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Source tracing of detrital serpentinite in the Oligocene molasse deposits from the western Alps (Barrême basin): implications for relief formation in the internal zone.

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Abstract. We present the first contribution of tracing the source area of ophiolitic detritus in the Alpine molasses by Raman spectroscopy. The lower Oligocene molasse deposits preserved in the Barrême basin, in the SW foreland of the western Alpine arc, are known for the sudden arrival of the first "exotic" detritus coming from the internal Alpine zones. Among them, the pebbles of serpentinized peridotites have so far not been studied. We show that they only consist of antigorite serpentinite, implying that they originate from erosion of HT-blueschists. In contrast, the upper Oligocene/lower Miocene molasse, shows mixed clasts of serpentine including antigorite and lizardite without any evidence of chrysotile. This suggests that they were derived from a less metamorphosed unit such as the LT-blueschist unit. Taking into account the sediment transport direction in the basin and the varied metamorphic characteristics of the other ocean-derived detritus, we constrain the lithological nature of the source zones and the location of the relief zones, identified as the internal Alps, SE of the Pelvoux external crystalline massif. Available structural data and *in situ* thermochronological data allow reconstructing the Oligocene to early Miocene collisional geometry of the Paleogene subduction wedge. This phase corresponds to two major phases of uplift evolving from a single relief zone located above the Ivrea body during the early Oligocene and persisting up to the early Miocene; then during the late Oligocene/early Miocene a second relief zone developed above the Briançonnais zone. At that time, the internal western Alps acquired its double vergency.

Keywords: serpentinite pebbles, Raman spectroscopy, source tracing, western Alps, Barrême basin

1. Introduction

Continental collision leads to formation of a convergent mountain belt and to surface uplift. Erosion of this mountain belt feeds clastic sediments to the adjacent basins (e.g. Burbank, 2002). Tracing the different sources of synorogenic detritus provides information on the nature and extent of litho-tectonic units exposed during the successive stages of continental collision. Classic provenance analysis based on sandstone petrography led to identifying several remarkable pebble lithologies, which are of particular importance for discussing the nature and location of potential source areas (e.g. Dickinson & Suczek, 1979; Garzanti *et al.* 2007). More recently, low-temperature thermochronology of detrital grains has developed into a powerful tool to trace the thermal and exhumation history of orogens (see references in Carter & Bristow, 2000; Bernet & Spiegel, 2004; Bernet & Garver, 2005). In the Alps as in other collision belts, the search of ophiolite clasts in synorogenic sediments is traditionally of particular importance, especially if Cr-spinel is abundant as this heavy mineral is characteristic of an ophiolitic source (e.g. Najman & Garzanti, 2000; Garzanti *et al.* 2000). When Cr-spinel are rare or absent, which is the case in the western Alpine synorogenic sediments (Evans & Mange-Rajetzky, 1991), serpentinites mainly derived from hydrated peridotites can be useful to depict ophiolitic sources as serpentinites are particularly resistant to fluvial transport (McBride & Picard, 1987). Usually, serpentinite clasts are seldom studied because of difficulties in determining their mineral species, mostly distinguishing between chrysotile, lizardite and antigorite. However, the mineralogy of serpentinite provides direct information on the ophiolite metamorphic history. The use of Raman spectroscopy allows overcoming the difficulties of serpentine characterization (Rinaudo *et al.* 2003; Auzende *et al.* 2004; Groppo *et al.* 2006). Applied to different ophiolite-bearing thrust sheets along the Queyras-Monviso transect, this method shows that the nature of serpentine minerals evolves along the subduction wedge of the western Alps, in accordance with the different P-T conditions estimated from petrologic metamorphic studies (Auzende *et al*. 2006). On this basis, we propose here to apply the same method to sand grains and pebbles of serpentinite from synorogenic molasse deposits in the foreland basin remnants of the western Alps.

We chose the Barrême basin, a small piggy-back basin with well preserved lower Oligocene to lower Miocene molasse formations (e.g. Evans & Elliott, 1999; Callec, 2001 with references therein). The sampled syn-collisional molasse deposits are known to bear the very first detrital input from the internal Alpine zone, often termed “exotic clasts” (historical references in Chauveau & Lemoine, 1961; Morag *et al*. 2008). The early Oligocene is also the time when these internal zones underwent a major orogenic phase related to what may be regarded as the climax of the collision in the western Alps enhancing relief formations (Lardeaux *et al*. 2006; Morag *et al*. 2008; Bernet & Tricart, 2011). Considering these constrains allows us discussing the possible origin of the analysed detrital serpentinites in the context of the Alpine relief formation, from ~ 34 to 23 Ma ago.

2. Geologic setting

The Alps are located on the boundary between the European and African plates. Alpine evolution along the Eurasia-Africa boundary was initially dominated by plate divergence, which induced Mesozoic continental rifting and ocean opening. Since the Late Cretaceous, plate convergence has resulted in subduction and collision (Rosenbaum & Lister, 2005 and references therein). The structural geometry of the western Alps classically resulted from indentation of the southern margin of Europe by the Adriatic microplate or African promontory after the closure of the intervening Tethyan Ocean (e.g. Coward & Dietrich, 1989; Platt *et al*. 1989; Schmid & Kissling, 2000), associated with strain partitioning in oblique collision context between Adria and Europe (Malusà et al. 2009), and possibly with slab retreat (Vignaroli et al. 2008). In this study we only consider the southern part of the western Alpine arc (Fig. 1a and 1b). The internal zone consists of a pile of metamorphic nappes with dominant Paleogene north- or northwest- directed structures (Choukroune *et al.* 1986). The structure of metamorphic internal zone (Fig. 1b and c) consists of a refolded stack of units derived from the ocean (Piedmont zone) and its European margin (Briançonnais zone) (Tricart *et al*. 2006).

The Briançonnais zone mainly displays late Paleozoic to Mesozoic sediments and pre-Alpine basement rocks. A remarkable nappe stack involves the pre-, syn- and post-rift Tethyan sediments originating from a stretched margin (e.g. Claudel & Dumont, 1999). This stack of cover nappes was shortened during collision and, being a mechanically contrasted multilayer, gave rise to regional west- and east-verging folds and associated thrusts. The latter are known as the Briançonnais backfolds and backthrusts and correspond to the present-day alpine fan-shaped structure (Tricart, 1984) metamorphosed under greenschist facies conditions (Goffé *et al*. 2004; with references therein). The age of backthrusting is debated, and is either ascribed to the Oligocene (Tricart 1984) or to the Eocene (Malusà *et al.* 2005a). The Briançonnais basement consists of pre-Alpine magmatic and metamorphic rocks. Its Permo-Carboniferous sedimentary cover is variably re-worked during the Alpine orogeny. The metamorphic evolution of these basement slices, where upper blueschist and/or eclogite facies conditions have been deciphered (Goffé *et al*. 2004), contrasts with the evolution of the cover nappe pile. These significant metamorphic gaps are consistent with the existence of severe tectonic decoupling between the Briançonnais units.

The composite Piedmont zone (Fig. 1b and 1c) corresponds to the present-day juxtaposition of different structural levels of the paleo-subduction wedge from the blueschist accretionary wedge (Schistes lustrés) to the eclogitized internal crystalline massif. The Schistes lustrés (Fig. 1c) consist of high pressure units including metamorphic marls, clays, and limestones. Marls dominate (calc-schists) and enclose hectometric to kilometric tectonic boudins of

Triassic dolomites or Jurassic ophiolites sometimes interpreted as olistholiths (Tricart & Lemoine, 1986; Deville *et al.* 1992). Burial under blueschist-facies conditions during the Late Cretaceous to early Eocene times built an accretionary wedge that was strongly redeformed when collision relayed subduction in late Eocene time (Agard *et al*. 2002; Lardeaux *et al*. 2006; Tricart & Schwartz, 2006). In details, the metamorphic conditions evolved from low temperature (LT) blueschist conditions in the western part to high temperature (HT) blueschist in the eastern part of the complex (Tricart & Schwartz, 2006; Schwartz *et al*. 2009; Bousquet *et al*. 2008).

The Monviso ophiolitic unit, squeezed between the Schistes lustrés complex and the Dora-Maira internal crystalline massif contains major remnants of the Tethyan oceanic lithosphere that were strongly deformed and metamorphosed under eclogite-facies conditions (Lombardo *et al*. 1978; Schwartz *et al*., 2000; Rubatto & Hermann, 2001) during the Eocene (Duchêne *et al*. 1997). Contrasted eclogitic conditions (e.g., Schwartz *et al*. 2000) indicate that the Monviso massif is an imbricate of units rapidly exhumed (1 cm/yr) within the subduction channel during the Eocene and accreted beneath the Schistes lustrés complex under blueschist-facies conditions at 20–35 km depth (Schwartz *et al*. 2000; 2001). The Monviso eclogites are separated from the Dora-Maira massif by a ductile normal fault (Blake & Jayko, 1990; Philippot, 1990; Schwartz *et al*. 2001).

Located in the lowermost structural position, the Dora-Maira internal crystalline massif corresponds to a stack of more or less deeply subducted continental basement slices involved in a ‘domelike’ structure (Fig. 1c). Here again, significantly contrasted metamorphic conditions have been inferred (Chopin *et al*. 1991; Compagnoni & Rolfo, 2003). Quartz-bearing eclogite facies rocks outcrop at the top of the Dora-Maira dome and overlie a coesite bearing eclogitic unit. This pile of thin (<1 km) high to ultra-high pressure metamorphic units overlies the lowermost Pinerolo-Sanfront blueschist unit along a thrust contact. The latter unit is similar, with respect to their lithologies, structural position and metamorphic evolution to the Briançonnais basement slices.

In contrast to the internal zone, the external zone of the western Alps consists of pre-Alpine crystalline basement (Pelvoux and Argentera massifs) of the proximal European margin and its detached sedimentary cover of the Southern Subalpine Chains (Fig. 1b). The Alpine external zone is less shortened (Gratier *et al*. 1989) and only slightly metamorphosed, ranging up to greenschist facies conditions (Goffé *et al*. 2004). Polystage shortening mainly occurred from the Oligocene onwards (e.g. Ford *et al.* 2006; Dumont *et al.* 2008 with ref. therein). Non-metamorphic and gentle fold-thrust structures first developed in the Provence area until the Eocene in response to N-S compression. Superimposed Alpine structures *sensu stricto* developed from the Oligocene onwards when the effects of the Apulian plate indentation, first restricted to the internal arc, spread outwards through the external arc (Ford *et al*. 1999). From the Middle Miocene onwards up to the Present the external zone remains subject to active shortening while the internal zone undergoes extension (Sue & Tricart, 2003; Tricart et al. 2004).

In front of the internal zone, a flexural basin developed since the middle Eocene, and being the locus of deep sea turbidite sedimentation. The main witnesses to this flysch episode are the Grès d'Annot in the south and the Grès du Champsaur in the north (e.g. Sinclair, 2000; Sissingh, 2001; Ford & Lickorish, 2004) whose sedimentation started in middle and late Eocene times respectively. Everywhere, sedimentation ended abruptly in early Oligocene times, due to emplacement of the Helminthoid flysch nappe (exotic flysch in Fig. 1c; Kerckhove, 1969) coming from the internal zone. The structuring of the western Alpine belt in early Oligocene times was accompanied by enhanced mountain building (see discussion in Morag *et al*. 2008; Bernet & Tricart, 2011). Rapid erosion of this mountain belt might explain the fast regional cooling of the western Alpine belt during the Oligocene and the sudden supply of coarse metamorphic detritus along the northwestern flank of the western Alps (e.g. Morag *et al.* 2008; Bernet *et al.* 2009), and in small basins developed on top of the belt (e.g. Polino *et al.* 1991; Carrapa *et al.* 2003). Nevertheless, the volume of these clastic sediments is globally modest, and the Adriatic foredeep on the internal side of the western Alps was starved of orogenic detritus (Garzanti & Malusà, 2008). As matter of fact, the early Oligocene is also the time of the flysch-to-molasse transition in the European foreland basin when flysch basins closed and were incorporated into the collision wedge. From that time onwards small molasse basins formed in more and more external locations, as the Alpine deformation front migrated outward through the external arc (e.g. Ford *et al*. 1999). This is well illustrated in the Barrême basin (Fig. 1b), where thin Paleogene marine detrital formations (Fig. 2) represent a shallow water equivalent to the more proximal Grès du Champsaur and Grès d'Annot flysch formations (e.g. Joseph & Lomas, 2004). The overlying molasse sediments in the Barrême basin were deposited as the basin was transported on the Digne thrust sheet (de Graciansky, 1972; Artoni & Meckel, 1998; Evans & Elliot, 1999). Subsequently, in late Oligocene the stack of the metamorphic Briançonnais and Piedmont nappes was exhumed in brittle conditions (e.g. Malusà *et al.* 2005b) and transported along the Briançonnais thrust front (fig. 1). Possible modifications of detrital pathways at this stage will be investigated in this paper.

3. The sampled material

The Eocene to early Oligocene so called "Nummulitic" transgressive sequence covered the European foreland of the outward migrating Alpine belt (Sinclair, 1997; Ford *et al*. 1999). The "Nummulitic" series was mainly deposited in a deep and underfilled flexural basin and it generally overlies a continental erosional surface. This sequence always shows the succession of three sedimentary facies of increasing water depth (so called "Nummulitic trilogy"): (1) nearshore, Nummulites-bearing platform limestones; (2) hemipelagic foraminiferal (globigerina) marls (Blue marls Formation), and (3) clastic sediments. In the Barrême, the latter first appeared within the Blue marls Formation as thin turbiditic sandstones, the Grès de Ville Formation. The latter formation is capped by lower Oligocene marine conglomerates (Clumanc and Saint-Lions conglomerates) interpreted as channelized fan delta slope deposits (Artoni & Meckel, 1998; Fig. 2).

In the Barrême basin, the Grès de Ville (Fig. 2), consist of silty mudstone and thin-bedded fine-grained sandstone with an erosive base, graded bedding and cross-stratification. They recorded north-directed paleocurrents thanks to various sedimentary structures (flute cast, grooves crescent and prod marks) and their clastic material originates from the Provence-Corsica-Sardinia domain (Evans & Elliott, 1999; Callec, 2001). By contrast, the Clumanc and Saint-Lions conglomerates are interpreted as a Gilbert delta with west- to SW-directed transport (Callec, 2001). They are dominantly composed of coarse pebbles (generally 5 to 20 cm) with a sandy and fine-grained pebbly matrix (Callec, 2001) (Fig. 3). Different pebble populations were recognized early on by geologist (Termier, 1895 *in* Chauveau & Lemoine 1961; Graciansky *et al.* 1971) and especially Bodelle (1971), and are summerized in *Table* 1.

The Barrême thrust-top west verging syncline, accommodating W-E shortening, was fed by a deltaic system providing local Cretaceous limestone (lowermost Clumanc conglomerate beds) and farther internal Alpine clasts, including metamorphic and eruptive rocks (upper Clumanc beds and Saint-Lions conglomerates) (Fig. 2 and Table 1). The metamorphic clasts of internal origin were initially mentioned by Termier (1895, *in* Chauveau & Lemoine, 1961). These internal (or Penninic) pebbles mainly consist of ophiolitic clasts (oceanic crust or mantle) and of associated pelagic sediments, in particular the easily recognizable red radiolarian cherts (Fig. 3). The ophiolitic clasts are mainly massive or brecciated basalts but some variolithic fragments of pillow-lava cortex and rare gabbros have also been found. Morag et al. (2008) described garnet-blueschist pebbles. The $^{40}Ar/^{39}Ar$ geochronology of phengites from these metamorphic pebbles yielded a 34 ± 3 Ma ages suggesting a short time lag between exhumation of these rocks and their deposition in the lower Oligocene (34-28 Ma) basin. The Penninic provenance of the exotic pebbles in the Clumanc and Saint-Lions conglomerates (Fig. 2 and Table1) is consistent with the observed western to south-western sedimentary transport directions. The latter strongly differs from the previous (Eocene to lowermost Oligocene) north-directed transport which fed longitudinally the Barrême basin from the uplifted Provence-Corsica-Sardinia domain (e.g. de Graciansky, 1982; Evans & Mange-Rajetzky, 1991; Callec, 2001; Joseph & Lomas, 2004). The high amount of serpentinite pebbles we found was unexpected considering the literature (e.g. Chauveau & Lemoine, 1961).

The Grès verts are dated as Aquitanien (Evans & Mange Rajetsky, 1991), which is younger than the Clumanc and Saint-Lions conglomerates (Fig. 2 and Table 1). These fine silty-sandstone and siltstones show crossed-bedding typical for low energy fluvial channels with ephemeral over-bank deposits. The heavy mineral content of the Grès verts is varied, but includes many serpentinite grains (Fig. 3) with a suspected source corresponding to the Embrunais-Ubaye nappes (Evans & Mange-Rajetsky, 1991).

We analysed 38 small serpentinite pebbles, 1 to 3 cm in diameter each, sharing pronounced flat and blunt shape**,** sampled in the “La Poste” conglomerate at Clumanc (Table 2, Fig. 3). More precisely the majority of our serpentinite pebbles come from the lowermost layers, while historically serpentinite pebbles were essentially described in the middle layers of the La Poste conglomerate (e.g. Chauveau & Lemoine, 1961) (Table 1). Therefore, we sampled the very earliest deposits of exotic clasts in the molasse basin, which is important for the subsequent interpretation. In addition, we sampled the stratigraphically younger continental deposits of the Aquitanian “Grès vert” in the same Barrême molasse series. This sandstone formation is exceptionally rich in serpentinite grains and 45 single grains were analyzed for this study (Table 2, Fig. 3).

4. Methodology: Raman spectroscopy

Serpentinite minerals are hydrous phyllosilicates (up to 13 wt% water) formed during the hydration of basic and ultrabasic rocks. Serpentinites, shown here with a simplified structure formula of $(Mg, Fe^{2+})_3\ Si_2O_5(OH)_4$, are made of superposed tetrahedral and octahedral layers. The structures of the three serpentinite species lizardite, chrysotile and antigorite are distinguished by different distortions and spatial arrangements of the tetrahedral and octahedral layers. Layers are flat in lizardite, rolled up in chrysotile and corrugated in antigorite (e.g. Wicks & O'Hanley, 1988). Natural serpentinites sampled in high-grade metamorphic environments show antigorite as the most abundant species (Scambelluri *et al*. 1995, Guillot *et al*. 2000; Auzende *et al*. 2002; Groppo & Compagnoni, 2007). Experimental studies have also shown that antigorite is the stable variety under high pressure conditions (Ulmer & Trommsdorff, 1995; Wunder & Schreyer, 1997; Auzende *et al*., 2004). Lizardite and chrysotile are the major varieties of pseudomorphic textures observed in low-grade metamorphic serpentinites from oceanic lithosphere or ophiolites (Andréani *et al*. 2007). In this study, the different serpentinite species were characterized by Raman spectroscopy, coupled with petrographic observations on polished thin section of clasts. Raman spectroscopy was performed at the Laboratoire des Sciences de le Terre, Ecole Normale Supérieure de Lyon (ENS-Lyon), France, with a Horiba Jobin-Yvon LabRam HR800 apparatus. The light source was an argon-ion laser to achieve 514.5 nm wavelengths. An Olympus™ BX30 open microscope equipped with ×50 and ×80 long-working distance objectives was coupled to a spectrometer to focus the laser beam to a 3 μm spot diameter. The Raman signal was collected in the backscattered direction. Acquisition time was about 60s, distributed over three accumulation cycles, with a laser output power on the sample adjusted between 10 and 60 mW, depending on the intensity of the signal. Raw or polished serpentinite clast samples were indifferently used. The spectral resolution was 1cm^{-1} using 1800 gr/mm grating. The spectral regions from 150 to 1150 cm^{-1} and from 3600 to 3720 cm^{-1} (Fig. 4) were investigated because they include lattice vibrational modes characteristic for serpentinite species (Rinaudo *et al*. 2003; Auzende *et al*. 2004; Groppo *et al*. 2006). The bands detected in these spectral regions are indicative of the molecular and crystalline structure of the sample. For all spectra the assignments of the band position and band width were determined using the Peakfit© software.

In the low frequency region, four main peaks near 230, 390, 690 and 1100 cm^{-1} characterize the spectra of lizardite and chrysotile (Rinaudo *et al*. 2003; Groppo *et al*. 2006). At lower frequencies, intense antigorite peaks occur at 226, 373, 680 and 1043 cm^{-1}. Therefore, the peak positions are different in comparison to other serpentines, and the peaks are much broader (Fig. 4). The peak at 1043 cm^{-1} allows distinguishing easily the antigorite variety. Although weak, differences between chrysotile and lizardite can be clearly identified because of the sharpness of the Raman lines. In particular, a single high frequency band at 1100 cm^{-1} is observed in chrysotile whereas several convoluted bands are observed between 1060 and 1100 cm^{-1} in lizardite (Rinaudo *et al*. 2003 ; Groppo *et al*. 2006). In the high frequency region (Fig. 4), the OH vibrational mode shows a spectrum with an intense single peak at 3697 cm^{-1} in the case of chrysotile and a bimodal shape spectra for the lizardite and antigorite (Fig. 4). The spectrum of lizardite is characterized by a fine bimodal shape with a primary band at 3685cm^{-1} and a secondary band at 3703 cm^{-1}. The antigorite species presents a spectrum broader than the lizardite with a primary band near 3365 cm^{-1} and a secondary band centered at 3697 cm^{-1}. One to twenty Raman spectra were acquired on each lower Oligocene pebbles and one spectrum was acquired on each single grain coming from the upper Oligocene/lower Miocene sandstones (Table 2).

5. Serpentinite mineralogy

For the 38 serpentinite clasts from the Clumanc conglomerate that we analyzed, all the Raman spectra are typical for antigorite (Fig. 5a). No other variety of serpentinite was observed. This observation is confirmed by petrographic thin section analysis. The initial ultrabasic rocks were fully recrystallized and the antigorite minerals form a typical interlocking texture that replaces olivine. The original magmatic pyroxene was totally replaced by oriented antigorite flake aggregates constituting bastite (Fig. 6).

In the Aquitanian sandstone “Grès vert” 45 single grains were analyzed, the Raman spectroscopy analysis of single grains (Table 2) shows a mixture of dominant lizardite and subordinate antigorite. More precisely, lizardite is the only serpentine species presents (sample MB153, Fig. 5c and Table 2) in the fine grained, well sorted sandstone layers. In contrast, the moderately to poorly sorted sandstone layers, with a larger variety of grain sizes in the fine to coarse grained range, display 20% of antigorite in the coarser fraction (sample MB154, Fig. 5c and Table 2) but lizardite still dominates (sample MB154, Fig. 5d and Table 2). Nowhere chrysotile and talc were detected.

6. Discussion

6.1. Origin of serpentinite pebbles and sand grains

Despite the difficulty of precisely establishing the P-T stability field of serpentine species (e.g. Evans, 2004) (Fig. 7a), an empiric approach has been developed by comparing the worldwide occurrence, including the western Alps, of serpentinite species with the P-T conditions recorded by associated metamorphic rocks in HP-LT metamorphic environments

(Guillot *et al*. 2000; 2009; Auzende *et al*. 2002; 2006; Groppo *et al*. 2006). In summary, chrysotile and lizardite form under greenschist facies conditions in oceanic crust or ophiolites, while lizardite alone testifies to upper greenschist and low-temperature blueschist facies conditions (Fig. 7a). Antigorite progressively replaces lizardite under intermediate blueschist facies conditions. Antigorite becomes the exclusive serpentinite variety under high-temperature blueschist to eclogite facies conditions (Fig. 7a).

Thus, antigorite-bearing serpentinite pebbles from the lower Oligocene Clumanc conglomerate are only derived from HP-LT ophiolitic rocks metamorphosed under high temperature blueschist or eclogite facies conditions, i.e. the internal zone (Fig. 1 and Fig. 7b). As paleocurrent indicators favour a NE provenance for the detrital grains, the source of these pebbles has to be more precisely searched in the SE vicinity of the Pelvoux massif and not in more southerly HP-LT metamorphic assemblages such as the Voltri massif, presently exposed to the south of the Tertiary Piedmont basin (Fig. 1b). Pure antigorite serpentinites are an important component of the HT-blueschist Schistes lustrés unit along the drainage divide between France and Italy (Fig. 7b). Pure antigorite serpentinites also crop out further east, in the Monviso eclogitic complex in Italy (Schwartz *et al*. 2000; Auzende *et al*. 2006).

The occurrence of both lizardite and antigorite grains in the upper Oligocene/lower Miocene serpentinite-bearing sandstones (Grès verts) contrast with the sole occurrence of antigorite pebbles in the Lower Oligocene conglomerates. This suggests that the coarser antigorite fraction in the upper Oligocene/lower Miocene sandstone could derive from the same source, i.e. the HT-blueschits Schistes lustrés (or the Monviso eclogites) (Fig. 7a and 7b). Nevertheless, the occurrence of lizardite suggests a different source area, a less metamorphosed ophiolitic source, having escaped subduction and collision processes and only recording oceanic hydrothermal metamorphism. In the present-day Alpine framework, only the upper ophiolitic unit in the Chenaillet massif (Montgenèvre pass, upper Durance valley) escaped the subduction-collision metamorphic imprints, representing a part of the Tethyan oceanic lithosphere obducted onto the subduction wedge, towards the European continental margin (Goffé *et al*. 2004; Chalot-Prat, 2005; Schwartz *et al*. 2007). An alternative interpretation is that both grains (antigorite and lizardite) were derived from an unique source in where lizardite and antigorite coexisted. This has been observed in the present-day eastern part of the Piedmont Schistes lustrés complex (Fig. 7b), which was metamorphosed only under low-temperature blueschist facies conditions (Agard *et al*., 2001; Goffé *et al*. 2004; Tricart & Schwartz, 2006). Concerning the antigorite-bearing Upper Oligocene/Lower Miocene sands, one cannot preclude the possibility that they partly result from recycling of Oligocene detrital sediments.

6.2. The heterogeneity of sources in terms of metamorphic grade

Contrary to the serpentinite clasts, other Penninic pebbles within lower Oligocene conglomerates have been analysed in the lower 70s (e.g. de Graciansky *et al*. 1971). The basalts and their derivatives (“prasinite”, “diabases”) essentially bear zeolite or greenschist facies metamorphic assemblages, more rarely a blueschist facies imprint. The rare gabbros bear different types of blueschist assemblages, “calcschist” pebbles that likely originate from the Schistes lustrés complex are also observed (Morag *et al*. 2008). The abundant red-purple pebbles of radiolarian cherts only show a very low-grade metamorphic imprint as radiolarites are preserved (Cordey *et al.* 2012).

The upper Oligocene/lower Miocene sandstones contain a mixture of grains of serpentinites, unmetamorphosed radiolarites and LT (lawsonite bearing) blueschist basalts. As for the lower Oligocene molasse, the upper Oligocene/lower Miocene molasse shows detrital grains were derived from source areas that experienced contrasted metamorphic conditions from those of the LT-blueschist facies to low grade metamorphic conditions.

This heterogeneity both in the lower Oligocene and upper Oligocene/lower Miocene molasse deposits implies the erosion of a stack of ophiolite nappes that recorded contrasted P-T conditions, having the lithologic and metamorphic signatures of the Paleogene alpine accretionary wedge (Agard *et al*. 2002; Lardeaux *et al*. 2006; Yamato *et al*. 2007; Guillot *et al*. 2009).

The provenance of detrital sediments can be discussed. Indeed, it is classically admitted that the source of ophiolitic clasts in the Lower Oligocene molasse of the Barrême basin was located in the Embrunais-Ubaye nappes (e.g. Evans & Mange- Rajetzky, 1991). In the present-day situation, these nappes represent the closest outcrops of the internal or Penninic units, in an E to NE direction from the Barrême basin, which is the provenance direction of exotic detritus according to sedimentological analyses (Callec, 2001). Because these nappes are devoid of any HP-LT metamorphic imprint, they cannot represent the source of HP-LT metamorphic detritus like the antigorite-bearing serpentinite we sampled in the Barrême basin.

6.3. The sudden arrival of mixed metamorphic clasts into the Barrême molasse basin

The sudden erosion in early Oligocene times of the stack of variably metamorphosed nappes inherited from the Paleogene accretionary wedge fits well with two observations concerning the internal zone, SE of the Pelvoux massif.

First**,** structural analysis allows identifying a severe early Oligocene shortening. The corresponding post-nappe folds and thrusts are associated with the second and third regional schistosities in the Briançonnais zone. They display respectively outward and inward vergence, resulting in the so-called Briançonnais fan-shaped structure (Tricart *et al*., 2006 with reference therein). In the whole internal zone, it has been proposed that this major syncollision shortening was responsible for the bending of the arc (Thomas *et al*. 1999; Collombet *et al*. 2002).

Second, detrital thermochronology analysis of Barrême Oligocene molasse recently allowed to estimate very short lag times from fission-track ages for zircons grains (Bernet &Tricart, 2011; Jourdan *et al.* 2011), in consistence with $^{40}Ar/^{39}Ar$ ages of phengites in pebbles from the same sediments (Morag *et al*. 2008). Comparison to *in situ* thermochronological data acquired from the internal zone led Bernet &Tricart (2011) and Jourdan *et al.* (2011) to propose erosion-driven fast exhumation (1-3 mm/y) in early Oligocene times. Tectonic exhumation is likely to have been enhanced by erosional processes. Consequently, at that time, these internal zones are supposed to have formed an elevated and narrow mountain range. In summary**,** converging approaches allow us proposing that the source of mixed metamorphic and very low-grade metamorphic ophiolitic detritus suddenly fed the Barrême molasse basin during early Oligocene times was an elevated and possibly narrow mountain range (Fig. 8a). The volume of Oligocene sediments preserved in the proximal alpine foreland (Barrême basin) is quite low but a much larger amount of sediments is pounded within the Oligocene grabens of the Liguro-Provençal rift, the Rhône valley and the Gulf of Lion (Séranne, 1999).

6.4. Implications for the evolution of the internal western Alps

Looking for the potential source areas of the sand and pebbles of antigorite-bearing serpentinites in the Barrême basin molasse deposits we took into account different constrains relative to the metamorphic grade, structural and orogenic history of the internal zone, but also sedimentological constrains relative to the Barrême basin stratigraphy. The source we are looking for, must present lithologies such as those presently exposed in the Piedmont zone. As a working hypothesis we consider a generally N-S trending, elevated, subaerially exposed accretionary wedge (Platt, 1986; 1987; 1993) which experienced syncollisional shortening and unroofing during early Oligocene times (Fig. 8a). Its shape probably corresponded to the present-day internal zone, but extended outwards to the presently exposed exotic flysch nappes (Fig. 1b and 1c). The pile of shallow-dipping nappes displays intense fold and thrust structures, due to Lower Oligocene shortening, especially westwards directed structures (Fig. 8). Nevertheless, the whole structure was inherited from the Paleogene accretionary wedge and displays a general metamorphic gradient. The base of the nappe pile corresponds to Subbriançonnais and Briançonnais nappes with high pressure (lawsonite-bearing) greenschist metamorphism, being covered by the Piedmont nappes inherited from the Paleogene accretionary wedge. In the frontal part, the LT-blueschist facies unit cropped out, while HT-blueschists were already exhumed (Fig. 8) in the steep rear part of the internal wedge (Yamato *et al.* 2007).

The location of eclogitic units within the early Oligocene structure deserves a specific discussion. Geometrically, they possibly could have topped this wedge, outcropping along the highest parts of the corresponding mountain range, knowing that the antigorite-bearing clasts may as well originate from both blueschist- and eclogite-bearing nappes. Up to now no evidence for the erosion of eclogites was found in the Barrême basin. Presently, eclogites are only exposed along the Italian flank of the Alps forming the present Monviso massif (Lombardo *et al*. 1978; Schwartz *et al*. 2000). Further to the north they outcrop in the high Maurienne valley (Rolland *et al*. 2000) and in the Zermatt-Saas zone (Barnicoat, 1985). At the opposite and also very far away, eclogites outcrop in the Voltri group of Liguria, where their first detrital reworking in the Tertiary Piedmont basin is stated at early Oligocene times (Federico *et al*. 2004). Concerning the timing of exhumation along our transect, eclogites boulders do not appear in the Torino Hills conglomerates, at the internal foot of the mountain range, before the early Miocene (Polino *et al*. 1991). Fission-track analyses in the Monviso massif confirm that these eclogites were exhumed later than Miocene times and were structurally located beneath the Schistes lustrés accretionary wedge (Schwartz *et al*. 2007; Yamato *et al*. 2007). By early Oligocene times the eclogitic unit forming the serpentinite subduction channel was not yet being exhumed along the studied transect (e.g. Guillot *et al*. 2009). The only exposed HP-LT metamorphic rocks were the blueschists coming from the Schistes lustrés accretionary subduction wedge. The variation of serpentine species from antigorite to lizardite between early Oligocene and late Oligocene/early Miocene in the Barrême basin is interpreted as a shift of the paleo-relief through time enhancing drainage system modifications. During the early Oligocene the prominent relief was probably located above the HT-blueschist zone (Fig. 8a) progressively a second zone of reliefs developed westward above the LT-blueschist zone (Fig. 8b). Nevertheless, in the lower Miocene Torino Hills conglomerates, the abundant arrival of decimetric to metric blocks of blueschists, with serpentinites representing more than 50% of the detrital sediments and the first arrival of eclogites (2%) (Polino *et al*. 1991), suggests that rapid surface uplift enhanced erosion and created high relief persisting in the vicinity of the present-day Torino Hills.

Thus we propose the following scenario to explain the mineralogical evolution recorded on both flanks of the young Alpine orogenic system during Oligocene and early Miocene times. The transition from subduction to collision by early Oligocene time induced the rapid and prominent aerial exposure of the eastern part of the HP-LT accretionary wedge (Fig. 8a) and predominantly fed the external molasse basins. From the early Oligocene to the early Miocene, the relief shifted westward due to the uplift of the frontal part of the collisional wedge (Briançonnais zone) in response to the activation of the Briançonnais thrust front (Fig. 8b). This renewed topography and dynamics enhance erosion of the external part of the accretionary wedge (LT-blueschist). In the eastern part, the vertical indentation at the rear of the accretionary wedge related to the emplacement of the Ivrea body (Schmid and Kissling, 2000) allows the progressive exhumation of eclogites and focused erosion fed the internal molasse basins in the Po plain (Torino Hills and Tertiary Piedmont basin in Fig. 1b). At that time (late Oligocene to early Miocene), the western Alps progressively acquired its doubly vergent structure (Fig. 8b).

7. Conclusions

Sands and conglomerates derived from blueschist facies metamorphic rocks have some characteristic minerals such as glaucophane that are poorly resistant to transport or diagenesis in a sedimentary basin. Serpentinite, hitherto little studied, may be a more reliable marker of HP-LT source rocks in provenance studies. In order to quickly identify serpentine species in sedimentary rocks we propose to use Raman spectroscopy on single detrital clasts. We successfully applied this method to detrital serpentinite from the Barrême basin, in the western Alpine foreland. Mineralogical analysis by Raman spectroscopy shows that serpentinite pebbles from the lower Oligocene conglomerates in the Barrême basin consist entirely of antigorite, indicative of high-temperature blueschist facies metamorphism.

During the early Oligocene, paleocurrent indicators indicate a shift from a southern provenance (Provencal shelf and the Corso-Sardinian massif) to a north-eastern provenance of detrital grains in the Barrême molasse basin. Thus the source of the lower Oligocene pebbles has to be searched in the internal part of the Alps, SE of the Pelvoux massif. The arrival of such exotic clasts reveals the erosion of a suddenly raised mountain range in the internal Alpine zones along the same transect, the Piedmont cordillera, as a consequence of major shortening pulses. The coexistence of metamorphic pebbles (abundant antigorite clasts but also rare blueschist gabbros) with non-metamorphic radiolarite clasts constrains the lithologic composition and structure of the sediment source area. The presence of metamorphic ophiolitic material contradicts the classical view that the only source of exotic clasts in the lower Oligocene Barrême molasses basin is located in the Embrunais-Ubaye flysch nappes that glided in front of the Alpine wedge.

On the contrary we propose that the supply of exotic clasts supply to the Barrême basin is a direct consequence of the major syncollision shortening experienced by the Paleogene accretionary wedge in early Oligocene times, by 34-28 Ma, to the SE of the Pelvoux massif. The remnants of this refolded subduction wedge corresponds to the present-day deeply eroded internal zone. The absence of eclogite clasts on both flanks of the early Oligocene collidera means that eclogitic units were not exhumed during early Oligocene times along the Monviso transect, contrary to what is known along the more southerly Liguria transect.

The same mineralogical analysis by Raman spectroscopy was also conducted on serpentinite sand grains from the upper Oligocene/lower Miocene Grès verts of the Barrême basin. The analyzed clasts consist of antigorite and dominant lizardite, to some part depending on grain size, indicative of a change through time in source rock lithology. This observation is compatible with a westward propagation of the cordillera with erosion of lesser metamorphic units during ongoing collision. Simultaneously, the increasing arrival of HP ophiolitic materials in the Po plain suggest the persistence of high reliefs in the internal zone, maintained by deep vertical indentation of the Ivrea body.

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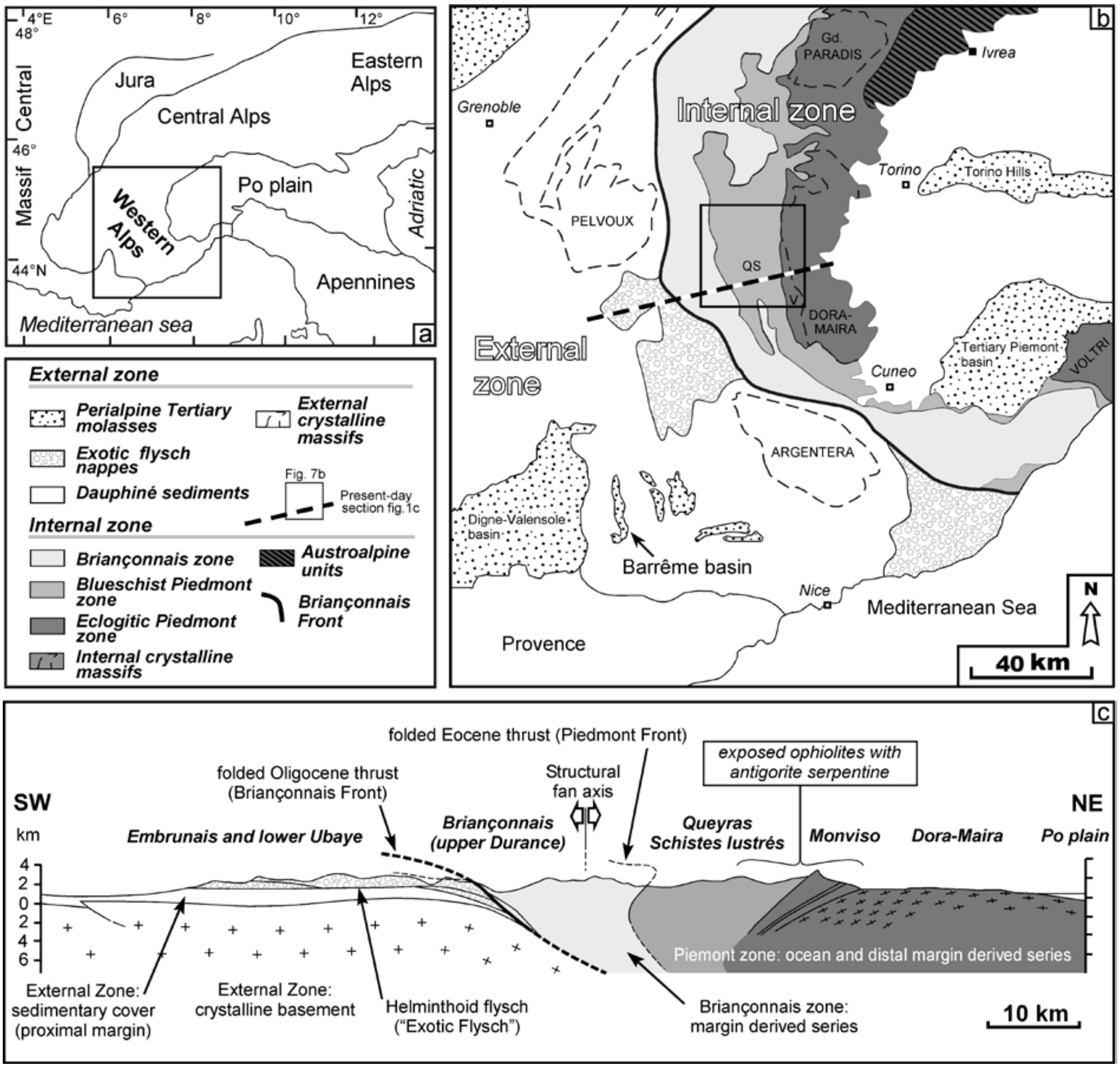

Figure 1. – Geological setting of the study area. (a) Geographical setting of the western Alps showing location of (b). (b) Tectonic sketch map of the western Alps showing the Barrême basin in the external zone. The internal zone is bounded by the Penninic deep crustal thrust to the west with: QS-Queyras Schistes lustrés; V-Monviso. The rectangle locates Figure 7b. (c) Sketch-section of the present-day structure. Presently, metamorphic serpentinite with antigorite but without lizardite (HT-blueschists and eclogites) crops out along both sides of the watershed (Eastern Queyras Schistes lustrés, Monviso).

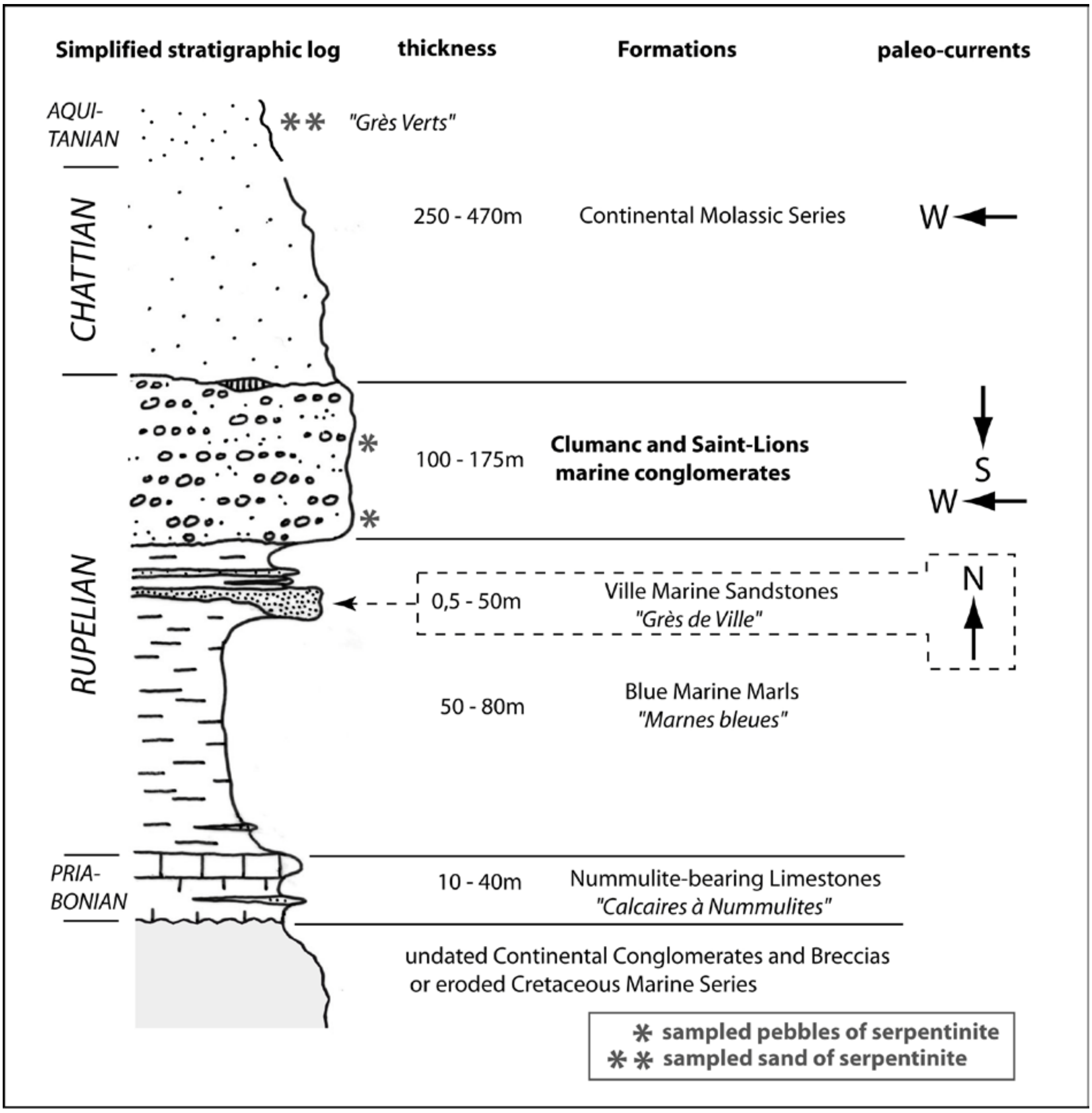

Figure 2. – Simplified stratigraphic log of the Barrême basin molasse series. Depositional ages are from Callec (2001).

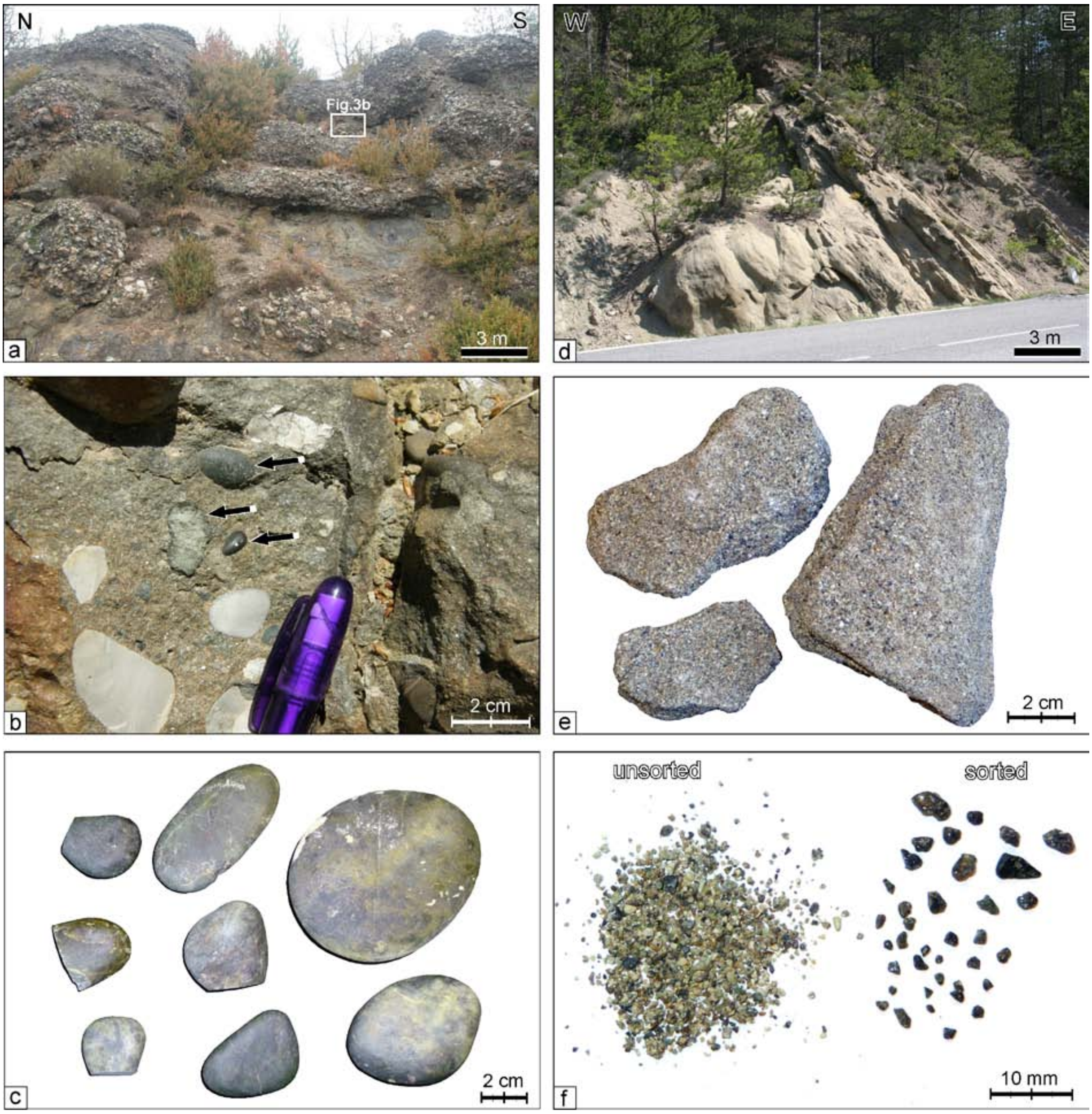

Figure 3. – Conglomerates and sandstones from the Oligo-Miocene Barrême basin. (a) Outcrop view of the upper layer of the Clumanc Formation showing coarse conglomerate channels with erosive base and graded bedding incised in sandstones. The latter are interpreted as turbiditic deposits (Artoni & Meckel, 1998; Evans & Elliott, 1999; Callec, 2001). (b) Close view of matrix-supported pebbles in coarse sandstones. Centimetric serpentinite pebbles are underlined by black arrows, others are limestones. (c) Investigated serpentinite pebbles: size ranges from 3 cm to 6 cm. (d) Outcrop view of the Grès verts formation: coarse sandstone-filled channels interbedded in flood-plain fine-grained silty-sandstones are interpreted as fluvial channels and over-bank deposits, respectively (Evans & Elliott, 1999). (e) Facies of the Grès verts sandstones. (f) Isolated grains of the Grès verts Formation, bulk composition to the left and sorted serpentinite grains to the right.

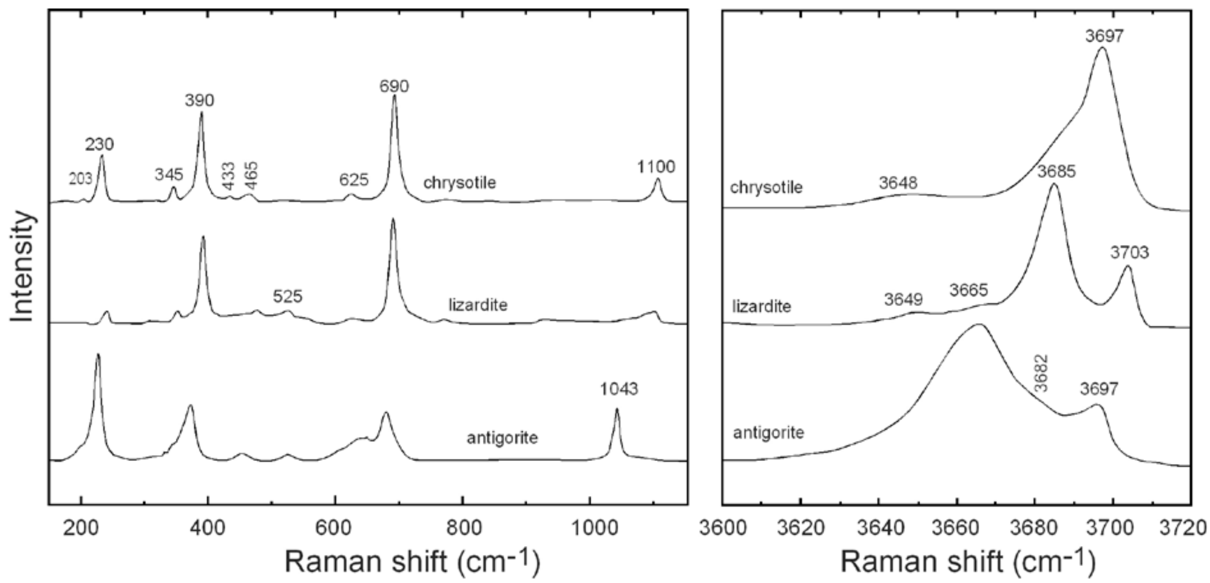

Figure 4. – Characteristic Raman spectra for the different varieties of Alpine serpentinite with band assignments according to Rinaudo *et al*. (2003), Groppo *et al*. (2006), and Auzende *et al*. (2006).

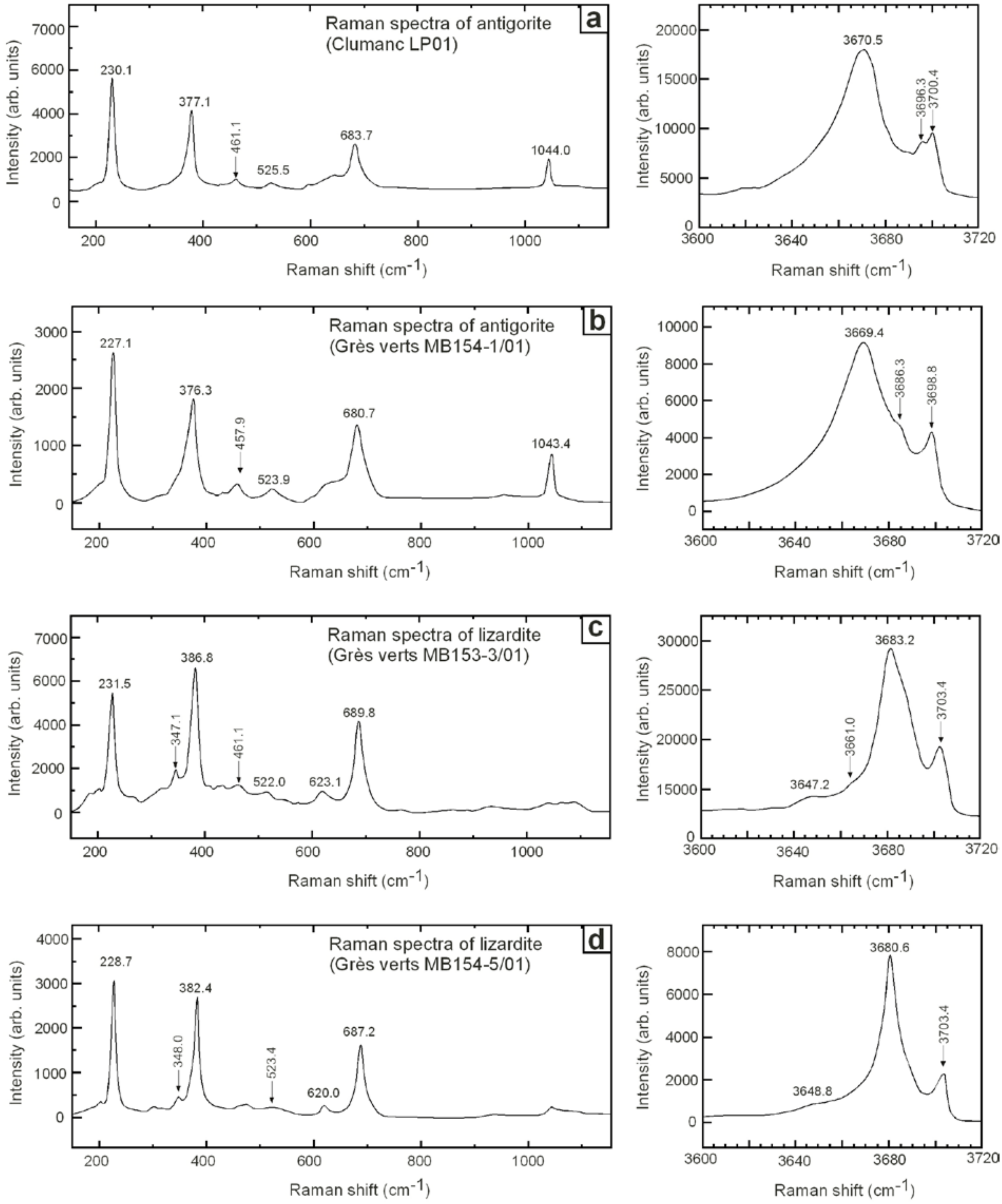

Figure 5. –Typical Raman spectroscopy spectra of serpentinite varieties obtained from detrital ultramafic clasts (LP01) or sand grains (MB153 and MB154) from the Oligocene-Miocene Barrême basin. For each Raman spectra the signal was analyzed on two acquisition spectral windows and the characteristic bands were indicated. (a) and (b)-Raman spectra of antigorite obtained respectively from the Clumanc conglomerate (sample LP01) and from the coarse to fine grained sand fraction of the Grès verts (sample MB154). (c) and (d)- Raman spectra of lizardite obtained respectively from the coarse grained fraction of sample MB154, and from the homogeneously fine grained sandstone sample MB153.

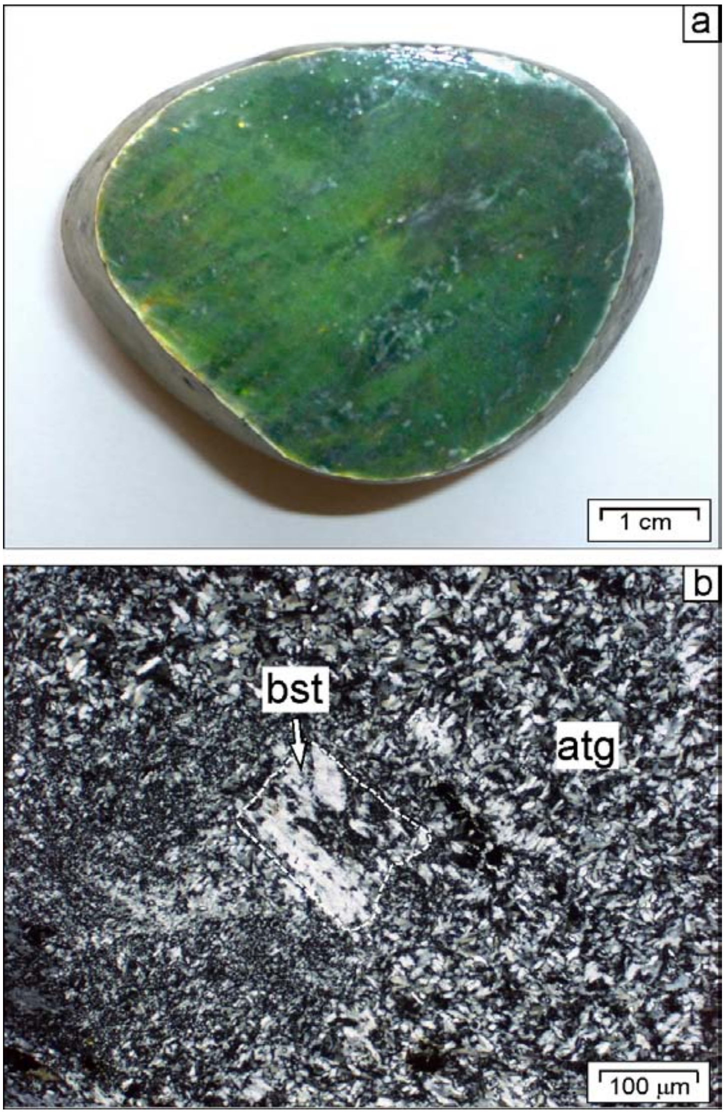

Figure 6. – (a) macroscopic view of the serpentinite pebble from Clumanc conglomerates (LP01 sample). (b) Typical microstructures observed in thin section under a polarizing optical microscope (crossed polarized). The matrix was full recrystallized into flakes of antigorite (atg) with an interlocking structure. The primary peridotitic pyroxene was replaced by oriented antigorite flake aggregates corresponding to “bastite” antigorite (bst).

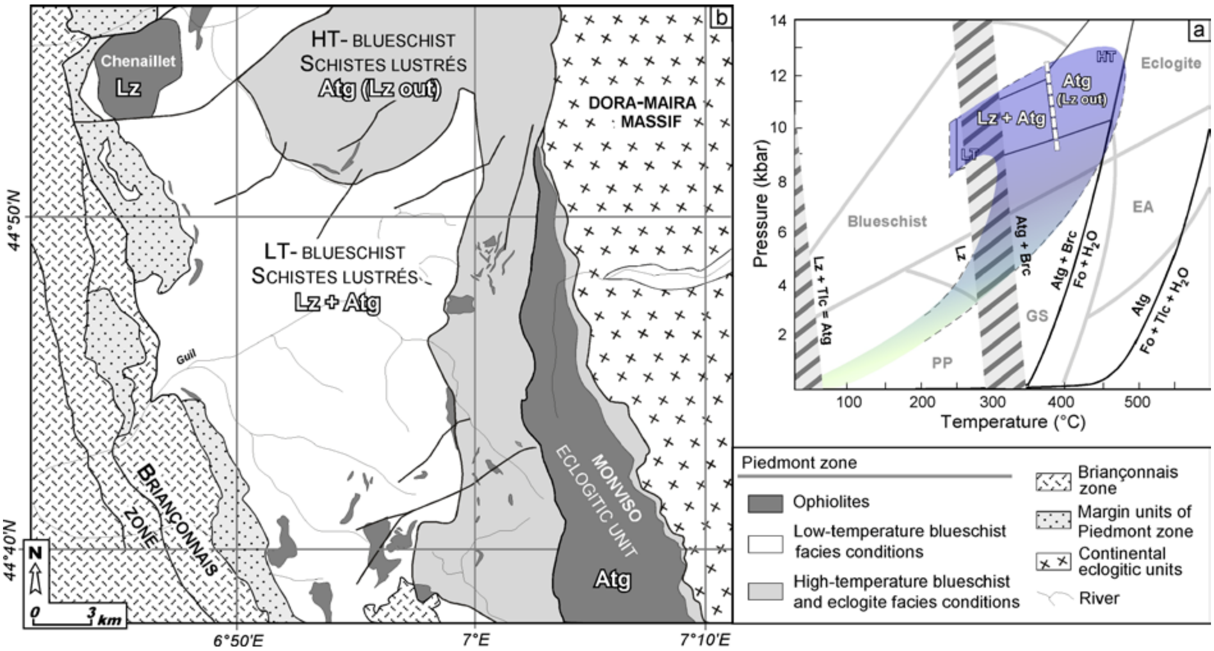

Figure 7. – (a) Phase diagram of antigorite and lizardite (after Evans, 2004) with Lz-lizardite, Atg-antigorite, Tcl-talc, Brc-brucite, Fo-forsterite. Up to 380°C the lizardite is entirely destabilized. The Blueschist pressure-temperature (*P*-*T*) conditions in the Queyras Schistes lustrés are indicated. Metamorphic facies are from Spear (1993): PP-Prehnite Pumpellyite, GS-greenschist, EA-epidote amphibolite. (b) Simplified tectonic of the Schistes lustrés of the Piedmont zone. This domain is bounded by the Briançonnais zone to the west and by the internal crystalline massif of Dora Maira and by the Monviso eclogitic unit to the east. The Schistes lustrés complex corresponds to a calcschist-rich accretionary wedge, enclosing scattered ophiolitic bodies, metamorphosed under HP facies conditions, from LT-blueschist facies conditions (Lizardite and antigorite bearing serpentinite) to the west to HT-blueschist facies conditions (only antigorite bearing serpentinite) to the east.

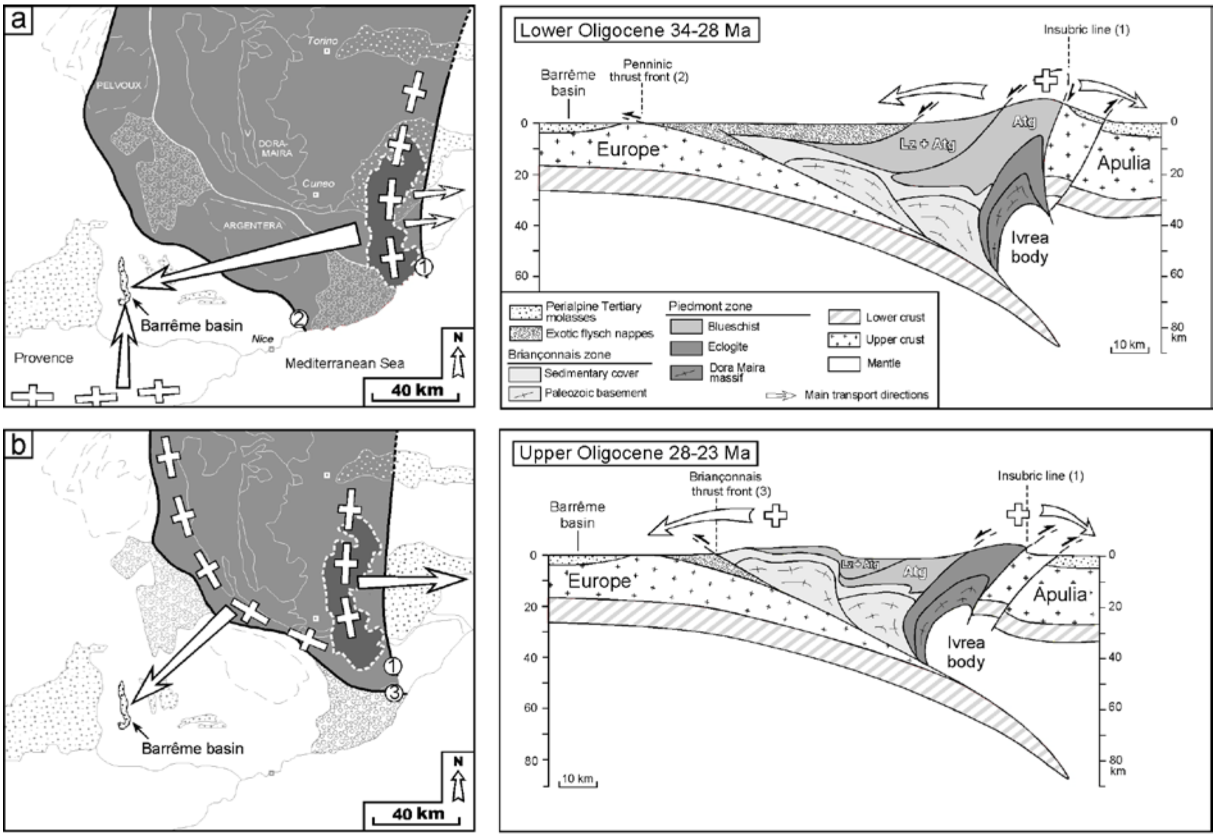

Figure 8**.** – Palinspastic maps and cross sections reconstructions showing the evolution of the relief zones from early to late Oligocene (based on Schmid & Kissling, 2000; Ford et al. 2006). (a) During the early Oligocene the main relief zones (white cross) were located above the Dora Maira massif and in Provence. These two main reliefs zones feed the Barrême basin while the Tertiary Piedmont basin and Torino Hills are only feed by the internal alpine relief in blueschist clasts. (b) During the late Oligocene the activation of the Briançonnais thrust front allows the formation of a westward relief zone feeding the Barrême basin while in the internal alpine zone the persistence of high relief feeds the Tertiary Piedmont basin and the Torino Hills in eclogitic clasts.

sampling locality	Sedimentary rock	Metamorphic rock	Andesite	Eruptive rock
Clumanc's lower and middle layers	99%, essentially Cretaceous limestone (generally 15 cm)	Quartzite, serpentinite	rare	**gabbro, dolerite**, granite, mircodiorite, rhyolite
Clumanc's upper layer	80% silt and quartz, 5-15% of Cretaceous limestone**. Some radiolarite**, sandstone	>1%, Migmatite, quartzite, serpentinite	15%	4%, **gabbro (there where euphotide), dolerite**, granite, microdiorite, microgranite, pillow-lavas cortex
Saint-Lions conglomerate	85%. Essentially calcareous sandstone, 2-3% Cretaceous limestone, **radiolarite**	Rare	3%	12%; granite, **pillow-lavas cortex, microdiorite, microgranodiorite**

Table 1. – Pebbles population density of Clumanc and Saint-Lions conglomerates, modified after Bodelle (1971). The lower layers of the lower Oligocene Clumanc conglomerate contain mainly limestone pebbles, and a lower proportion of metamorphic and eruptive rock. The upper layers contain more exotic rocks such as granitoïds and various ophiolititic pebbles (gabbros, pillow-lavas cortex, dolerites and serpentinites).

Samples	nature	formation *(see figure 2)*	latitude	longitude	Number of spectra	Serpentinite species
PT6011.1	1 pebble	Clumanc conglomerate	44°02'10" N	06°22'26" E	20	100% Atg
PT7064A.1 *to* PT7064A.4	4 pebbles	Clumanc conglomerate	44°02'10" N	06°22'26" E	80	100% Atg
LP01 *to* LP03	3 pebbles	Clumanc conglomerate	44°02'10" N	06°22'26" E	60	100% Atg
LP10 *to* LP13	4 pebbles	Clumanc conglomerate	44°02'10" N	06°22'26" E	80	100% Atg
PT9012.01 *to* PT9012.26	26 pebbles	Clumanc conglomerate	44°02'10" N	06°22'26" E	30	100% Atg
MB153	20 fine grains	"grès verts" (green sandstones)	43°57'17" N	06°24'03" E	20	100% Lz
MB154	25 coarse grains	"grès verts" (green sandstones)	43°57'17" N	06°24'03" E	25	80% Lz / 20% Atg

Table 2. – Location and petrologic nature of serpentinite pebble samples in the Barrême basin. The proportion of antigorite (Atg) vesrus lizardite (Lz) present in pebbles is statistically indicated.