



Volatile (Li, B, F and Cl) mobility during amphibole breakdown in subduction zones



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ABSTRACT

Amphiboles are ubiquitous minerals in the altered oceanic crust. During subduction, their breakdown is governed by continuous reactions up to eclogitic facies conditions. Amphiboles thus contribute to slab-derived fluid throughout prograde metamorphism and continuously record information about volatile exchanges occurring between the slab and the mantle wedge. However, the fate of volatile elements and especially halogens, such as F and Cl, in amphibole during subduction is poorly constrained. We studied metagabbros from three different localities in the Western Alps: the Chenailet ophiolite, the Queyras Schistes Lustrés and the Monviso meta-ophiolitic complexes. These samples record different metamorphic conditions, from greenschist to eclogite facies, and have interacted with different lithologies (e.g. sedimentary rocks, serpentinites) from their formation at mid-oceanic ridge, up to their devolatilization during subduction. In the oceanic crust, the initial halogen budget is mostly stored in magmatic amphibole (F = 300–7000 ppm; Cl = 20–1200 ppm) or in amphibole corona (F = 100–7000 ppm; Cl = 80–2000 ppm) and titanite (F = 200–1500 ppm; Cl < 200 ppm) formed during hydrothermal seafloor alteration. It is thus the fate of these phases that govern the halogen fluxes between the crust and the overlying mantle and/or the plate interface in subduction zones. Li and B are poorly stored in the oceanic crust (<5 ppm). In subduction zones, prograde metamorphism of metagabbros is first marked by the crystallization of glaucophane at the expense of magmatic and amphibole coronas. This episode is accompanied with a decrease of halogen concentrations in amphiboles (<200 ppm of F and Cl) suggesting that these elements can be transferred to the mantle wedge by fluids. In the Queyras Schistes Lustrés complex, the intense deformation and the abundant devolatilization of metasedimentary rocks produce large fluid flows that promote rock chemical hybridization (metasomatic mixing with hybrid composition between metasedimentary rock and metagabbro) at the metasedimentary rock/metagabbro contacts. Such fluid/rock interactions result in a strong addition of Li in glaucophane (up to 600 ppm) whereas halogen concentrations are unaffected. At eclogite facies conditions, metagabbros display low halogens concentrations (<20 ppm of F and < 100 ppm of Cl) relative to altered oceanic crust (F = 40–650 ppm; Cl = 40–1400 ppm) suggesting that these elements are continuously released by fluids during the first 30–80 km of subduction whatever the tectonic environment (e.g. slab, plate interface) and the considered fluid/rock interactions.

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1. Introduction

Magmas erupted at arc volcanoes are generated by partial melting of metasomatized mantle wedge peridotites, and are enriched in H₂O and volatile elements relative to Mid-Oceanic Ridge Basalts (MORB) or Oceanic Island Basalts (e.g. Le Voyer et al., 2010; Rose-Koga et al., 2014; Straub and Layne, 2003). The distinct geochemical signatures and physical properties of arc lavas result from volatile transfer from the slab to the mantle wedge (e.g. Bouilhol et al., 2009; Dalou et al.,

2012; Kendrick et al., 2012). Quantitative assessments of these chemical fluxes require knowledge about the pressure and temperature conditions at which slab-derived fluids are released and transferred to the mantle wedge and the volatile composition evolution of the slab during prograde metamorphism. Slab dehydration occurs between 40 and 200 km (e.g. Magni et al., 2014; Schmidt and Poli, 2014) and is related to the stability of hydrous phases such as serpentine (Ulmer and Trommsdorff, 1995; Wunder and Schreyer, 1997), chlorite (Grove et al., 2006, 2009), amphiboles, lawsonite, and/or mica (Pawley and Holloway, 1993; Poli and Schmidt, 1995). The evolution of volatile abundance, during the breakdown of hydrous phases in the slab, is critical to establish the mass balance of the fluxes between the slab, the

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surface and the deep mantle. However, it remains a significant challenge because multiple metamorphic reactions or fluid/rock interactions occur during subduction.

The composition of primitive arc magmas, recorded in olivine-hosted melt inclusions, is enriched in halogens, such as F and Cl, relative to MORB demonstrating that these elements are mobile in slab derived fluids (e.g. Le Voyer et al., 2010; Rose-Koga et al., 2014; Straub and Layne, 2003). Unlike many fluid mobile elements, such as B or Li (e.g. Bebout et al., 2007, 2013; Marschall et al., 2007, 2009; Penniston-Dorland et al., 2010, 2012; Scambelluri et al., 2004a,b; Vils et al., 2010; Zack et al., 2003; Scambelluri and Tonarini, 2012), the behavior of halogens during subduction is poorly constrained (see Kendrick et al., 2012). Halogens are present in OH⁻ sites within hydrous minerals, such as sheet silicates or amphibole (e.g. Anselmi et al., 2000; Oberti et al., 1993; Volfinger et al., 1985). Those minerals, which are generally formed after seawater-lithosphere interactions at mid-oceanic ridges, can contain significant concentrations of halogens (up to several thousand of ppm; Barnes and Sharp, 2006; Coogan et al., 2001; Ito et al., 1983; Vanko, 1986). During prograde metamorphism, the devolatilization of serpentinites and metagabbros is accompanied with a decrease in halogen concentration suggesting that this process can trigger the transfer of a significant amount of halogens in fluids to the mantle wedge (Bonifacie et al., 2008; John et al., 2011; Kendrick et al., 2011; Philippot et al., 1998). On the other hand, Cl seems to be retained in metasedimentary rocks during subduction (Selverstone and Sharp, 2015). At depth, the devolatilization of slab serpentinites is believed to be a significant source of water and fluid mobile elements for arc magmas (e.g. Kendrick et al., 2011; Scambelluri et al., 2004a; Ulmer and Trommsdorff, 1995; Wunder and Schreyer, 1997; Scambelluri and Tonarini, 2012). However, mass balance calculation suggests that this process, alone, cannot explain the observed halogen anomalies of arc magmatism, notably F concentrations. As such additional components in the subducted oceanic lithosphere must act as significant halogen reservoirs during subduction (Debret et al., 2014).

Amphibole is a common component in the altered oceanic crust and its crystal structure can incorporate a wide range of lithophile and volatile elements (e.g. Coogan et al., 2001; Gillis et al., 2003; Messiga et al., 1995; Volfinger et al., 1985). In oceanic settings, amphibole-bearing metagabbros are formed by crystallization of silicate melt at temperature greater than 850 °C, and by continuous metamorphic reactions during lithosphere cooling and hydration between 850 to 300 °C (Coogan et al., 2001; Debret et al., 2013a; Gilbert et al., 1982; Messiga and Tribuzio, 1991; Spear, 1993; Tribuzio et al., 1995; Vanko, 1986). During subduction, amphibole breakdown in the slab is governed by continuous reactions occurring below 3 GPa (Poli and Schmidt, 1995) that release a significant amount of water (up to 2 wt%), mostly to the forearc region. It is clear that the breakdown of amphibole during subduction prograde metamorphism could be responsible for delivering a large flux of slab-derived fluid to the overlying mantle wedge. This mineral thus plays a significant role during element fractionation between the fluid and residual slab, and records continuous information about volatile transfer from slab to mantle wedge. However, while there are an increasing number of *in situ* studies focusing on the fate of volatile elements in the hydrous phases composing the subducted lithosphere (e.g. Bebout et al., 2007, 2013; Debret et al., 2013b, 2014; Lafay et al., 2013; Scambelluri et al., 2004a; Vils et al., 2010; Vitale Brovarone et al., 2014), most of them have examined micas, serpentines and/or lawsonites and the potential volatile, especially halogens, losses from amphiboles during subduction remain poorly constrained.

This study aims to determine the volatile (F, Cl, Li and B) fluxes between the different lithologies composing the slab and the mantle wedge. The approach taken is an *in situ* (Secondary ion mass spectrometry and microprobe analyses) and detailed study of amphiboles major and volatile element composition in metagabbros from a suite of Western Alps meta-ophiolites. The micro analytical methods employed here aim to separate oceanic, prograde and retrograde metamorphic evolution of

amphiboles and thus better decipher the mobility of volatile elements during metamorphism than bulk rock studies. Metagabbros from 6 different Western Alps meta-ophiolites, which metamorphic conditions span from greenschist to eclogite facies, were selected for a detailed petro-geochemical study. These samples have interacted with different lithologies (e.g. sedimentary rocks, serpentinites) during subduction leading to the released of halogens (F and Cl) to fluids.

2. Geological setting and petrographic observations

Western Alps meta-ophiolites represent hectometric to kilometeric size mafic and ultramafic bodies formed at magma poor settings, and are related to the opening of the Ligurian Ocean in the middle Jurassic (Lagabrielle and Cannat, 1990; Müntener et al., 2010). These meta-ophiolites represent the first few kilometers of the Ligurian oceanic lithosphere, which structure is similar to the lithosphere generated at slow- or ultra-slow spreading ridges (Bernoulli et al., 2003; Lagabrielle et al., 2014; Manatschal et al., 2011; Tricart and Lemoine, 1986). This study focuses on the Piemont zone of the south-western Alps (Fig. 1a and b) where ophiolitic units were metamorphosed and juxtaposed during alpine subduction and collision in Late Cretaceous to Tertiary (e.g. Tricart, 1984). We sampled metagabbros from 6 meta-ophiolites on a transect of ~20 km through the Queyras Schistes Lustrés complex, from the Montgenevre to the Monviso (Fig. 1b).

The studied meta-ophiolites belong to two different domains. (i) The Queyras Schistes Lustrés complex consists of 10% of meta-ophiolites and 90% of metasedimentary rocks and is interpreted as a paleo-sedimentary wedge (Tricart and Schwartz, 2006). The Queyras meta-ophiolites form metric to kilometeric bodies embedded in dominant Jurassic to Lower Cretaceous clastic metasedimentary rocks (Lagabrielle et al., 1984; Lemoine et al., 1987). They have experienced strong interactions between mafic lithologies and sediment derived fluid during subduction (Lafay et al., 2013). In this complex, three tectono-metamorphic units record variable P-T conditions (Fig. 1b, c), increasing from low-temperature blueschist facies conditions (LT-blueschist; 320–360 °C and 9–11 kbar) in western Queyras to medium-temperature (MT-blueschist; 340–390 °C and 10–12 kbar) and high-temperature blueschist (HT-blueschist; 380–470 °C and 12–18 kbar) conditions toward to the East (Agard et al., 2001; Ballèvre et al., 1990; Lagabrielle et al., 2014; Schwartz et al., 2013; Tricart and Schwartz, 2006). (ii) The Monviso meta-ophiolite corresponds to a remnant of the oceanic lithosphere preserving multiple slices of the oceanic crust embedded in serpentinites and re-equilibrated at eclogitic facies P-T conditions (Angiboust et al., 2011; Schwartz et al., 2001). Unlike the Queyras meta-ophiolites, the massif evolved during subduction without interacting with external fluids (Lafay et al., 2013).

For reference, we also studied the Chenaillet massif. This massif is considered to be representative of the pre-subduction alpine oceanic lithosphere since it has been poorly affected by alpine subduction/collision and related metamorphism (e.g. Caby, 1995; Lafay et al., 2013; Lagabrielle et al., 2014; Manatschal et al., 2011; Mével et al., 1978; Schwartz et al., 2013). The massif is interpreted as an obducted portion of the Tethyan oceanic lithosphere (e.g. Caby, 1995). It preserves a complete section of the oceanic lithosphere composed of serpentinite intruded by metagabbros and topped by basalts and rare sediments (Manatschal et al., 2011). The massif mostly records an oceanic metamorphism, ranging from granulite to greenschist facies conditions, during lithosphere cooling and hydration (Mével et al., 1978).

2.1. The Chenaillet ophiolite

The Chenaillet ophiolite (Fig. 1b) is a thin tectonic nappe resting upon the Queyras Schistes Lustrés complex. The massif preserves pre-Alpine structure and basement-cover relationship between serpentinitized mantle, metagabbros, sediments and metabasalts. The

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