

# Zeolite assemblages from Northern Patagonian Andes, Argentina: A review

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## ABSTRACT

*In this work very low-grade metamorphic assemblages found in the Northern Patagonian Andes (Argentina) are summarized. On the basis of previous studies, several occurrences of zeolites have been delimited. According to their mineralogy, textures and structures, three different alteration stages that evidence a progressive decrease in temperature have been established. The first one (stage I) was the consequence of an event of regional metamorphism that reached the greenschist facies (350 °C and 2 kbar). During this stage, pyroxene, amphibole and feldspar primary phenocrysts broke down to produce an assemblage of actinolite, grossular-andradite, chlorite, albite, prehnite, titanite, clinozoisite, epidote and calcite. In the subsequent stages, direct hydrothermal precipitation took place as the temperature decreased. Thus, zeolites, calc-silicates, calcite, quartz and cristobalite started to precipitate as cavity fillings. During stage II, temperature decreased below 220 °C and wairakite, yugawaralite, laumontite, pectolite, dachiardite, celadonite, thomsonite, pumpellyite and interstratified chlorite/smectite crystallized. Prehnite, adularia, titanite and albite were also deposited but only as minor species. These minerals mainly alter feldspar phenocrysts and fill amygdalites of basalts and andesites. Stage III is characterized by a temperature drop (below 180 °C) and by the crystallization of hydrothermal secondary minerals within open spaces. Most of the alkaline zeolites were deposited during this last event of alteration, filling joints in metabasites and granitoids. Although Ca-stilbite is the most abundant alteration mineral, analcime, natrolite, barrerite, offretite, chabazite, stellerite, heulandite, mordenite, scolecite, mesolite, quartz, calcite, cristobalite and smectites were also produced.*

*Key words: zeolites, low-grade metamorphism, metabasites, hydrothermal alteration, Northern Patagonian Andes, Argentina.*

## RESUMEN

*En esta contribución se compilan las paragénesis de bajo grado metamórfico que se hallaron en los Andes Patagónicos Septentrionales (Argentina). Sobre la base de estudios previos, se delimitaron numerosas localidades caracterizadas por la abundancia de zeolitas. De acuerdo con sus composiciones mineralógicas, sus texturas y sus estructuras se reconocen tres estadios diferentes de alteración que reflejan un progresivo decrecimiento de la temperatura. El primero de ellos fue consecuencia de un metamorfismo regional que alcanzó condiciones de esquistos verdes (350 °C y 2 kbar). Durante este evento, la alteración de fenocristales primarios de feldespatos, piroxenos y anfíboles produjo actinolita, grosularia-andradita, clorita, albita, prehnita, titanita, clinozoisita-epidoto y calcita. En los estadios sucesivos, se incrementa la precipitación directa de minerales a partir de soluciones hidrotermales a medida que la temperatura*

disminuye. Así, comenzaron a precipitar zeolitas, calcosilicatos, calcita, cuarzo y cristobalita rellenando cavidades. Durante un segundo evento, la temperatura descendió por debajo de 220 °C, ocasionando la cristalización de wairakita, yugawaralita, laumontita, pectolita, dachiardita, celadonita, thomsonita, pumpellyita y estratificados de clorita/esmectita. Sólo en cantidades subordinadas se formaron prehnita, adularia, titanita y albita. Estos minerales alteran fenocristales de feldespato y llenan vesículas en basaltos y andesitas. El último evento tuvo lugar cuando la temperatura descendió por debajo de 180 °C y se caracteriza por la cristalización tardía de minerales secundarios de baja temperatura dentro de cavidades. Durante este período precipitó el mayor número de zeolitas alcalinas, llenando cavidades en metabaltos y granitoídes. Si bien la estilbita-Ca es el mineral más abundante del tercer evento de alteración, también se encuentran analcima, natrolita, barrerita, offretita, chabazita, estellerita, heulandita, mordenita, escolecita, mesolita, cuarzo, calcita, cristobalita y esmectitas.

*Palabras clave:* zeolitas, bajo grado metamórfico, metabasita, alteración hidrotermal, Andes Patagónicos Septentrionales.

## INTRODUCTION

Very low grade metamorphism is a widespread phenomenon associated with major processes that take place in the shallower levels of the Earth's crust (Frey and Robinson, 1999). Temperatures and pressures up to 400 °C and 4 kbar produce changes in the textures, chemistry and mineralogy of the rocks. As zeolites can be produced not only by metamorphism but also by hydrothermal alteration processes, they become one of the most important group of minerals to understand the record of changes imprinted in altered rocks. On the other hand, over the past 40 years, some zeolites have been used in agriculture, waste water treatment, gas separation, building constructions, as catalysts, energy storage collectors and in medicine (Deer *et al.*, 2004). Even though they have been described in different provinces of Argentina, Patagonian Andes offer the greatest varieties and some areas where they can become a profitable resource.

It has long been recognized that the uplift of the Northern Patagonian Andes is the result of the subduction of oceanic crust beneath the South American western margin, from the Late Paleozoic to present times. Changes in the rate of plate convergence and ridge collisions produced the main structural and petrological characteristics, which led to a division into two different sectors (Figure 1a): A northern one (Lat 38° - 43° S), where the study area is located, is characterized by a set of Paleogene igneous rocks with superimposed very low grade metamorphic assemblages, and a southern one dominated by Cretaceous volcanism (Ramos, 1999).

In the northern sector, several studies have been carried out by different authors to determine the thermal anomaly and the hydrothermal fluids associated with metamorphism. The study of the secondary assemblages allows to determine the temperature and pressure of the process that affected the rocks of this area. These data are useful to understand the geological evolution of the region, as well as to determine zones where zeolites may be a valuable resource not only for the industry, but also for further studies on mineralogy and petrology.

In light of the reasons presented above, during the last

fifteen years, secondary minerals present in the Northern Patagonian Andes have been the focus of numerous research studies. Previous surveys allowed us to characterize each assemblage, their mineralogy (specially the zeolites species) and the *P-T* conditions. Due to the fact that enough information about the secondary assemblages of many different occurrences have been already obtained, the aim of the present study is to make the first review of the sub-greenschist metamorphism between Lat 38°30' and 44°00' S. Thus, all the secondary minerals, their textures, structures and assemblages, as well as the characteristics of their wall rock, are here summarized. By analysing these data, changes in the temperature and pressure and in the amount of hydrothermal fluids over the time are estimated.

## METHODOLOGY

Although different instruments were used for each one of the contributions considered in this review, the methodology employed was the same. Preparation of thin and polished sections, X-ray diffraction analysis and microanalyses were accomplished in order to identify each mineral species. X-ray diffraction patterns were collected between  $2\theta = 4^\circ - 70^\circ$  in 0.05 steps using Cu-K $\alpha$  radiation (50 kV, 30 mA). Most of the analyzed samples had to be concentrated, separating minerals by hand-picking under a binocular microscope. Special care was taken to obtain pure samples to avoid mineral mixtures. Microanalyses were mainly carried out with a scanning electron microscope (SEM - Phillips 9100), at 20 kV, coupled to an energy-dispersive X-ray spectrometer (EDS), which allowed to obtain qualitative chemical analysis.

## GEOLOGICAL SETTING

The main morphotectonic units exposed in the Patagonian Andes are the consequence of two main periods of subduction along the western margin of Gondwana. The

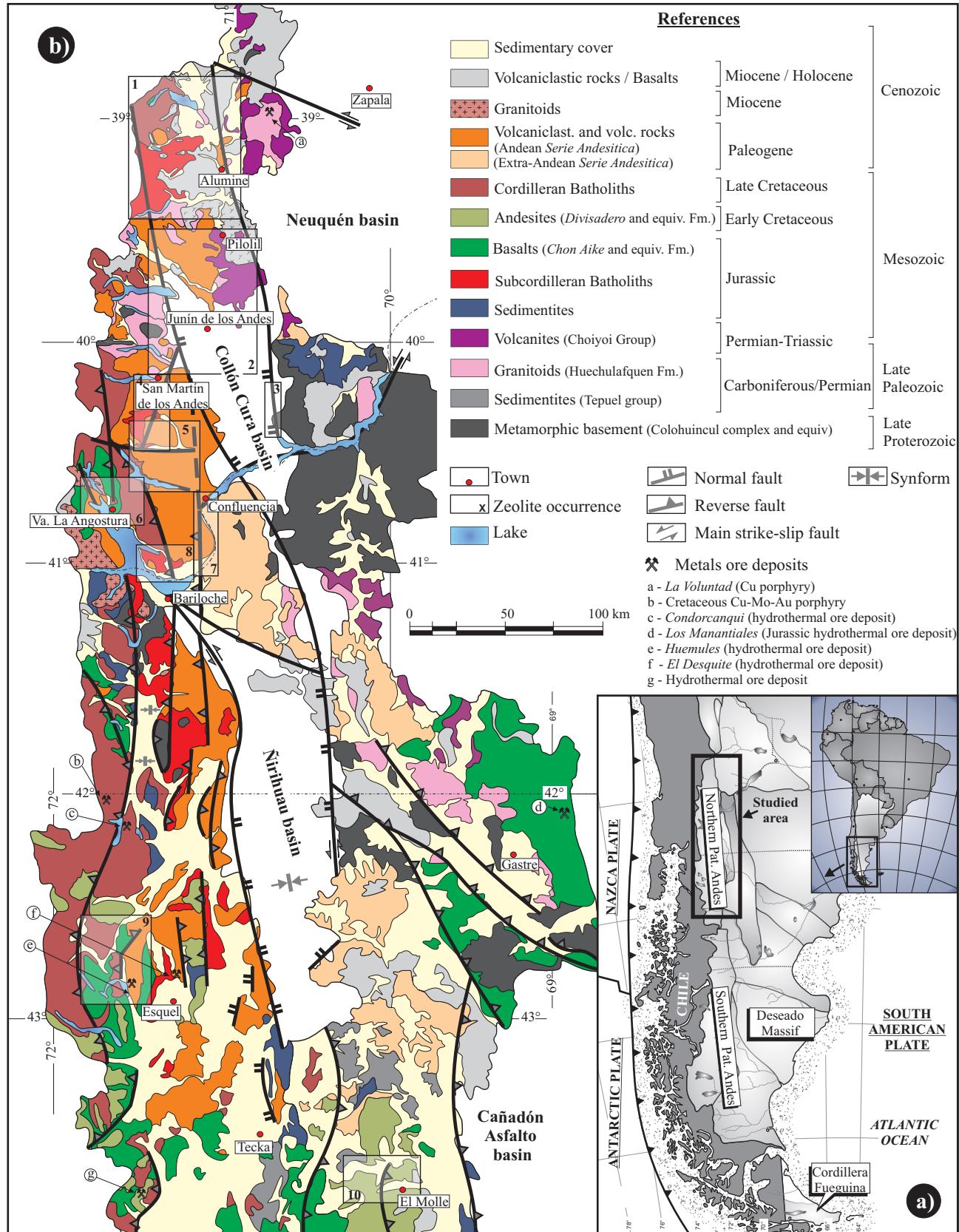


Figure 1. Outline map of the study area showing its different geological units, the major structures and the most important localities where the secondary assemblages described in this paper were sampled (indicated by numbered rectangles); localities are listed in Table 1.

oldest one, known as Gondwanic Magmatic Cycle, affected the central part of northern Patagonia (easternmost study area), whereas the youngest one was developed along the western margin of South America producing the present cordillera, where the study area is included. The Andean Orogeny (as this last tectono-magmatic cycle is known) could have started around the Late Paleozoic according to the oldest magmatic rocks related to the subduction of proto-Pacific oceanic crust below the South American western margin (Hervé *et al.* 1998).

Igneous rocks formed by arc volcanism were widely distributed and emplaced on a metamorphic basement composed of gneisses, schists, migmatites, amphibolites and tonalites (Dalla Salda *et al.*, 1999). These metamorphic rocks are grouped into the Colohuincul Complex, whose ages are constrained into the Late Proterozoic (Turner, 1965; Linares *et al.*, 1988; Dalla Salda *et al.*, 1999). As this metamorphic basement and the overlying Phanerozoic sedimentary cover are not associated with the mineralogy studied in this paper, their detailed characteristics are not going to be described in this work. In the following section, we will focus on the most significant features related to volcanic and intrusive units which are associated with very low grade metamorphic assemblages.

### Volcanic units

From Late Permian to Middle Triassic, extensional processes in the central part of South America produced bimodal magmatism, which is represented by the Choiyoi Group (Groeber, 1946; Kay *et al.*, 1989; Llambias, 1999). Despite the fact that this unit covers approximately 200,000 km<sup>2</sup> of the Argentinian territory, in the study area it only appears as small outcrops surrounded by metamorphic and igneous rocks. In the Patagonian Andes, the Choiyoi Group is restricted to the northern sector (Figure 1b), where it mainly consists of andesitic plateaus and volcanic breccias (Turner, 1976).

Jurassic volcanic rocks (Chon Aike, Lonco Trapial, Huemul, Montes de Oca, Piltriquitrón, Lago la Plata, El Quemado formations and equivalents) mainly occur in the southern and central part of the Patagonian Andes (Figure 1b). According to Uliana *et al.* (1985), this volcanism was a consequence of the steepening of the subducting oceanic crust that produced extension throughout the outer part of the plate, coetaneously to arc volcanism, along the western border of South America. Thus, basalts and andesites located at the eastern slope of the Andes, along the study area, were produced during this period.

Cretaceous andesites and rhyolites (Divisadero formation) overlie the previous units from Lat 44°33' S towards the south (Figure 1b). These volcanic rocks are locally separated from the Jurassic volcanism by sedimentary rocks of the Rio Mayo basin, which represent a back-arc basin formed during regional extension (Ramos

*et al.*, 1982; Franzese *et al.*, 2003). Jurassic and Cretaceous volcanism belong to the same magmatic arc, and hence both units have calc-alkaline affinities (Ramos *et al.*, 1982; Uliana *et al.*, 1985).

Andesites, basalts, dacites and ignimbrites (Auca Pan, Ventana, Nahuel Huapi and Huitrera formations), grouped in the Serie Andesítica, were formed from the Paleocene to the Oligocene. This unit mainly crops out in the northern sector of the study area (Figure 1), with more than 1,500 m thickness, and has calc-alkaline affinity (Dalla Salda *et al.*, 1981; Rapela *et al.*, 1984). According to Rapela *et al.* (1984), two different units are comprised in this group: Andean Serie Andesítica, composed of andesites generated during the Eocene, and extra-Andean Serie Andesítica which is 200 m thick and is mainly composed of Paleocene ignimbrites. Their geochemical signatures evidence that they are related to the magmatic arc formed by the subduction of the Nazca Plate beneath the South America western margin (Dalla Salda *et al.*, 1981).

In Miocene times, the magmatic arc moved towards the west, producing ignimbrites and tuffs (Collón Curá Formation) as a consequence of repeated felsic explosive events (Mazzoni and Benvenuto, 1990). During the Pliocene, the magmatic arc migrated westward from the Northern Patagonian Andes (Rapela *et al.*, 1984). Today, the Quaternary volcanic front is associated with the Liquiñe-Ofqui fault system located in Chile (Muñoz and Stern, 1989; Lara *et al.* 2001). In the study area, Quaternary basalts only occur as small basaltic plateaus placed at the top of the ranges.

### Intrusive units

Carboniferous granitoids (Huechulafquen complex) appear as small outcrops in the northern sector of the study area (Figure 1b). These granitoids might indicate the beginning of tsubduction in the western margin of the Patagonian Andes, or could be considered as the consequence of the subduction-collision in the Patagonia-Gondwana proto-margin (Ramos 1984; Dalla Salda *et al.*, 1991).

During Jurassic and Cretaceous times, meanwhile andesites, dacites and rhyolites had been erupted onto the surface, plutonic rocks were being emplaced beneath them (Haller and Lapido, 1982). The Andean Patagonian Batholith, composed of granitoids and minor diorites and gabbros, was the result of this igneous activity (Ramos *et al.*, 1982; Bruce *et al.*, 1991; Pankhurst *et al.*, 1999, Lizuain, 1999). Although the ages of the batholith vary between 160 and 8 Ma, Rapela and Kay (1988) recognized five major pulses that tend to be younger westwards (González-Díaz, 1982). According to their ages, compositions and geographic position, Gordon and Ort (1993) recognized two different batholiths: Sub-Cordilleran Batholith and Cordilleran Batholith (Figure 1b). The former presents Jurassic ages, smaller sizes and mainly occurs towards the east of the

foothill axis. Conversely, the latter presents Cretaceous ages and comprises the core of the Patagonian foothills throughout 2,200 kilometers. However, despite their differences, both batholiths present similar magmatic arc signatures (Gordon and Ort, 1993; Pankhurst *et al.* 1999; Rapela and Kay, 1988).

Finally, Miocene granitic stocks (Coluco unit) were found to the west of the Northern Patagonian Andes. These rocks represent the last intrusions generated by the Tertiary Andean arc magmatism, and are emplaced along the axis of the Liquiñe Ofqui fault system (González-Díaz, 1982; Lavenu and Cembrano, 1999) (Figure 1b).

## DISTRIBUTION AND GENERAL ASPECTS OF ZEOLITES OCCURRENCES

Along the Northern Patagonian Andes the alteration processes that produced zeolite-rich assemblages seem to be concentrated in some restricted areas (Figure 1b). During the last years, several studies were carried out in these localities in order to determine the mineral assemblages and the crystallization sequences. Table 1 summarizes the names of the localities affected, their geographic coordinates, the

composition of the host rocks and the main contributions published about each ones. The aim of this section is to establish the common features presented by the secondary assemblages in all these areas.

There is a clear relationship between the composition of the host rock and the mineralization that took place in each occurrence (Vattuone and Latorre, 1990). Granitoids only present some joints lined by alteration minerals of 1 to 5 mm wide. The breakdown of plagioclase originated albite and calcium-rich zeolites, whereas chlorite, epidote and prehnite were produced by the alteration of primary amphibole and biotite. On the contrary, in basic and intermediate volcanic rocks, the secondary minerals completely fill cavities and replace primary minerals (Figure 2). The groundmass, as well as phenocrysts, are pervasively altered to albite, Ca-zeolites, pumpellyite, phyllosilicates, quartz and cristobalite. Nevertheless, in these volcanic rocks, secondary minerals mostly occur concentrated in joints (forming veins of few centimeters wide by several centimeters long; Figures 2c-2e), in vesicles (Figure 2f, 2g), or in cavities that can reach more than 10 centimeters (Figure 2a, 2b).

Even though all Jurassic, Cretacic and Tertiary volcanic rocks present alteration assemblages, Paleogene

Table 1. Characteristic features of the studied localities. Geographic coordinates, wall rock compositions and contributions for each one of the localities indicated in the Figure 1.

Localities	Lat S	Long W	Wall Rocks	Main References
1 Alumine Rucachoroi	39°14'14" 39°13'41"	70°55'03" 71°10'36"	Jurassic and Cretaceous volcanites (Aluminé Formation) and Miocene basalts (Rancahue Formation)	Lagorio <i>et al.</i> (2001); Latorre and Vattuone (1996); Vattuone (1990); Vattuone and Latorre (1990); Vattuone <i>et al.</i> (1996b); Martínez Dopico <i>et al.</i> (2004); Gallegos <i>et al.</i> , (2008)
2 Pilo Lil Junín de los Andes	39°38'28" 39°56'36"	70°56'49" 71°04'28"	Miocene basalt and andesites (Chimehuin Formation and equivalents)	Vattuone <i>et al.</i> (1999); Vattuone <i>et al.</i> (2005b; 2008)
3 Collón Cura	40°23'44"	70°38'31"	Oligocene basalts (Collón Cura Formation)	Latorre and Vattuone (1994a)
4 Pio Proto San Martín de los Andes Chapelco	40°06'50" 40°09'10" 40°18'25"	71°10'36" 71°21'17" 71°23'11"	Paleogene basalts (Serie Andesítica Formation)	Vattuone and Latorre (1994, 1996a); Vattuone <i>et al.</i> (1997; 1999; 2001b); Tourn and Vattuone (2002); Vattuone and Tourn (2002)
5 Meliquina Paso Córdoba	40°19'18" 40°36'50"	71°21'01" 71°10'18"	Paleogene andesites and tuffs (Serie Andesítica Formation)	Latorre and Vattuome (1995); Vattuone <i>et al.</i> (1996a)
6 Villa La Angostura	40°38'42"	71°37'35"	Jurassic basalts (Montes de Oca Formation), Paleogene andesites (Serie Andesítica) and Cretaceous granitoids (Los Machis Formation)	Depine <i>et al.</i> (2003); Leal (1999); Latorre and Vattuone (1994b)
7 Confluencia	40°54'13"	71°01'20"	Tertiary basalts and andesites (Serie Andesítica Formation)	Vattuone <i>et al.</i> (2001a, 2001c)
8 Huemul	40°57'49"	71°19'19"	Metabasites of Jurassic age (Montes de Oca Formation) and Paleogene basalts (Ventana formation)	Gargiulo (2006); Gargiulo y Vattuone (2008)
9 Futalaufquen Cholila Nahuel Pan	42°49'18" 42°29'55" 42°57'20"	71°36'41" 71°30'01" 71°12'43"	Cretaceous volcanites (Divisadero Formation)	Vattuone <i>et al.</i> (2000b, 2002, 2005a, 2006); Vattuone and Latorre (2002a, b)
10 El Molle	43°43'50"	70°02'06"	Cretaceous basalts (Tres Picos Formation)	Vattuone <i>et al.</i> (2000a); Vattuone and Latorre (1999)

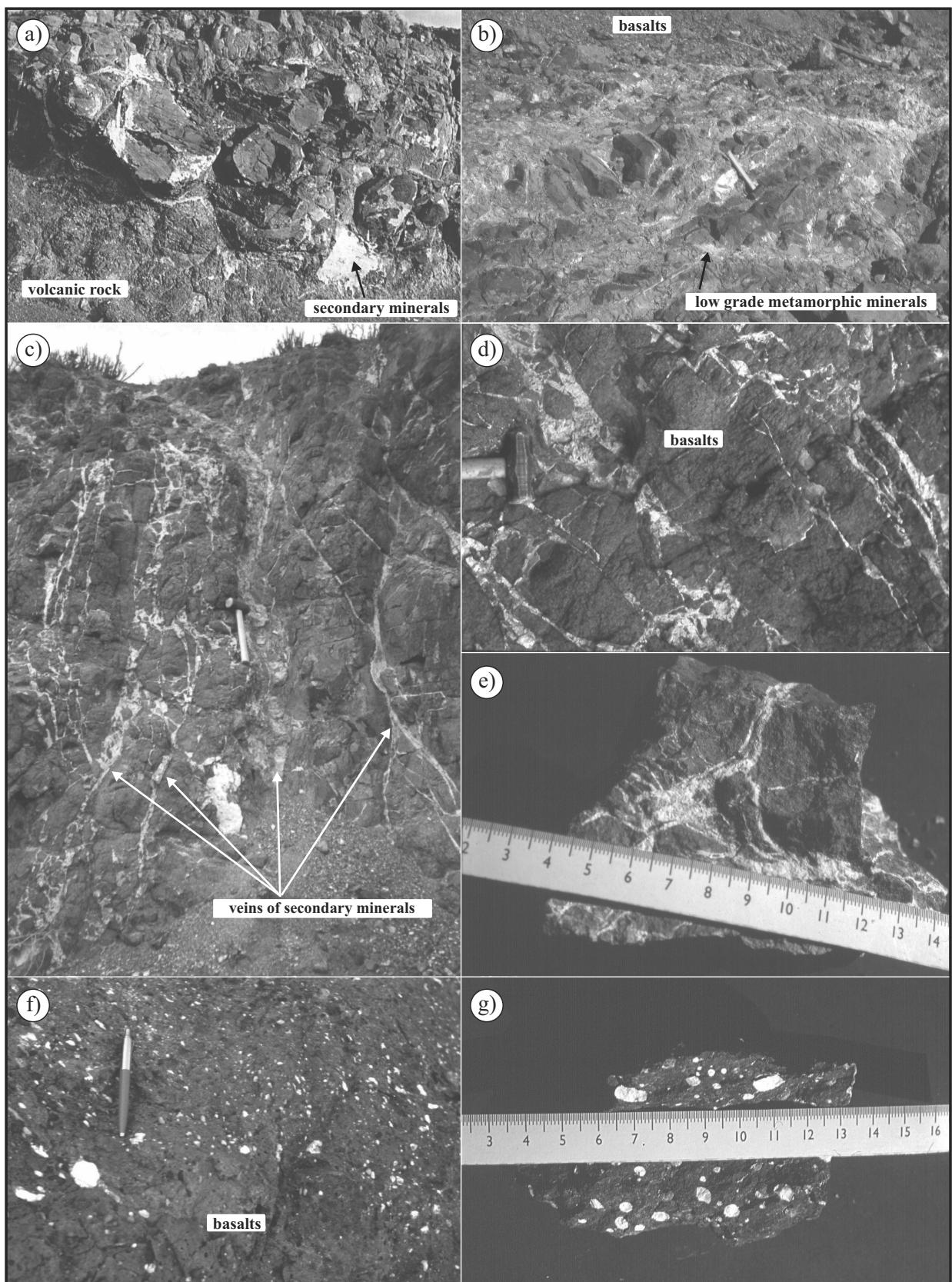


Figure 2. Photographies of the most common secondary textures. a, b: Basalt outcrops from localities 1 and 7 with advanced alteration to low grade minerals; c-e: veins filled with zeolite assemblages, from localities 1 and 10; f, g: vesicles filled with secondary minerals from locality 2.

tuffs, basalts and andesites are the most altered units in the Northern Patagonian Andes (Figure 1b, localities 5 and 7). These volcanic rocks present interstratified smectite/chlorite (s/c) produced by olivine alteration, whereas albite, zeolites and epidote were generated by the breakdown of plagioclase (Vattuone and Latorre, 1990). Thus, the greatest concentrations of zeolites and other secondary minerals are associated with volcanic rocks, which have higher permeability than the granitoids.

The relationship between these very low grade metamorphic assemblages and ore deposits in the Northern Patagonian Andes has not been clearly established. Ore deposits that occur in this sector of the Andes have been objects of few studies. Garrido and Domínguez (1999) studied La Voluntad porphyry copper (*a* in Figure 1b), which is related to Permian stocks (281±4 Ma). According to Ametrano *et al.* (1979) and Pezzutti and Genini (1999), the Condorcanqui deposit represents another copper anomaly and consists of quartz veins with bornite, chalcopyrite, chalcocite, digenite and pyrite hosted by Cretaceous andesites (*c* in Figure 1b). Near this last deposit, a Mo-Cu-Au porphyry occurs hosted in the metamorphic basement (Genini, 1999) (*b* in Figure 1b). Towards the south, a Paleogene porphyry copper (Huemules deposit) composed of pyrite, galena, sphalerite, chalcopyrite, covellite, pirrhotite and bornite associated with gold and silver occurs (Viera and Hughes, 1999) (*e* in Figure 1b). Additionally, gold and silver anomalies associated with sulfide mineralization, hosted in andesitic rocks of Jurassic age, constitute an hydrothermal deposit named El Desquite (Soechting, 2002) (*f* in Figure 1b). Finally, Pesce (1978) reported hydrothermal mineralization rich in pyrite, chalcopyrite and galena hosted in volcanic rocks but linked to Cretaceous granitoids (*g* in Figure 1b).

It is worth emphasizing that zeolite assemblages are associated with ore mineralization only in some restricted localities (number 6, 7 and 8, see Figure 1b). In these areas, native copper, pyrite and chalcopyrite appear disseminated in the volcanic rocks or lining joints. Besides, close to locality 4, zeolites coated by native copper and cuprite were found (Figure 3a, Vattuone *et al.*, 1996b; Tourn and Vattuone, 2002). Further studies focusing on the ages of ore deposits and their secondary minerals would be necessary to conclude about any possible linkage between zeolite precipitation and the metal mineralizations of the Northern Patagonian Andes.

## ZEOLITES ASSEMBLAGES

As far as we know, nearly 20 different zeolites and many other secondary minerals occur in several occurrences of the Northern Patagonian Andes. Due to the fact that its evolution started as regional metamorphism and ended up as small thermal anomalies, secondary minerals were produced by metamorphic reactions or by direct precipitation from hydrothermal fluids. Their mineral assemblages

and their textures allow us to recognize three main different stages (Figure 4). It is worth emphasizing that even though each one of these stages represents a specific range of *P-T* conditions it does not mean that all the minerals listed in each column of the Figure 4 are in equilibrium. These stages only represent groups of minerals formed by the same geological process that produced similar, but not equal, conditions along the study area, and therefore more than one assemblage constitute each column, according to the wall-rock composition and the chemistry of the fluids ( $PO_2$ ,  $PCO_2$ , pH, etc.) in each locality. In this section, these assemblages, their textures, structures and reactions will be described.

### Stage I

Regional metamorphism was the first process that affected the igneous rocks of the study area (Figure 4, stage I). This event affected all rocks, but in different ways according to their chemical composition. Minerals generated during this stage only occur replacing few phenocrysts and represent the oldest secondary assemblages. It is worth emphasizing that although the variety of mineral species produced by the metamorphism is broad, their number is scarce. The assemblages associated with this first stage evidence a gradual temperature decrease from under greenschist conditions to zeolites facies.

Vattuone and Latorre (1990) and Lagorio *et al.* (2001) described hedenbergite-diopside, grandite garnet and orthoclase as the highest metamorphic assemblages generated by breakdown of primary minerals under amphibolite facies conditions (Figure 3b). These minerals occur in few localities and were only recognized in Jurassic volcanic rocks.

Epidote + chlorite + albite + quartz + magnetite + titanite + grandite garnet + K-feldspar assemblages suggest that the metamorphic grade decreased to greenschist facies. Associations like these were determined by Vattuone and Latorre (1990) and Vattuone *et al.* (2005a) in Cretaceous volcanic rocks around the localities 1 and 9 (Figure 1). According to Deer *et al.* (1986) an association of grandite garnet, magnetite and epidote allowed us to estimate temperatures below 325 °C and pressures above 2 kbar.

Even though pumpellyite-actinolite facies are less common, tremolite-actinolite + epidote-clinozoisite + pumpellyite + chlorite assemblage occurs in the locality 9 (Figure 1) (Vattuone *et al.*, 2005a). Temperatures and pressures for mineral associations like these were established by Schiffman and Liou (1980) at around 250 °C and 1.5 kbar, respectively.

Prehnite-pumpellyite facies occurs in the locality number 4, indicating a temperature decrease. Under this condition, prehnite + pectolite + chlorite + laumontite + pumpellyite + albite crystallized inside Jurassic and Paleogene volcanites (Figures 3c, 3d).

Finally, zeolites facies characterizes the end of the

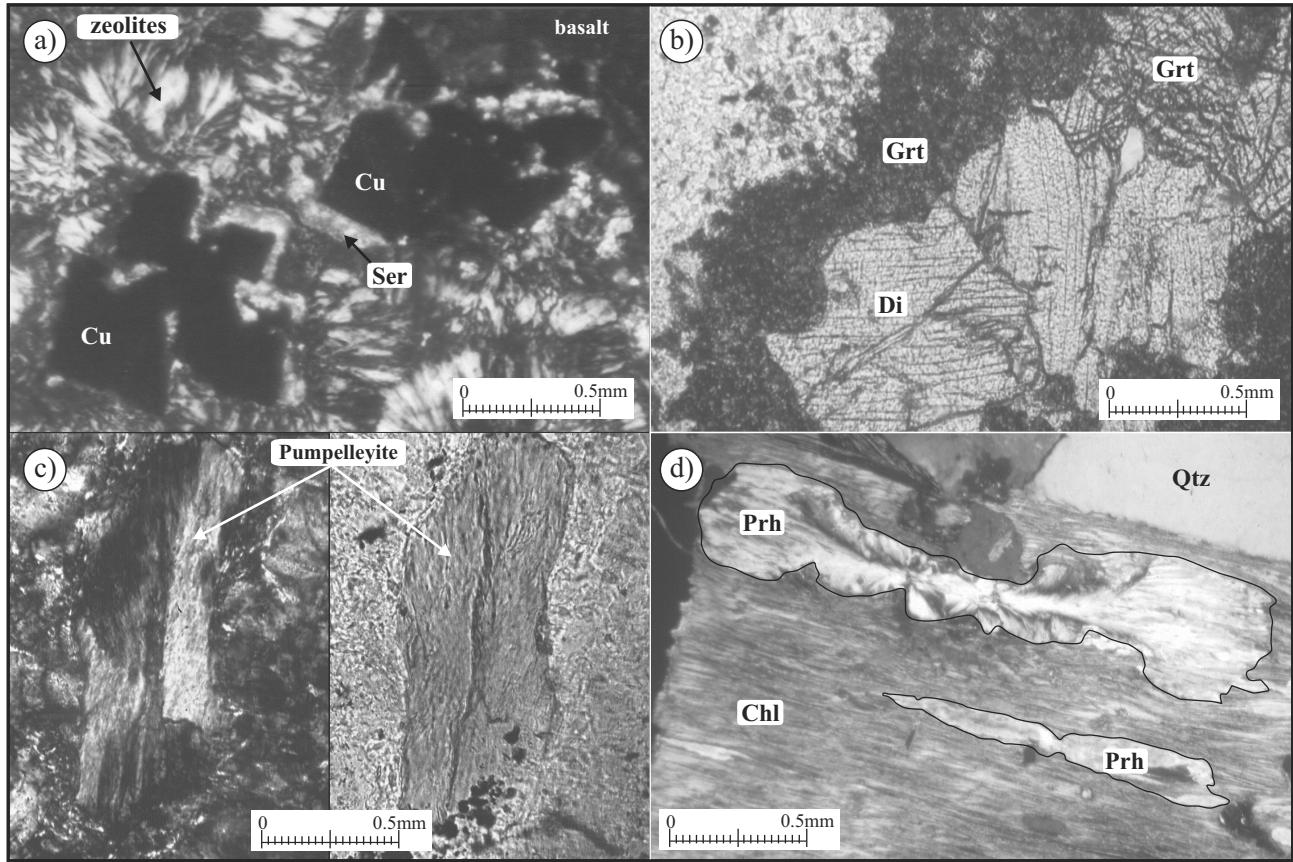


Figure 3. Microphotographies of the most characteristic assemblages. a: Basalt with native cooper, sericite and zeolites found in locality 1; b: diopside and garnet crystals in a metabasite from locality 1; c: twinned crystal of pumpellyite; d: prehnite and chlorite formed by complete alteration of a primary biotite.

metamorphic process. Epidote + albite + chlorite + interstratified c/s + laumontite and, to a lesser extent, adularia + pumpellyite + prehnite + heulandite + mordenite occur affecting the same volcanic units (localities 4, 7 and 9, Figure 1).

Thus, regional metamorphism could have reached temperatures of approximately 350 °C and pressures below 2 kbar. Scarce aggregates of zeolites and phyllosilicates, which fill cavities in the groundmass of basalts and andesites, suggest that at the end of this metamorphic event small amounts of fluids could have taken part of this first process.

## Stage II

Superimposed to regional metamorphism, hydrothermal processes produced very low grade assemblages rich in zeolites altering the wall rocks. These processes generated most of the secondary minerals (mainly zeolites; Figure 4, stage II), but they only affected restricted areas associated with the rise of fluids. Localities 5, 7 and 10 (see Figure 1b) show the best outcrops of hydrothermally altered Cretaceous and Tertiary volcanic rocks (Vattuone and Latorre, 1996a,

1999; Vattuone *et al.* 1996b, 2005a).

Stage II is mainly found as amygdale filling and as veinlets in basalts and andesites. Epidote, wairakite, yugawaralite, laumontite, pectolite, prehnite, pumpellyite, da-chiardite, thomsonite, interstratified c/s, celadonite, quartz, albite and adularia were precipitated when hydrothermal fluids reached temperatures within zeolites facies (Vattuone and Latorre, 1994, 1996b; Vattuone *et al.*, 2001b, 2006; Figures 5a-5e). As a result, a large number of Ca-zeolites were generated, being laumontite the most abundant (Figure 5a). The greatest diversity of alteration minerals occurs in basalts and andesites, whereas in granitic rocks actinolite, prehnite, epidote and chlorite only appears pseudomorphically altering biotite crystals (Vattuone and Latorre, 1990). During this stage the wall-rock alteration must have been less abundant because the fluids were mainly in contact with previous secondary minerals formed in stage I.

The upper limit of laumontite-wairakite-yugawaralite stability field allows us to estimate pressures around 0.6 kbar and temperatures between 220 and 250 °C (Miyashiro and Shido, 1970; Zeng and Liou, 1982; Frey *et al.* 1991) (Figure 6a). Whereas, calc-silicate minerals in equilibrium with the zeolites generated in this stage suggest temperatures below 230°C (Vattuone *et al.* 1999).

### Stage III

During the last stage (Figure 4, stage III) minerals of very low temperature precipitated from hydrothermal fluids. These hydrothermal assemblages vary from the edge to the centre of the cavities evidencing an evolution in fluid conditions over time. In some localities, this assemblage is not preceded by any other secondary minerals, which suggests that hydrothermal fluids affected a larger area during the last stage (Figure 5f). The most characteristic assemblages generated during this stage can be found in localities 1, 4, 6 and 7 (Figure 1b).

These secondary minerals fill cavities in basalts and andesites as well as joints in granitic rocks, which suggests that the chemistry of the hydrothermal fluids was not primarily affected by the composition of the wall rocks. However it is worth emphasizing that the highest concentrations of zeolites were always observed in volcanic rocks, where they appear as thin coatings, filling amygdules and joints of several centimeters wide, or replacing primary igneous minerals.

Minerals generated during this stage are summarized in Figure 4. Although stilbite is the most abundant, Ca-heulandite, mordenite, scolecite, mesolite, chabazite, Ca-K-Na phillipsite, analcime, natrolite, barrerite, offretite, stellerite, quartz, calcite, fluorite, cristobalite and smectites were also found as late fillings in some cavities (Vattuone and Latorre, 2002a; Vattuone and Tourn 2002; Vattuone *et al.* 2001a, 2002, 2008; Montenegro and Vattuone 2008; Figures 5f-5i). Beidellyite composition of the smectites was found in the aggregates that fill the cavities of the groundmass, whereas saponite composition occurs when they are the result of olivine phenocrysts alteration (Vattuone *et al.*, 1997). It is important to emphasizing that alkaline zeolites became more abundant during this last stage. Offretite, barrerite and chabazite, together with fluorite and fluorapatite suggest an increase of alkaline elements in the hydrothermal fluids.

In the localities 5 and 6 some stilbites and heulandites also could have been generated after laumontite when temperature decreased. According to Liou (1971) and Miyashiro and Shido (1970) laumontite breaks down to form stilbite or heulandite at temperatures below 200 °C. Therefore, the assemblages found in the studied area must have been produced at lower temperatures and pressures below 2 kbar.

Magnetite, sphalerite, pyrite, chalcopyrite, native copper and cuprite probably precipitated from the same hydrothermal fluids that formed the very low grade metamorphic assemblages (Figure 4) (Vattuone *et al.*, 1996a; Tourn and Vattuone, 2002). These ore minerals occur as disseminated specks of less than two millimeters (localities 6 and 8; Figure 1b), or filling thin veinlets and amygdules associated with quartz (localities 1, 2 and 4; Figure 1b). Even though all these minerals occur scattered in the entire study area, sulfides are concentrated in localities 6 and 8,

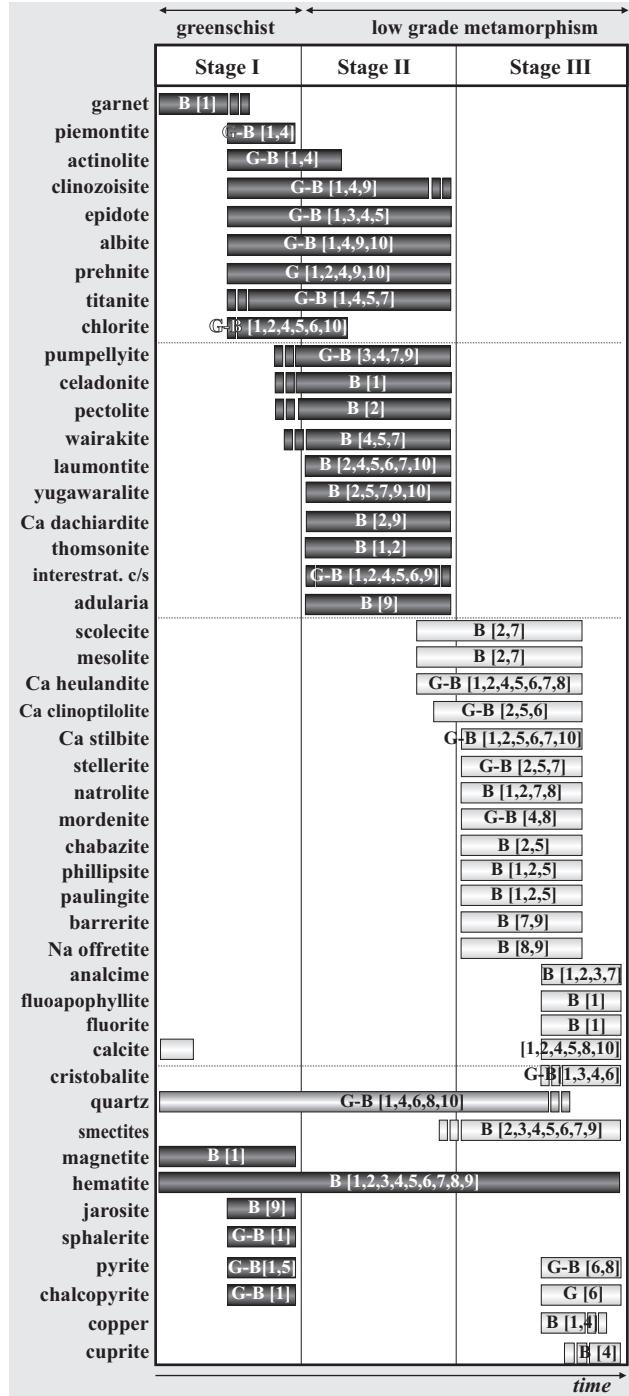


Figure 4. Paragenetic sequence scheme. Wall-rock composition (B=bas-andesites, G=granitoids) and localitiy number ([x]) are indicated inside each area.

whereas native copper and cuprite were mainly found in locality 4 (see Figure 1b).

Finally, as it was assumed that the stages II and III represent the evolution of the first metamorphic event, changes in pressure and temperature over time can be schematized. On the basis of Zeng and Liou (1982), Liou *et al.* (1985) and Frey *et al.* (1991), who established the most

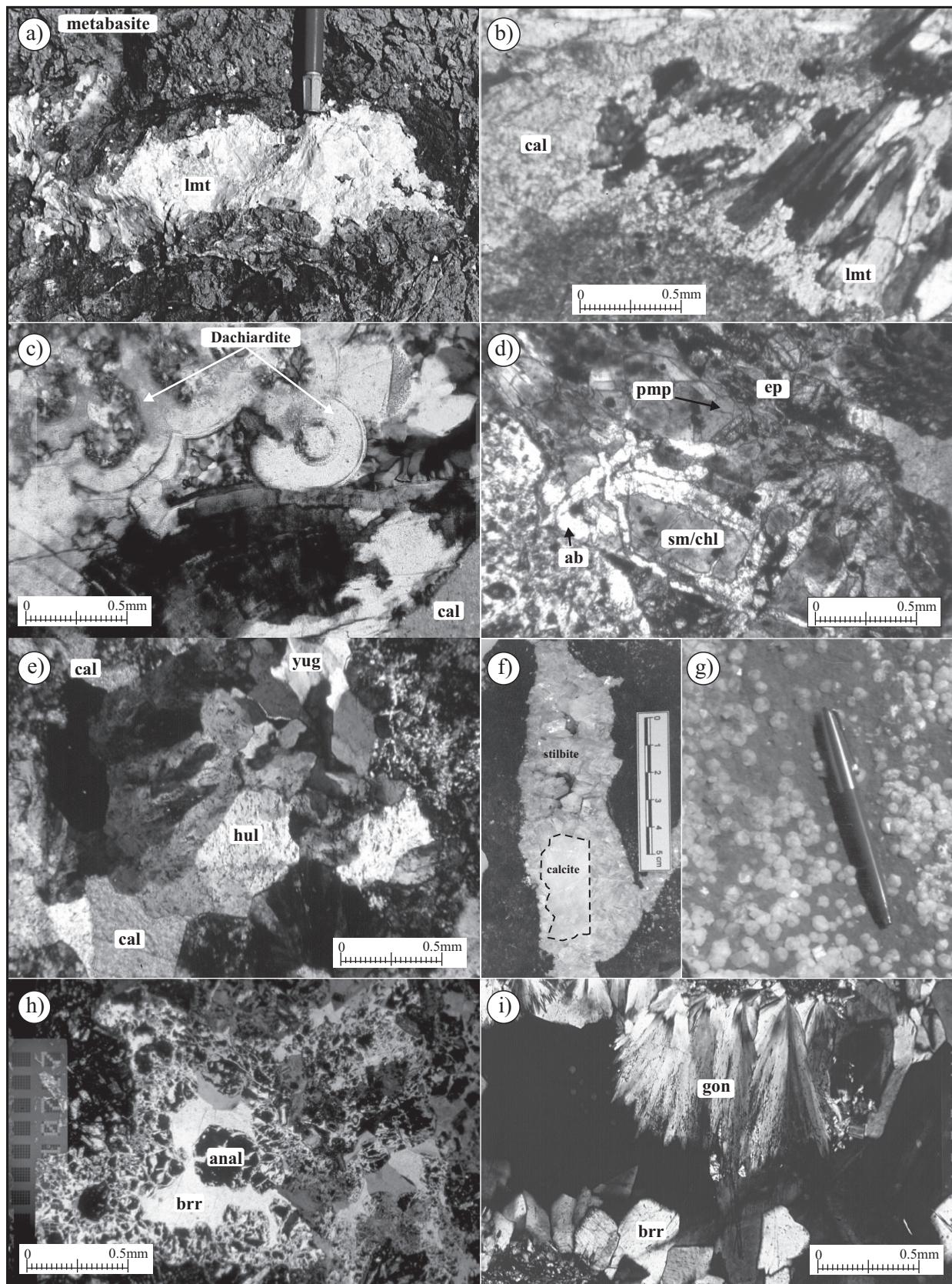


Figure 5. Microphotographies of the most common secondary minerals generated during stages II and III. a, B: Laumontite and calcite assemblages in metavolcanites; c: dachiardite aggregates; d: albite, epidote, chlorite, smectite and pumpellyite assemblages; e: calcite, heulandite and yugawaralite; f: basalt cavities filled by stilbite and calcite (locality 6); g: small crystals of analcime covering joint planes; h: analcime and barrerite; i: vesicles filled by barrerite and gonardite.

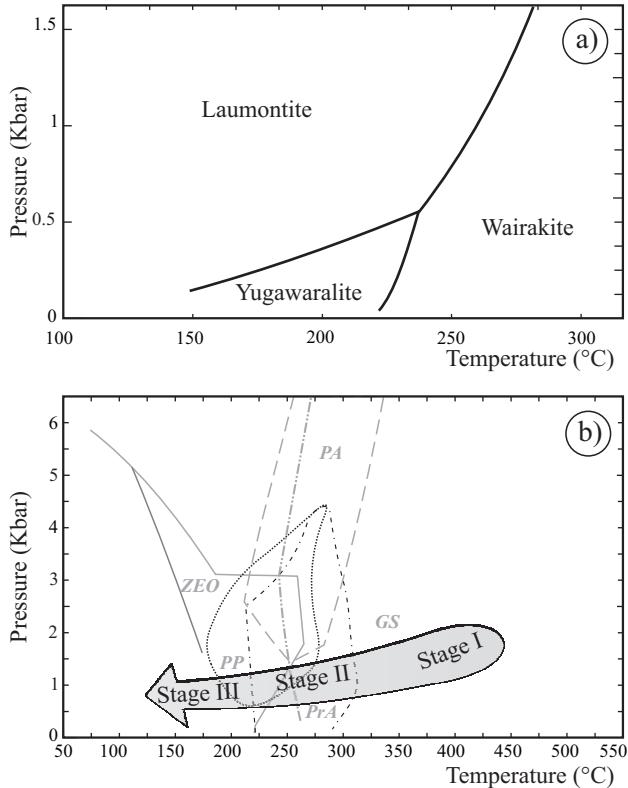


Figure 6. a)  $P$ - $T$  diagram showing the stability relationship among yugawaralite, laumontite and wairakite after Zeng and Liou (1982), Liou *et al.* (1985) and Frey *et al.* (1991); b) Schematic diagram of the  $P$ - $T$  conditions of each stage.  $P$ - $T$  fields for low grade metamorphic facies are from Frey *et al.* (1991); ZEO: zeolites facies, PP: prehnite-pumpellyite facies, PA: pumpellyite-actinolite facies, PrA: prehnite-actinolite facies and GS: greenschist facies.

common  $P$ - $T$  diagrams for low-grade metamorphism in metabasites, Figure 6b shows the evolution of temperature and pressure deduced from the alteration assemblages of the study areas.

## DISCUSSION AND CONCLUSIONS

As secondary assemblages have not been dated, the geologic setting associated with them is still subject of controversy. However, there are two features that enable us to suggest some general conditions for the zeolite mineralization studied in this paper.

In regard to the tectonic setting associated with the zeolite mineralization in Northern Patagonian Andes, if a thermal anomaly triggering hydrothermal activity was the result of the Cretaceous or Miocene granitic intrusions, secondary assemblages should define a zonation increasing their temperature of formation towards these units. However, the geographic distribution of secondary minerals does not vary following a specific trend. On the contrary, the thermal anomaly that produced these alterations does not seem to be restricted to the plutonic bodies, which suggests that the

mineralization was mainly controlled by the development of the volcanism and the lineaments of the uppermost sector of the lithosphere.

On the other hand, the relationship between the secondary assemblages and rocks of specific ages suggests that these mineralizations must have been generated from the Jurassic to the Miocene. The assemblages produced during the first stage mainly occur in volcanic rocks of Jurassic age and, subordinately, in the Tertiary volcanites (Depine *et al.*, 2003). Thus, this regional metamorphism should have taken place after the Jurassic; perhaps during the Cretaceous, when the relative motion between Nazca plate and South America increased, producing a high thermal anomaly and afterwards hydrothermal activity (Rapela *et al.*, 1983, 1984). On the other hand, Quaternary basalts do not present secondary assemblages, which suggests that these alteration processes must have finished before this period. Hence, the  $P$ - $T$  conditions of the alteration processes accords with the period in which the volcanic arc was more active in the study area. This fact allows us to explain why the Mesozoic volcanic rocks present higher metamorphic grade than the Tertiary basalts. In a regional study of low-grade metamorphism on the Central Andes (northwards the study area) Levi *et al.* (1989) suggest that the grade of metamorphism is higher in the oldest rocks. According to all the data compiled for this contribution, we suggest that the same situation occurs southwards, along the study area.

On the basis of these facts, we suggest that the alteration processes were related to the thermal anomaly that produced the Cretaceous-Miocene volcanic arc development. The coincidence between the most important magmatic period and the mineralization event supports this hypothesis.

Finally, as a result of the present study, the following conclusions can be drawn:

1. The rocks found in the study area present three main assemblages that were generated during three different stages of alteration. The oldest one was the result of a Cretaceous regional metamorphism, whereas during the last two stages the hydrothermal alteration was more pervasive. Although the last hydrothermal processes were spatially restricted in comparison to the previous regional metamorphism (due to the fact that it was constrained by fluid circulation) the major concentration of zeolites is associated with these last processes.

2. The oldest metamorphic assemblage could have started under amphibolite facies conditions, but temperature gradually decreased to the greenschist facies. The mineralogical assemblages associated with grandite garnets suggest that this first event reached temperatures of about 350 °C and pressures near 2 kbar. As temperature decreased, most of the high grade metamorphic minerals suffered retrograde metamorphism until they turned into the secondary assemblages that nowadays can be found.

3. Hydrothermal precipitates filled cavities of the volcanic rocks with zeolites, phyllosilicates, cristobalite and

calcite. This assemblage indicates a decrease in temperature, and high concentrations of alkaline elements,  $\text{SiO}_2$  and  $\text{CO}_2$  in the fluids (Vattuone and Latorre, 1990). At the beginning, the fluids must have had high temperature and therefore their composition could have been controlled by the wall rock. During this stage the fluids could have leached Ca, Fe and Mg from basalts and andesites. Thus, smectites and zeolites with high Ca and low  $\text{SiO}_2$  must have been produced during the second stage and seem to be strongly linked to the wall rock composition. Over time, secondary minerals lined the walls of cavities (isolating the fluids from the wall-rocks), meanwhile the temperature of the system decreased. As a result, the last fluids (stage III) could have produced minor wall-rock alteration, and zeolites with alkaline elements and higher  $\text{SiO}_2$  formed.

4. Zeolite assemblages generated by hydrothermal precipitation (during the last stages) occur in specific localities. Even though many species were recognized, laumontite and stilbite are the most common. These minerals are mainly found as open space fillings in the most permeable rocks. Basalts, andesites and ignimbrites host zeolites in different amounts according to their permeability. In the study area, the highest abundance of zeolites was found in locality 7, where they fill veins and amygdules of a Tertiary ignimbrite.

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