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Carbonation by fluid–rock interactions at high-pressure conditions: Implications for carbon cycling in subduction zones

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ABSTRACT

Carbonate-bearing lithologies are the main carbon carrier into subduction zones. Their evolution during metamorphism largely controls the fate of carbon, regulating its fluxes between shallow and deep reservoirs. Recent estimates predict that almost all subducted carbon is transferred into the crust and lithospheric mantle during subduction metamorphism via decarbonation and dissolution reactions at high-pressure conditions. Here we report the occurrence of eclogite-facies marbles associated with metasomatic systems in Alpine Corsica (France). The occurrence of these marbles along major fluid-conduits as well as textural, geochemical and isotopic data indicating fluid–mineral reactions are compelling evidence for the precipitation of these carbonate-rich assemblages from carbonic fluids during metamorphism. The discovery of metasomatic marbles brings new insights into the fate of carbonic fluids formed in subducting slabs. We infer that rock carbonation can occur at high-pressure conditions by either vein-injection or chemical replacement mechanisms. This indicates that carbonic fluids produced by decarbonation reactions and carbonate dissolution may not be directly transferred to the mantle wedge, but can interact with slab and mantle-forming rocks. Rock-carbonation by fluid–rock interactions may have an important impact on the residence time of carbon and oxygen in subduction zones and lithospheric mantle reservoirs as well as carbonate isotopic signatures in subduction zones. Furthermore, carbonation may modulate the emission of CO_2 at volcanic arcs over geological time scales.

1. Introduction

Subduction exerts a key role in the long-term carbon cycle by regulating the fluxes of carbon between the Earth's surface and the deep Earth. The redistribution of carbon between the exogenic and endogenic reservoirs largely depends on the evolution of carbonate-bearing lithologies in subduction zones. Carbonates are present in sedimentary, mafic and ultramafic lithologies, and constitute the dominant reservoir of carbon in the subducting lithosphere (Alt and Teagle, 1999; Kelemen and Manning, 2015). There is growing evidence that carbonates can be extremely reactive during subduction metamorphism via devolatilization reactions (Cook-Kollars et al., 2014; Kerrick and Connolly, 2001), as well as carbonate dissolution via fluid–rock interactions at high-pressure conditions (Frezzotti et al., 2011; Ague and Nicolescu, 2014; Kelemen and Manning, 2015), carbonate reduction reactions (Galvez et al., 2013a; Malvoisin et al., 2012), or melting of the subducting crust (Poli, 2015). Nonetheless, key questions remain regarding the mechanisms of carbonic fluid transfer from the slab to the lithospheric mantle in the sub-arc region, their role in mantle wedge metasomatism, and their contribution to the CO_2 degassing at volcanic arcs.

To address these questions, a growing number of studies have been carried out using both experimental petrology (Molina and Poli, 2000; Poli et al., 2009) and thermodynamic modeling (Kerrick and Connolly, 2001; Gorman et al., 2006). All these studies agree that, considering a "closed system" (no external fluid supply), significant carbon transfer to the mantle wedge is feasible only at shallow depth in the forearc region. This transfer is made possible via devolatilization reactions that, based on experimental and thermodynamic results, are much more limited at deeper, subarc conditions (Connolly, 2005; Poli et al., 2009). Nevertheless, the role of

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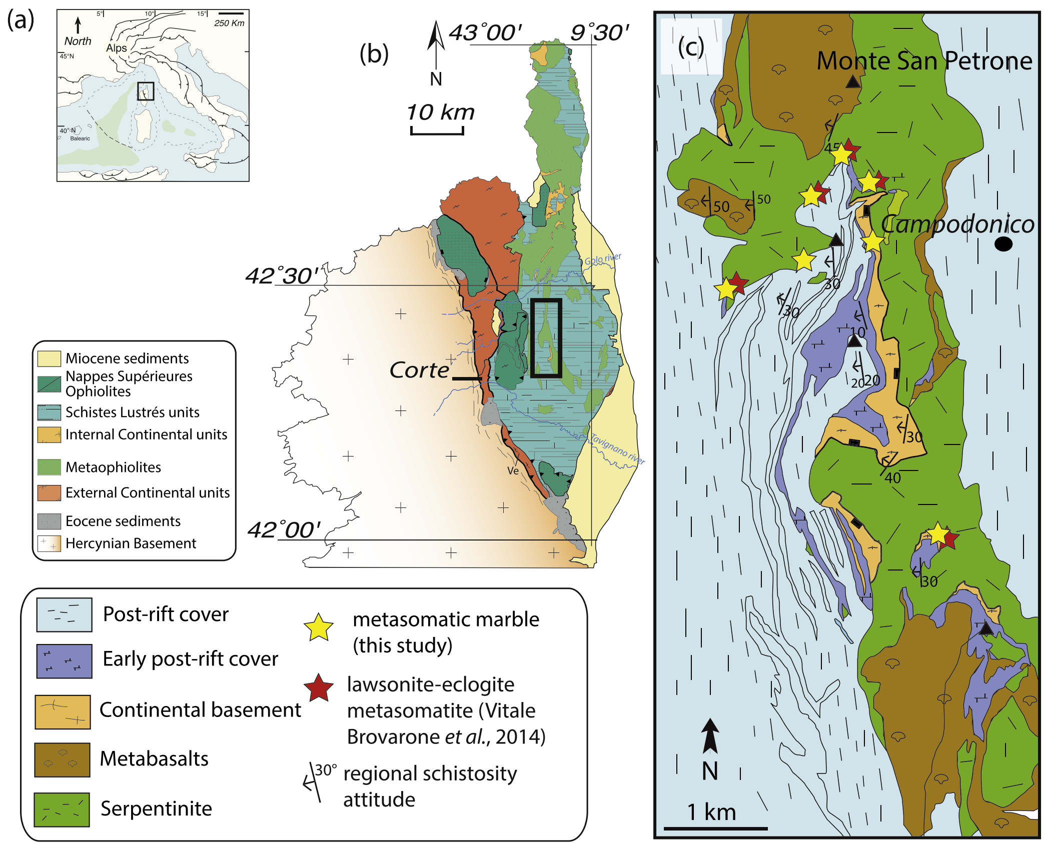

Fig. 1. Geological background of the study area. a) Simplified regional setting of Alpine Corsica (France) in the Western Mediterranean region. Modified after Molli and Malavieille (2011). b) Simplified tectono-stratigraphic map of Alpine Corsica. The black box indicates the position of the study area. c) Simplified geological map of the San Petrone unit. Figures modified after Vitale Brovarone et al. (2011b, 2013).

fluid–rock interactions appears to be critical for the stability of carbonates. Recently, field based studies (Ague and Nicolescu, 2014; Frezzotti et al., 2011) as well as theoretical and experimental works (Facq et al., 2014; Sverjensky et al., 2014) have pointed out that massive carbonate dissolution in fluids may occur at high-pressure-low temperature conditions and can generate large amounts of carbonic fluids (see Kelemen and Manning, 2015 for review). Accounting for carbonate dissolution at high-pressure conditions in comprehensive budgets overturns older paradigms on carbonate stability with respect to carbon mobility in subduction zones. The most recent budgets actually predict that carbonate dissolution allows almost all subducted carbon to be transferred to the mantle wedge (Kelemen and Manning, 2015). Owing to the very recent discovery of these processes, much remains to be learned about the fate of these carbonic fluids and their interaction with slab- and mantle-forming rocks.

Here, we report the occurrence of eclogite-facies marbles formed by fluid–rock interaction processes (metasomatism) occurring along intensely metasomatized lithological interfaces (Alpine Corsica, France). We present and discuss the occurrence, textures, mineralogy and geochemistry of these metasomatic marbles. We then propose a mechanism of carbonates formation by precipitation and mineral carbonation by carbonic fluid–rock interactions at high-pressure conditions. Finally, the implications and contribution of rock carbonation to the deep carbon fluxes and cycling are discussed.

2. Geological setting

Alpine Corsica (France) is a branch of the Alpine orogenic system (Jolivet et al., 1991; Molli and Malavieille, 2011) (Fig. 1a). The belt mainly includes remnants of subducted Mesozoic slow-spreading oceanic and passive margin lithosphere, which formed part of the Tethys Ocean basin. This rock package is classically referred to as the Schistes Lustrés Complex (Fig. 1b; Jolivet et al., 1990; Fournier et al., 1991; Malavieille et al., 1998; Vitale Brovarone et al., 2013). The exceptional preservation of prograde-to-peak mineral assemblages, including widespread lawsonite, makes the Schistes Lustrés of Alpine Corsica an excellent site for field investigations related to subduction. These units underwent various metamorphic overprints during the Alpine subduction ranging from subgreenschist-facies conditions of about 300 °C and 0.5 GPa to lawsonite blueschist and lawsonite eclogite-facies conditions which reached 500–550 °C and ~2.3 GPa (e.g. Fournier et al., 1991; see Vitale Brovarone et al., 2013 for review).

Carbonate-bearing rocks are widespread in the Schistes Lustrés of Alpine Corsica. The most typical carbonate-bearing lithologies are metamorphosed oceanic sediments (referred to as calcschists hereafter), ophicarbonate, and carbonated metabasalts (Miller et al., 2001; Ravna et al., 2010; Vitale Brovarone et al., 2011b). In each lithology, primary carbonates and various generations of carbonate veins are observed (Miller et al., 2001; Ravna et al., 2010; Vitale Brovarone et al., 2011a). These veins have been shown to be in most cases in isotopic equilibrium with the host rocks with little effect of external fluid infiltrations and metasomatism (Cartwright and Buick, 2000). On the other hand, evidence for high-pressure-low temperature fluid–rock interactions and metasomatism in the Schistes Lustrés of Alpine Corsica is widespread, most typically localized at lithological interfaces. In this work, we focus on processes occurring where serpentinites are in contact with metasedimentary rocks.

Here, past results on high-pressure metasomatism in Alpine Corsica are briefly summarized. In the blueschist-facies zone, reactions between serpentinites and overlying metasediments led