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► **To cite this version:**

Rémi Coltat, Philippe Boulvais, Yannick Branquet, Antonin Richard, Alexandre Tarantola, et al.. Moho carbonation at an ocean-continent transition. *Geology*, 2022, 50 (3), pp.278-283. 10.1130/G49363.1 . insu-03469219

HAL Id: insu-03469219

<https://insu.hal.science/insu-03469219v1>

Submitted on 10 Dec 2021

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1 Moho carbonation at an ocean-continent transition

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12 **ABSTRACT**

13 Carbonation of mantle rocks during mantle exhumation is reported in present-day
14 oceanic settings, both at mid-ocean ridges and ocean-continent transitions (OCTs).
15 However, the hydrothermal conditions of carbonation (i.e., fluid sources, thermal
16 regimes) during mantle exhumation remain poorly constrained. We focus on an
17 exceptionally well-preserved fossil OCT where mantle rocks have been exhumed and
18 carbonated along a detachment fault from underneath the continent to the seafloor along a
19 tectonic Moho. Stable isotope (oxygen and carbon) analyses on calcite indicate that
20 carbonation resulted from the mixing between serpentinization-derived fluids at ~175 °C
21 and seawater. Strontium isotope compositions suggest interactions between seawater and
22 the continental crust prior to carbonation. This shows that carbonation along the tectonic

23 Moho occurs below the continental crust and prior to mantle exhumation at the seafloor
24 during continental breakup.

25 INTRODUCTION

26 At magma-poor ocean-continent transitions (OCTs), mantle exhumation occurs
27 along detachment faults (e.g., the Iberia margin; Boillot et al., 1987), in a comparable
28 way to (ultra)slow spreading ridges (Tucholke et al., 1998; Cannat et al., 2006).
29 Associated hydrothermal fluid circulation triggers serpentinization of mantle rocks, high-
30 temperature (>350 °C) mineralization (Cu-Fe-Co-Zn-Ni; Coltat et al., 2019b), and
31 carbonation of ultramafic rocks (Lemoine et al., 1987). While the tectonic processes
32 responsible for mantle exhumation are well investigated at present-day OCTs (Peron-
33 Pinvidic et al., 2013) and fossil OCTs (Epin et al., 2019), syn-exhumation carbonation of
34 mantle rocks remains poorly constrained so far (e.g., Coltat et al., 2019a, 2020). This is
35 mainly due to the fact that present-day OCTs are buried and accessible only by drilling,
36 whereas fossil analogues are commonly overprinted by metamorphic events during late
37 orogenesis disturbing the former hydrothermal systems.

38 We focus on the exceptionally well-preserved Tasna OCT section exposed in
39 southeastern Switzerland. The continuous 6-km-long section shows mantle rocks
40 exhumed along a detachment fault that defines a tectonic Moho in the south and the
41 exhumed seafloor sealed by deepwater post-rift sediments in the north. Hydrothermal
42 carbonates occur in mantle rocks all along the detachment. Hence, the Tasna section
43 offers a unique opportunity to unravel, through coupled field work and oxygen, carbon,
44 and strontium isotope geochemistry, the conditions of carbonation (i.e., structural setting,
45 fluid sources, thermal regimes) during mantle exhumation.

46 **GEOLOGICAL SETTING**

47 The Tasna OCT belongs to the Penninic Tasna nappe exposed in the Engadine
48 window near Scuol in southeastern Switzerland (Ribes et al., 2020, and references
49 therein; Figs. 1A–1C). It has been interpreted to represent the most distal Briançonnais
50 **and/or** European margin of the Alpine Tethys (Florineth and Froitzheim, 1994; Ribes et
51 al., 2020). The Tasna outcrops preserve, across 6 km, an exhumed subcontinental mantle
52 that underlies a wedge of thinned mid- to lower continental crust composed of pre-rift
53 migmatites and Permian gabbros (Florineth and Froitzheim, 1994). Both the contact
54 separating the crust and the mantle as well as the top of the crust represent detachment
55 faults, namely the Lower and Upper Tasna detachments (LTD and UTD, respectively;
56 Florineth and Froitzheim, 1994). The UTD was crosscut by the LTD (Figs. 1D and 1E),
57 and both show a southeast-northwest-oriented stretching lineation in present-day
58 coordinates (Manatschal et al., 2006). The detachments show well-developed damage
59 zones formed by cataclasites whereas core zones are characterized by foliated cataclasites
60 and gouges (Manatschal et al., 2006) and are sealed by Upper Jurassic to Lower
61 Cretaceous deep-marine post-rift sediments (Ribes et al., 2020). Ar-Ar phlogopite dating
62 and stratigraphic constraints suggest late Middle to early Late Jurassic exhumation
63 (Manatschal et al., 2006; Ribes et al., 2020), predating the Alpine orogeny.

64 During the Eocene–Oligocene, the Tasna OCT was emplaced onto the Lower
65 Tasna unit (Ramosch **and** Roz-Champatsch zones; Figs. 1C–1E) along a north-vergent
66 thrust. Thus, the Tasna OCT discussed here remained little affected by Alpine
67 deformation and metamorphism and preserves its pre-Alpine, exhumation-related
68 structures (Fig. 1E).

69 **RESULTS**

70 Structural relationships between carbonation and deformation were characterized
71 by both field and microscopic analyses. We collected 32 samples of ophicalcites and
72 carbonated rocks along the LTD (Fig. 1E) for C, O, and Sr isotope geochemistry (see
73 details in the Supplemental Material¹).

74 **Field and Petrographic Data**

75 At the Tasna OCT, spinel lherzolites display an advanced serpentinization,
76 although **relics** of primary minerals are locally preserved. Along the LTD,
77 serpentinites display a cataclastic fabric, locally crosscut by green serpentine veins.
78 Toward the top of the LTD, cataclasites are foliated and contain rounded serpentinite
79 clasts. Carbonation in the LTD shows a gradient across 20 m, reaching a maximum in the
80 core zone where it occurs both as discrete calcite + chlorite veins (Fig. 2A) and as
81 pervasive replacement of serpentinites (foliated ophicalcites; Fig. 2B).

82 In the overlying thinned crust, migmatites and Permian gabbros display high-
83 temperature mylonitic fabrics. At their base, the gabbros show carbonated, matrix-
84 supported cataclasites with pervasively carbonated gabbro clasts (Fig. 2C; ~~App. 2~~ Fig. S1
85 in the Supplemental Material). The cataclasites are surrounded by chlorite-rich gouge
86 zones with discrete calcite veins. Southeast-northwest striations on chlorite shear planes
87 show the same orientations as kinematic indicators measured along the LTD. Foliated
88 gabbros are locally crosscut by high-angle structures with respect to the LTD (Fig. S1). A
89 greenschist-assemblage alteration (epidote + chlorite + albite + quartz) is commonly
90 observed both in the gabbro groundmass and in high-angle structures, together with
91 calcite veins (Fig. S1). Migmatites consist of quartz, feldspars, micas, garnet, and minor

92 zircon, monazite, and opaque minerals. This assemblage is commonly altered into epidote
93 + chlorite + quartz with graphite flakes locally associated with epidote (Fig. S1).

94 **Stable (C and O) and Sr Isotope Compositions of Carbonates**

95 The O, C, and Sr isotope compositions of carbonates are reported in Figures 3A
96 and 3B and in ~~App. 3~~ **Table S1 in the Supplemental Material**. Along the LTD, the $\delta^{18}\text{O}$
97 values of carbonates range from 13.5‰ to 25.5‰ and the $\delta^{13}\text{C}$ values are from -4.75‰
98 to 1.49‰, being comparable to the isotopic signatures of Alpine Tethys opicalcites (Fig.
99 3A). The $\delta^{18}\text{O}$ values of carbonates from the northern domain are between 16.8‰ and
100 25.5‰, higher than in the southern domain (13.5‰–16.6‰). The $\delta^{13}\text{C}$ values are
101 between -0.69‰ and 1.36‰ in the northern domain, whereas they vary between -4.75‰
102 and 1.49‰ in the southern domain. Variations in both $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values also exist
103 within samples (Fig. 3C).

104 Along the LTD, the Sr isotope compositions range from 0.707043 to 0.710738
105 (Fig. 3B), with carbonates from the southern domain (with low $\delta^{18}\text{O}$ values)
106 **[[generally?]]** being more radiogenic than in the northern domain. In addition, at a given
107 site, the Sr isotope compositions of the carbonates are heterogenous (i.e., site Tas. 1; Fig.
108 3B).

109 **DISCUSSION**

110 **A Preserved Syn-Exhumation Hydrothermal Carbonation**

111 At the Tasna OCT, several arguments **can be made** that carbonation was
112 synchronous with Jurassic extension. First, undeformed Mesozoic post-rift marine
113 sediments seal carbonated extensional structures (Manatschal et al., 2006), indicating that
114 carbonation occurred during oceanic conditions. Secondly, southeast-northwest striations

115 measured on chlorite shear planes surrounding matrix-supported cataclasites (Fig. 2C) are
116 consistent with those measured along the LTD (Manatschal et al., 2006), suggesting that
117 both chlorite gouge zones and cataclasites were formed during Jurassic detachment
118 faulting. **[[Unless the following sentence is a continuation of the “Secondly” idea,**
119 **please preface it with “Thirdly” for consistency]]**The presence of carbonate veins
120 either found as clasts in matrix-supported cataclasites or crosscutting the latter indicates
121 that carbonation occurred during the formation of the cataclasites and, by inference,
122 during detachment faulting. Finally, high-angle structures characterized by a calcite-
123 bearing greenschist alteration crosscut the Permian mylonitic gabbros and hence postdate
124 this deformation. Such a greenschist alteration might be the consequence of oceanic
125 hydrothermal alteration (Richardson et al., 1987 **[[Richardson et al., 1987 is not in the**
126 **reference list.]]**; Cann and Gillis, 2004 **[[Cann and Gillis, 2004 is not in the reference**
127 **list.],** previously recognized in mafic rocks along Jurassic detachment faults in Jurassic
128 Tethyan OCTs (Coltat et al., 2019a, 2020). Also, this alteration could be inherited from
129 oceanic conditions during the Jurassic rifting.

130 Even if carbonation along the LTD occurred during the Jurassic rifting, the
131 preservation of the isotopic compositions of the carbonates might be questioned (see
132 Bernoulli and Weissert [2020], the comment of Coltat et al. [2021], and the reply of
133 Bernoulli and Weissert [2021]). Here, given the lack of Alpine compressional structures
134 able to channelize fluids and the range of 12‰ observed for $\delta^{18}\text{O}$ values of carbonates
135 along the LTD (Figs. 3A and 3B) as well as in a given sample (Fig. 3C), we infer that
136 isotopic equilibration did not occur during Alpine metamorphism and that the isotopic
137 compositions are indicative of Jurassic oceanic hydrothermal conditions.

138 **Multiple Isotope Reservoirs for Carbonation**

139 At the Tasna OCT, the C and Sr isotope compositions of carbonates are very
140 heterogenous (Figs. 3A, 3B), suggesting different fluid sources and/or, for carbon,
141 processes inducing isotopic fractionation. In the northern domain, the $\delta^{13}\text{C}$ values of
142 carbonates are centered at 0‰–1‰, indicative of a marine origin (Bach et al., 2011). The
143 low Sr isotope compositions measured in the northern domain (centered around ~ 0.7075),
144 comparable to those of Jurassic seawater, also support a marine origin. In the southern
145 domain, the Sr isotope compositions are higher (as high as 0.710738). Only the
146 continental crust has $^{87}\text{Sr}/^{86}\text{Sr}$ values as radiogenic as those measured in the carbonates of
147 the southern domain, suggesting interactions between seawater and the continental crust
148 prior to carbonation. In the southern domain, some carbonates display $\delta^{13}\text{C}$ values as low
149 as -4.75% . Such negative $\delta^{13}\text{C}$ values have been reported in carbonates at the Lost City
150 hydrothermal field (Mid-Atlantic Ridge; Früh-Green et al., 2003). There, they were
151 attributed to the involvement of C-bearing species with a low $\delta^{13}\text{C}$ value (like methane)
152 produced during Fischer-Tropsch-like reactions in serpentinization-derived fluids (SFs;
153 Ludwig et al., 2006). Serpentinization is known to generate CH_4 and H_2 (Charlou et al.,
154 2002), compounds that have been sampled in venting fluids at the Lost City hydrothermal
155 field (Kelley et al., 2001). At the Tasna OCT, the circulation of such fluids during
156 Jurassic mantle exhumation is expected to have occurred as well. Indeed, the graphite
157 accompanying the greenschist facies alteration of migmatites (Fig. S1) is likely derived
158 from the reduction of CO_2 -bearing species reacting with H_2 , a process proposed for other
159 settings (Mastalerz et al., 1995). Hence, at the Tasna OCT, the involvement of SFs, which
160 were affected by fluid–continental crust interactions prior to serpentinization, is deduced

161 from the lowest $\delta^{13}\text{C}$ values and the highest $^{87}\text{Sr}/^{86}\text{Sr}$ values of carbonates, whereas
162 seawater mostly controlled the carbon and strontium budgets during the formation of
163 carbonates with $\delta^{13}\text{C}$ values close to 0‰–1‰ and low $^{87}\text{Sr}/^{86}\text{Sr}$ values.

164 **Conditions of Fluid-Rock Interactions Leading to Carbonation**

165 In the southern domain, carbonates display $\delta^{18}\text{O}$ values centered around ~15‰
166 (Fig. 3A). The limited range of the $\delta^{18}\text{O}$ values indicates precipitation from a unique fluid
167 at constant temperatures. On the other hand, the $\delta^{18}\text{O}$ values of the Tasna serpentinites
168 are centered around 8.4‰ (Engström et al., 2007). The ~6.6‰ difference is the
169 qualitative sign of isotopic equilibrium between calcite and serpentine under
170 hydrothermal conditions. The oxygen isotope fractionation factor between calcite and
171 serpentine (Zheng, 1993) indicates a temperature of ~175 °C for equilibration, i.e., for
172 both carbonation in the southern domain and serpentinization at depth.

173 Because seawater contains a higher concentration of dissolved carbon species
174 than SFs (Tivey, 2007), even minor contribution of seawater may drastically change the
175 $\delta^{13}\text{C}$ values of carbonates whereas the $\delta^{18}\text{O}$ values remain unaffected. Thus, in the
176 southern domain, the C and O isotope compositions are better explained by a carbonation
177 mechanism involving the mixing of small amounts of seawater with SFs.

178 The increase of the $\delta^{18}\text{O}$ values of the carbonates toward the northern domain is
179 due to progressive involvement of larger amounts of cold seawater during carbonation.
180 The progressive mixing of SFs with seawater may be modeled by simple isotopic mass-
181 balance calculation. Whereas the carbonates with the lowest $\delta^{18}\text{O}$ values form from
182 almost pure SFs at 175 °C mixing with minute amounts of seawater, the highest $\delta^{18}\text{O}$
183 values measured (25.5‰) can form under a SF/seawater ratio of ~1:2 at a temperature of

184 60 °C. The global temperature range (60–175 °C) encompasses the **temperatures**
185 estimated in ophicalcites formed during mantle exhumation at oceanic core complexes
186 (Kelley et al., 2001; Bach et al., 2011; Schroeder et al., 2015). In this context, the variable
187 O, C, and Sr isotope values measured in a given sample from Tasna (Fig. 3C; **Table S1**)
188 would be the record of superimposed carbonation events during mantle exhumation, the
189 carbonates with low $\delta^{18}\text{O}$ values being formed at depth prior to those with high $\delta^{18}\text{O}$
190 values near the seafloor.

191 **A Hydrothermal Model for the Moho Carbonation at Tasna**

192 In Figure 4, we propose a hydrothermal model for syn-exhumation carbonation of
193 the tectonic Moho at the Tasna OCT. During Jurassic rifting, seawater circulated
194 downwards along normal faults cutting through the continental crust, increasing the Sr
195 isotope composition of the modified seawater (Fig. 4A). At depth, serpentinization
196 reactions occurred by alteration of the subcontinental mantle. During their way back to
197 the seafloor along the detachment plane, the SFs (former seawater) were focused along
198 the uppermost permeable damage zone of the detachment and progressively mixed with
199 ambient or downgoing seawater, inducing carbonation (Fig. 4A). High fluid fluxes
200 occurred in high-permeability fault zones of the detachment, leading to the formation of
201 foliated ophicalcites, whereas less-permeable cataclasites were mostly affected by
202 fractures sealed by calcite (Fig. 4B).

203 Carbonate precipitation from the mixing of SFs with seawater is predicted by
204 numerical modeling (Palandri and Reed, 2004) and is proposed to explain carbonation at
205 the Lost City hydrothermal field (Ludwig et al., 2006). Early carbonation in deep zones
206 occurred from the mixing of low- $\delta^{13}\text{C}$, ^{87}Sr -rich, carbon-bearing SFs with minute

207 amounts of seawater. During mantle exhumation, the proportion of seawater
208 progressively increased in the hydrothermal ~~mixed~~ fluid (i.e., decrease of the SF/seawater
209 ratio), explaining the increase of $\delta^{18}\text{O}$ and decrease of $^{87}\text{Sr}/^{86}\text{Sr}$ values at constant $\delta^{13}\text{C}$
210 values. The increasing amount of seawater induced a decrease of the formation
211 temperatures of carbonates toward the northern domain.

212 CONCLUSIONS

213 Carbonation of the tectonic Moho interface is observed at the Tasna ocean-
214 continent transition in the Swiss Alps. The exceptional preservation of the structural and
215 isotopic systems allows highlighting the following major points:

- 216 (1) Carbonation occurred during syn-serpentinization mantle exhumation beneath the
217 continent wedge and lasted after mantle exhumation at the seafloor.
- 218 (2) Carbonation resulted from the mixing of serpentinization-derived fluids (**temperature**
219 of $\sim 175\text{ }^\circ\text{C}$) with cold seawater.
- 220 (3) The contribution of unmodified seawater increased consistently with progressive
221 mantle exhumation toward the seafloor.

222 ACKNOWLEDGMENTS

223 **We thank** X. Lecoq and D. Vilbert for thin-section preparation and Sr isotope
224 analyses, respectively. This **study was granted from** **[[“study was funded by the”?]]**
225 CNRS-INSU-CESSUR to P. Boulvais and **[[from? (Current wording implies that**
226 **CNRS-INSU-CESSUR granted money to the FMR)]]** the French Ministry of Research
227 (R. Coltat’s Ph.D.). We thank A. Zagorevski and I. Bindeman for constructive reviews,
228 and W. Clyde for editorial handling.

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345 **FIGURE CAPTIONS**

346 Figure 1. (A) Simplified geological map of the Alpine system in Western Europe (from
347 Schaltegger et al., 2002). (B) Geological map of the Engadine window in the southeastern
348 Alps, **Switzerland**. (C) North-south–striking cross section along the Tasna nappe
349 **(modified after Ribes et al., 2020). See B for location. [[Briefly explain the diagram at**
350 **the lower right corner]]** **OCT—ocean-continent transition**. (D,E) Southwest-northeast–
351 oriented field view of the Tasna section (from Ribes et al., 2020) (D) and corresponding
352 line drawing after field analysis (E). Samples from sites Tas. 4 and **Tas. 8** were collected
353 along blocks slipped down from the cliff. Their corresponding location is projected.

354 **[[In the figure, panel A, replace “&” with “and”; replace “units/ophiolites” with,**
355 **e.g., “units (green) and ophiolites (black)”. In panel B, put “N” and “E” at the end**
356 **of latitude/longitude labels instead of at the beginning; in the symbol explanation,**
357 **insert a space on either side of the plus symbol (+). In panel C, label the vertical axis**
358 **with a description—e.g., “Elevation (m)”;** **make instances of “unit” lowercase; spell**
359 **out instances of “P.”. In panel E, make instances of “unit” lowercase; spell out**
360 **instances of “P.”; insert a space after each instance of “Tas.” (before the numeral) to**

361 **match text and other figures; in the symbol explanation, insert a space on either side**
362 **of the plus symbol; change “Samples” to “Sample”]].**

363

364 Figure 2. Field pictures from the Tasna section along the the Lower Tasna
365 detachment. **[[Provide location information (latitude/longitude) for each photo]]** (A)
366 Calcite (Cc) + chlorite (Chl) veins cutting through serpentinites from the northern domain
367 (site Tas. [4](#)). (B) Foliated opicalcites at contact with overlying gabbro along tectonic
368 Moho interface from the southern domain (site Tas. [2](#)). (C) Matrix-supported cataclasite
369 with clasts of gabbro at the top of the Lower Tasna detachment in the southern domain
370 (site Tas. [3](#)).

371

372 Figure 3. (A,B) $\delta^{13}\text{C}$ (**Vienna Peedee belemnite**, VPDB) (A) and $^{87}\text{Sr}/^{86}\text{Sr}$ versus $\delta^{18}\text{O}$
373 (**Vienna standard mean ocean water**, VSMOW) (B) isotopic diagrams of carbonates from
374 carbonated rocks from the Tasna section plotted together with **opicalcites in the Alpine**
375 **realm** **[[Clarify how these are represented in the figure]]** (Weissert and Bernoulli,
376 1984; Schwarzenbach et al., 2013; Lafay et al., 2017; Coltat et al., 2019a, 2019b).
377 Samples from Engström et al. (2007) were collected at the vicinity of sites Tas. 3 and
378 Tas. [4](#). **[[Define “SW” (seawater?)]]** (C) Sawed sections of opicalcites showing variety
379 of carbonate textures and corresponding oxygen and carbon isotope values.

380 **[[In the figure, panel A, make “eastern” lowercase; capitalize and correct the**
381 **spelling of “Apennines”]]**

382

383 Figure 4. Schematic tectonic-hydrothermal model for carbonation at the Moho interface
384 at the Tasna ocean-continent transition. (A) Mantle exhumation was accompanied by
385 upward circulation of serpentinization-derived fluids along the detachment (geometry
386 adapted from Ribes et al., 2020). **[[Explain the color gradient labeled “T°C”]] Sr—**
387 **[[strontium?]]**; LTD—Lower Tasna detachment; UTD—Upper Tasna detachment. (B)
388 Serpentinization-derived fluids progressively mixed with seawater, leading to carbonate
389 precipitation. See text for details.

390 **[[In the figure, panel A, insert a space between “175” and “°C”; change “T°C” to**
391 **just “*T*” (with italics) and define in the caption as “temperature”? (see query**
392 **above)]]**

393

394 ¹Supplemental Material. **[[Please provide a brief caption here]]**. Please visit
395 <https://doi.org/10.1130/XXXXX.1> to access the supplemental material, and contact
396 editing@geosociety.org with any questions.