

## Crustal Deformation Effects on the Chemical Evolution of Geothermal Systems: Case Studies from Southern Andes

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### ABSTRACT

A better understanding of the chemical evolution of fluids in geothermal and hydrothermal systems requires data-based knowledge regarding the interplay between active tectonics and fluid flow. The Southern Andes volcanic zone is one of the best natural laboratories to address this issue because of the occurrence of numerous geothermal areas, recent seismic activity generated by regional fault systems, and intense volcanic activity. Geothermal systems have been understudied in this area, and limited scientific information exists about the role of local kinematic conditions on fluid flow and mineralization during the development and evolution of geothermal reservoirs. In this study, we provide data for a 1:200,000 scale geological and structural map of the Villarrica–Chihuío area as a setting in which to perform a structural analysis of active geothermal areas. This structural analysis, combined with geochemical modelling of hot spring data, allows the identification of two magmatic-tectonic-geothermal domains based on fault systems, volcanic activity, and lithologies. The Liquiñe–Ofqui fault system (LOFS) domain encompasses geothermal areas located either along the master or subsidiary faults. These are favourably orientated for shear and extension, respectively. In the LOFS domain, the geochemistry of hot spring discharges is controlled by interaction with the crystalline basement, and is characterized by low B/Cl conservative element ratios and high pH. In marked contrast, the arc-oblique long-lived fault systems (ALFS) domain includes geothermal occurrences located on the flanks of volcanoes forming WNW-trending alignments; these systems are built over faults that promote the development of crustal magma reservoirs. Unlike the first domain, the fluid chemistry of these geothermal discharges is strongly controlled by volcanic host rocks, and is typified by lower pH and higher B/Cl ratios. Reaction path modelling supports our model: chemical evolution of geothermal fluids in the Villarrica–Chihuío area is strongly dependent on structurally controlled mechanisms of heat transfer. Within this framework, heat transfer by conduction is responsible for the LOFS domain, whereas magmatically enhanced advective transport dominates heat flow in the ALFS domain. Although more studies are needed to constrain the complex interplay between tectonics and fluid flow, results from this study provide new insights towards efficient exploration strategies of geothermal resources in Southern Chile.

### 1. INTRODUCTION

Geothermal activity is dependent upon the interaction between a heat source, circulating fluids, and permeable pathways. The conceptual models considering this interaction guide the exploration and exploitation of geothermal resources. The main inputs of the geothermal conceptual models are the permeability architecture and the fluid geochemistry (Goff and Janik 2000). The permeability architecture in geothermal systems is defined by the geometry and kinematics of fault–fracture networks (e.g. Sibson 1996). Faults may act as impermeable barriers to cross-fault flow or as high permeability conduits, although their permeability relative to the host rock depends on fault displacement, host rock lithology, hydrothermal mineral precipitation, and the seismic cycle. Fault–fracture networks including their damage zone damage zones are likely to develop directional permeability in the medium stress direction ( $\sigma_2$ ) (e.g. Sibson 1996). Correspondingly, the chemical evolution of geothermal fluids (Arnórsson et al. 2007) is defined by (1) transport and absorption of magmatic components (e.g. SO<sub>2</sub>, HCl, HF, H<sub>3</sub>BO<sub>3</sub>) producing acidic and reactive fluids; (2) heat–fluid–rock interaction processes which neutralize fluids and hydrothermally alter rocks; and (3) surface processes such as boiling, mixing, and dilution with meteoric fluids. The continuous hydrothermal mineral precipitation seals both intrinsic permeability related to rock porosity and open fracture networks (Cox 2010). Therefore, the processes defining the chemical evolution of fluids are facilitated or inhibited by the dynamical permeability architecture, which creates a highly anisotropic system (Rowland and Simmons 2012). A key question arising is how crustal deformation affects the chemical evolution of geothermal systems. The few works addressing this question have only analysed veins in fossil geothermal systems, which allows the determination of deformation (stress, strain) and thermodynamical (P–T–X) conditions of vein formation. However, the chemical analysis of fluids trapped in minerals as fluid inclusions has some analytical limitations, which have been only recently overcome. Yet, vein fluid inclusion analysis is not as precise and accurate as the routine analysis of geothermal waters. The few studies on active geothermal systems addressing the interplay between deformation and chemical evolution of fluids, emphasize the relevance of fault and fracture network geometry and kinematics (Rowland and Simmons 2012). However, the conceptual models proposed based on specific geodynamical context still need to be specified. The Southern Andes volcanic zone (SVZ) provides one of the best natural laboratories to address the interplay of fault–fracture networks and chemical evolution on geothermal systems, because of the occurrence of numerous geothermal areas (25% of Chilean geothermal areas), recent seismic activity generated by regional fault systems, and highly active volcanism. Tectonic activity is represented by two regional scale fault systems: the arc-parallel Liquiñe–Ofqui fault system (LOFS) and the arc-oblique WNW-striking long-lived basement fault systems (ALFS) (Cembrano et al. 1996). Furthermore, these fault systems are genetically and spatially related to the magmatic evolution in the SVZ, forming two categories of volcano–tectonic associations (Cembrano and Lara 2009). (1) The LOFS, with NNE-striking master faults favourably orientated for dextral shear with respect to the prevailing stress field and NE-striking tension fractures likely to form under relatively low differential stress. Volcanic activity comprises NE-striking volcanic alignments containing mainly

basaltic to basaltic–andesitic lithologies in either stratovolcanoes or minor eruptive centres. (2) The ALFS with WNW-striking faults severely misorientated with respect to the prevailing stress field. The volcanic activity comprises WNW-striking alignments of stratovolcanoes and displays a more evolved magma series (basaltic to rhyolitic). Our study case is located in the Villarrica–Chihuio area (39°S–40°S in Chile), where both categories of volcano–tectonic associations are present and there is a high density of geothermal areas (Figure 1). Moreover, the geothermal areas are spatially associated with the two different volcano–tectonic associations, being located either (1) over the NNE-striking master fault of the LOFS volcano–tectonic associations or (2) in the flanks of the volcanoes of the ALFS volcano–tectonic associations (Figure 1(B)). These two volcano–tectonic associations allow analysis of the role of regional faults and volcanism in defining the chemical evolution of geothermal fluids. The objective of this research is to get insights into the role of different fault systems in the chemical evolution of the geothermal fluids of the Villarrica–Chihuio area. Our hypothesis is that the interplay of volcanism and tectonics defines the occurrence of the major processes (e.g. magmatic gas absorption, fluid–rock interaction, fluid mixture) in the chemical evolution of geothermal fluids. This article aims to (1) interpret the role of fault systems in geothermal fluid flow through a structural analysis of published and new field data (geometry and kinematics) and (2) establish the processes that define the chemical evolution of fluids through sampling of thermal and meteoric fluids and the geochemical modelling. The details on the methods used are presented in Sánchez et al., 2013. Our results show the occurrence of two distinctive magmatic–tectonic–geothermal domains, which define the heating mechanism and chemical evolution of geothermal fluids in the SVZ. Although more studies are needed to constrain the often-complex interplays between tectonics and fluid flow, results from this study show that exploration of geothermal resources should focus on the long-lived arc-oblique magmatic–tectonic geothermal domain.

## 2. GEOLOGICAL SETTING

The main tectonic features in the Southern SVZ are the LOFS and the ALFS (Figure 1) (e.g. Cembrano and Lara 2009). The LOFS is a major intra-arc fault system that dominates the SVZ between 38°S and 47°S. The LOFS accommodates strain along the intra-arc by dextral strike shearing along the NNE-striking master fault and normal and dextral strike–slip in subsidiary ENE-striking faults (Rosenau et al. 2006). Fault–slip data and stress tensors for Pleistocene deformation along the northern portion of the LOFS consistently show a subhorizontal maximum principal compressive stress ( $\sigma_{Hmax}$ ) trending N60°E (Rosenau et al. 2006). NNE-striking master faults are favourably orientated for dextral shear with respect to the prevailing stress field and ENE-striking tension fractures likely form under relatively low differential stress (Cembrano and Lara 2009). In the above-described tectonic setting, the WNW-striking faults of the ALFS are severely misorientated with respect to the prevailing stress field and have been interpreted as crustal weaknesses associated with pre-Andean faults reactivated as sinistral-reverse strike–slip faults during arc development. These fault systems are recognized as the contact of basement rocks with younger units and in some cases present mylonites as evidence of ductile deformation.

## 3. RESULTS

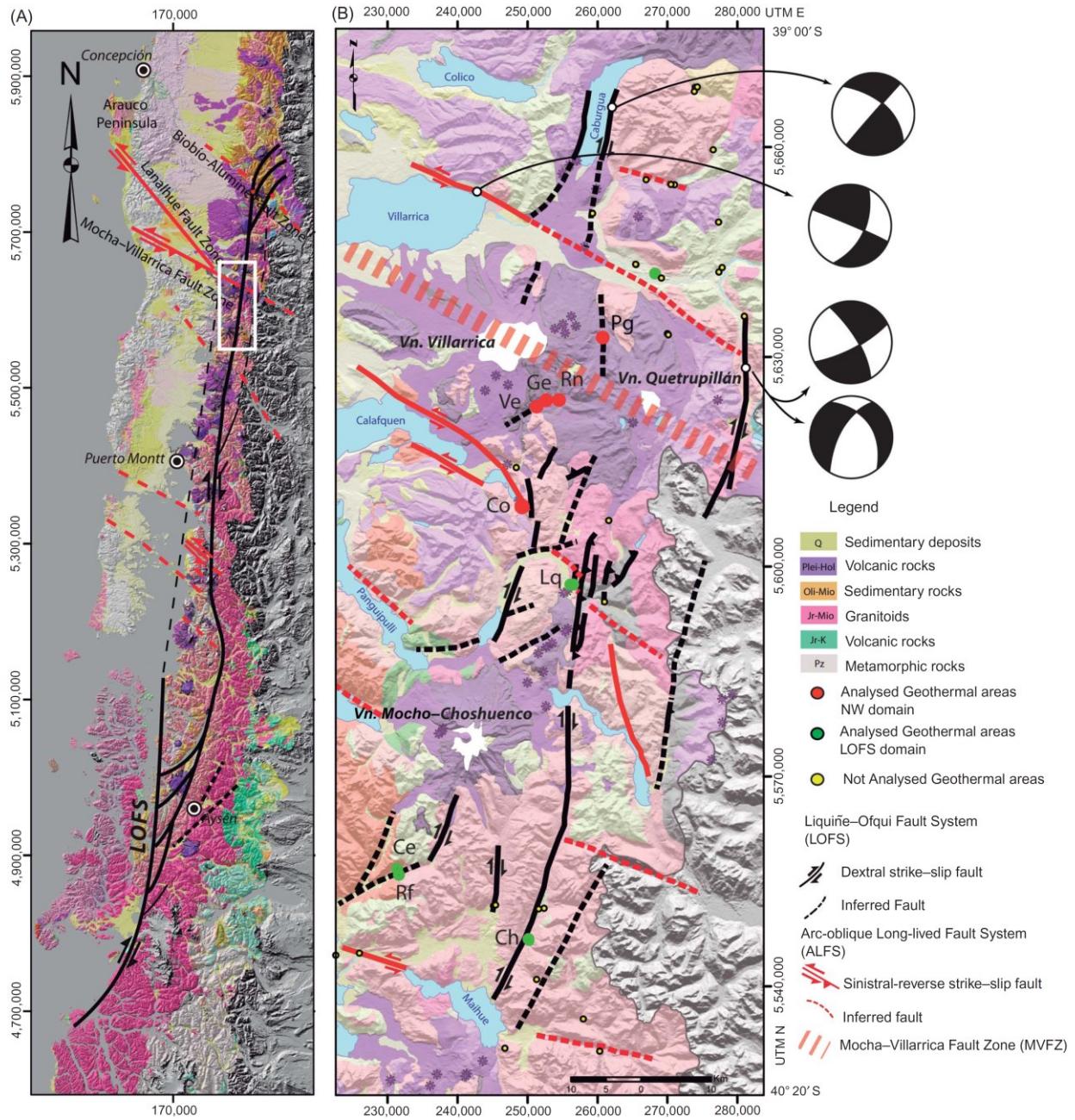
### 3.1 Fault systems

The geological and structural map of the Villarrica–Chihuio area (Figure 1(B)) shows the location of the main stratovolcanoes (Villarrica, Quetrupillán, Mocho–Choshuenco), geothermal areas, and fault systems. Within this area, two contrasting lithology assemblages exist: a crystalline basement of low intrinsic permeability, overlain by relatively permeable Pliocene to Holocene volcanoclastic units. The geothermal areas are spatially correlated to fault systems, and some of them are in the flanks of the Villarrica volcano (Figure 1(B)). To assess the interplay of volcano–tectonic associations and geothermal systems we define two magmatic–tectonic geothermal domains: the LOFS domain and the ALFS domain. The LOFS domain includes geothermal areas located along either master or subsidiary faults of the LOFS. There, thermal water emerges from fractures in granitoids or sediments overlying them. Main structural features of this domain are the arc-parallel NNE-striking dextral strike–slip master faults and subsidiary NE-striking dextral and normal faults which are favourably oriented for shear and/or extension with respect to the prevailing stress field. The LOFS cuts and displaces the ALFS in the vicinities of Liquiñe (Lq) and Coñaripe (Co), but locally, the opposite cross-cutting relationship is observed (Figure 1(B)). Volcanic systems linked to these faults exhibit primitive magmas, which are transported through tension cracks with no magmatic chamber development (Cembrano and Lara 2009). The ALFS domain hosts geothermal areas, which are located on the WNW-trending aligned volcano flanks where hot springs discharge from volcanic rocks or sedimentary deposits overlying them. The condition for reactivate faults of ALFS promotes long residence of magma in crustal reservoirs, which may serve as heat sources for geothermal systems. Most of the studied geothermal areas consist of several hot springs, which in some cases form well-defined alignments. These alignments are mainly NE and coincide with the faults mapped in the vicinities of both magmatic–tectonic–geothermal domains. In the Geométricas (Ge) and Vergara (Ve) geothermal areas from the ALFS domain (see Figure 1), hot springs are aligned N55E and N62E, respectively, spatially associated with a NE inferred fault affecting Pleistocene–Holocene volcanic units. In the Liquiñe (Lq) geothermal area, which belongs to the LOFS domain, the hot springs trend N15E and lie within fractures in granites, close to one NNE master fault of the LOFS.

### 3.2 Chemistry of thermal fluids

The thermal waters have low content of dissolved solids (TDS <550 mg/l) and are classified as Na–SO<sub>4</sub> type based on major ion concentration. The thermal waters of the LOFS domain are alkaline (pH 8.9–9.7) with surface temperatures between 37 and 82°C, whereas those of the ALFS domain are sub-alkaline (pH 7.8–8.7) and have temperatures from 36°C to 70°C. The Cl–SO<sub>4</sub>–HCO<sub>3</sub> diagram is commonly used to classify geothermal fluids and interpret the main geochemical processes occurring. In this diagram, the low chloride and relatively high sulphate concentrations place thermal waters of Villarrica–Chihuio close to the steam-heated water field. The ratios between conservative elements such as chorine (Cl), boron (B), and lithium (Li) normally do not get modified and remain unchanged even with dilution, which allows them to be used as tracers of the geothermal fluid sources (Giggenbach, 1991). In the B–Cl–Li triangular plot the two domains are plotted as different clusters (Figure 2). Also, the absolute B concentrations in the ALFS domain (0.4–7 mg/l) are higher than the background levels detected in rivers and cold springs nearby (B < 0.03 mg/l; e.g. Aihue river, San Luis cold spring; Pérez 1999). The ratio-based geothermometers (Na–K, K–Mg) are considered to be less affected by dilution than single constituent-based geothermometers. The Na–K–Mg ternary diagram allows the estimation of temperatures in subsurface and approach to equilibrium state (Giggenbach 1991). Fully equilibrated samples have all the phases saturated and both geothermometers whereas partially equilibrated waters suffer re-equilibration during fluid ascent,

but both are suited for temperature estimations. In the study area, Ch, Lq, Ge, Rf, and Co samples are in the partially equilibrated field. Considering the geothermal areas least affected by dissolution, as indicated by higher temperature, and Silica and Cl concentrations of the geothermal discharges, we obtained estimations for reservoir temperatures of 100–150°C (Lq, Ch) for the LOFS domain and 140–180°C (Ge) for the ALFS domain.



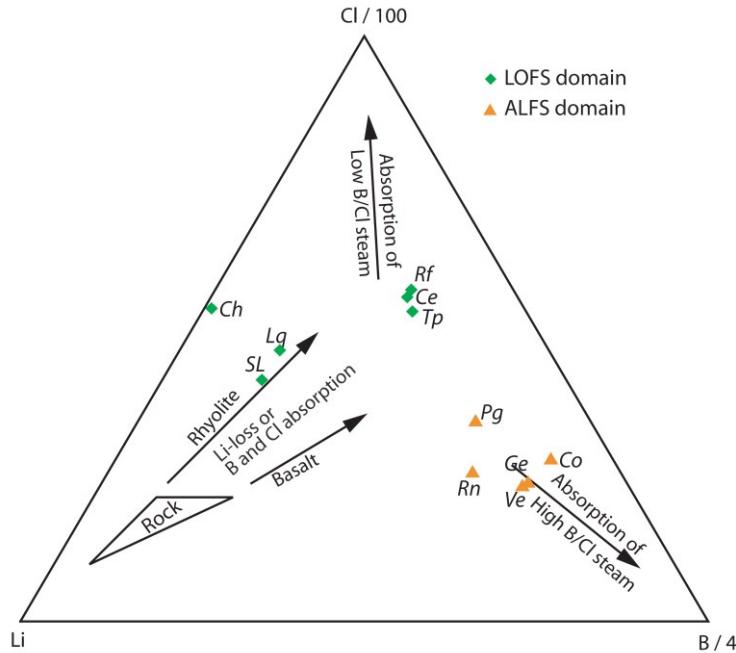
**Figure 1: (A) Regional geological map of the Southern Volcanic Zone showing the LOFS and the ALFS fault systems. (B) Geological map of the Villarrica–Chihui area with the compilation and reinterpretation of fault systems based on Lara and Moreno (2004); Moreno and Lara (2008); Cembrano and Lara (2009); Rosenau et al. (2006); Rosenau (2004); and Potent (2003). Also shown are the chemically analysed geothermal areas of the two magmatic-tectonic-geothermal domains: LOFS domain (green dots) and ALFS domain (orange dots). The rest of the geothermal areas are in yellow. The P/T dihedra related to faults are from Potent (2003), showing dextral strike-slip in the LOFS and sinistral strike-slip in the ALFS. Figure from Sanchez et al., 2013.**

## 4 DISCUSSION

### 4.1 Interplay of faults system and fluid flow

From the analysis of fault systems, volcanic activity, and hydrothermal systems, we have identified two distinctive magmatic-tectonic-geothermal domains: the LOFS domain and the ALFS domain (Figure 1(B)). Although these domains are not completely independent of each other, they reflect the different natures of the geothermal systems, which are defined by their heat sources. We propose that the nature of the heat source for the geothermal systems of the LOFS domain is the high heat flux in an intra-arc region, which is transferred by conduction to the deep circulating fluids (Figure 2). In the ALFS domain, in turn, magmatically

enhanced advective transport dominates heat and mass flow (Figure 2). The WNW-striking ancient faults, which are misorientated with respect to the prevailing stress field, provide conditions for magma reservoir development. Although these two distinctive domains show contrasting features, the ultimate heat source for both of them is the melt in the MASH zone (Hildreth and Moorbath 1988).



**Figure 2: Conservative element diagram Cl–B–Li.** Fields defined by Giggenbach (1988). LOFS domain samples in green diamonds and ALFS domain in orange triangles. Note: SL = San Luis; Pg = Palguín; Geométricas = Ge; Rincón = Rn; Vergara = Ve; Coñaripe = Co; Trifupán = Tp; Liquiñe = Lq; Río Florín = Rf; Cerrillos = Ce; Chihuío = Ch. Figure from Sanchez et al., 2013.

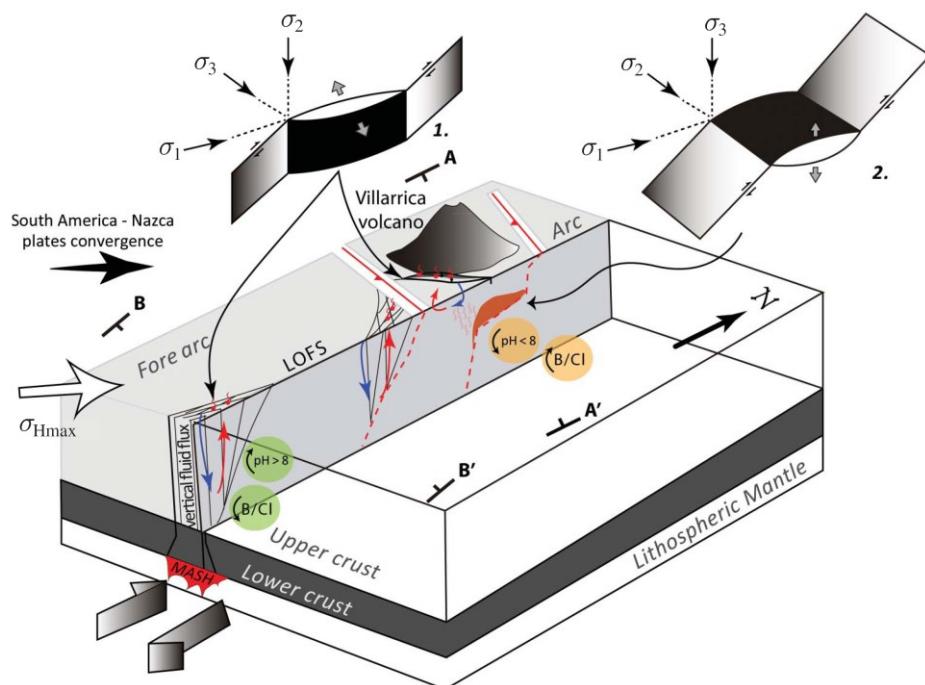
Most of the geothermal areas are spatially associated with regional fault systems, and the hot spring alignments exhibit a similar trend. This is consistent with the idea of a first order control exerted by brittle deformation on hydrothermal fluid flow (e.g. Sibson 1996). The latter suggests that geothermal fluid flow is concentrated in fracture networks once the initial porosity is destroyed by hydrothermal alteration (e.g. Cox 2010). Thus, the secondary architecture permeability overprints the intrinsic permeability anisotropies originated by stratification in porous rocks. Fault–fracture networks, consisting of faults, extensional fractures, and extensional shear-fractures are conduits for hydrothermal fluid flow. Activation and preservation of fault–fracture networks as highly permeable conduits require the condition of fluid pressure  $P_f \sim \sigma_3$  (Sibson 1996). In these networks, the directional permeability follows a direction parallel to  $\sigma_2$ , which is perpendicular to the slip vector (Sibson 1996). Therefore, in the damage zone of the strike-slip LOFS domain ( $\sigma_1 \sim N60E$ ; Lavenu and Cembrano 1999; Rosenau et al. 2006), the vertical permeability is enhanced. These are the ideal conditions to form a deep convection cell. However, in the ALFS domain the permeability is only enhanced under fluid overpressure conditions, i.e. when fluid pressure ( $P_f$ ) is higher than lithostatic pressure ( $P_l$ ), because  $\sigma_3 = \sigma = \rho gh = P_l$ . When this condition is met, fault–fracture networks mainly promote lateral circulation of fluids. At shallow levels ( $< 2$  km) and hydrostatic conditions, it is more likely to activate tension fractures or shear fractures instead of misorientated faults (Rowland and Simmons 2012). That is the case of geothermal fluids at shallow levels in both domains, transported through ENE-tension fractures as reflected by ENE-aligned hot springs (Figure 3). The recharge of the systems with meteoric water is also likely to occur through ENE-tension fractures of fault–fracture networks.

We propose that these magmatic-tectonic-geothermal domains are also represented in the rest of the SVZ. Evidence to support this idea is the high geothermal resource potential areas, genetically related to WNW volcanic chains, such as Nevados de Chillán (250°C; Lahsen et al. 2010), Tolhuaca volcano (250°C; Melosh et al. 2012), and Puyehue Cordón–Caulle (300°C, Sepúlveda et al. 2007). Thus the ALFS domain hosts geothermal systems heated by a shallow magmatic reservoir and favours the development of high enthalpy resources. However, detailed structural and chemical analysis should be combined to further quantify the effect of faults and fractures on those geothermal systems. Emphasis is required in those fluids least affected by shallow dilution (geothermal reservoirs) with the determination of the open fracture disposition and the local stress fields.

#### 4.2 Chemical evolution of geothermal fluids

The origin of water in Villarrica–Chihuío thermal fluids at the surface is practically only meteoric water without a significant contribution of magmatic fluids, as shown by isotope data (Sánchez et al., 2013). Moreover, thermal waters display very small or no  $\delta^{18}\text{O}$  shift with respect to GMWL, despite this shifting normally occurring due to fluid–rock interaction at geothermal temperatures. The lack of  $\delta^{18}\text{O}$  shift may be produced by an insufficient temperature for isotopic exchange and/or may be indicative of a system with very high water/rock ratios or a system that does not have enough time to equilibrate with the surrounding rocks, together with the effect of mixing of meteoric water at shallow depths.

In geothermal systems associated with volcanism, the mixed volcanic condensate ( $\text{H}_2\text{O}$ ,  $\text{CO}_2$ ,  $\text{HCl}$ , and  $\text{SO}_4$ - rich) and heated meteoric waters are separated in two phases through adiabatic decompression, allowing the ascent of a gaseous phase rich in sulphur ( $\text{H}_2\text{S}$ ). The condensation of the primary magmatic volatiles or the secondary steam phase into ground-surface waters forms steam-heated waters (Giggenbach 1991). The clustering of the geothermal discharges of the ALFS domain in the  $\text{Cl}-\text{SO}_4-\text{HCO}_3$  anion ternary diagram, together with decreasing  $\text{SO}_4/\text{Cl}$ ,  $\text{SO}_4/\text{HCO}_3$  ratios away from the Villarrica volcano, is consistent with a steam-heated interpretation for these discharges. However, steam-heated waters are in general acid fluids ( $\text{pH} < 4$ ) (Giggenbach 1991), with only a few exceptions. The prominent degassing from the crater lake of the Villarrica volcano ( $460 \pm 260$  tons/day  $\text{SO}_2$ ; Witter et al. 2004) validates this interpretation. Absorption and condensation of the gaseous phase rich in sulphur species and low in chloride causes the  $\text{NaSO}_4$  type waters, with a higher  $\text{B}/\text{Cl}$  ratio for the discharges of the ALFS domain. Therefore, the chemical evolution of fluid from the ALFS domain is consistent with a magmatic contribution of heat and mass and neutralization by water–rock interaction and fluid mixing with groundwater. The contribution of a magmatic source of fluids is not evident in the LOFS domain data. The lower  $\text{B}/\text{Cl}$  ratio,  $\text{B}$  and  $\text{Cl}$  absolute concentrations, and higher  $\text{Na}/\text{H}$  and  $\text{K}/\text{H}$  activity ratios may reflect the complete lack of magmatic contribution and considerable fluid–rock interaction. The geological features of the LOFS domain, emplaced in the granitic rocks farther than 25 km from the nearest stratovolcano, where migration of the magmatic fluids is unlikely, supports this idea. Both conceptual models proposed for the ALFS and LOFS domain contrast with the seawater origin of thermal water in SVZ proposed by Risacher et al. (2011). Alternatively, we propose that the interplay of volcanism and tectonics defines the nature and origin of geothermal systems and that the contribution of a seawater signature through rain is negligible, at least when geothermal systems are as far as 150 km from the ocean.



**Figure 3: Illustration summarizing the physical and chemical processes governing the two magmatic-tectonic-geothermal domains in the Villarrica–Chihui area. The nature of the heat source for the geothermal systems of the LOFS domain is the high heat flux in an intra-arc region, which is transferred by conduction to the deep circulating fluids. In the ALFS domain, magmatically enhanced advective transport dominates heat and mass flow. The chemical signature of fluids is shown in circles for the LOFS domain (green) and ALFS domain (orange). Note: Blue arrows = cold meteoric water; red arrows = thermal fluids;  $\sigma_{\text{Hmax}}$  = direction of the maximum horizontal stress. Figure from Sanchez et al., 2013.**

## 5 CONCLUSIONS

We have identified two magmatic-tectonic-geothermal domains based on the nature and kinematics of fault systems, volcanic activity, and rock types: the ALFS domain and the LOFS domain. The chemistry of the fluids shows contrasting signatures in these domains in conservative elements ( $\text{Cl}-\text{B}-\text{Li}$ ) and activity diagrams. We propose that the role of fault systems on the geochemical evolution of geothermal fluids occurs through the development of magmatic-tectonic-geothermal domains, which ultimately defines the heat source.

- In the LOFS domain, fault–fracture networks related to the damage zone of the deep seated NNE-striking master fault increases vertical permeability in the crystalline basement. These fracture networks promote development of deep (<3 km) convection cells and heat–fluid–rock interaction after the infiltration of meteoric water (Figure 3). The ultimate heat transfer mechanism of fluids is by conduction from the crystalline host rock in the high heat flow realm of the intra-arc (Figure 3). The heat–fluid–rock interaction and the lack of direct magmatic contribution imprints a signature of low  $\text{B}/\text{Cl}$  ratios and high pH in the composition of the geothermal fluids. The hot springs discharge from the faulted crystalline basement.

● In the ALFS domain, the WNW-striking inherited basement faults, which are strongly disorientated with respect to the prevailing stress field, provide suitable conditions for the development of magma reservoirs. These crustal magmatic reservoirs are the source of heat and mass for the geothermal systems. The mass transfer results in a signature of higher B/Cl ratios and neutral pH in the chemistry of the geothermal fluids compared to those of the LOFS domain. Therefore, the geochemical evolution in the ALFS domain can be represented as meteoric water absorption of magmatic gases, interaction with volcanic rocks, and dilution (Figure 3). The discharge of these systems is through volcanic units, which may promote lateral fluid flow.

● At shallow levels (<2 km) under hydrostatic conditions, the fluid flow is through NE-tension fractures optimally orientated for reactivation where discharge (hot springs) and recharge (meteoric waters) of fluids in the geothermal systems is likely to occur. Our conceptual model of the effects of crustal deformation on the geochemical evolution of fluids might be applicable to the rest of the Southern Volcanic Zone, where the most prominent geothermal resources are genetically related to WNW volcanic chains.

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