Differentiation of mantle-derived calc-alkaline magmas at mid to lower crustal levels: experimental and petrologic constraints

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ABSTRACT. — This contribution compares experimentally derived liquid-lines-of-descent on primary, hydrous calc-alkaline magmas ranging from picrobasaltic to basaltic andesite at pressures of 1bar to 15 kbar with geochemical evolutionary trends of two contrasting, subduction related, continental arc systems, namely the Adamello batholith and the Cascadian Arc system.

Field, petrologic and geochemical data of both systems are indicative for polybaric crystallization over basically the entire crustal column. This is clearly supported by experimental data that reveal that the majority of basic to intermediate rock compositions are not only close to the liquid-line-of-descent indicating that crystal retention in plutonic systems is not very efficient in basaltic to intermediate magmas, but also that the mean crystallization pressures obtained from CaO-MgO diagrams are in the range 5-10 kbar consistent with deep-level (crust-mantle boundary) and intermediate crustal level (15-25 km) magma storage areas where the principal crystallization-driven differentiation took place. In the case of the southern Adamello, subtle differences in the liquid-lines-of-descent are fully consistent with experimental data and trace element geochemistry indicating that different rock suites (plutons) have acquired their major and trace element characteristics at different crustal levels. The principal differences in crystallization-differentiation with increasing depth is the decreasing plagioclase stability leading to delay of the onset of plagioclase crystallization and the enhancement of both earlier clinopyroxene and amphibole crystallization with increasing depth shifting derivative liquids closer to the metaluminous / peraluminous limit or even within the peraluminous field with increasing pressure, fully consistent with the geochemical evolution of the southern Adamello granitoids that partly evolve to peraluminous compositions that cannot be linked to extensive crustal assimilation.

RIASSUNTO. — In questo lavoro sono messe a confronto i) le linee d'evoluzione di liquidi magmatici, dedotte sperimentalmente nell'intervallo di pressione da 1 bar a 15 kbar per magmi idrati primari di tipo calc-alcalino e composizione variabile da picrobasaltica ad andesite basaltica, e ii) gli andamenti evolutivi di natura geochimica riscontrati in due diversi sistemi subduttivi di arco continentale: il batolite dell'Adamello nelle Alpi meridionali e le vulcaniti della *Cascades Province*, esposte lungo la costa occidentale del continente nordamericano.

I dati geologici, petrologici e geochimici suggeriscono, per entrambi i sistemi, una cristallizzazione polibarica avvenuta verosimilmente nell'intera sezione crostale. Ciò è chiaramente

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indicato dai dati sperimentali secondo i quali: i) la maggioranza delle rocce di composizione da basica ad intermedia non solo si colloca in prossimità delle linee d'evoluzione dei liquidi (suggerendo che la ritenzione delle fasi solide nei sistemi plutonici non è molto efficace nei magmi da basaltici ad intermedi); ii) gli intervalli barici nel sistema CaO-MgO si collocano mediamente attorno a 5-10 kbar, coerentemente con i livelli crostali profondi (alla transizione crosta-mantello) ed intermedi (15-25 Km) delle zone di accumulo dei magmi in cui, per cristallizzazione, avviene la maggior parte della differenziazione magmatica. Nel caso dell'Adamello meridionale, minime -ma significative- differenze nella posizione delle curve di evoluzione dei liquidi sono perfettamente compatibili con i dati sperimentali e la distribuzione degli elementi in traccia. Ciò suggerisce che differenti serie intrusive hanno acquisito la loro segnatura geochimica, sia in termini di elementi maggiori che in traccia, a differenti livelli crostali.

All'aumentare della profondità, le differenze più significative nei processi di cristallizzazione/differenziazione sono: i) la ridotta stabilità del plagioclasio, con il conseguente ritardato inizio della sua cristallizzazione, e ii) un aumento nella precoce formazione sia del clinopirosseno che dell'anfibolo, con conseguente spostamento dei liquidi residuali verso il limite metalluminoso/peralluminoso, o perfino nel campo peralluminoso con l'aumento della pressione. Tale modello è perfettamente consistente con l'evoluzione geochimica dei granitoidi dell'Adamello meridionale che evolvono parzialmente verso composizioni peralluminose non riconducibili ad estesi fenomeni di assimilazione crostale.

KEY WORDS: *Calc-alkaline magmatism, differentiation, Cascades, Adamello, experimental petrology, active continental margins*

INTRODUCTION

A fundamental topic in volcanic – plutonic systems in general and in subduction-related calc-alkaline systems in particular is the relationship between plutonic and volcanic rocks and directly relates to the question if the magmas that produced the plutonic rocks differentiate 'in situ' at the depth of final emplacement or, alternatively, did they acquire their chemical composition at greater depth or even during ascent? This, in turn, leads to a series of related topics such as: do gabbroic to granitoid plutonics emplaced at rather shallow levels (5-15 km) represent subvolcanic magma reservoirs? Are common dioritic to granodioritic or even granitic plutonic rocks cumulates or do they represent (near) liquid composition, i.e. are their compositions closely following liquid-lines-of-descent of typical mantle-derived parental magmas? Additional questions not addressed in this contribution are: (1) what is the potential role of magma mingling/mixing and/or assimilation of country rocks by mafic to acidic magmas, and, do these processes that operate in addition to crystallization-differentiation significantly alter the liquid-lines-of-descent?

For tholeiitic rift or plume related basaltic magmatism such as for example the Deccan traps or the Permian Ivrea-Verbano and Malenco systems, the role of deep-seated magma storage reservoirs, as the principal areas where magma differentiation of mantle-derived basaltic magmas takes place, has been advocated already since considerable time (e.g. Rivalenti *et al.*, 1975; Cox, 1980; Bergantz, 1989; Lightfood *et al.*, 1990; Müntener *et al.*, 2000). In subduction-related calc-alkaline systems, several studies have suggested that these systems operate in a similar way. Differentiation of mantle-derived, hydrous basaltic magmas occurs in deep-seated magma reservoirs by a variety of processes; fractional-crystallization and, hence, the associated production of ultramafic to mafic cumulates being the most prominent one, but additional processes such as replenishment with less differentiated magmas, assimilation of country rocks and mixing of variably differentiated magmas may operate simultaneously with variable importance for the overall differentiation (e.g. Hildreth and Moorbath, 1988; Grove *et al.*, 2005; Annen *et al.*, 2006).

This contribution attempts to constrain the location (depth) where arc rocks acquire their major and trace element compositions by combining (1) field evidence from the plutonic rocks exposed in the Tertiary Adamello intrusion located in the Southern Alps that represents an active continental margin setting; (2) major and trace element geochemistry of typical plutonic and volcanic arc rocks using two contrasting examples: the Cascadian volcanics and the Adamello plutonic and dike rocks; and (3) experimental phase equilibria relevant for the differentiation of primary and derivative parental magmas under conditions spanning the entire range

from the surface to the deep roots of the igneous system (up to 50 km depth) by combining liquid-lines-of-descent derived from both experiments and natural systems.

FIELD, GEOCHEMICAL AND EXPERIMENTAL DATA

Field Evidences: Adamello Intrusion

The Tertiary Adamello batholith, located in the Eastern Southern Alps, represents igneous products generated in an arc-related continental margin setting. The batholith is predominantly composed of intermediate (tonalite to granodiorite) plutonic rocks, associated with minor ultramafic, gabbroic and granitic varieties (Bianchi *et al.*, 1970; Callegari and Dal Piaz, 1973). The final emplacement depth of the granitoids is estimated to 2-5 kbar in accordance with the absence of igneous epidote (< 6 kbar) and the occurrence of amphibole/biotite assemblages throughout the intermediate plutonic rocks (>2 kbar). The field relations and geochemistry of various plutons forming the southern part of the Adamello batholith have been extensively studied over several decades (Bianchi and Dal Piaz, 1937a, 1937b; Callegari and Dal Piaz, 1973; Brack, 1984, 1985; Del Moro *et al.*, 1985; Macera *et al.*, 1985; Ulmer *et al.*, 1985; Blundy and Sparks, 1992). These studies reveal that single plutons of kilometric sizes (3-10 km) generally lack any zonation with respect to lithology and composition. This renders it very unlikely that any significant differentiation of the magmas that formed these plutons has occurred at the level of final emplacement. In addition, one of the ultramafic/gabbroic complexes that formed in-situ at shallow levels from basaltic to basaltic andesite magma (Cornone di Blumone Complex) evidenced by crescumulate and exceedingly high contact metamorphic temperatures (>850°C) (Ulmer, 1982, 1986), does not show any significant differentiation with respect to the mineral compositions of the cumulate rocks. This complex, located at rather shallow depths (2-3 kbar), most probably represents the exceptional case of a remnant of a subvolcanic conduit that fed an overlying andesitic volcano. There is no evidence that the magmas passing through this conduit suffered any significant differentiation at this shallow level. The principal mineralogy of these mafic to ultramafic cumulate rocks (olivine clinopyroxenites, olivine gabbros to amphibole-bearing cpx-magnetite gabbros) are consistent with crystallization at rather shallow levels (2-3 kbar, 7-10 km) basing on known phase equilibria of hydrous, calc-alkaline magmas under fO_2 conditions close to the Ni-NiO equilibrium (e.g. Sisson and Grove, 1993a, 1993b).

Two additional series of basaltic (gabbroic) to andesitic and dacitic (tonalitic) rocks occur in the Southern Adamello that exhibit contrasting crystallization – differentiation behavior, not consistent with simple fractional crystallization at shallow level:

(i) A suite of dike rocks, ranging in composition from picrobasalt to high-silica andesite/dacite crosscuts the entire plutonic complex. Geochemistry, including isotope geochemistry, clearly indicates that these dike rocks are genetically linked to the magmas that formed the plutonic rocks (Ulmer *et al.*, 1985; Kaghami *et al.*, 1991). The crystallization sequence in these dike rocks differ from the shallow level plutonics of the Cornone di Blumone series by having amphibole much earlier on the liquidus, shortly after or even concomitantly with plagioclase following olivine and cpx (Ulmer, 1988). The most primitive varieties of these dike rocks (picrobasalts) fulfill all requirements, based on bulk and mineral chemistry, for parental magmas of the entire igneous rock suite of the (Southern) Adamello.

(ii) The mafic-ultramafic rock associations of the so called Val Fredda Pluton (Monte Mattoni, Monte Cadino, Monte Frerone) that are associated with amphibole-poor, biotite-bearing leuco-tonalites (formerly called Val Fredda quartz diorites, Bianchi *et al.*, 1970) exhibit yet another crystallization sequence characterized by early amphibole cumulus crystals containing inclusions of forsteritic olivine (up to Fo_{88}) and aluminous cpx set in a matrix of strongly zoned plagioclase (An_{95}-An_{10}) and salitic (low-Al) cpx showing evidence for boiling (formation of so called 'occelli') at the shallow level of intrusion. The ultramafic parts of this complex are composed of hornblendites, sometimes called cortlandites, associated with porphyritic (cumulus) amphibole gabbros and acicular hornblende gabbros free of any cpx. These cumulate rocks have attracted considerable attention and strongly diverging models have been proposed to explain their rather unusual assemblages and textures.

Ulmer *et al.* (1985), Ulmer (1986), Nimis and Ulmer (1998), Blundy and Shimizu (1991) and Blundy and Sparks (1992) using different methods and approaches arrived basically at the same conclusion: the primocrysts forming the cumulus phases (amphiboles with their inclusions) originate from a deep-seated magma reservoir located in the lower crust around 30 km depth (8-10 kbar), whereas the matrix crystals forming a typical shallower level gabbroic assemblage formed during emplacement of the crystal mush at the final level of intrusion around 2-3 kbar pressures.

Tiepolo and Tribuzio (2005) have explained the two contrasting assemblages and clinopyroxenes associated with them as the product of magma mingling of mantle- and slab-derived melts respectively. However, their inference for a slab-derived melt, primarily basing on cpx-liquid partitioning calculations, can be explained as an artifact of extreme changes in crystal-liquid partition coefficients as a function of melt and crystal chemistry and temperature. Melts with very similar geochemical characteristics (very high Sr/Y values, see further down) have recently been produced in a series of experiments conducted to study the differentiation of andesitic magmas in deep-seated magma reservoirs (8-12 kbar, Alonso Perez *et al.*, 2008a) by extensive amphibole fractionation and the absence of plagioclase in the crystallization assemblage. The occurrence of forsteritic olivine coexisting with high-Mg, pargasitic amphibole and delayed plagioclase crystallization are the typical features of high-pressure crystallization in hydrous basaltic systems as clearly revealed by the experimental studies of Ulmer (1988), Kägi (2001) and Grove *et al.* (2005).

In summary, the Southern Adamello plutonic and dike rocks clearly reveal that differentiation of picrobasaltic, basaltic and andesitic magmas that are parental to the tonalitic to granodioritic rocks building the bulk of the Adamello batholith at the current level of exposure (>98% intermediate to acidic plutonics) mostly did not take place at the shallow level of final emplacement, but must have occurred at deeper levels ranging from the crust-mantle boundary (estimated at the time of igneous activity to 35-45 km) to mid-crustal levels (10-15 km). There are, in addition, strong evidences (e.g. from the Val Fredda Complex) that crystallization-differentiation occurred polybarically starting in deep-seated reservoirs and en-route during ascent to the final location of intrusion and emplacement.

Some geochemical constraints

An immediate question following the short outline of principal features of the southern Adamello plutonic rocks is: does this situation, fractionation-differentiation mainly occurring at deeper levels below the shallow magma storage areas that are potential reservoir feeding overlying volcanoes, apply also to volcanic systems? In order to provide a rough answer to this question, a comparison of the bulk rock chemistry of the plutonic and dike rocks of the Adamello batholith with the volcanic rocks of the Cascadian arc (Western USA to SW Canada, N-California to southernmost British Columbia) has been conducted. Both systems form 'active continental margins' and are characterized by low- to medium K, calc-alkaline igneous rocks and share many features that, in my opinion, allow a comparison of the deeper, plutonic Adamello system with the shallow, volcanic system of the Cascadian Arc. Bulk major and trace element data for the Adamello batholith (focusing on the southern and central parts, including the Monte Re di Castello and Adamello superunits (Bianchi *et al.*, 1970) have been compiled from a variety of sources including Italian sources (Di Colbertaldo, 1941, 1942, 1950; Zanettin, 1956; Callegari, 1963; Bianchi *et al.*, 1970; Macera *et al.*, 1985; Zantedeschi *et al.*, 1989), data from Blundy and Sparks (1992), and data from ETH Zurich, both published (Ulmer *et al.*, 1985; Kaghami *et al.*, 1991) and unpublished PhD (Ulmer, 1986), and many unpublished diploma thesis (Brack, Schellhorn, Sonderegger, Beiss, Ulmer, Mäder, and Küng). Geochemical data for the Cascadian volcanics have kindly been made available to the author by Jon Blundy, Bristol University, who compiled over 1600 analyses from a variety of sources for all major volcanic centers of the Cascadian arc ranging from Lassen Peak in the South (Northern California) to Mount Garibaldi in the North (southernmost British Columbia) spreading over 1000 km.

Comparing the geochemistry of these two contrasting systems reveals some striking similarities between the plutonic rocks and their volcanic counterparts: with the exception of (some) ultramafic and mafic cumulates, the

major and trace element compositions of the two contrasting systems, both forming medium- to low- K calc-alkaline trends, agree very well and superimpose each other. This indicates that not only plutonic rocks most probably acquire their chemical composition at considerable depths, but also volcanic rocks. This is further substantiated by the fact that in both systems basic and intermediate rocks generally lack any significant Eu anomalies, advocating that plagioclase did not play a decisive role as a major fractionating phase in these systems at early stages. Many trace element patterns of the plutonic rocks of the Adamello are actually indicative for early amphibole fractionation in the absence of plagioclase co-precipitation consistent with the occurrence of hornblendites associated with granitoid rocks exhibiting trough-like REE (see also Fig. 7) patterns with very low concentration of MREE. Similar phase assemblages from enclaves/xenoliths at the Cascadian volcano Mt. Shasta / Medicine Lake have been associated with differentiation processes operating in magma reservoirs located in the lower crust or at the crust-mantle boundary (Grove *et al.*, 2003). The superposition of plutonic and volcanic rocks in the case of the Adamello and the Cascadian rocks implies that (at least) intermediate to acidic plutonic rocks closely resemble liquid compositions and that accumulation of phenocrysts/primocrysts only marginally affects their composition. If true, phase equilibria constraints and experimentally or theoretically derived liquid-lines-of-descent could be utilized to constrain magma differentiation processes; this will be attempted in the next section.

Experimental constraints

In order to quantify the phase assemblages and the composition of derivative liquids, a compilation of published and not yet published experimental data from our laboratory and literature data on the crystallization-differentiation of primary and derivative parental magmas in hydrous, mostly H_2O-undersaturated, and in dry systems relevant for the evolution of arc magmas has been performed. Our laboratory has conducted several series of experiments on hydrous picrobasaltic and high-Mg basaltic andesite primary and basaltic andesite parental magmas under conditions of

the lower crust and uppermost mantle (5-15 kbar, Ulmer, 1986, 1988, 1992; Kägi, 2001; Kägi *et al.*, 2005; Müntener and Ulmer, 2006; Alonso Perez *et al.*, 2008a, 2008b), as well as on dry tholeiitic primary liquid at 7-10 kbar (Villiger *et al.*, 2004; Villiger *et al.*, 2007b). Equilibrium and fractional crystallization experiments were performed mostly under oxidizing (NNO) but also under more reducing (FMQ-1 to -3) conditions to explore the effect of oxygen fugacity in addition to pressure, temperature and H_2O-content. The results of these higher pressure experiments were supplemented with 1-bar experiments from our laboratory and literature data on hydrous melting of basaltic compositions at 1-12 kbar (Sisson and Grove, 1993a, 1993b; Kawamoto, 1996; Müntener *et al.*, 2001; Sisson *et al.*, 2005; Feig *et al.*, 2006).

The principal results presented here have been conducted on two contrasting types of primary magmas (Fig. 1): (i) a hydrous pricrobasalt dike from the Adamello containing 2.6 wt.% H_2O representing a deep mantle extraction depth of about 90 km (28 kbar) inside the garnet-peridotite stability field reflecting the situation of an active continental margin with thick crust and lithosphere (such as the Alps), and (ii) a high-Mg basaltic andesite from Mount Shasta containing 4.5 wt.% H_2O (California, Baker *et al.*, 1994) that represents mantle extraction pressures of about 10 kbar (35 km) typical for a juvenile continental margin with shallow crust and nearly absent lithosphere as typical for the Cascadian Arc. Both, classical equilibrium crystallization experiments on a fixed bulk composition at variable temperature and pressure (and to a lesser extend fO_2 conditions) and step-wise fractional crystallization experiments (Kägi, 2001; Villiger *et al.*, 2004) more closely representing natural conditions of magma differentiation, were conducted.

The results of these studies are exemplified here by a selection of bar-diagrams displaying the mineralogy and weight fraction of solid phases formed during crystallization at a given temperature (Fig. 1). This type of representation illustrates the crystallization assemblages and increasing amount of crystalline phases for each step of decreasing temperature in equilibrium crystallization experiments (Fig. 1a) and the solid fraction renormalized to 100% for each individual step in fractional crystallization experiments conducted on
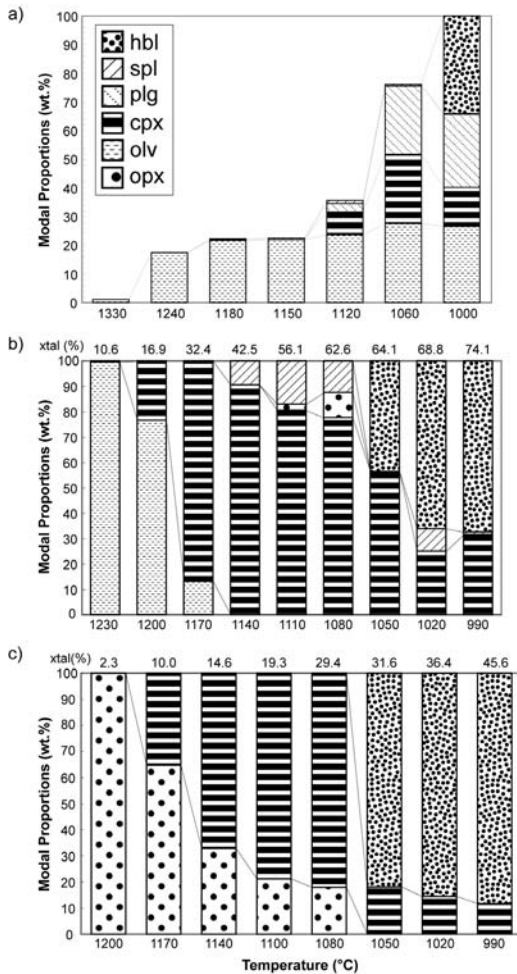
Fig. 1 – Schematic representation (weight fractions) of modal compositions of solids as a function of temperature for: **a)** equilibrium crystallization experiments on a picrobasalt at low fO_2 (close to C-CO-CO$_2$ buffer) at 5 kbar (P. Ulmer, unpublished experimental data). The difference to 100% corresponds to the liquid fraction at each temperature; at 1000°C the solidus of this composition is basically reached; **b)** each fractionation step as a function of temperature for fractional crystallization experiments on a picrobasalt at high fO_2 (close to Ni-NiO buffer) at 10 kbar (Kägi, 2001). Top labels indicate accumulated amounts of solids crystallized at a given temperature; **c)** each fractionation step as a function of temperature for fractional crystallization experiments on a high-Mg basaltic andesite at high fO_2 (close to Ni-NiO buffer) at 10 kbar (Kägi, 2001). Abbreviations: hbl – hornblende (amphibole); spl – spinel (Fe-Ti-Cr oxide phases); plg – plagioclase; cpx – clinopyroxene (Ca-rich pyroxene); opx – orthopyroxene (low-Ca pyroxene); olv – olivine.

the composition of the liquid phase measured in the previous, higher temperature experiment, thereby simulating perfect fractional crystallization (Figs. 1b and c).

The principal results from these phase equilibria studies are: (1) at pressures exceeding 8 kbar (Figs. 1b and c), plagioclase is a very late to absent phase in hydrous, calc-alkaline mantle-derived melts; cpx (± opx) and amphibole are the dominant phases controlling the evolution of derivative liquids. At lower pressure (1-5 kbar, Fig. 1a), amphibole is late and plag, cpx and Fe-Ti-oxide (mainly magnetite) are the dominant fractionating phases; (2) at depths exceeding 20 km (7 kbar) >50 % ultramafic cumulates (dunite, wehrlite, pyroxenite, hornblendite) are crystallized from primary liquids under both, hydrous and dry conditions, prior to the formation of any gabbroic cumulates; and (3) at pressures exceeding 8 kbars, garnet might become an igneous phase in differentiated liquids and at pressures exceeding 10 kbar garnet crystallizes even from primary liquid compositions (Alonso Perez *et al.*, 2008a, 2008b.) consistent with field observations from the Kohistan batholith (N-Pakistan) not discussed in this contribution as garnet crystallization is most probably not relevant for the Adamello batholith and the Cascadian Arc.

The phase relations of these systems are used to construct and ultimately control the location of the liquid-lines-of-descent in 'chemical space'. There are, of course, differences related to the starting, primary or parental, magma compositions of a particular rock suite, particularly affecting more incompatible elements such as Na_2O, K_2O, TiO_2, P_2O_5, etc. However, one can demonstrate that two rather contrasting starting compositions, a picrobasalt from the Adamello with 17 wt.% MgO (Fig. 2b) and a high-Mg basaltic andesite from Mt. Shasta (Cascades) with 10 wt.% MgO (Fig. 2c) follow very similar liquid-lines-of-descent for major elements.

Liquid-lines-of-descent for the picrobasaltic (Figs. 2a and b) and basaltic andesite (Fig. 2c) composition are simplified by recalculating melt compositions into mineral endmember components (olivine, silica, clinopyroxene, plagioclase, Fe-Ti-Cr oxide, K-feldspar) after the method of Grove *et al.* (1992) and projecting them onto the ternary diagram olivine-diopside-silica. Starting from a picrobasaltic compositions, liquids evolve at lower

pressures of 1 bar to 5 kbar and under both, relatively oxidizing (FMQ+1, 1 bar) and relatively reducing (close to C-CO-CO_2 buffer in graphite-Platinum double capsules, 5 kbar) conditions, first towards the cpx apex due to olivine crystallization (Figs. 1a and 2a), followed by a sharp bend away from olivine and cpx towards silica-saturated conditions resulting in metaluminous (diopside-normative) andesitic to dacitic derivative liquids. Due to bulk composition constraints, equilibrium crystallization at 5 kbar evolves to less silica-rich compositions than fractional crystallization. Dominant cumulate assemblages produced by shallower level crystallization of basaltic to picrobasaltic mantle melts are dunites, followed by olivine gabbros and late amphibole gabbros.

At 10 kbar (Fig. 2b), in all but one series, the liquids evolve from olivine-normative to quartz-normative compositions, but the extent of silica enrichment and the phase relations are quite different. Equilibrium crystallization experiments at 10 kbar conducted in graphite-Platinum double capsules that exert relatively low fO_2 conditions (near the C-CO-CO_2 buffer) drives the liquid to nepheline-normative compositions (Fig. 2b), whereas for fractional crystallization, slight silica enrichment is obtained (similar to experiments conducted by Villiger *et al.*, 2004 and Villiger *et al.*, 2007b on an anhydrous tholeiitic basalt). Crystallization at relatively high fO_2 (~FMQ +1) drives resulting melt compositions towards the opx-qtz join. For equilibrium crystallization, the composition of resulting liquids changes from olivine to quartz ± corundum normative, while for fractional crystallization liquids remain diopside normative (metaluminous). The liquid-lines-of-descent drawn in Fig. 2b demonstrate that oxygen fugacity exerts an important control on obtaining derivative liquids relevant for deep arc rocks. A comparison with experimental data on a primary basaltic andesite starting composition obtained at 10-12 kbar for fractional and equilibrium crystallization respectively (Kägi, 2001; Müntener *et al.*, 2001; Figs. 1c and 2c) shows that derivative liquids follow similar fractionation trends, irrespective of the primary liquid composition, despite quite different crystallizing assemblages and/or modal proportions of cumulates formed: the picrobasaltic composition produces appreciable amounts of olivine (about 20%) in the first crystallization steps,

followed by dominantly cpx with very minor opx and large amounts of amphibole when andesitic liquid compositions are reached (Fig. 1b), whereas the basaltic andesite starting composition produces none (fractional) to very minor (equilibrium crystallization) olivine, but considerable amounts of opx, followed by opx plus cpx, followed by amphibole plus cpx. This indicates that the starting compositions control the early cumulate paragenesis in the lower crust (dunites, wherlites, olivine-clinopyroxenites for picrobasalts versus orthopyroxenites and websterites for the basaltic andesite), but followed in both cases by pyroxene-bearing hornblendites and, at lower pressures and/or temperatures, by cpx-hornblende gabbros. These are the typical successions of ultramafic to mafic cumulates observed in shallow (e.g. Cornone di Blumone) and deep-seated (e.g. Kohistan arc, Pakistan: Miller and Christensen, 1994; Burg *et al.*, 1998; Tonsina arc, Alaska: DeBari and Sleep, 1991; Greene *et al.*, 2006; Val Fredda pluton, Adamello: Ulmer *et al.*, 1985; Blundy and Sparks, 1992) plutonic series, testifying the applicability of these experimental studies to actual plutonic rock series.

Fig. 2d reveals an interesting relationship between pressure of crystallization and normative diopside content using the aforementioned olivine-cpx-qtz projection by Grove *et al.* (1992): from 1 bar to 10 kbar, derivative andesite to dacite to rhyodacite (diorite – tonalite – granodiorite) liquids become increasingly depleted in cpx-component and change from metaluminous (cpx-normative) to peraluminous (corundum normative) at 10 kbar. This shift is a direct consequence of the suppressed plagioclase stability and enhanced amphibole crystallization with increasing pressure ultimately leading to peraluminous intermediate granitoid liquids. Such relationships have already been proposed on the basis of experimental data and theoretical considerations by Cawthorn and Brown (1976) and Cawthorn and O'Hara (1976), but many researchers still attribute a shift from metaluminous to peraluminous intermediate to acidic granitoid magmas to enhanced assimilation of pelitic country rocks that is evidently not required! Increasing the pressure further to 15 kbar leads to an inversion of the trend: derivative liquids become again metaluminous (Fig. 2d); this is a consequence of the crystallization and fractionation of garnet from andesitic liquids (i.e. at temperatures < 1100°C) that
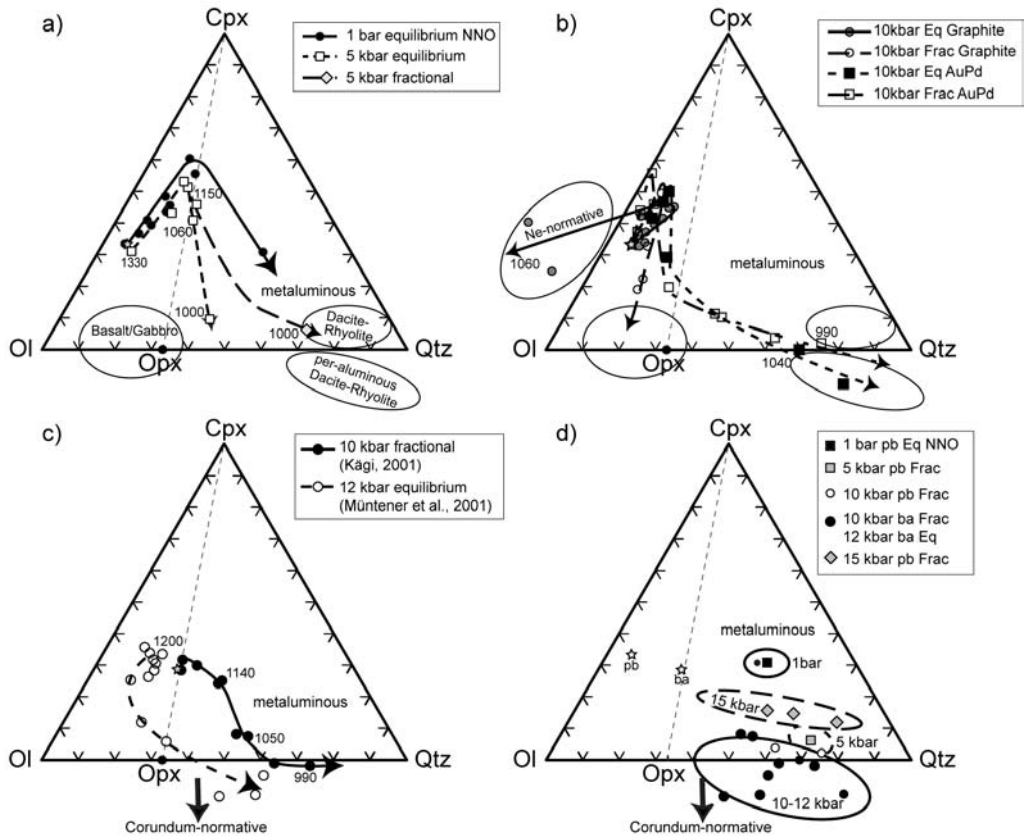
Fig. 2 – Normative, pseudoternary olivine – cpx – quartz projection of: **a)** the liquid-lines-of-descent for equilibrium and fractional crystallization experiments at 1 bar and 5 kbar pressure under oxidizing (1 bar, NNO) and reducing (5kbar, CCO-buffer) conditions. Starting material (grey star symbol) is a natural picrobasalt from the Adamello batholith (Ulmer, 1988) containing 2.6 wt.% H_2O. Normalization into six components (olivine, cpx, plg, qtz, K-fsp, oxide) after the projection scheme of Grove *et al.* (1992). Elliptical fields indicate locations of basalt/gabbro and metaluminous and peraluminous acidic (dacite-rhyolite) liquids. Numbers close to symbols refer to experimental run temperatures; **b)** the liquid-lines-of-descent for equilibrium and fractional crystallization experiments at different fO_2 at 10 kbar on the picrobasalt. Note that the silica-rich liquids containing appreciable amounts of normative quartz that are typical for subduction related magmatism are only obtained by experiments at relatively high fO_2 (~NNO); **c)** the liquid-lines-of-descent for equilibrium and fractional crystallization experiments under relatively oxidizing conditions (FMQ to NNO) at 10 (fractional; Kägi, 2001) and 12 kbar (equilibrium; Müntener *et al.*, 2001) on a high-Mg basaltic andesite from Mount Shasta (Baker *et al.*, 1994) containing 4.5 wt. H_2O (grey star symbol); **d)** derivative intermediate to acidic liquids obtained by fractional and equilibrium crystallization from hydrous picrobasaltic (pb) and basaltic andesite (ba) starting compositions at 1bar, 5, 10, 12 and 15kbar; 15 kbar data from Alonso Perez *et al.* (2008b). Abbreviations in legends: pb = picrobasalt starting material; ba = high-Mg basaltic andesite starting material; Eq = equilibrium crystallization; Frac = fractional crystallization; NNO = experiments conducted at an fO_2 corresponding to the Ni-NiO equilibrium; Graphite = experiments conducted in graphite-Pt double capsules (reducing, C-$CO-CO_2$); AuPd = experiments conducted in AuPd double capsules (oxidizing, NNO).

drives the liquids back into the metaluminous field. Such liquids can, however, easily be distinguished from shallow level fractionates by their rather unusual trace element patterns expressed for example by extremely high Sr/Y ratios and very

fractionated REE patterns (high La/Yb and Sm/Yb ratios).

Interestingly, tonalitic to granodioritic plutonics from the southern Adamello show two distinct evolutions/populations: the intermediate plutonics

associated with hornblende-rich cumulates (Val Fredda pluton, Corno Alto and Sostino quartz diorites, and Malga Ervina trondhjemites) are predominantly (>80%) peraluminous, whereas the intermediate plutonics associated with the shallow level, amphibole-poor cumulates of the Cornone di Blumone complex are nearly exclusively metaluminous (diopside-normative) further supporting deep and intermediate level fractionation for the parental magmas forming these plutonics. It should be noted that the Val Fredda plutonics are among the Adamello rocks yielding the least crustal contamination as evidenced by low [87]Sr/[86]Sr initial, low $\delta^{18}O$ and high [143]Nd/[144]Nd initial values (Del Moro *et al.*, 1985; Kaghami *et al.*, 1991), basically indistinguishable from the Cornone di Blumone plutonic rocks; hence variable crustal assimilation cannot be advocated to explain their large differences in normative cpx and corundum respectively.

Comparison of experimental and natural liquid compositions

In the case of anhydrous tholeiitic liquids, simple oxide-oxide relationships, such as the CaO versus MgO variation can be utilized to quantify the crystallization depths of cpx-bearing parental magmas (e.g. Grove *et al.*, 1992; Yang *et al.*, 1996; Herzberg, 2004; Villiger *et al.*, 2007a) and has successfully been applied to the crystallization depths of MOR basalts. A similar approach is attempted here for hydrous calc-alkaline systems separating basaltic to basaltic andesitic and picrobasaltic primary liquids, and clearly stating that this is a first attempt! In the case of anhydrous liquids, the onset of cpx and/or plagioclase crystallization is strictly pressure controlled: increasing pressure leads to an earlier onset of cpx crystallization (closer to the liquidus) and hence to a lower absolute value CaO and a shift of the CaO peak towards higher MgO contents. Utilizing available experimental data, published (see figure captions for references) and unpublished data from our laboratory, preferably from fractional crystallization experiments, Fig. 3 for basalt to basaltic andesites and Fig. 4 for picrobasalts have been constructed. It is clearly evident that for the picrobasalt the system performs as anticipated: with increasing pressure the peak in CaO, i.e. the onset of cpx crystallization, is shifted

to higher MgO contents and the height of the CaO peak is decreasing. At lower pressures, a crossover of the 5 kbar curve with the 10 and 15 kbar curves is observed. This is most probably related to the onset of plagioclase crystallization in the 5 kbar experiments that strongly depletes CaO at rather high MgO levels.

For the basaltic system (Fig. 3) the situation is similar but data have been obtained for rather different bulk compositions with variable H_2O contents and fO_2 conditions. In general, the same succession is observed: decreasing CaO peak and shift to higher MgO contents with increasing pressure and crossover of the 1-2 and 5 kbar curves and the 10 kbar curve at lower MgO contents. Unlike the tholeiitic system, where olivine basalts of rather constant composition (MOR basalt or similar) have been used as starting materials, in the calc-alkaline system a large variety of primary liquids with silica contents ranging from 45 to nearly 60 wt.% and MgO contents from less than 10 to over 17 wt.% have been identified. Therefore, a single CaO-MgO diagram cannot be employed to decipher the crystallization depths of all or even a large part of calc-alkaline plutonic and volcanic systems.

In the following discussion, the behavior of the two selected reference systems are confronted with the experimentally-derived CaO-MgO trends just discussed, i.e. (1) the Cascadian Arc system thought to be derived mainly from rather high silica, low pressure primary basaltic andesite magmas (e.g. Grove *et al.*, 2003), and (2) the Adamello batholith inferred to be derived from rather high-pressure ol-tholeiitic to picrobasaltic primary magmas (e.g. Ulmer, 1988). Fig. 5 compares the CaO-MgO relationship of bulk rock compositions of >1600 analyses of Cascadian volcanics with the liquid-lines-of-descent derived from hydrous experiments on calc-alkaline compositions from 1 to 10 kbar (Fig. 3): the Cascadian volcanics occupy a rather narrow field that branches at MgO contents >4 wt.% and > 6 wt.% MgO into different evolutionary trends consistent with dominant crystallization around 1-2 kbar, 5-7 kbar and around 10 kbar pressure. Volcanic rock compositions with relatively high MgO contents (>4 wt% MgO) that follow the lower pressure evolutionary trend (1-2 kbar) are rather rare, indicating that the principal differentiation from primitive basaltic andesite magmas to intermediate
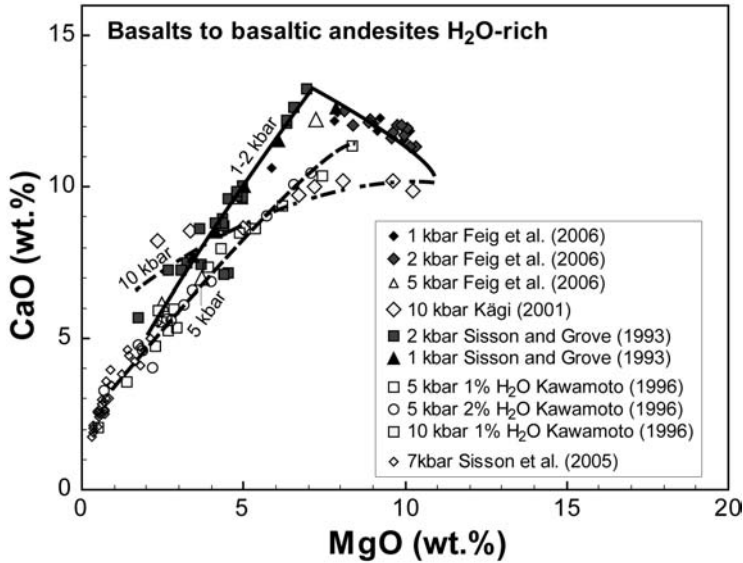
Fig. 3 – CaO – MgO diagram (in wt.%) displaying experimental liquids at 1 – 10 kbar conducted with hydrous basalt to basaltic andesite starting compositions from Sisson and Grove (1993a), Kawamoto (1996), Kägi (2001), Sisson *et al.* (2005), and Feig *et al.* (2006). Solid and broken lines are eyeball fits through the experimental data points for 1-2, 5, and 10 kbar respectively.
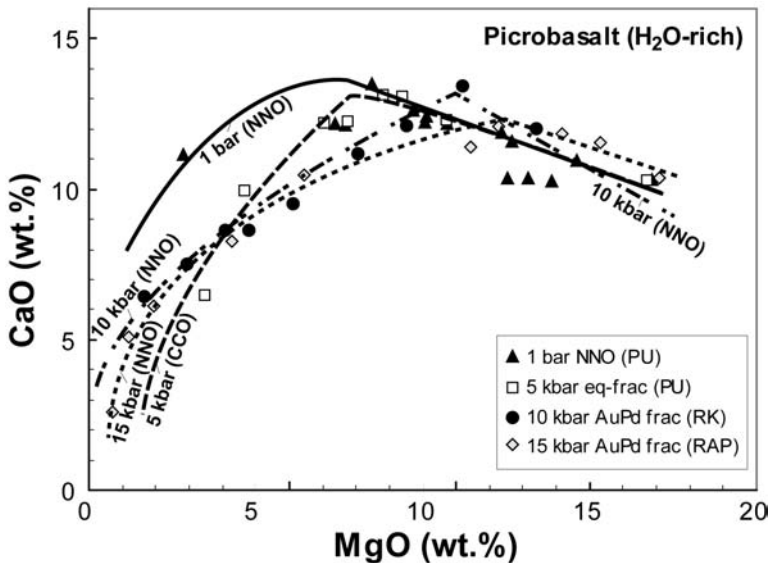

Fig. 4 – CaO – MgO diagram (in wt.%) displaying experimental liquids at 1bar – 15 kbar conducted with hydrous picrobasalt starting compositions from Ulmer (PU; unpublished data at 1bar and 5 kbar); Kägi (RK; Kägi, 2001) and Alonso Perez (RAP; Alonso Perez *et al.*, 2008b). Solid and broken lines are eyeball fits through the experimental data points for 1bar, 5, 10 and 15 kbars respectively. Note the shift in the CaO maximum to lower absolute values and higher MgO contents with increasing pressure and the crossover of the 5 kbar liquid-line-of-descent with the 10 and 15 kbar lines at low MgO (high differentiation).

volcanic compositions (andesites to dacites) did not take place predominantly in subvolcanic magma reservoirs, but the majority of these higher MgO lavas clearly follows the trend consistent with 5-7 and 10 or even more kbars advocating for both: polybaric and deep-seated magma crystallization. To date, it is however not clear if the two trends consistent with 5-7 and 10 kbar crystallization pressure are indeed significant and not biased by sampling as the bulk rock data utilized in this study encompass a large number of volcanic centers spreading over 1000 km length of the arc. It should also be noted that it is clearly known from the Cascadian system (e.g. Mount Shasta) that even a single volcanic center can contain nearly dry olivine-tholeiitic primary magmas contemporaneously with H$_2$O-rich high-silica basaltic andesite primary magmas. The use of a single set of liquid-lines-of-descent is definitely an oversimplification for such complex systems; nevertheless the outcome is consistent with the inferred magma evolution in the well studied Shasta/Medicine Lake system (e.g. Grove *et al.*, 2002), where polybaric differentiation with three dominant magma storage sites located at

the crust-mantle boundary (around 30 km), at mid-crustal levels (around 15-20 km) and in a shallow level sub-volcanic magma reservoir (around 7-10km) have been proposed to be responsible for the phenocryst assemblage and the overall chemical evolution of this volcanic system.

Fig. 6 compares the experimentally derived liquid-lines-of-descent for a picrobasaltic primary liquid with a selection of plutonic and dike rocks of the southern part of the Adamello batholith in the CaO-MgO diagram. Plutonic rocks display much more scatter than the volcanic rocks of the Cascades, which is primarily the expression of cumulate processes operating in addition to fractional crystallization (and assimilation) leading to rocks exhibiting high and low CaO contents (cpx- and plag-rich, and olivine-rich cumulates, respectively). However, intermediate plutonics ranging up to 7 wt.% MgO and the dike rocks occupy a rather narrow field overlapping with the Cascadian volcanics. There are, however, interesting details that relate directly to the principle crystallization depths of the gabbroic to acidic plutonics:
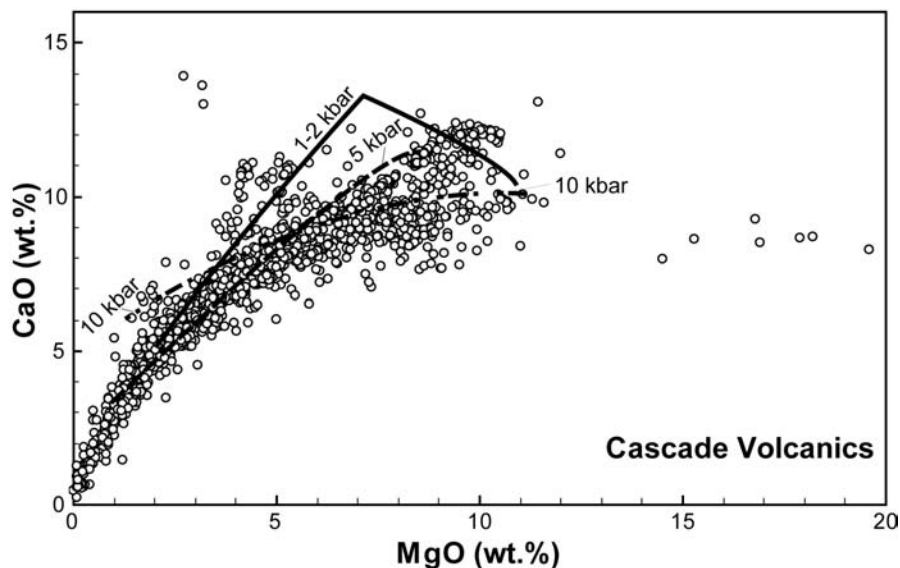

Fig. 5 – CaO – MgO diagram (in wt.%) displaying >1600 bulk rock data of the Cascadian Arc volcanics (N-California/USA to southernmost British Columbia/Canada; data compilation made available to the author by Jon Blundy) with superimposed lines depicting experimentally derived liquid-lines-of-descent at 1-2 kbar, 5 kbar and 10 kbar for basaltic to basaltic andesite primary magmas (taken from Fig. 3). Note bifurcation of data at MgO contents >6 wt.% consistent with crystallization-differentiation at approximately 5 and 10 kbar pressure respectively.
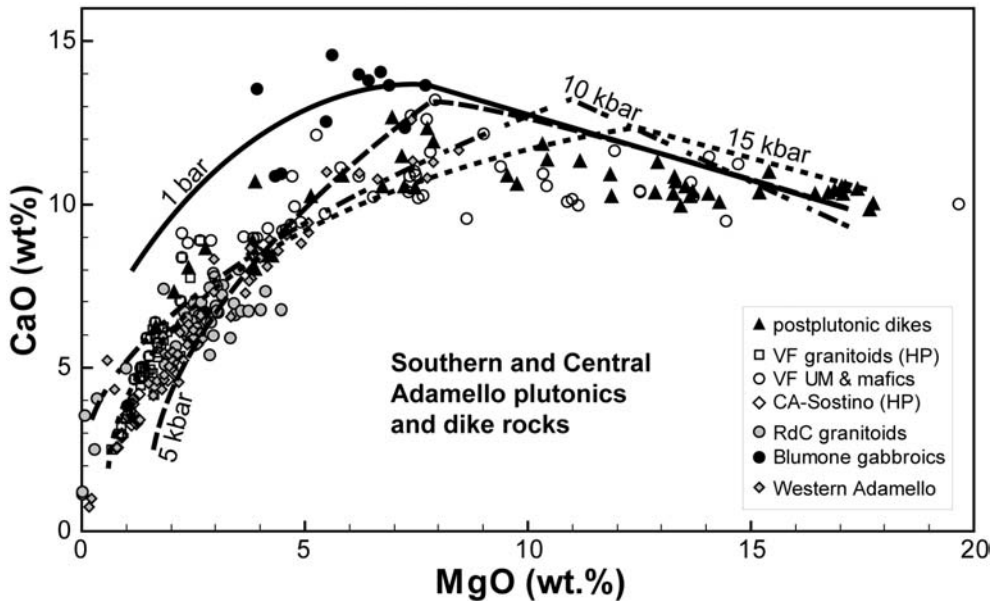
Fig. 6 – CaO – MgO diagram (in wt.%) displaying bulk rock data of plutonic and dike rocks from the southern Adamello (data compiled by the author, for references see text) with superimposed lines depicting experimentally derived liquid-lines-of-descent at 1 bar, 5, 10 and 15 kbar for picrobasaltic primary magmas (taken from Figure 4). Abbreviations: VF granitoids (HP) = Val Fredda leuco-tonalites evidencing derivation by crystallization at high pressures; VF UM & mafics = Val Fredda hornblendites and hornblende-gabbros; CA-Sostino (HP) = Corno Alto and Monte Sostino quartz diorites with high-pressure signatures; RdC granitoids = undifferentiated quartz diorites, tonalites and granodiorites from the Re di Castello superunit (S-Adamello); Blumone gabbroics = gabbro to quartz diorite rocks of the Cornone di Blumone complex evidencing low pressure crystallization at < 2-3 kbar; Western Adamello = gabbros to tonalites and leuco-tonalites of the Western Adamello superunit (Central Adamello, younger than Re di Castello superunit). For details see text.

Val Fredda ultramafic to gabbroic rocks plot at lower CaO contents than the Blumone rocks and, rather subtle but consistently, the intermediate to acidic rocks of the Val Fredda pluton plot at lower MgO for a given CaO than the derivative rocks of the Blumone complex and the majority of the southern Adamello granitoids. This is fully consistent with the derivation of the Val Fredda granitoids and similar tonalitic to granodioritic rocks associated with hornblende-rich cumulates (Corno Alto – Sostino and Malga Ervina trondhjemites) by high-pressure, amphibole dominated crystallization contrasting the cpx-plagioclase dominated fractionation assemblage of the Blumone intermediate compositions. These rather subtle differences are also clearly reflected in the trace element geochemistry of these two contrasting gabbroic to acidic plutonic systems: Fig. 7 displays a summary of REE data on tonalitic to granodioritic plutonic rocks from the southern Adamello ((Kaghami *et al.*, 1991; and unpublished data) distinguishing granitoids associated with hornblende-rich cumulates (e.g. Val Fredda) from granitoids associated with gabbroic cumulates dominated by cpx (e.g. Blumone complex). The granitoids that have formed by extensive amphibole fractionation and delayed plagioclase fractionation display lower middle to heavy REE abundances and trough-like patterns consistent with the removal of amphiboles that fractionate middle and heavy REE from light REE (e.g. Tiepolo *et al.*, 2007) and lack of any significant Eu anomaly consistent with delayed plagioclase crystallization and/or high fO_2 conditions.

An additional example of the influence of the crystallizing assemblage on the trace element geochemistry is provided in Fig. 8 displaying the rather well known (and often misused) Sr/Y
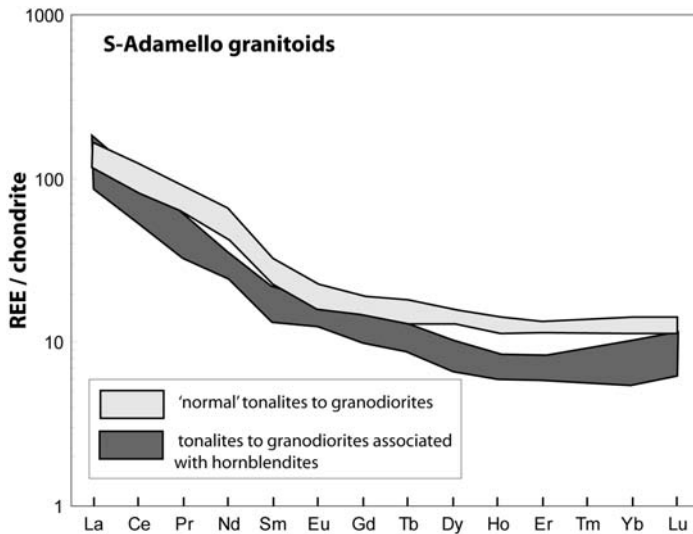
Fig. 7 – Rare Earth Element (REE) distribution diagram (C1 chondrite normalized) for southern Adamello (Re di Castello superunit) intermediate granitoids distinguishing between normal tonalites/granodiorites associated with hornblende-cpx gabbros and tonalites/granodiorites associated with hornblendites (cortlandites) displaying low concentrations of middle to heavy REE inferring a dominant role of amphibole as the dominant mafic phase and delayed plagioclase crystallization consistent with derivation from a magma reservoir located at greater depths than for the 'normal' tonalites/granodiorites.
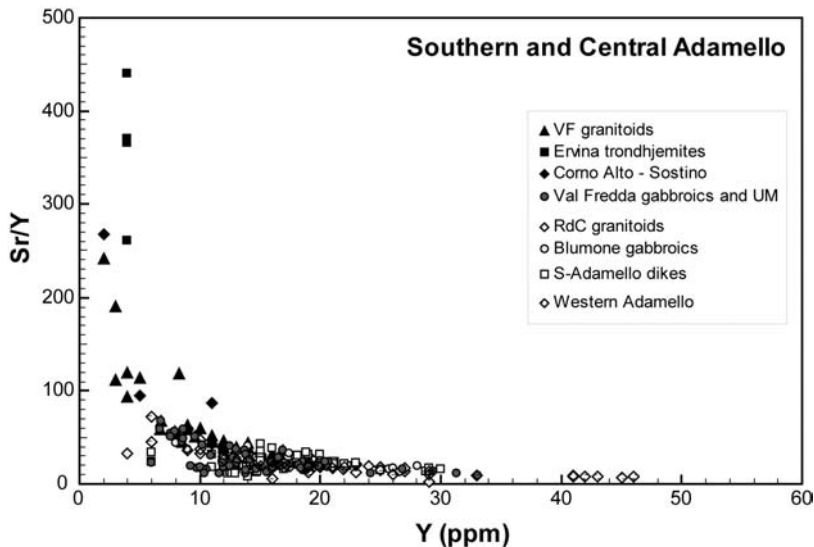
Fig. 8 – Sr/Y versus Y (ppm) diagram for southern Adamello plutonic and dike rocks. Solid symbols refer to intermediate to acidic plutonics associated with hornblendite cumulates of the Val Fredda, Corno Alto and Monte Sostino (Eastern Adamello) and the Malga Ervina trondhjemites (Upper Daone Valley); grey symbols refer to the hornblende-rich cumulates of the Val Fredda pluton; and open symbols refer to 'normal' granitoids associated with hornblende-pyroxene-bearing gabbroic cumulates (Blumone). Very high Sr/Y ratios at low Y concentrations are commonly associated with derivation from slab melts (garnet in the residue), but can alternatively be generated by abundant and early amphibole fractionation and delayed plagioclase crystallization typical for higher pressure crystallization-differentiation of andesite magmas in the lower arc crust.

versus Y diagram that is commonly employed to distinguish slab-derived melts ('adakites') with high Sr/Y and low Sr contents from liquids derived from mantle-melts (lower Sr/Y, higher Y). In general, high Sr/Y is attributed to the presence of garnet either in the source (e.g. eclogite) or during differentiation. However, some of the intermediate to acidic granitoids of the southern Adamello display very high Sr/Y ratios. Inspection of these samples reveals that they all belong to the aforementioned plutonic units that are associated with hornblenditic cumulates. Experimental data on the crystal-liquid partitioning of amphibole and acidic liquids at high pressures (R. Alonso Perez, *personal communication* 2007) indicate that an amphibole-rich assemblage, even without garnet, but in the absence of plagioclase at pressures of 8-12 kbar can indeed produce such high Sr/Y and low Y values because amphibole accommodates Y, but not Sr. Some of the data points located at very low Sr/Y ratio and Y contents of about 10 ppm in Fig. 8 are indeed hornblenditic cumulates that represent the cumulates extracted from andesitic liquids (100 ppm of Sr, 10 ppm of Y) to produce the leuco-tonalites and trondhjemites exhibiting high Sr/Y (500-1000 ppm Sr, 2 ppm Y) and that would normally been termed 'adakites'.

SUMMARY AND CONCLUSIONS

This contribution is a preliminary attempt to combine petrologic (field and geochemical) and experimental constraints to decipher the principal locations of differentiation of mantle-derived, hydrous, calc-alkaline magmas in continental margin settings. The results obtained by this combination is fully consistent with the proposal that basaltic to intermediate to acidic volcanic and plutonic rocks acquire their major (and most probably also their trace) element composition predominantly in deep-seated magma reservoirs located either close to the crust mantle boundary (30-45 km) or in deeper parts of the arc crust (15-25 km). This, in turn, is consistent with the absence of zoned igneous complexes in shallower level plutonic rocks that typically form calc-alkaline batholiths associated with subduction zone magmatism. However, the relationships at the lower end of MgO contents, i.e. for more

differentiated calc-alkaline rocks (granodiorites / rhyodacites) indicate that some (limited) shallow level differentiation in subvolcanic magma reservoirs (2-3 kbar) occurs in addition. This is clearly shown in the study presented by Grove *et al.* (2005) for Mt. Shasta, where both, deep seated and rather shallow liquid-lines-of-descent have been identified for a particular (high-Mg andesite) suite of volcanics and for some of the Southern Adamello tonalites formed as the product of shallow level crystallization of basaltic magmas in the Cornone di Blumone area.

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