



# The composition of Alpine marine sediments (Bündnerschiefer Formation, W Alps) and the mobility of their chemical components during orogenic metamorphism

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## ABSTRACT

The Bündnerschiefer of the Swiss-Italian Alps is a large sedimentary complex deposited on the Piemonte–Liguria and Valais oceans and associated continental margins from the upper Jurassic to Eocene. It is made of a large variety of sequences associated or not with an ophiolitic basement. The Bündnerschiefer makes an accretionary prism that developed syn-tectonically from the onset of alpine subduction, and it records orogenic metamorphism following episodes of HP metamorphism. The Bündnerschiefer shares important similarities with the Otago schists of New Zealand and with the Wepawaug schists of Connecticut, both of which form accretionary prisms and have an orogenic metamorphic imprint.

With the aim of testing the hypothesis of mobility of chemical components as a function of metamorphic grade, in this work I present fifty-five bulk chemical analyses of various lithological facies of the Bündnerschiefer collected along the well-studied field gradient of the Lepontine dome of Central Switzerland, in the Prättigau half window of East Switzerland, and in the Tsaté Nappe of Valle d'Aosta (Italy). The dataset includes the concentration of major components, large ion lithophile elements (Rb, Sr, Ba, Cs), high field strength elements (Zr, Ti, Nb, Th, U, Ta, Hf), fluid-mobile light elements (B, Li), volatiles (CO<sub>2</sub>, S), REEs, and Y, V, Cr, Co, Sn, Pb, Cu, Zn, Tl, Sb, Be, and Au. These data are compared against the compositions of the global marine sediment reservoir, typical crustal reservoirs, and against the previously measured compositions of Otago and Wepawaug schists. Results reveal that, irrespective of their metamorphic evolution, the bulk chemical compositions of orogenic metasediments are characterized by mostly constant compositional ratios (e.g., K<sub>2</sub>O/Al<sub>2</sub>O<sub>3</sub>, Ba/Al<sub>2</sub>O<sub>3</sub>, Sr/CaO, etc.), whose values in most cases are undistinguishable from those of actual marine sediments and other crustal reservoirs. For these rocks, only volatile concentrations decrease dramatically as a function of metamorphic temperature, and significant deviations from the reservoir signatures are evident for SiO<sub>2</sub>, B, and Li. These results are interpreted as an indication of residual enrichment in the sediments, a process taking place during syn-metamorphic dehydration from the onset of metamorphism in a regime of chemical immobility. Residual enrichment increased the absolute concentrations of the chemical components of these rocks, but did not modify significantly their fundamental ratios. This poor compositional modification of the sediments indicates that orogenic metamorphism in general does not promote significant mass transfer from accretionary prisms. In contrast, mass transfer calculations carried out in a shear zone crosscutting the Bündnerschiefer shows that significant mass transfer occurs within these narrow zones, resulting in gains of H<sub>2</sub>O, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, K<sub>2</sub>O, Ba, Y, Rb, Cu, V, Tl, Mo, and Ce during deformation and loss of Na<sub>2</sub>O, CO<sub>2</sub>, S, Ni, B, U, and Pb from the rock. These components were presumably transported by an aquo-carbonic fluid along the shear zone. These distinct attitudes to mobilize chemical elements from orogenic sediments may have implications for a potentially large number of geochemical processes in active continental margins, from the recycling of chemical components at plate margins to the genesis of hydrothermal ore deposits.

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## 1. Introduction

The geological evolution of orogenic belts is strongly influenced by the flux of fluids liberated at depth by dehydrating subducting slabs. Field-based petrological and geochemical studies (Scambelluri et al.,

2007) and experimental studies (Kessel et al., 2005) show that the composition and physical state of these fluids evolve from those of hydrous melts to those of aqueous fluids, or less commonly of transitional solute-rich fluids. Subduction-zone fluids act as solvents and transfer solutes from the slab to the overlying mantle wedge during a more or less continuous process of dehydration that starts relatively early in the subduction process. Moving from their source regions, these fluids trigger the metasomatism of slab and wedge, which in turn mediates the global cycling of the elements, yields the generation of large volumes

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of arc magmas, and ultimately forms the continental crust (Manning, 2004). One important example of this role of subduction-zone fluids is the evidence for enrichment of incompatible elements in arc lavas (e.g., K, Sr, Rb, Ba, U, and Pb) with respect to the typical altered oceanic crust (Becker et al., 2000; Churikova et al., 2001). Other relevant geological processes depending on, or generated by, the transfer of subduction-zone fluids are the genesis of ore deposits (Halter et al., 2002) and the evolution of crustal metamorphic systems (Spear, 1995).

Ocean marine sediments contain about 7 wt.% of mineral-bound H<sub>2</sub>O before subduction (Plank and Langmuir, 1998), and hence represent a fluid-rich subducting layer. Such layer reduces however its H<sub>2</sub>O content to 2–4 wt.% at a depth of about 30 km along the slab due to early dehydration (Nakamura and Iwamori, 2009), indicating that the influence of marine sediments in the production of subduction-zone fluids reduces strongly with depth. An independent feature that suggests a reduced influence of the sediments is the documented thickness of marine successions. In most basins, a relatively thin layer of sediments deposits on the oceanic lithosphere before subduction, resulting in an average thickness of 50–500 m at trenches (only few trenches, e.g. Makran and Andaman of the Indian Ocean, host several km of sediment successions: Plank and Langmuir, 1998). In volumetric terms, current sediment subduction is estimated to be globally between 0.5 and 0.7 km<sup>3</sup> a<sup>-1</sup>, but a variable (and unconstrained) fraction of this volume is skimmed off the downgoing slab and frontally accreted to the active buttresses of accretionary prisms (von Huene and Scholl, 1991). All these data together suggest that the role of marine sediments in the global bulk production of subduction-zone fluids is probably secondary when compared to the role of the much thicker oceanic crust.

The efficiency of sediment subduction with respect to the transfer of chemical elements in subduction-zone fluids has been the subject of a number of studies in the last decades, but a detailed knowledge of the physical and chemical properties of this “sedimentary component” is a matter of debate. Early trace element (Saunders et al., 1991) and radiogenic isotope studies (Hawkesworth et al., 1991) indicated a relatively minor contribution of marine sediment in the global budget of subduction-zone fluid. On the other hand, the first global compilation on marine sediments (Plank and Langmuir, 1993) showed a direct correlation between sediment flux into a number of trenches for a suite of major and trace elements (Ba, Sr, K, Rb, Cs, La, Th and U) and the abundance of the same elements in the associated volcanics. This correlation led to the conclusion that some of the geochemical characteristics of arc volcanics can be traced back to the sediments at the trench. Later field (Busigny et al., 2003; Sadofsky and Bebout, 2003; Spandler et al., 2003), computational (Kerrick and Connolly, 2001), and experimental (Hermann and Rubatto, 2009; Hermann et al., 2006; Kessel et al., 2005) studies of subduction-related metamorphism of marine sediments indicated a decoupling between the release of volatiles (i.e., H<sub>2</sub>O, CO<sub>2</sub>, CH<sub>4</sub>) and that of trace elements in subducting slabs. Accordingly, while release of H<sub>2</sub>O and CO<sub>2</sub> from dehydration starts relatively early in the subduction history, the liberation of key trace elements is controlled by the thermodynamic stability of a number of accessory minerals (i.e., allanite, monazite, rutile, apatite, and zircon) up to sub-arc depth. Supercritical liquids (sensu: Kessel et al., 2005) and hydrous melts (Hermann and Rubatto, 2009; Hermann et al., 2006) are identified as fundamental carriers of a suite of trace elements like Rb, Cs, Sr, Ba, Nb, Ta, La, Ce, Pb, U, Th, Be, and B, from the slab sediments, and their formation constrains the onset of efficient chemical transport at a depth of about 120–180 km (corresponding to pressures of 4–6 GPa) and at a temperature of 750–800 °C. At these conditions, thin layers of subducting sediments are inferred to detach from the downgoing slab and rise buoyantly into the mantle wedge, within which they form small diapirs (thickness: 250–500 m; diameter: 3–4 km) that melt partially and deplete the original sediment in incompatible elements like Sr, Pb, Th, Nd, and La (Behn et al., 2011).

One important corollary of these recent studies is that marine sediments subjected to typical conditions of regional metamorphism

(T < 700 °C and P < 1.5 GPa) should have a weak potential to mobilize chemical elements. This is a fundamental conclusion that contrasts with a large number of studies based on high-pressure metamorphic vein mineral assemblages, chemical and isotopic composition of regional metamorphic rocks, fluid inclusion data from metamorphic minerals, and petrologic models of the genesis of exhumed deep crustal rocks. Accordingly, fluids generated by Barrovian metasediments would transport a variety of rock- and also ore-forming elements (Ague, 2003, for a review). Calculations carried out on amphibolite facies rocks estimated a large regional, time-integrated flux of tens of thousands of m<sup>3</sup> of aqueous fluid for each m<sup>2</sup> of rock (Ague, 1994b), which would control the stability of key metamorphic minerals (e.g., feldspars, micas, aluminum silicates, and garnet) and transport major rock- and ore-forming elements.

In this work, I present a dataset of fifty-five bulk chemical analyses of the Bündnerschiefer of the Swiss-Italian Alps (BdS), a sedimentary complex deposited on the Piemonte–Liguria and Valais oceans and associated continental margins from the upper Jurassic to Eocene and subjected to regional metamorphism during a cold subduction. I use this dataset to test the hypothesis of mobility of its chemical components during orogenic metamorphism. Data are compared against the global reference compositions of marine sediments (GLOSS: Plank and Langmuir, 1998) and also against other datasets from orogenic belts in North America and New Zealand. Results show that most fundamental compositional ratios of the BdS are remarkably uniform within the considered range of metamorphic conditions (350 °C/0.3 GPa; 700 °C/1.0 GPa), and compare well with those of the reference GLOSS and other crustal reservoirs, with the exception of volatiles (CO<sub>2</sub>, H<sub>2</sub>O) and few fluid-mobile species (SiO<sub>2</sub>, B, Li). This compositional similarity is evident also in the sediments from North America and New Zealand, suggesting that orogenic metamorphism of average subducting sediments is probably unable to mobilize significant proportions of chemical elements from marine sediments, including several ore-forming elements.

## 2. The GLOSS reference composition

Determined to evaluate global geochemical fluxes associated with subducted marine sediments, the reference GLOSS composition is based on a weighted compilation of the extensive DSDP and ODP database from twenty-five trench locations (70% of the trenches worldwide) and on additional ~250 chemical analyses (Plank and Langmuir, 1998). As such, the GLOSS is not a true rock composition, but the most representative and actualistic global reservoir of marine sediments, as well a proxy of the upper Continental Crust (Rudnick and Gao, 2003). GLOSS is part of the “Geochemical Earth Reference Model”<sup>1</sup>, and has been a reference frame for experimental work (Hermann and Rubatto, 2009; Kessel et al., 2005) and computational studies of subduction-zone metamorphism (Kerrick and Connolly, 2001).

There are two good reasons to choose the GLOSS as a relevant precursor rock for sedimentary complexes. First, the concentration ranges of its components are a measure of the inherent lithological heterogeneity of marine sediments, which is typically large. The second reason is the strong evidence for a correlation between the concentrations of some important chemical components (e.g., K<sub>2</sub>O, Al<sub>2</sub>O<sub>3</sub>, REEs, etc.) and the proportions of the detrital, biological, carbonatic, and hydrothermal lithological constituents. This correlation represents a first order compositional constraint on marine sediments, and together with the characterization of lithological heterogeneity is necessary for mass transfer calculations in general. The lithological heterogeneity of a precursor rock (i.e., protolith) is a parameter that must be evaluated when mass transfers between geological fluids and rocks are calculated at the large (Grant, 1986) and small scale (Garofalo, 2004). In the case of syn-metamorphic mass

<sup>1</sup> See: [http://earthref.org/cgi-bin/germrd-s1-advanced.cgi?database\\_name=germrd&search\\_start=reservoir&res\\_id=19](http://earthref.org/cgi-bin/germrd-s1-advanced.cgi?database_name=germrd&search_start=reservoir&res_id=19)

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