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1 Syn-tectonic, low-temperature meteoric water-derived  
2 carbonation of the New Caledonia Peridotite Nappe

3 **Benoît Quesnel<sup>1\*</sup>, Pierre Gautier<sup>1\*</sup>, Philippe Boulvais<sup>1\*</sup>, Michel Cathelineau<sup>2\*</sup>, Pierre**  
4 **Maurizot<sup>3</sup>, Dominique Cluzel<sup>4</sup>, Marc Ulrich<sup>2,5</sup>, Stéphane Guillot<sup>5</sup>, Stéphane Lesimple<sup>6</sup>, and**  
5 **Clément Couteau<sup>7</sup>**

6 *<sup>1</sup>Géosciences Rennes, Université Rennes 1, UMR 6118 CNRS, 35042 Rennes Cedex, France,*

7 *<sup>2</sup>Georessources, Université de Lorraine, UMR CNRS 7359, CREGU, 54506 Vandoeuvre-lès-*  
8 *Nancy, France*

9 *<sup>3</sup>Bureau de Recherches Géologiques et Minières, 98845 Noumea, Nouvelle-Calédonie*

10 *<sup>4</sup>Pole Pluridisciplinaire de la Matière et de l'Environnement, Université de la Nouvelle-*  
11 *Calédonie, 98851 Nouméa, Nouvelle-Calédonie*

12 *<sup>5</sup>ISTerre CNRS, Université Grenoble 1, 38041, Grenoble, France*

13 *<sup>6</sup>Service Géologique de la Nouvelle-Calédonie, Direction de l'Industrie, des Mines et de*  
14 *l'Energie, 98851 Nouméa, Nouvelle Calédonie*

15 *<sup>7</sup>Service géologique, Koniambo Nickel SAS, 98883 Voh, Nouvelle Calédonie*

16 \*E-mails: benoit.quesnel@univ-rennes1.fr, pierre.gautier@univ-rennes1.fr,

17 philippe.boulvais@univ-rennes1.fr; michel.cathelineau@univ-lorraine.fr;

18 pierre.maurizot@gouv.nc; dominique.cluzel@univ-nc.nc; marc.ulrich@univ-lorraine.fr;

19 Stephane.Guillot@ujf-grenoble.fr; stephane.lesimple@gouv.nc; ccouteau@koniambonickel.nc.

20 **ABSTRACT**

21 Exceptional outcrops recently exposed in the Koniambo Massif allow the study of the  
22 serpentine sole of the Peridotite Nappe of New Caledonia. Many magnesite veins are observed,

23 with characteristics indicating that they were emplaced during pervasive top-to-SW shear  
24 deformation. The oxygen isotope composition of magnesite is homogeneous ( $27.4\text{‰} < \delta^{18}\text{O} <$   
25  $29.7\text{‰}$ ) while its carbon isotope composition varies widely ( $-16.7\text{‰} < \delta^{13}\text{C} < -8.5\text{‰}$ ). These  
26 new data document an origin of magnesite from low temperature meteoric fluids. Laterization on  
27 top of the Peridotite Nappe and carbonation along the sole appear to represent complementary  
28 records of meteoric water infiltration. Based on the syn-kinematic character of magnesite veins,  
29 we propose that syn-laterization tectonic activity has enhanced water infiltration, favoring the  
30 exportation of leached elements like Mg, which led to widespread carbonation along the  
31 serpentine sole. This calls for renewed examination of other magnesite-bearing ophiolites  
32 worldwide in order to establish whether active tectonics is commonly a major agent for  
33 carbonation.

## 34 **INTRODUCTION**

35 Carbonation of ultramafic rocks is the process by which  $\text{CO}_2$ -bearing fluids react with  
36 olivine and/or serpentine to form magnesite ( $\text{MgCO}_3$ ) (e.g., Klein and Garrido, 2011). Based on  
37 stable isotope and structural evidence, Kelemen et al. (2011) recently showed that present day  
38 carbonation of the Oman ophiolite is due to downward infiltration of meteoric waters in the  
39 absence of significant tectonic activity. Other stable isotope studies have also established the  
40 meteoric origin of the fluids from which magnesite has formed in a number of ophiolite  
41 occurrences (Jedrysek and Halas, 1990; Fallick et al., 1991; Gartzos, 2004; Jurkovic et al., 2012).  
42 Some of these ophiolites include laterites and associated iron-nickel ore deposits capping the  
43 ultramafic rocks (e.g., Eliopoulos et al., 2012).

44 The main ophiolite of New Caledonia, referred to as the Peridotite Nappe, has also  
45 undergone intense laterization since its emergence. This has led to supergene nickel ore

46 formation, a process which implies a well drained percolation system through the peridotites  
47 (Trescases, 1975). Recently exposed outcrops in the Koniambo Massif (Fig. 1A) show large  
48 surfaces of the serpentine sole that forms the base of the nappe (Figs. 1B and 2), providing  
49 unprecedented access to fresh samples. Numerous magnesite veins are observed along these  
50 outcrops, attesting for widespread carbonation.

51 Here we present oxygen and carbon isotope compositions of the magnesite veins and  
52 argue that they must originate from meteoric water. Furthermore, in contrast with the situation  
53 depicted in Oman (Kelemen et al., 2011), most veins appear to have formed syntectonically. This  
54 leads us to discuss possible genetic links between laterization, carbonation and tectonics.

## 55 **GEOLOGICAL SETTING**

56 New Caledonia lies 2000 km east of Australia, in the SW Pacific. About 40% of the  
57 island's surface consists of peridotite. Peridotites overlie rock units of the Norfolk Ridge  
58 microcontinent with a sub-horizontal contact materialized by a strongly deformed serpentine sole  
59 (Avias, 1967). This geometry results from the southwestward obduction of the Peridotite Nappe,  
60 initially rooted in the Loyalty Basin, sometimes between ~37 and 27 Ma (Cluzel et al., 2001,  
61 2012; Paquette and Cluzel, 2007).

62 On top of the nappe, laterites have developed at the expense of the peridotites (Trescases,  
63 1975). Several planation surfaces attest for distinct episodes of weathering during the Neogene  
64 and probably the Oligocene (Latham, 1986; Chevillotte et al., 2006; Sevin et al., 2012). This is  
65 consistent with biogeographic and phylogenetic studies indicating that New Caledonia was  
66 already emerged in the Late Oligocene (Grandcolas et al., 2008).

67 Magnesite is widespread in New Caledonia and occurs as veins within the serpentine sole  
68 of the Peridotite Nappe, and as nodular heaps in recent alluvial deposits and present-day soils.

69 Since Glasser (1904), the origin of the veins is supposed supergene, possibly linked to the  
70 laterization process.

## 71 **OBSERVATIONS AND SAMPLING**

72 The Koniambo Massif is one of the klippe of the Peridotite Nappe located along the  
73 West Coast (Fig. 1A). Recently, Koniambo Nickel SAS initiated a large industrial site for nickel  
74 production. As a result, new outcrops of exceptional quality and size have been created in the  
75 serpentine sole of this massif (Figs. 1B and 2).

76 In the serpentine sole, rocks are highly deformed, either finely schistose and/or intensely  
77 brecciated. A dense network of meter-thick shallow-dipping shear zones attests for pervasive  
78 non-coaxial deformation with a top-to-SW sense of shear. Magnesite occurs as veins, up to ~30  
79 cm thick and irregularly distributed. Two main vein types are recognized (Figs. 3A, 3B and 3C,  
80 and Figs. A and B in Data Repository). Type 1 veins are located within or along the margins of  
81 the main shear zones. Open to tight asymmetric folding of some of these veins indicates that they  
82 formed during, or possibly before, shearing. Type 2 veins are steeper and occasionally crosscut  
83 by the shallow-dipping shear zones, demonstrating they do not represent younger structures. The  
84 obliquity of these veins with respect to the shear zones (Fig. 3D) and the local occurrence of  
85 magnesite as coarse fibers orthogonal to vein walls (Fig. 3C) are consistent with their  
86 interpretation as tension gashes opened during top-to-SW shearing.

87 Both vein types have been sampled along two cross sections (BMS and VAV) located in  
88 the Vavouto peninsula, just above the basal contact of the Peridotite Nappe (Fig. 1B, Table  
89 DR1). Samples were also collected in other highly serpentized zones, in the Koniambo Massif  
90 (sample CONV, Fig. 1B) and in the Kopeto Massif, ~50 km to the south-east (samples NEP and  
91 GAIACS). In addition, sample DECH was collected from a series of nodular heaps in a soil. The

92 large size of the heaps (~15–30 cm) and their isolated character within an abundant and  
93 homogeneous clayey matrix suggest that they formed in situ in the soil (i.e., they are unlikely to  
94 represent reworked pebbles).

#### 95 **CARBON AND OXYGEN ISOTOPE DATA**

96 Isotopic analyses were performed at the stable isotope laboratory of the University of  
97 Rennes 1. Samples were finely crushed in a boron carbide mortar and reacted with anhydrous  
98 phosphoric acid at 75 °C for 24 h. The liberated CO<sub>2</sub> was analyzed on a VG SIRA 10 triple  
99 collector mass spectrometer. The experimental fractionation coefficient between magnesite and  
100 CO<sub>2</sub> is  $\alpha_{\text{CO}_2\text{-Magnesite}} = 1.009976$  at 75 °C (Das Sharma et al., 2002). In the absence of a magnesite  
101 standard, in-lab calcite standard samples were analyzed together with the magnesite samples  
102 under identical conditions in order to control the general reliability of the protocol. The  
103 analytical uncertainty is estimated at 0.3‰ for O and 0.2‰ for C.

104 Results are presented in Figure 4 and Table DR1. All magnesite samples display  
105 comparable and high oxygen isotope values, irrespective of their structural position or sampling  
106 site ( $27.4\text{‰} < \delta^{18}\text{O} < 29.7\text{‰}$ ); the carbon isotope composition is highly variable and negative  
107 ( $-16.7\text{‰} < \delta^{13}\text{C} < -8.5\text{‰}$ ). Focusing on the samples from the Vavouto peninsula (BMS and  
108 VAV), the  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values do not show any correlation.

#### 109 **METEORIC ORIGIN OF CARBONATION**

110 The isotopic compositions of magnesite reflect the conditions at which fluid/rock  
111 interactions occurred. As the  $\delta^{18}\text{O}$  values are homogeneous, the physical conditions of  
112 carbonation were constant, including water origin and temperature. In addition, the large spread  
113 of  $\delta^{13}\text{C}$  values indicates at least two sources of carbon. Based on the arguments to follow, we  
114 suggest that a strong interaction with meteoric fluids is the most likely process that led to

115 carbonation along the serpentine sole. Firstly, the largely negative  $\delta^{13}\text{C}$  values rule out marine  
116 water as  $\delta^{13}\text{C}$  values would be centered around 0‰. Secondly, in New Caledonia, the oxygen  
117 isotope composition of serpentine minerals is between 1.7 and 13.9‰ (Ulrich, 2010). If  
118 magnesite had precipitated in equilibrium with these serpentines at 200 °C (a minimum  
119 temperature for serpentine formation; e.g., O’Hanley and Wicks, 1995), it would display  $\delta^{18}\text{O}$   
120 values between 12.2 and 24.4‰ (Fig. 5). The range would be displaced toward even lower  $\delta^{18}\text{O}$   
121 values if higher temperatures were considered. Given the observed values are higher, we  
122 conclude that serpentinization and carbonation were not synchronous, even though serpentine  
123 and the magnesite veins share a common structural record of top-to-SW shearing. Therefore, at  
124 least part of the pervasive deformation observed along the sole post-dates serpentinization, a fact  
125 consistent with the mechanical softness of serpentine (e.g., Byerlee, 1978). We also considered  
126 the  $\delta^{18}\text{O}$  value of magnesite expected if formed at near-surface temperatures from meteoric water  
127 (Fig. 5). To date, no isotopic data is available for meteoric precipitations in New Caledonia. A  
128  $\delta^{18}\text{O}$  range between  $-1$  and  $-7$ ‰ is used, which corresponds to values of meteoric precipitations  
129 on isolated islands at inter-tropical latitude and low elevation. We obtain a theoretical  $\delta^{18}\text{O}$  range  
130 for the corresponding meteoric-derived magnesite that compares well with our data (Fig. 5).

131 Three independent observations confirm the meteoric origin of the fluids that led to  
132 carbonation. Firstly, the magnesite sampled in a soil (DECH) has isotopic compositions  
133 comparable to the magnesite veins from the serpentine sole. Secondly, the highest  $\delta^{13}\text{C}$  values  
134 from the veins are close to  $-8$ ‰, which coincides with the expected value for carbonates in  
135 which carbon is taken from the atmosphere (as illustrated by sample DECH). Additionally, the  
136 lowest  $\delta^{13}\text{C}$  values, around  $-15$ ‰, point to a biogenic contribution, either from surface soils, or  
137 from a deep-seated carbon source such as biogenic methane liberated from sediments buried

138 below the Peridotite Nappe. Part of the low  $\delta^{13}\text{C}$  signal may also reflect magnesite precipitation  
139 from high-pH fluids (e.g., Fourcade et al., 2007), which are commonly associated with  
140 carbonation of ultramafic rocks (Jurkovic et al., 2012). Thirdly, in Figure 4, we compare our data  
141 set with data from the literature on magnesite veins hosted by ultramafic rocks and for which a  
142 meteoric origin of the fluids has been proposed. Our data show a more restricted range in  $\delta^{18}\text{O}$   
143 values centered on the right side of the literature data cloud. This observation may reflect a  
144 difference in the initial  $\delta^{18}\text{O}$  value of rain water due to distinct paleogeographic position.

#### 145 **LINKS BETWEEN LATERIZATION, CARBONATION AND TECTONICS**

146 The record of meteoric waters through carbonation along the serpentine sole implies that  
147 water circulated downward through the peridotite pile. An efficient drainage system has likely  
148 been provided by the dense network of fractures that characterizes the New Caledonia  
149 peridotites. This network is also recognized to have played a major role in peridotite weathering  
150 and the distribution of nickel ore (e.g., Leguéré, 1976). Hence, laterization on top of the  
151 Peridotite Nappe and carbonation along the serpentine sole may correspond to complementary  
152 records of meteoric water infiltration, as anticipated by Glasser (1904). In practice, laterization  
153 involves the leaching of large amounts of magnesium, a highly mobile ion that can be viewed as  
154 a tracer of fluid circulation from the surface down to the serpentine sole where it precipitated to  
155 form magnesite veins. Correlatively, nickel, which is less mobile, has accumulated at the base of  
156 the lateritic profile (e.g., Trescases, 1975). Throughout New Caledonia, the richest nickel ores  
157 are associated with a couple of planation surfaces associated with laterites up to 30 m thick (e.g.,  
158 Chevillotte et al., 2006). They cap the Koniambo Massif at elevations between ~400 and ~800 m,  
159 in agreement with the island-scale mean elevation of 640 m reported by Chevillotte et al. (2006).  
160 The outcrops of the Vavouto peninsula, where most of our magnesite samples come from, lie



161 near sea level, and so does the basal contact of the Peridotite Nappe around much of the  
162 Koniambo Massif. Therefore, downward infiltration of meteoric waters likely occurred across a  
163 vertical distance of at least ~600 m before magnesite formed along the serpentine sole. Greater  
164 vertical distances are also possible since older laterite-bearing planation surfaces are locally  
165 preserved at elevations up to ~1250 m in the nearby Kopeto Massif and in southern New  
166 Caledonia (Latham, 1986; Chevillotte et al., 2006).

167       Highly efficient drainage of meteoric water through the peridotite pile is attested for by  
168 the low temperature conditions at which magnesite formed along the serpentine sole. Figure 5  
169 shows that a temperature of 25 °C fits well the data whereas 35 °C is hardly compatible. The  
170 minimum mean temperature at which laterites form is ~25 °C. Using 15 °C/km as a lower bound  
171 for the geothermal gradient in the Peridotite Nappe, surface water penetrating to depths of at  
172 least ~600 m would have reached temperatures of at least 34 °C if in thermal equilibrium with  
173 the host rocks. Our data are inconsistent with this hypothesis; instead, they imply rapid  
174 infiltration of meteoric water as far down as the serpentine sole. This is consistent with abundant  
175 morphological evidence documenting karst structures developed during laterization (Trescases,  
176 1975; Latham, 1986).

177       Because active slip typically increases the permeability of faults, water drainage through  
178 the peridotites could have been strongly enhanced during active faulting. Indirect evidence for  
179 deformation-assisted fluid circulations across the peridotite pile is provided by the syn-kinematic  
180 character of the studied magnesite veins along the sole (Fig. 3, see also Data Repository) and the  
181 observation that at least some of the nickel mineralizations underlying laterites developed during  
182 brittle deformation (Cluzel and Vigier, 2008; our own observations in the Koniambo Massif).

183 Most magnesite veins of the Koniambo Massif have been emplaced during top-to-SW  
184 shearing deformation. Southwestward shearing recorded along the basal contact of the Peridotite  
185 Nappe may reflect obduction (e.g., Cluzel et al., 2001) or post-obduction reactivation of the  
186 contact as a SW-dipping extensional detachment (Lagabrielle and Chauvet, 2008). Deformation  
187 occurred sometimes between ~37 and 27 Ma if related to obduction, or later, but before ~20 Ma,  
188 if related to post-obduction NE-SW extension (Chardon and Chevillotte, 2006). The main  
189 laterites of New Caledonia were also formed before ~20 Ma (Chevillotte et al., 2006; Sevin et  
190 al., 2012). Hence, available time constraints make it possible that carbonation and laterization  
191 occurred at the same time, during tectonic activity.

192 As a result, considering the further requirement of fast downward circulation of surface  
193 waters for maintaining low temperature conditions during magnesite formation, we propose that  
194 syn-laterization tectonic activity enhanced water infiltration and played a major role in the  
195 exportation of leached elements like Mg, leading to widespread carbonation along the serpentine  
196 sole.

#### 197 **POTENTIAL IMPLICATIONS FOR OTHER CARBONATED OPHIOLITES**

198 Syn-tectonic carbonation along the serpentine sole of the New Caledonia ophiolite  
199 contrasts directly with the well documented case of post-tectonic subsurface carbonation of the  
200 Oman ophiolite (Kelemen et al., 2011). Studies documenting meteoric water-derived magnesite  
201 in other ophiolite occurrences lack a structural description that would allow the syn- versus post-  
202 tectonic character of carbonation to be evaluated (Jedrysek and Halas, 1990; Fallick et al., 1991;  
203 Gartzos, 2004; Jurkovic et al., 2012). Nevertheless, syn-laterization tectonically-driven  
204 carbonation of ultramafic rocks, as proposed here for New Caledonia, may concern other areas  
205 worldwide. For instance, the ophiolites of the Dinaric-Hellenic segment of the Alpine orogen

206 include (i) large volumes of magnesite originated from meteoric water (e.g., Gartzos, 2004;  
207 Jurkovic et al., 2012), (ii) laterites capping the ultramafic rocks, with iron-nickel ore deposits  
208 (e.g., Eliopoulos et al., 2012) (iii) various time constraints showing that obduction occurred in  
209 the Late Jurassic, (iv) the unconformity of Late Jurassic sediments on at least some of the  
210 laterites (Robertson et al., 2012). These features strongly suggest that laterization occurred  
211 during obduction, which opens the possibility that carbonation occurred simultaneously, fostered  
212 by tectonic activity. This pleads for renewed examination of the Dinaric-Hellenic and other  
213 carbonated ophiolites worldwide in order to establish whether active tectonics is commonly a  
214 major agent for carbonation.

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### 311 **FIGURE CAPTIONS**

312 Figure 1. A) Simplified geological map of New Caledonia. B) Geological map of the  
313 southwestern margin of the Koniambo Massif, adapted from Maurizot et al. (2002). BMS, VAV  
314 and CONV indicate magnesite sampling sites. Laterites shown on this map belong to a planation  
315 surface that is younger than those having led to much thicker laterites at higher levels of the  
316 Koniambo Massif (at elevations between ~400 and ~800 m, see the text) (Latham, 1986;  
317 Chevillotte et al., 2006; Chardon and Chevillotte, 2006).

318 Figure 2. Field view along part of the ‘BMS’ cross-section (located in Fig. 1B), illustrating the  
319 exceptional size and freshness of the outcrops recently opened in the serpentine sole of the  
320 Koniambo Massif.

321 Figure 3. Field observations within the serpentine sole of the Koniambo Massif. A, B, C, field  
322 views illustrating the relations between deformation and the emplacement of magnesite veins.  
323 Numbers 1 and 2 refer to the two main vein types as described in the text. The two ellipses in B  
324 show sites where tight folding of a ‘type 1’ vein is well visible. In C, magnesite occurs as coarse  
325 fibers suborthogonal to the walls of this ‘type 2’ vein (sample BMS Gio 9). D, stereogram  
326 showing the orientation, along the ‘BMS’ cross-section, of major shear zones (shown as poles of  
327 planes) and of magnesite veins occurring in between the shear zones (‘type 2’ veins, shown as  
328 great circles).

329 Figure 4.  $\delta^{13}\text{C}$  versus  $\delta^{18}\text{O}$  diagram of magnesite veins hosted by ultramafic rocks in different  
330 regions worldwide. Black and gray symbols are from this study and the literature, respectively.

331 **[[Spell out versus or place period after vs. abbreviation in figure (Figs. 4 and 5). Citations  
332 should be: Fallick et al., 1991; Jurkovic et al., 2012; Jedrysek and Halas, 1990.]]**

333 Figure 5. Histogram of oxygen isotopic compositions of magnesite. In black, data from this  
334 study. In gray, composition range of theoretical magnesite that would develop in equilibrium  
335 with serpentines, using the serpentine  $\delta^{18}\text{O}$  data of Ulrich (2010). The dashed line shows the  
336 theoretical composition range of magnesite that would form at near-surface temperature from  
337 South-Pacific meteoric waters. The theoretical compositions are calculated using the serpentine-  
338  $\text{H}_2\text{O}$  and magnesite- $\text{H}_2\text{O}$  fractionation coefficients of Zheng (1993, 1999).



339 <sup>1</sup>GSA Data Repository item 2013xxx, xxxxxxxx, is available online at  
340 [www.geosociety.org/pubs/ft2013.htm](http://www.geosociety.org/pubs/ft2013.htm), or on request from [editing@geosociety.org](mailto:editing@geosociety.org) or Documents  
341 Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

342 **[[Please delete text below and insert into separate supplemental file.]]**

343 **SUPPLEMENTARY MATERIAL**

344 Table DR1. O and C isotope compositions of magnesite samples from this study. The  
345 results are given in ‰ versus SMOW for  $\delta^{18}\text{O}$  and ‰ versus PDB for  $\delta^{13}\text{C}$ .

346 Figure DR1. Additional field views in the serpentine sole of the Koniambo Massif. A, a  
347 low-dipping shear zone and associated ‘type 1’ magnesite veins. In this example, three subtypes  
348 of vein may be distinguished: (i) a planar vein running along, and parallel to, the roof of the  
349 shear zone, (ii) subplanar veins inside, and oblique to, the shear zone, representing Riedel- or C’-  
350 type shear planes, (iii) a folded composite vein inside, and broadly parallel to, the shear zone.  
351 Folding seems to result from both buckling and drag folding along the shear planes. B, a  
352 mylonite zone hosting ‘type 1’ magnesite veins. A first subtype consists of sheared  
353 (boudinaged?) veins paralleling the schistosity. A second subtype consists of thinner veins along  
354 Riedel- or C’-type shear planes.  
355