,91                                  | 23,24            | 0,02                                           | 0,12                                         | 0,00   | 0,05   | 0,29  |
| Na <sub>2</sub> O              | 7,27  | 0,51   | 0,02                                     | 0,03                                  | 0,00                                           | 0,00                                          | 0,03                                              | 0,00                                            | 6,65                       | 7,97                                   | 0,02             | 0,49                                           | 7,65                                         | 0,01   | 0,03   | 2,95  |
| K <sub>2</sub> O               | 0,02  | 0,02   | 0,11                                     | 0,00                                  | 0,00                                           | 0,01                                          | 0,00                                              | 0,00                                            | 0,01                       | 0,01                                   | 0,02             | 9,69                                           | 0,35                                         | 0,00   | 0,05   | 0,03  |
| Total                          | 98,63 | 98,41  | 87,41                                    | 87,31                                 | 100,62                                         | 101,07                                        | 100,03                                            | 99,44                                           | 100,91                     | 100,30                                 | 98,77            | 95,07                                          | 95,51                                        | 87,21  | 94,09  | 86,72 |
| O                              | 23    | 23     | 14                                       | 14                                    | 24                                             | 24                                            | 24                                                | 24                                              | 6                          | 6                                      | 12,5             | 11                                             | 11                                           | 7      | 11     | 29    |
| Si                             | 7,95  | 8,00   | 2,82                                     | 3,06                                  | 5,99                                           | 5,98                                          | 5,90                                              | 5,98                                            | 2,01                       | 2,01                                   | 3,07             | 3,45                                           | 3,01                                         | 2,00   | 3,97   | 6,05  |
| Ti                             | 0,00  | 0,00   | 0,00                                     | 0,00                                  | 0,02                                           | 0,01                                          | 0,02                                              | 0,01                                            | 0,00                       | 0,00                                   | 0,00             | 0,01                                           | 0,00                                         | 0,00   | 0,00   | 0,06  |
| Al                             | 1,70  | 0,09   | 2,37                                     | 1,91                                  | 3,91                                           | 3,96                                          | 3,98                                              | 3,99                                            | 0,29                       | 0,43                                   | 2,47             | 2,07                                           | 2,98                                         | 0,16   | 0,03   | 6,03  |
| Cr                             | 0,00  | 0,00   | 0,00                                     | 0,00                                  | 0,02                                           | 0,02                                          | 0,00                                              | 0,00                                            | 0,00                       | 0,00                                   | 0,00             | 0,00                                           | 0,00                                         | 0,02   | 0,01   | 0,01  |
| Fe tot                         | 1,24  | 0,69   | 2,22                                     | 0,84                                  | 3,37                                           | 3,75                                          | 2,65                                              | 3,52                                            | 0,29                       | 0,21                                   | 0,66             | 0,16                                           | 0,01                                         | 0,44   | 0,19   | 0,68  |
| Mn                             | 0,02  | 0,03   | 0,07                                     | 0,01                                  | 0,59                                           | 0,12                                          | 1,40                                              | 0,15                                            | 0,01                       | 0,00                                   | 0,01             | 0,00                                           | 0,00                                         | 0,01   | 0,00   | 0,00  |
| Mg                             | 2,19  | 4,25   | 2,50                                     | 4,15                                  | 0,45                                           | 0,72                                          | 0,34                                              | 0,67                                            | 0,47                       | 0,38                                   | 0,00             | 0,36                                           | 0,00                                         | 2,31   | 2,83   | 2,04  |
| Ca                             | 0,14  | 1,84   | 0,00                                     | 0,00                                  | 1,67                                           | 1,46                                          | 1,80                                              | 1,70                                            | 0,53                       | 0,46                                   | 1,98             | 0,00                                           | 0,01                                         | 0,00   | 0,00   | 0,05  |
| Na                             | 1,93  | 0,14   | 0,00                                     | 0,01                                  | 0,00                                           | 0,00                                          | 0,00                                              | 0,01                                            | 0,47                       | 0,55                                   | 0,00             | 0,06                                           | 0,94                                         | 0,00   | 0,00   | 0,92  |
| K                              | 0,00  | 0,00   | 0,01                                     | 0,00                                  | 0,00                                           | 0,00                                          | 0,00                                              | 0,00                                            | 0,00                       | 0,00                                   | 0,00             | 0,82                                           | 0,03                                         | 0,00   | 0,00   | 0,01  |
| Fe <sup>3+</sup>               | 0,17  | 0,04   | 0,01                                     | 0,00                                  | 0,08                                           | 0,04                                          | 0,12                                              | 0,03                                            | 0,18                       | 0,12                                   | 0,66             | 0,04                                           | 0,00                                         |        |        | 0,02  |
| Fe <sup>2+</sup>               | 1,07  | 0,65   | 2,21                                     | 0,84                                  | 3,29                                           | 3,71                                          | 2,53                                              | 3,49                                            | 0,11                       | 0,08                                   | 0,00             | 0,13                                           | 0,01                                         |        |        | 0,17  |
| Comp.                          | Gln   | Act    | Daph37.8<br>Clin42.6<br>Am16.2<br>Sud2.7 | Daph16.4<br>Clin81.4<br>Am0<br>Sud2.2 | Grs25.4<br>Prp7.4<br>Alm54.8<br>Sps9.9<br>Adr2 | Grs22.7<br>Prp12<br>Alm61.8<br>Sps2<br>Adr1.1 | Grs23.5<br>Prp10.3<br>Alm58.3<br>Sps6.1<br>Adr1.8 | Grs26.7<br>Prp5.6<br>Alm41.6<br>Sps23<br>Adr3.1 | Id27.9<br>Ae17<br>Quad55.1 | Jd42.3<br>Ae12.3<br>Zo41.8<br>Quad45.5 | Pi58.2<br>Zo41.8 | Tri6<br>Tri11.1<br>Pr12.7<br>Ms41.8<br>Cel34.6 | Tri1.1<br>Pr11.1<br>Pr12.7<br>Ms34.5<br>Cel0 |        |        |       |

1283

1284 *Table 3: Representative EMPA analyses (wt.%) of minerals in the reaction zones.*

1285

## 1286 Figure Captions

1287 *Figure 1: Geological maps of Syros, after (Laurent et al., 2016) and (Keiter et al., 2011), with*

1288 *localization of samples. mv: metavolcanic, mg: metagabbro, mr: marble, sp: serpentinite and*

1289 *msch: micaschists.*

1290 *Figure 2: (a) Satellite picture of the Kampos-Lia mélange zone with various field observations.*  
1291 *Histograms of blocks (b) length-to-width ratio and (c) volume. The 'other' category correspond to*  
1292 *mixte Fe-Ti and Mg-Al, metavolcanic or carbonate blocks. (d) Folded garnets layers from the Lia*  
1293 *shear zone. (e) Nature and localization of blocks. (f) Geological map of the Kampos-Lia mélange*  
1294 *zone with orientation of the blocks elongation axe.*

1295 *Figure 3: Deformation in the Kampos-Lia mélange zone. Metric (a, b) to centimetric (c) folds in*  
1296 *metavolcanic layers characterized by sub-horizontal W-E axes. (d) Folded succession of*  
1297 *metavolcanic layer, glaucophane-rich rind and tremolite-rich matrix. (e) Asymmetric boudinage*  
1298 *and folds in a metavolcanic layer compatible with top-to-the east shearing. (f) Rounded*  
1299 *metabasite block in a matrix top-to-the east shear band. (g) Asymmetric boudinage of a*  
1300 *metabasite layer in marble with top-to-the east shearing deformation. Sheath folds (h) and*  
1301 *asymmetric boudinage (i) of garnet rich layer associated with chlorite crystallization along the*  
1302 *Lia Shear zone.*

1303 *Figure 4: (a) Panorama of the Kampos-Lia mélange zone with (b) WE cross-section and (c) 3D*  
1304 *block diagram.*

1305 *Figure 5: Localization of the samples from transect L.*

1306 *Figure 6: Field pictures. Diversity of block-rind-matrix associations and mineralogic assemblages.*  
1307 *(a-f) metavolcanic layers, (g-j) Fe-Ti rich blocks, (k) Mg-Al rich blocks, (l) Block of intermediate*  
1308 *composition with associated rind and/or matrix.*

1309 *Figure 7: Rind systematics for the three major block and layer lithologies with microphotographs*  
1310 *of representative samples from (a,b) Metavolcanic layer, (d,e,g,h,i) Fe-Ti rich block, (j,k,l) Mg-Al*  
1311 *rich block and (c,f) matrix.*

1312 *Figure 8: Whole rock compositions of samples from transects L and G represented against*  
1313 *distance from the contact block/matrix. Double arrows to the side of the plot showing  $Al_2O_3$*   
1314 *concentration for transect L show the variation of initial compositions in Aluminum based on our*  
1315 *own data (samples L\_n290, L\_n57, L\_p165 and 581) , metavolcanics (zones I and II) from Pogge*  
1316 *von Strandmann et al. (2015) and unmetasomatized serpentinite from Cooperdock et al. (2018).*

1317 *Figure 9: Whole rock compositions from transects L, G and 581. (a) Th/La vs Sm/La diagram*  
1318 *after Plank (2005). (b) CaO vs MgO diagram with data compilation from Miller et al. (2009) for*  
1319 *reference. Harz: harzburgites (GEOROC database, MPI für Chemie, Mainz), AOC: altered oceanic*  
1320 *crust (Kelley et al., 2003), New Cal: New Caledonia (Spandler et al., 2008) and Catalina mélange*  
1321 *(King et al., 2006). The five zones of a reaction zone studied by Miller et al. (2009) are connected*  
1322 *from zone 1 to 5 (Miller et al., 2009). (c) Chondrite normalized Cr vs Eu anomalies ( $Eu/Eu^*$ )*  
1323 *diagram. (d, e, f) Cr vs  $Al_2O_3$ ,  $TiO_2$  and Zr diagrams. (g, h) Multi-element diagrams normalized to*  
1324 *chondrite (Sun and McDonough, 1989). Reference spectra are MORB (McDonough and Sun,*  
1325 *1995) and Syros serpentinites (Cooperdock et al., 2018).*

1326 *Figure 10: Mass ratio of whole rock compositions. (a) Element mobility in transect L along the*  
1327 *contact block/matrix during fluid rock interaction. Mass ratio of metasomatic matrix to*  
1328 *serpentinite ( $p5/p165$ ) against mass ratio of rind to metavolcanic ( $n1/n57$ ). Metasomatic*  
1329 *samples compositions are normalized to the protolith Al content. (b) Mass ratio of metasomatic*

1330 matrix to serpentinite (p125/p165) in transect L. As and Sb are below detection limit only in  
1331 L\_p165. Hf and Zr are below detection limit only in L\_p125. (c) Mass ratio of metasomatic matrix  
1332 to serpentinite (p5/p165) in transect L. Hf, In, Pb, Rb, Zr, Na<sub>2</sub>O and K<sub>2</sub>O content are below  
1333 detection limit only in L\_p165. As and Sb are below detection limit only in L\_p5. (d) Mass ratio of  
1334 rind to metavolcanic (n1/n57) in transect L. (e) Mass ratio of rind to Fe-Ti block (n2/n50) in  
1335 transect G. Ba, Cs, Pb, Rb, Na and K are below detection limit only in G\_n2.

1336 Figure 11: (a) Composition of amphiboles in a XNa (Na/(Na+Ca)) vs XMg (Mg/(Fe+Mg)) diagram  
1337 with an X-ray compositional map of tremolites from L\_p135. (b) Composition of chlorites in a 2Al  
1338 vs XMg diagram with two Si vs XMg diagrams for transects G and L.

1339 Figure 12: (a) Composition of clinopyroxenes in a Ca vs XMg diagram and a Di+Hd-Aeg+Jd  
1340 ternary diagram. (b) Composition of garnets in a Sps+Grs-Alm-Prp ternary diagram. (c)  
1341 Composition of phengites in a 2Al vs XMg diagram. (d) X-ray compositional map of a garnet from  
1342 L\_n290.

1343 Figure 13: (a-g) Chondrite normalized trace element profiles for minerals from samples from  
1344 transect L and G. (h) Chondrite-normalized Cr vs Eu anomalies (Eu/Eu\*) diagram for garnets.  
1345 Chondrite normalization data after Sun and McDonough (1989).

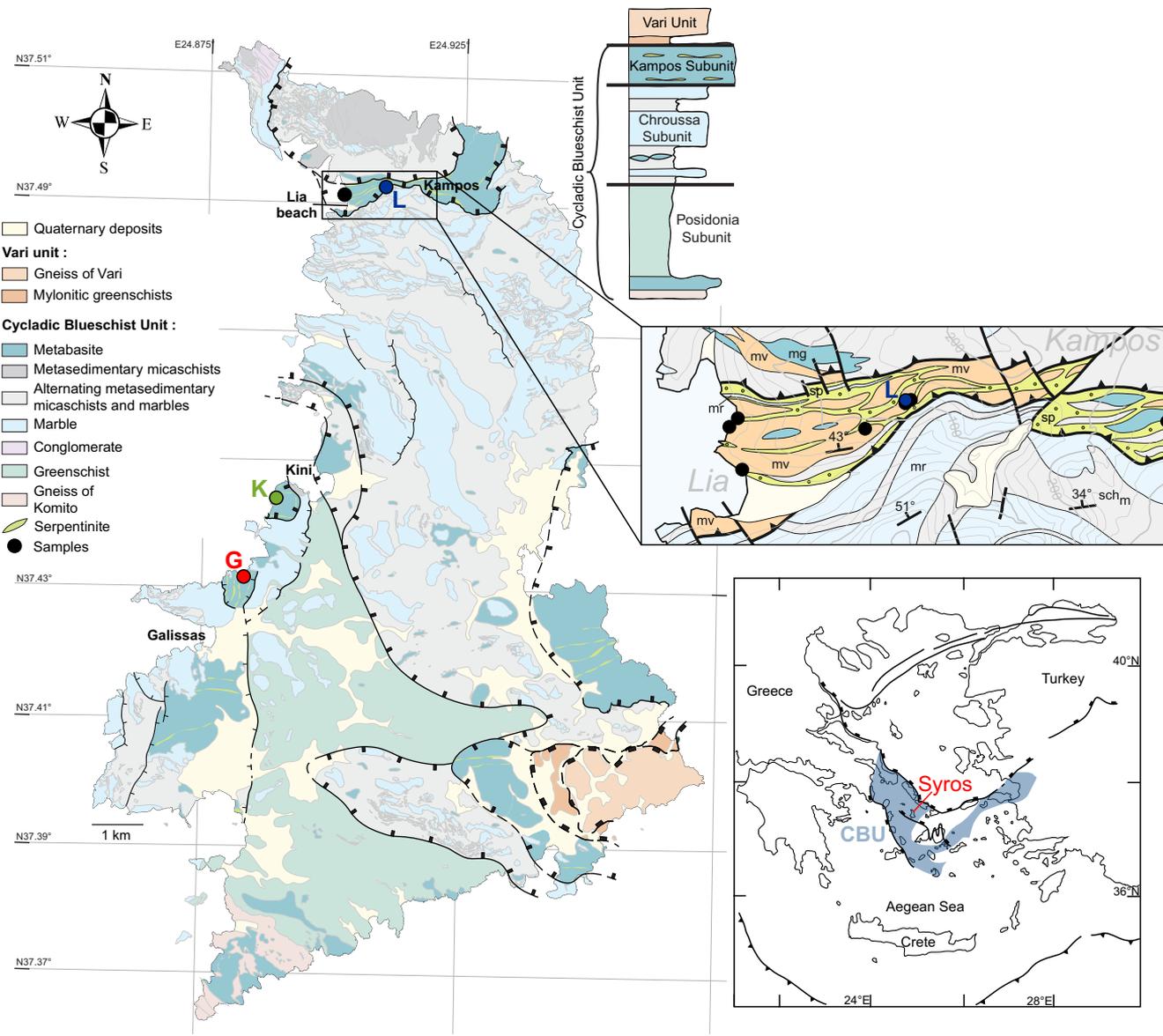
1346 Figure 14: Trace element compositions of amphiboles, chlorites and clinopyroxenes in transect L,  
1347 G and K. (a) Cr vs Li, (b) Cr vs Ni and (c) Cr vs Co diagrams.

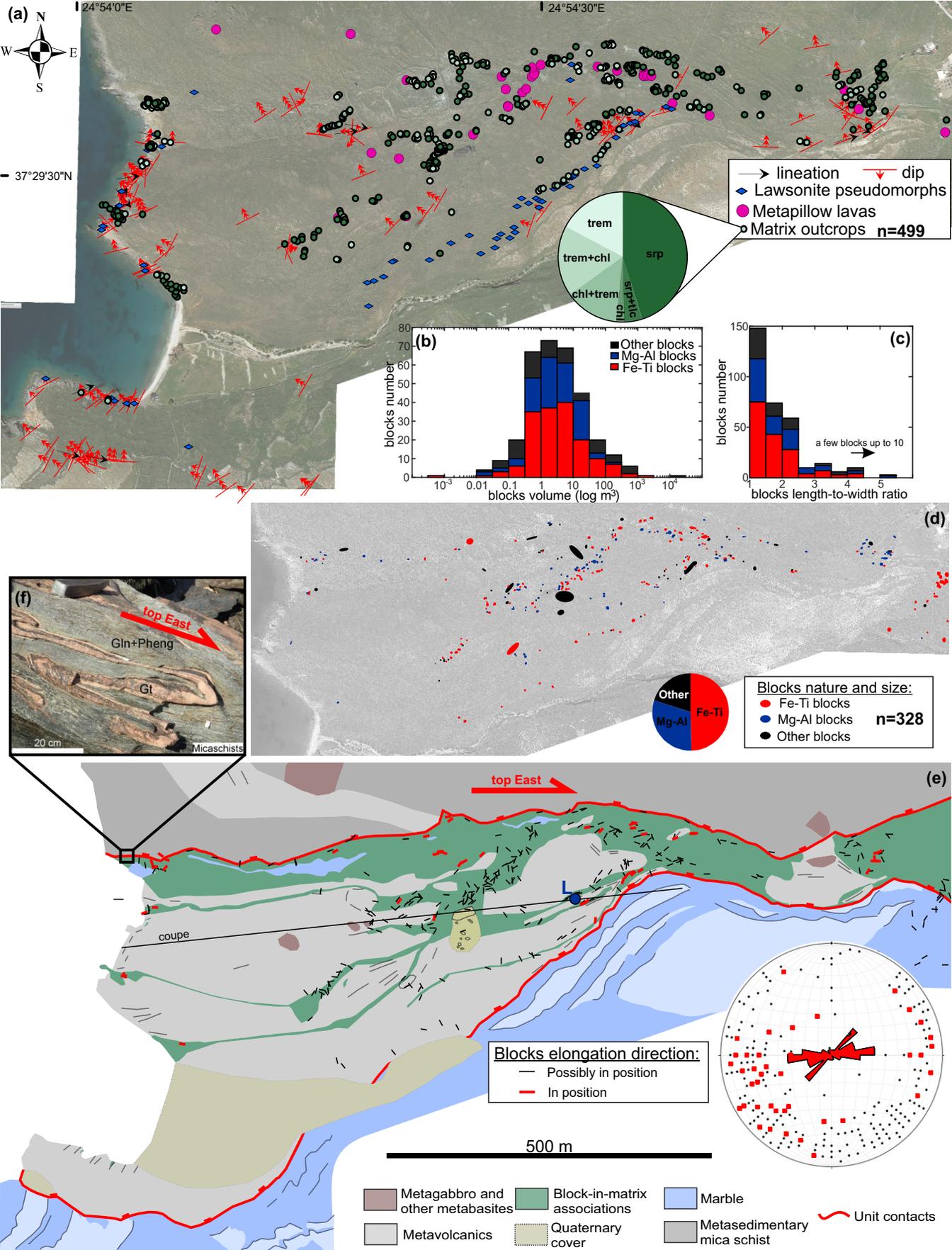
1348 Figure 15: Schematic diagrams showing the diversity of the block-matrix-fluid compositions and  
1349 interactions in Syros mélange zones.

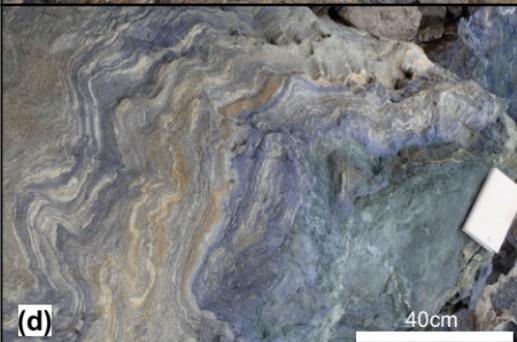
1350 *Figure 16: (a) Summary of P-T paths proposed by different researchers for Syros with estimated*  
1351 *P-T conditions for metasomatism. (b) Conceptual view of the Kampos-Lia mélange zone during*  
1352 *exhumation .(c) Schematic representation of the elements exchange during metasomatism in the*  
1353 *Kampos-Lia mélange zone with lithologies representativity.*

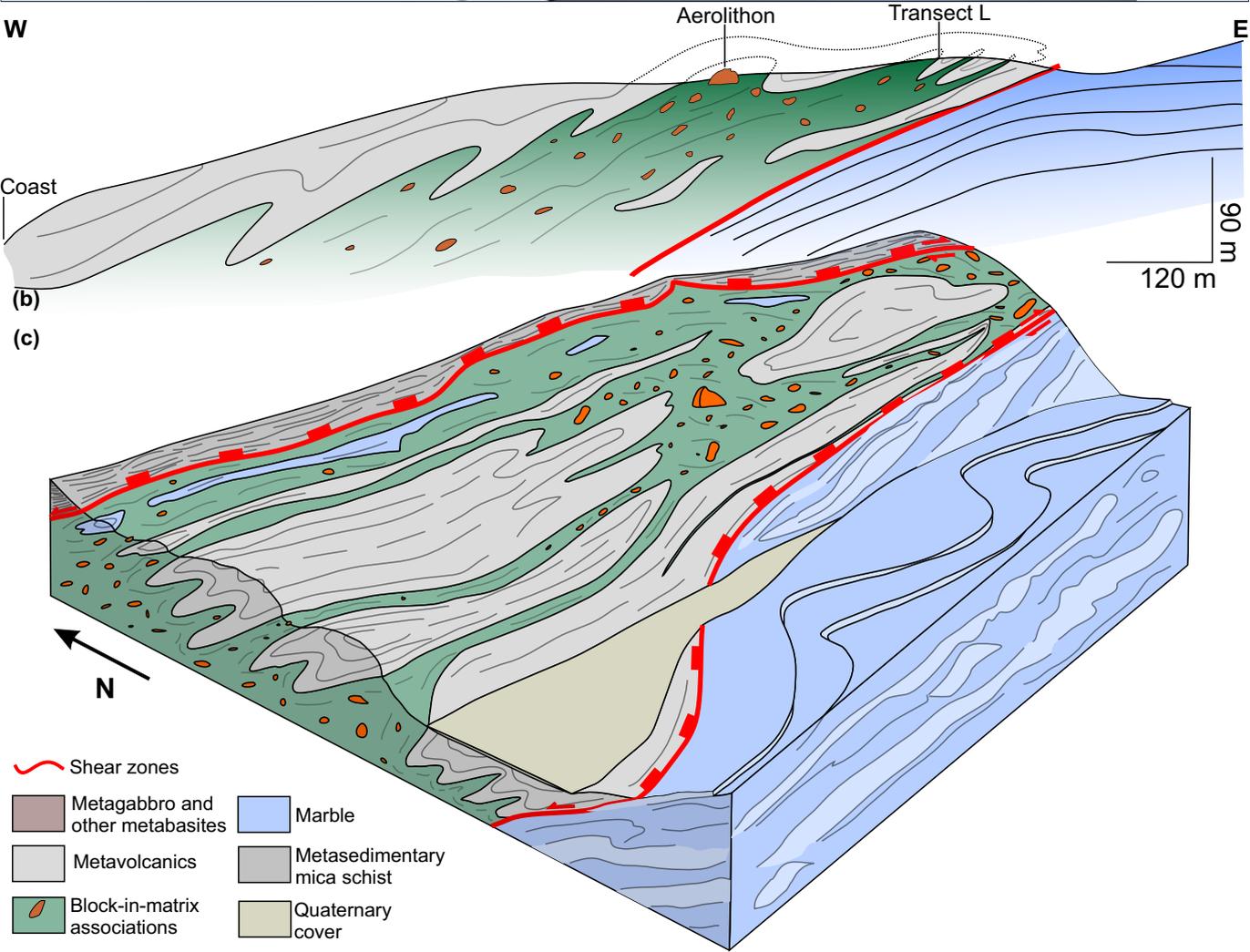
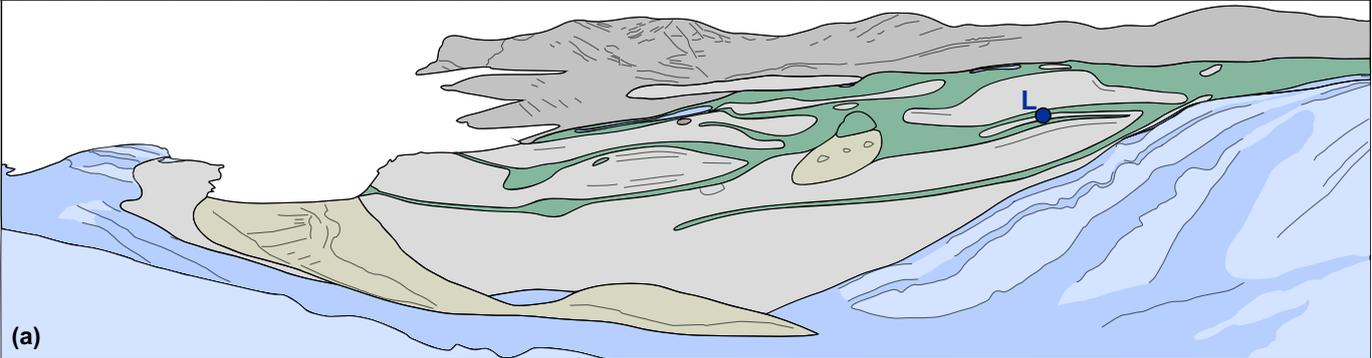
1354 *Figure 17: Three stage conceptual views of the Kampos-Lia mélange zone during the*  
1355 *tectonometamorphic evolution of the CBU*

1356









**Transect L**



p165

p135

p125

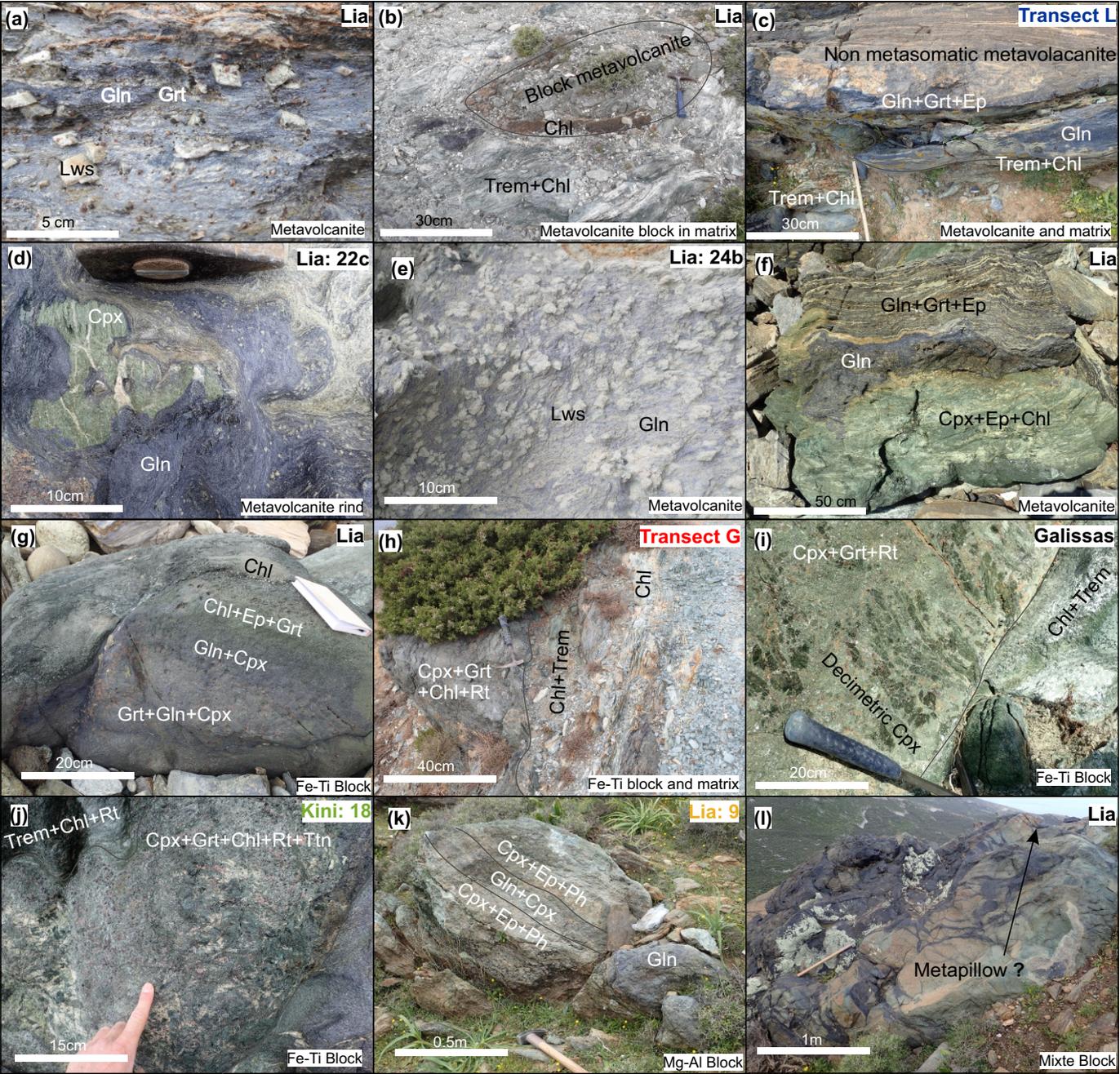
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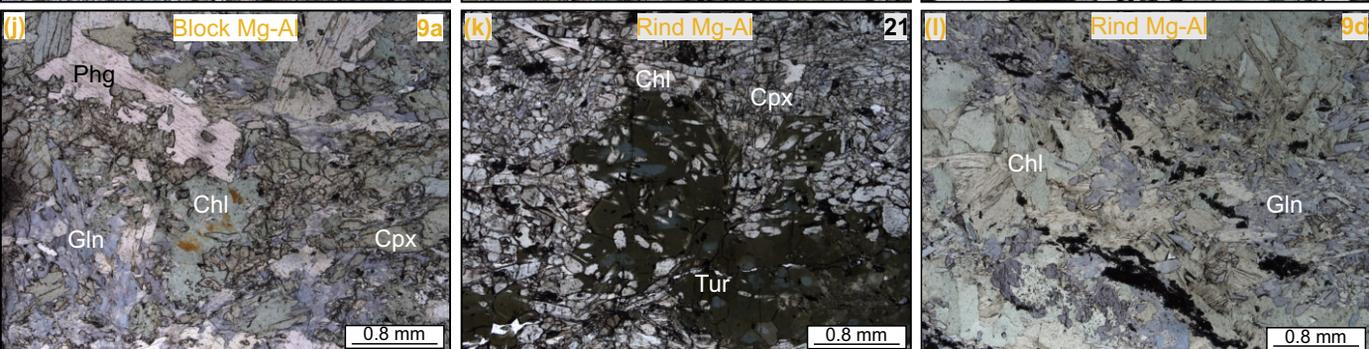
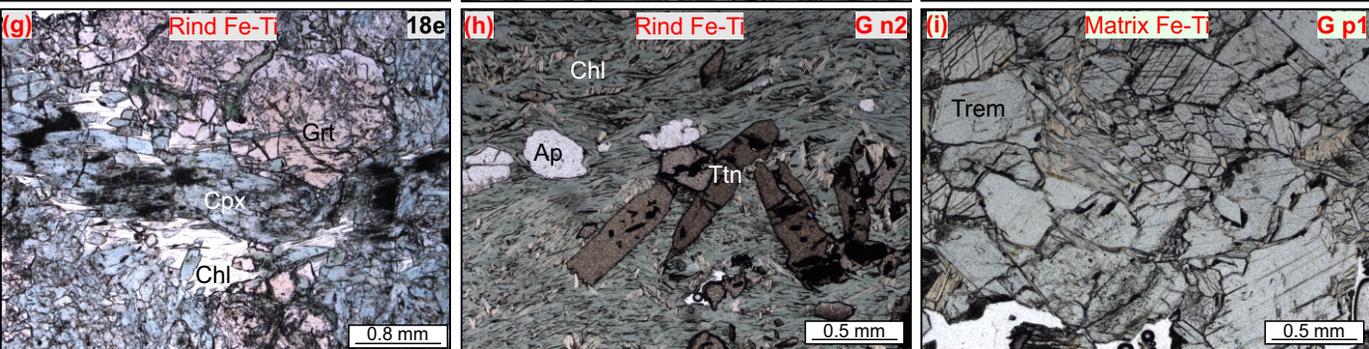
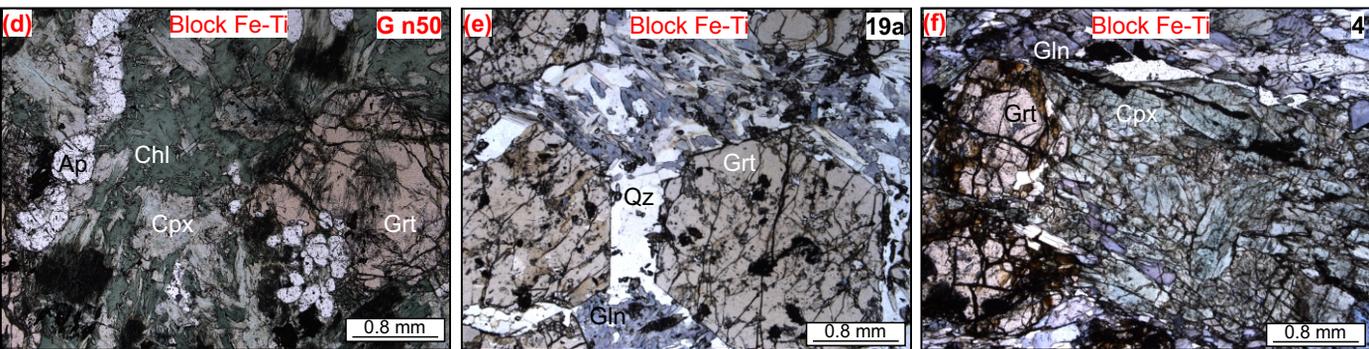
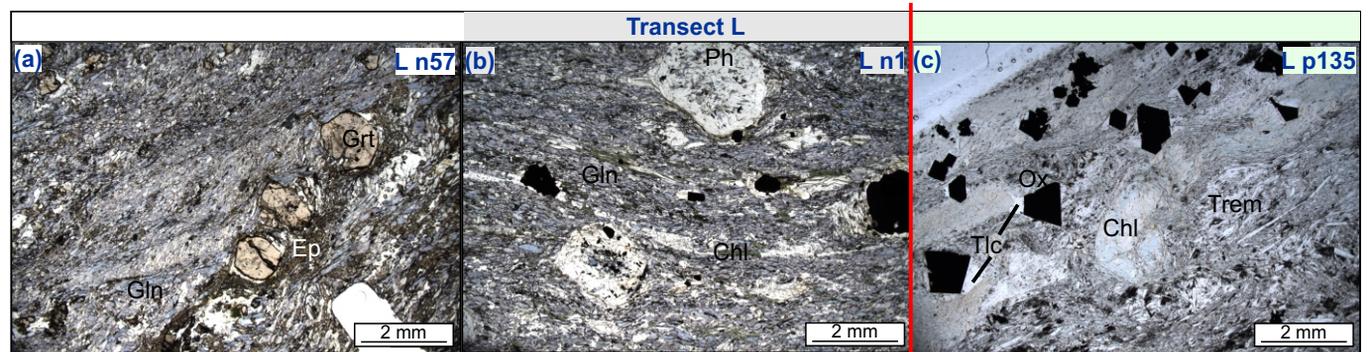
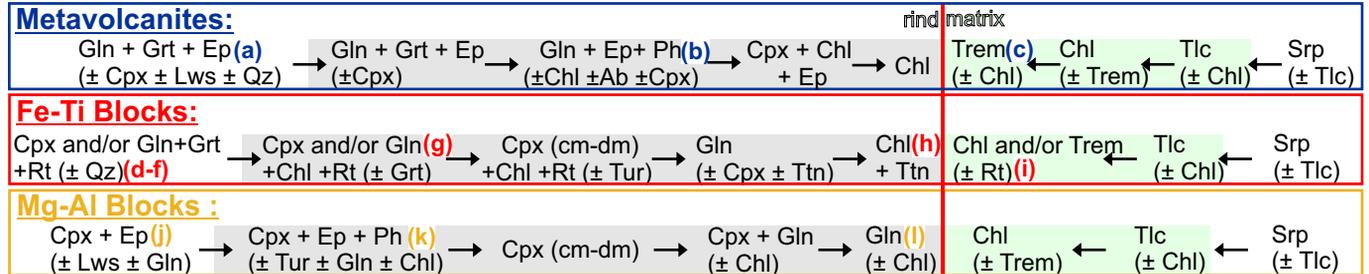
n1

n10

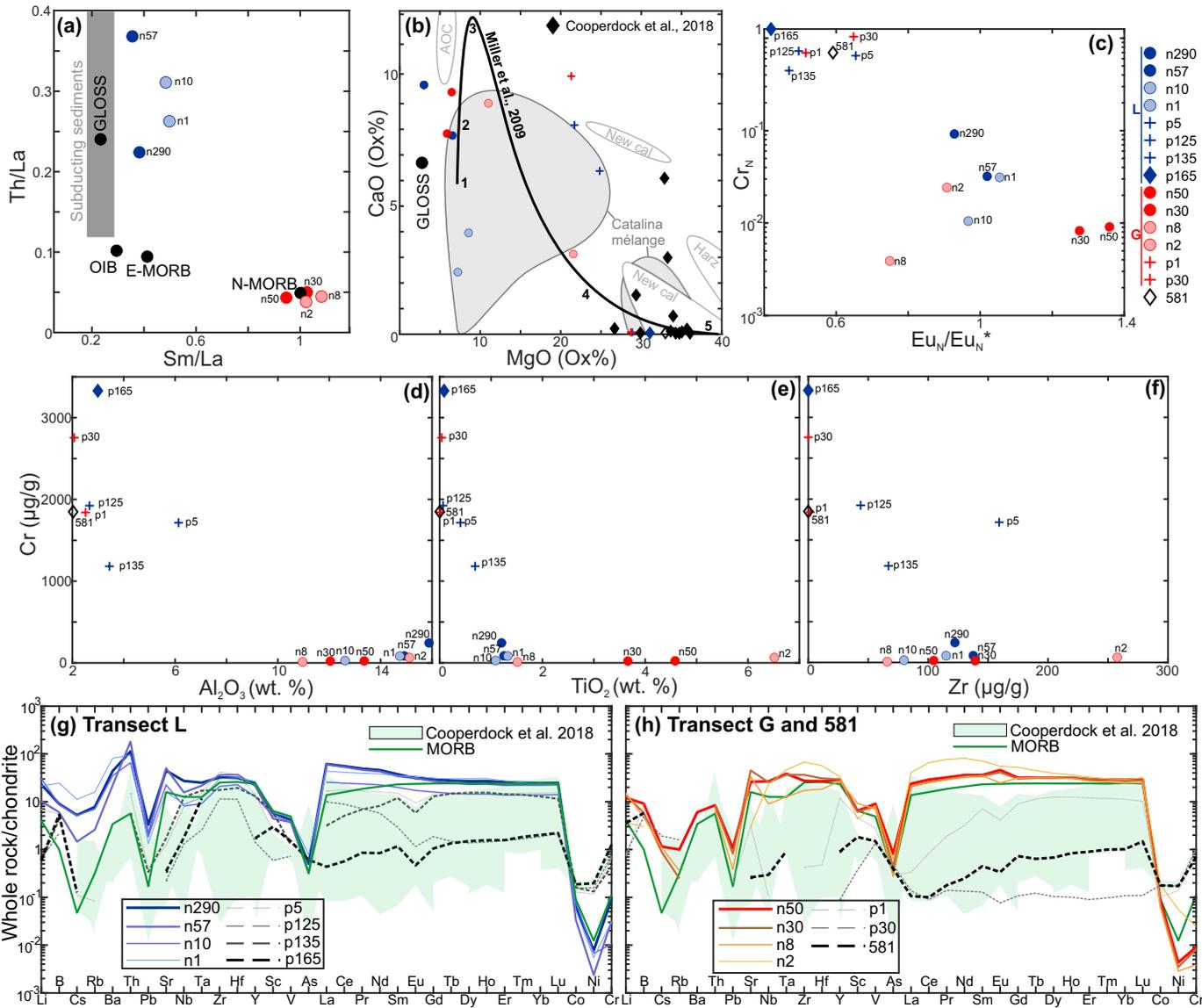
n57

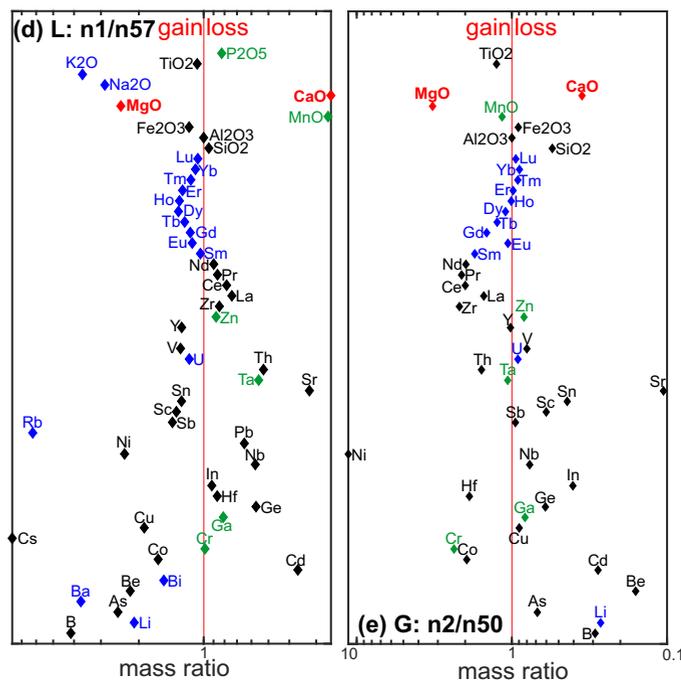
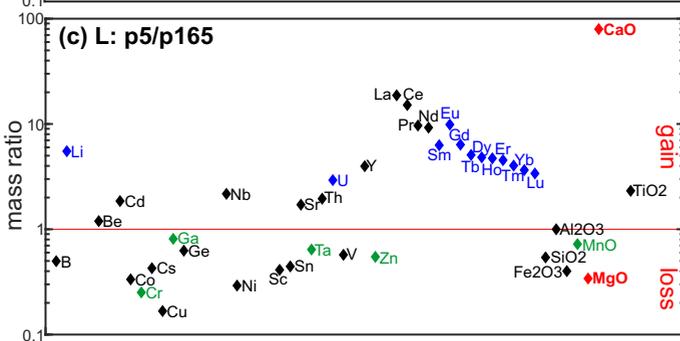
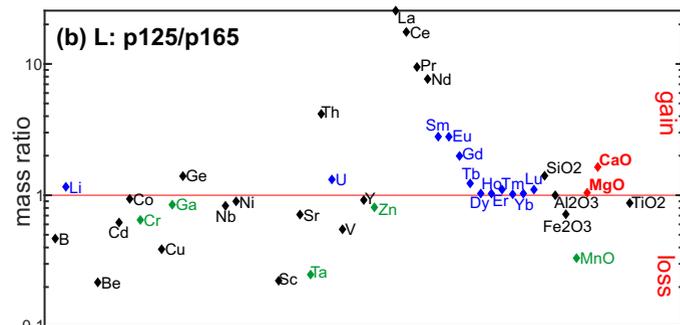
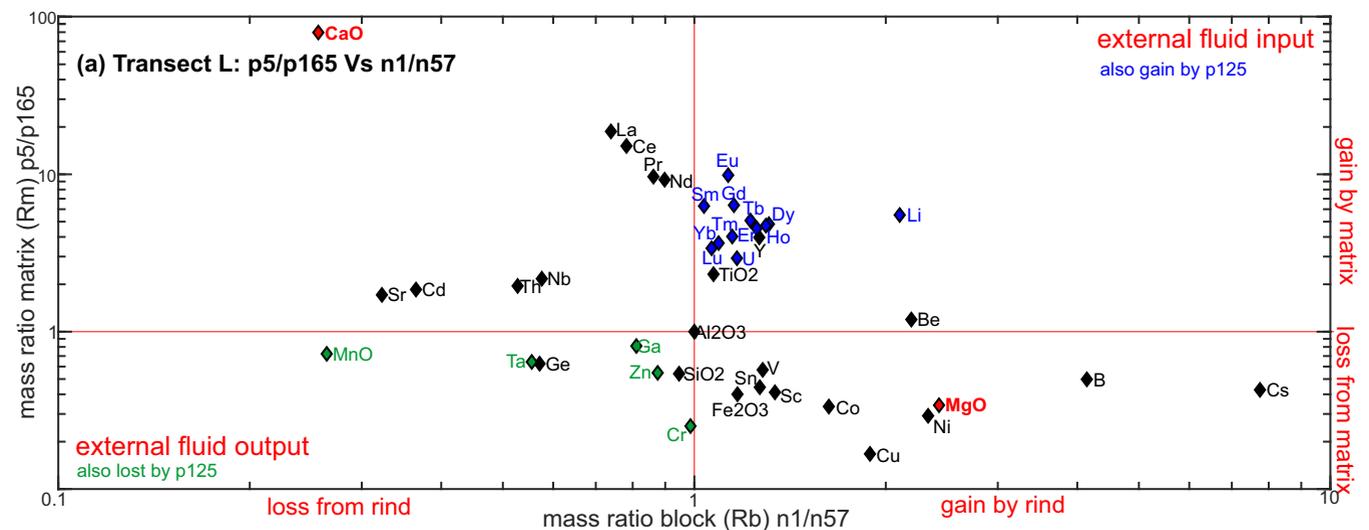
n290

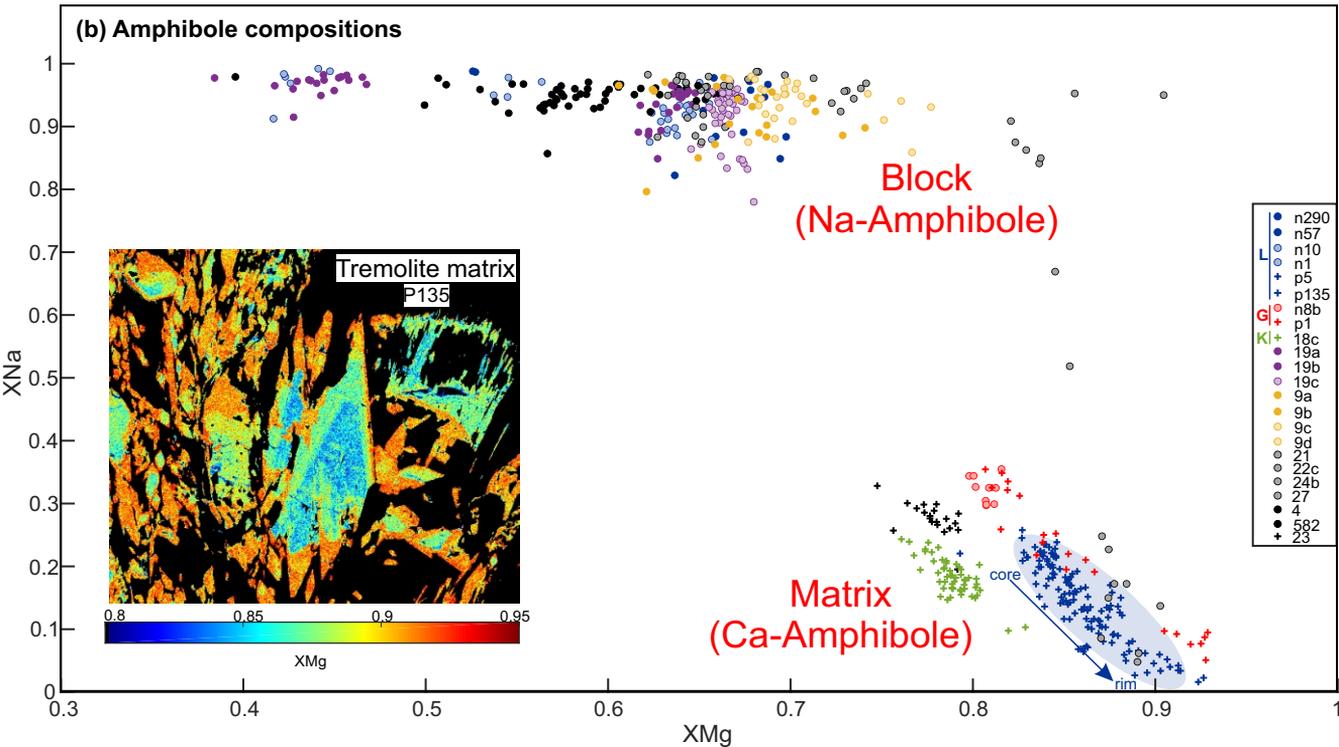
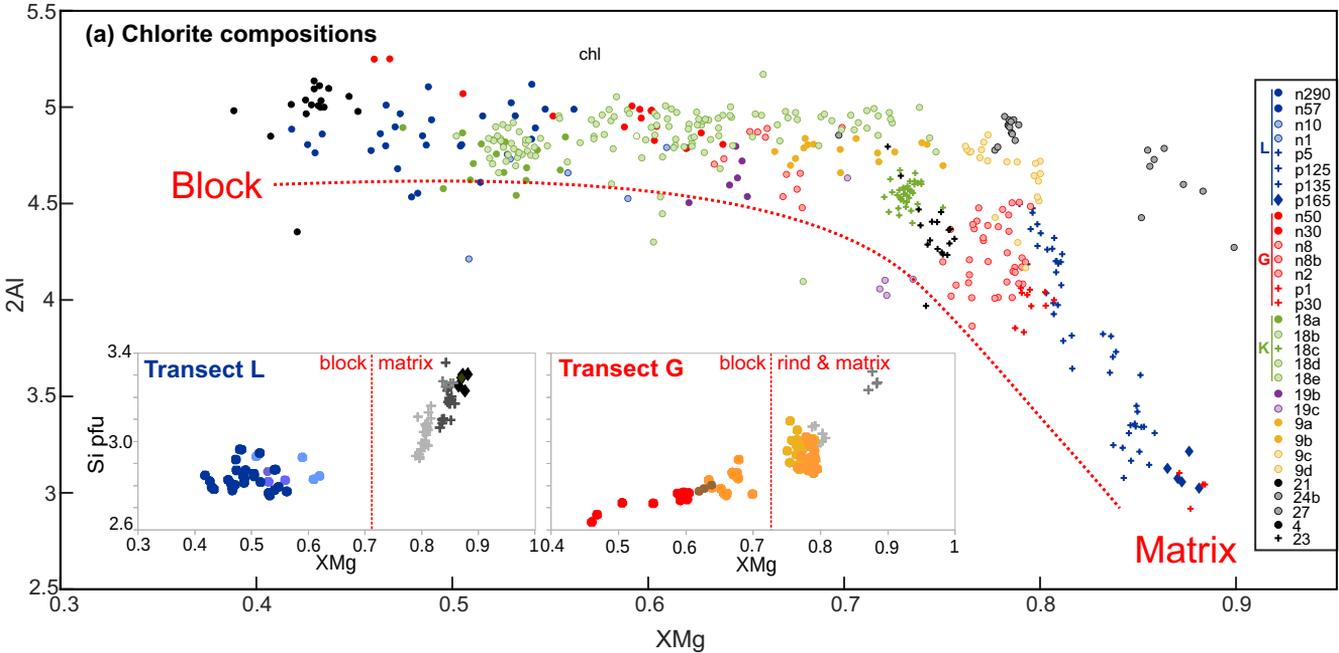


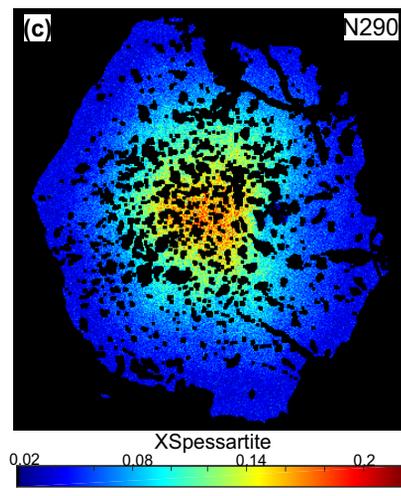
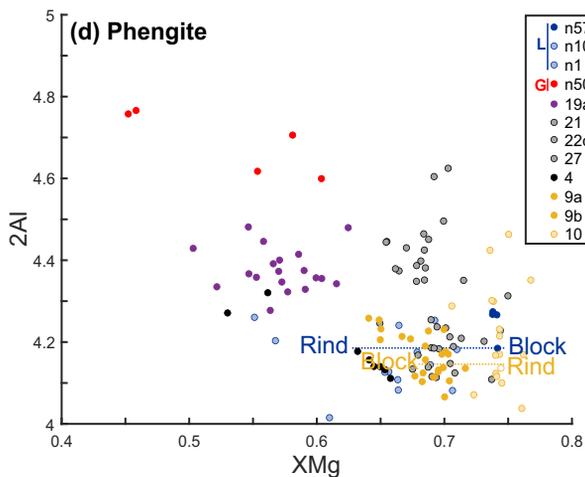
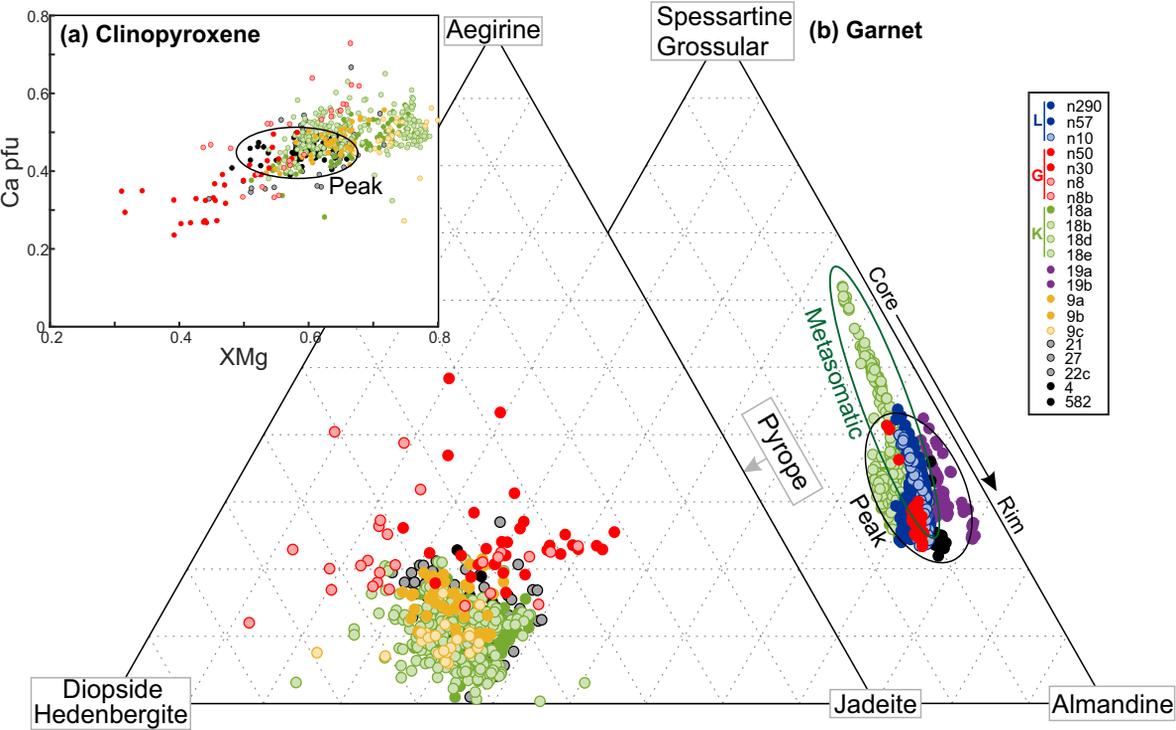




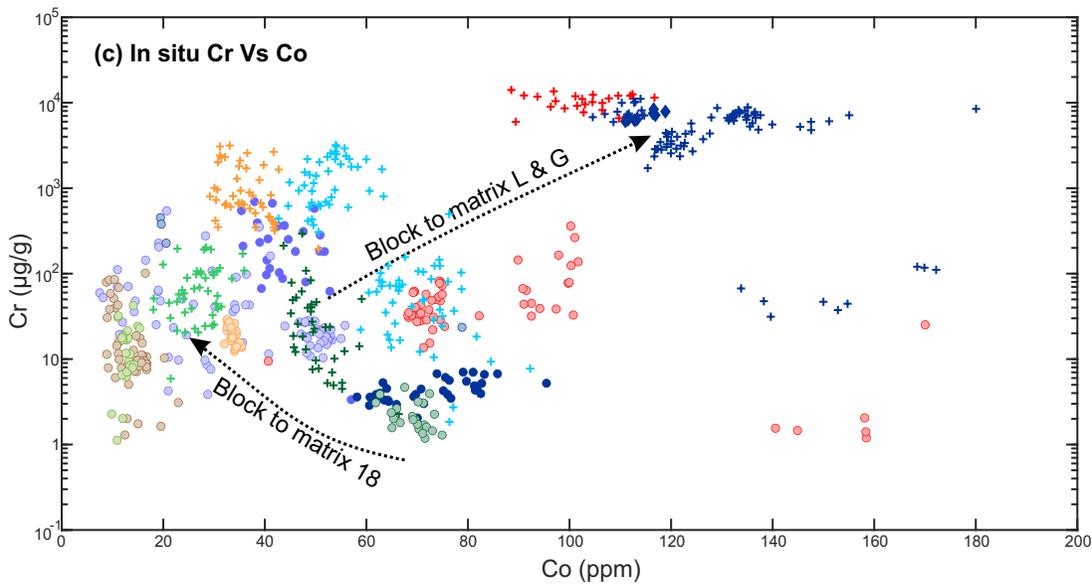
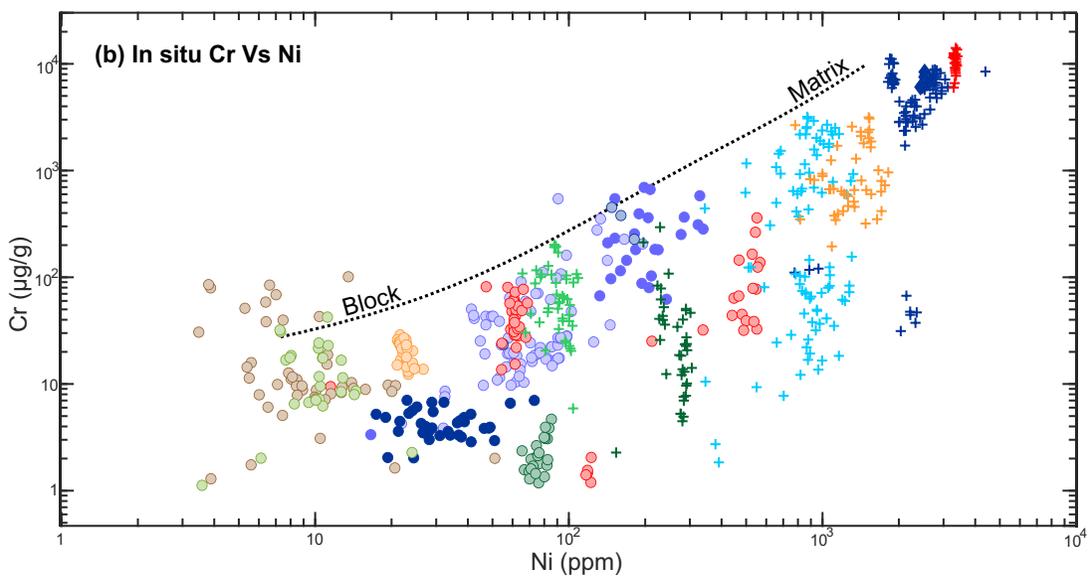
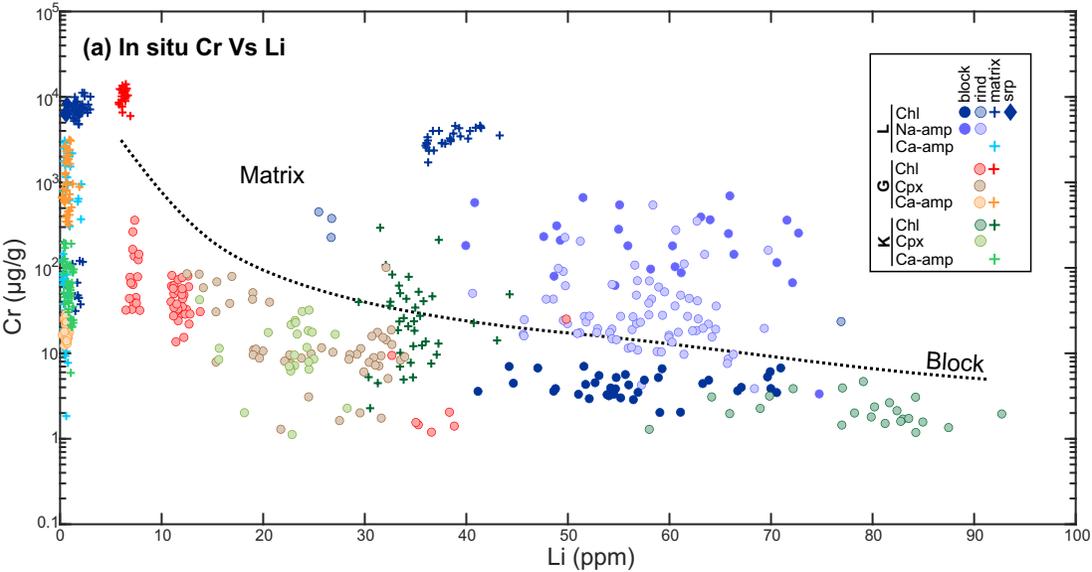


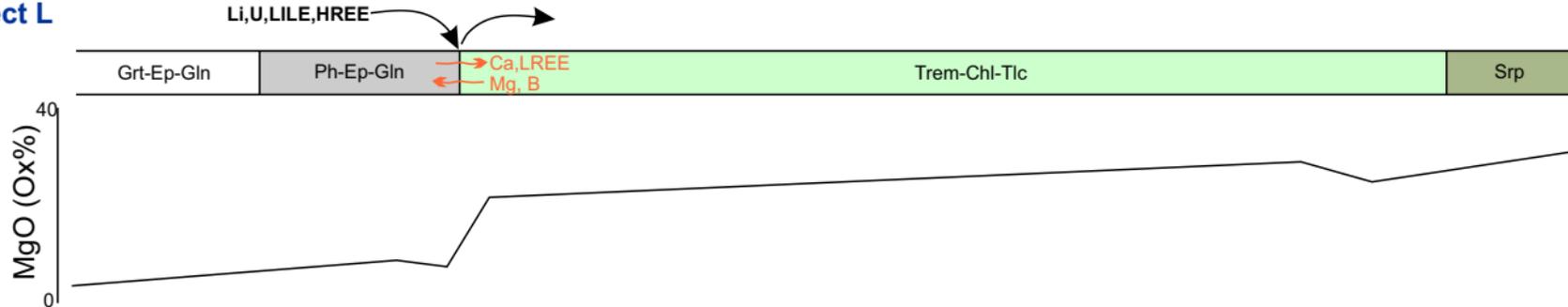
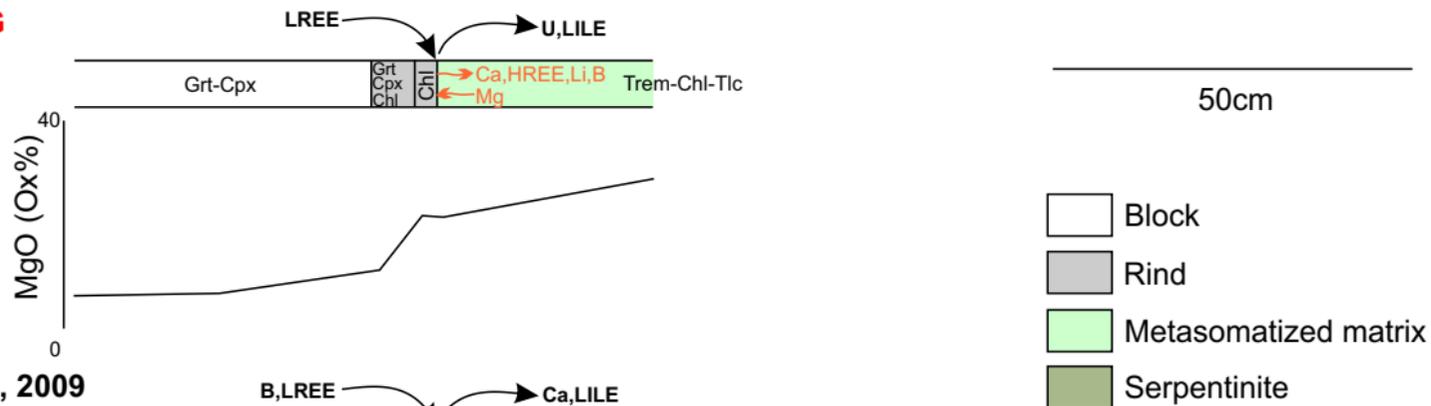
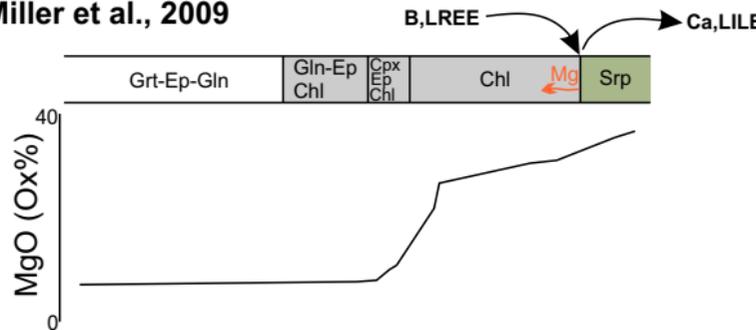




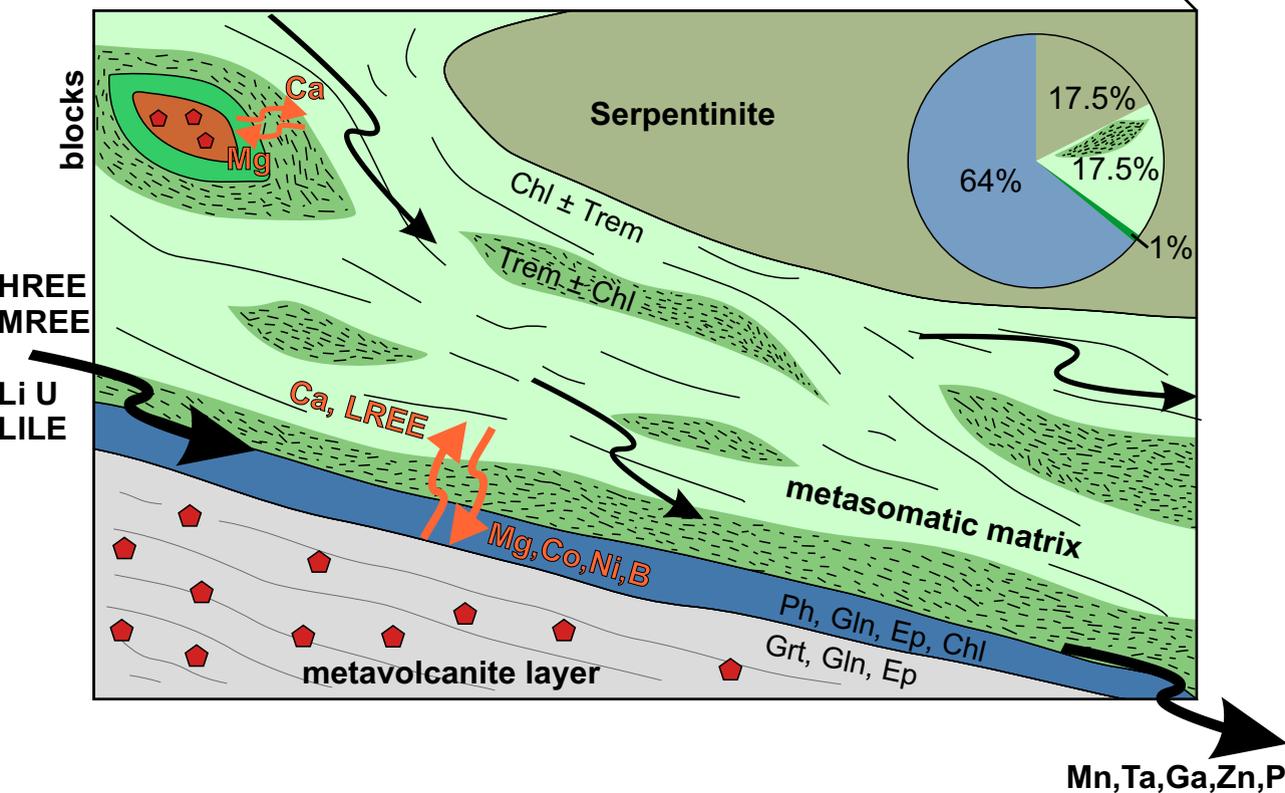
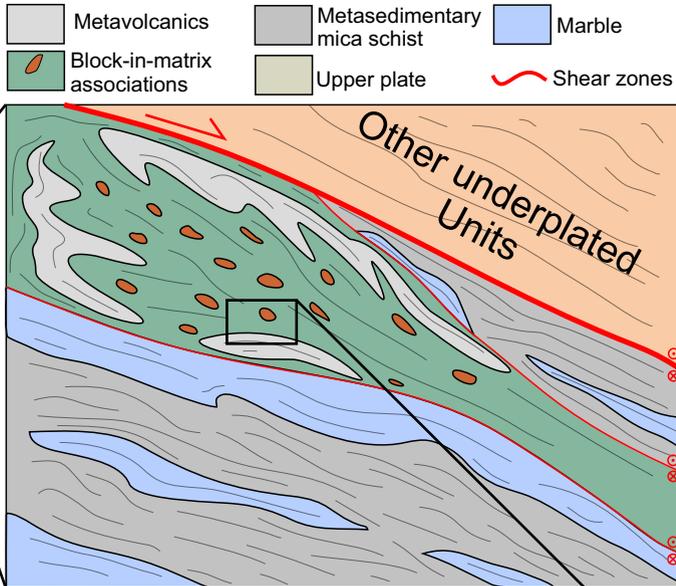
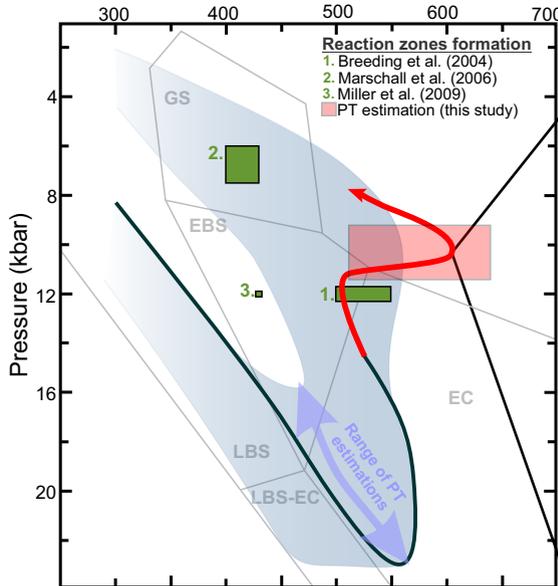




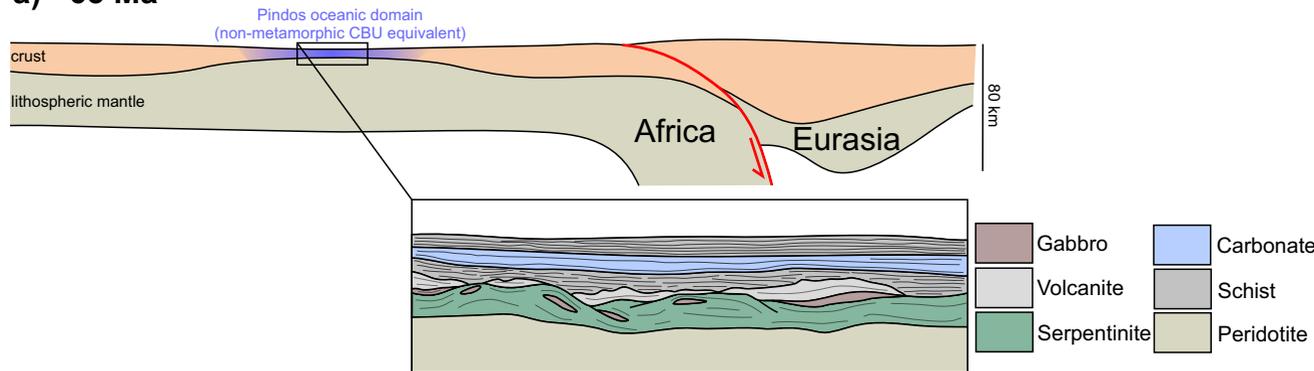


**a) Transect L****b) Transect G****c) Miller et al., 2009**

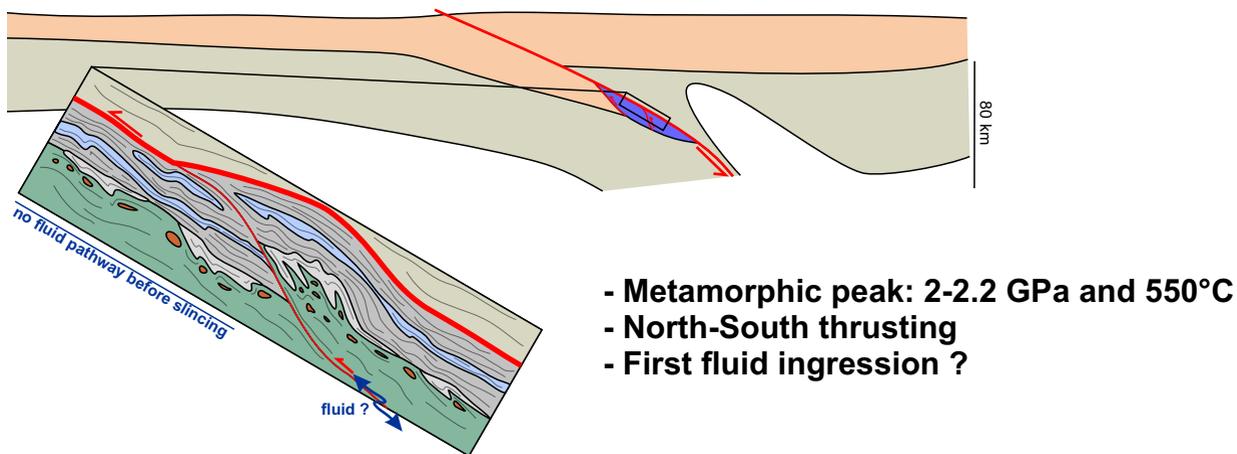
Temperature (°C)



a) ~65 Ma



b) 55-50 Ma: CBU peak burial



c) 30-35 Ma: transition from synorogenic to postorogenic exhumation

