HYDROGEOCHEMICAL CHARACTERIZATION OF THERMAL SPRINGS, LOS LAGOS, CHILE

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ABSTRACT

Los Lagos District is considered to be an area of interest for geothermal development because of the presence of numerous volcanic complexes and thermal springs as well as two major tectonic features: the NNE-SSW dextral-reverse Liquiñe-Ofqui Fault System (LOFS) and the WNW-ESE sinistral Arc-Oblique Long-lived Faults System (ALFS). Two identified groups of hot springs are believed to have formed based primarily on geographic location. The first group formed along the ALFS (Porcelana Chico, El Comau and Porcelana Grande), and is characterized as having a direct volcanic influence related to horizontal transport of fluid (expressed by higher B/Cl ratios), a geothermal source of chlorides and a minor cation exchange. These thermal waters record the highest temperatures. The second group is comprised of thermal waters along the LOFS and is subdivided into two subsets: (i) hot springs localized in the intertidal zone (Rollizos, Cochamó, Sotomó y El Yate) that have a slight interaction with saline water and a chloride mix origin; and (ii) those located in the continental zone, (Puyehue, Aguas Calientes, Rupanco, El Callao, Cayetué, Ralún, Puelo, Pichicolo, Llancahué, Cahuelmó y El Amarillo hot springs) which are especially far from estuaries and fjords and show an important cation exchange, this behavior is believed to reflect vertical transport of fluids that has ultimately led to an interaction with North Patagonian Batholith (NPB) rocks.

1. INTRODUCTION

Los Lagos District is located in southern Chile (40.5 – 44°S) (Figure 1), along the subduction margin of Nazca plate under the South American plate with rising magmatic fluids that have generated higher thermal gradients and a high amount of thermal waters. This district contains more than thirty springs distributed from north to south along structures belonging to the Liquiñe-Ofqui Fault System (LOFS) and the Arc oblique Long-lived basement Fault System (ALFS). Various types of volcanic complexes are also found along both of these fault systems. Los Lagos District, with thirty six identified thermal waters zones, represents an interesting area to analyze the potential influence of hot springs fluids chemistry (i.e. present lithology, fluids mixture), because springs are found in the intertidal zone, on the slopes of volcanoes and away from both the sea and volcanoes. The Continental Chiloé Miocene Gold Belt (SERNAGEOMIN-BRGM, 1995; Duhart *et al.*, 2001) is present in the same area as are several thermal springs, and parts of the stages of formation of these deposits could be related to the hot springs fluid transport through the same fault systems. Despite these known correlations, there is very little geochemical data about the geothermal systems of this part of the country and therefore limited knowledge on the potential use of these hot springs as a renewable energy source.

To understand the possible uses of geothermal energy from the local hot springs, geochemical analyses are necessary to constrain the system dynamics, features at depth and source of heat and geothermal fluids.

This study deals with the characterization of eighteen thermal waters recorded on the hot springs national cadastre (Hauser, 1997; Pérez, 1999; Flores, 2011), including their chemical composition as well as their field measurements of thermodynamic characteristics, estimated depth temperatures and basic geometry models created from Sánchez *et al.* (2013), where relations between hot springs chemical composition and adjacent structures are shown.

2. GEOLOGICAL BACKGROUND

Los Lagos District is located in the volcanically active Southern Volcanic Zone SVZ (Stern, 2004). The oblique convergence between the Nazca and South American plates produces varying stresses that generate numerous folds and faults with different orientations depending on the tectonic setting. Two major tectonic structures define the study area (Figure 1). The first tectonic feature is the NNE-SSW trending dextral-reverse faults of the LOFS (Cembrano *et al.*, 2000; Cembrano and Lara, 2009). Since this fault system is produced by oblique stress, there are transtensive basins that allow fluid ascent (Cembrano and Lara, 2009).

The second group consists of WNW-ESE trending, primarily sinistral faults of the ALFS that were formed by the reactivation of pre-Andean faults (López-Escobar *et al.*, 1995; Rosenau *et al.*, 2006; Sánchez *et al.*, 2013).

Lithological units are distributed along a N-S trend, subparallel to the main plate margin. Bahía Mansa Metamorphic Complex (BMMC) rocks outcrop in the Coast Range, Chiloé Island and the western continental part of the district (Duhart *et al.*, 2001), that are believed to have a Devonian-Permian protolith. The BMMC rocks are composed of pelitic to semipelitic schists, meta-graywackes, metabasites and some mafic to ultramafic plutons; with greenschist facies metamorphism.

North Patagonian Batholith rocks are also found at numerous locations in the eastern area of Los Lagos District (40 - 47°S) from the Andean Cordillera to the Coast Range (Adriasola *et al.*, 2005). The batholithic rocks consist of tonalites and granodiorites that formed from two geochronological groups led by changes in subduction dynamics: Upper Jurassic – Middle Cretaceous and Early Miocene – Pliocene.

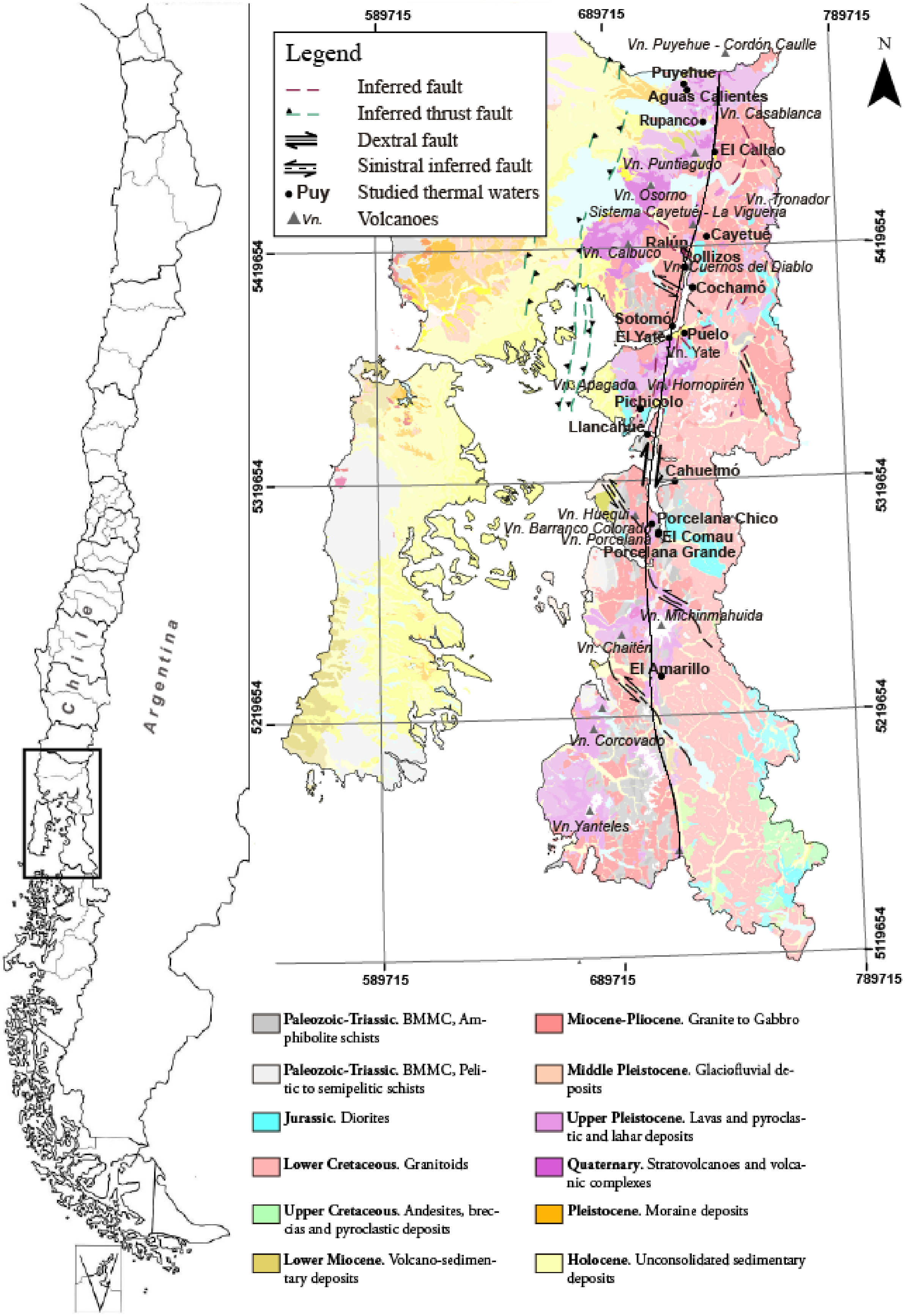

Figure 1: Position, geology, structures, volcanoes and studied hot springs of Los Lagos District. BMMC, Bahía Mansa Metamorphic Complex (SERNAGEOMIN-BRGM, 1995; Rosenau *et al.*, 2006; Aguilera *et al*, 2014).

There are numerous unconsolidated sedimentary deposits in the study area that formed by volcanogenic, glaciofluvial, moraine and modern fluvial processes (Adriasola, 2003) along with the presence of clays.

Different volcanic complexes (from basaltic to rhyolitic compositions) are present along these two fault systems based on the different permeability indexes of these structures. The volcanic systems aligned along a NNE-SSW trend correspond to Pleistocene-Holocene stratovolcanoes with a basaltic to andesitic composition, related to less magmatic differentiation during the ascent along structures of the LOFS (e.g. Cembrano and Moreno, 1994; López-Escobar *et al.*, 1995; Cembrano and Lara, 2009). Examples of this volcanic type that have affected the studied geothermal systems are Casablanca, Yate and Apagado volcanoes. In addition to the LOFS volcanoes, there are volcanic complexes localized along ALFS structures that have a NW-SE orientation and a variable chemical composition from basaltic to rhyolitic. Calbuco, Huequi and Barranco Colorado volcanoes are some examples of this group.

3. SAMPLING AND METHODS

Samples of eighteen hot springs and twelve surface waters were collected from three fieldwork campaigns. The sampling was done in late summer in March 2013, February 2014 and March 2014 following the sampling protocol of the Chilean National Geology and Mining Service (SERNAGEOMIN). All samples were collected and stored in high-density polypropylene bottles. Sample volumes and treatments of waters varied based on the chemical analysis used: (1) water samples for analysis of major elements concentration and for obtaining H and O isotopic ratios were raw waters only; and (2) those for analysis of trace elements concentration were filtered through a 0.45 µm syringe filter and acidified with the addition of 2 ml of HNO_3. Physicochemical parameters were measured in situ during field sampling with a portable multiparameter: temperature, pH, conductivity, salinity and the total dissolved solids index.

These samples were sent to the SERNAGEOMIN Chemical Laboratory, where the following methods were applied: (1) acid-base titration for alkalinity measurement; (2) Atomic Absorption Spectrophotometer (Perkin Elmer, AAS-4000, USA) for cation and SiO_2 composition, with an analytical error (AE) $\leq$2%; (3) Ion Chromatography (Dionex, ICS 3000, USA) for anion composition, with an AE $\leq$5%; (4) trace elements composition were obtained by Inductively Coupled Plasma and Mass Spectrometry (ICP-MS, Agilent 7500a, Agilent, USA), with an analytical error (AE) $\leq$2%; and (5) H and O isotopic ratios were detected by Isotope-Ratio Mass Spectrometry (IsoPrime100, Isoprime, UK), where analytical precision is $\leq$0,15 ‰ for $\delta^{18}O$ and $\leq$2,0 ‰ $\delta^{2}H$.

4. RESULTS

4.1 Hydrochemical features of selected hot springs

The chemical concentrations and isotopic ratios of sampled hot springs generated different classifications as shown below.

4.1.1 Geographic classification

A classification of studied hot springs by their geographical location was made, where main components of a geothermal system, as permeability and heat and water sources present distinct possible characteristics:

(a) Springs related to volcanic systems (VS): This hot springs are situated close to some eruptive centers, however they do not show an acidic behavior as it could be expected, but they have some of the highest surface temperatures (Table 1). This group is represented by red symbols in the geochemical diagrams of this study.

(b) Springs located along LOFS structures in the continental areas (LOFS-C): These springs are far from both volcanic centers and marine and estuary waters. The pH values are slightly alkaline and the temperatures are all similar. This group is plotted as green symbols in the geochemical diagrams.

(c) Springs located along LOFS structures in the intertidal zone (LOFS-I): these thermal waters are situated adjacent to LOFS structures that are exposed in intertidal areas, so estuary and seawater cover the hot springs outputs at high tide. Their pH and surface temperatures values cover a wider range, including the coldest hot springs. These thermal waters are represented by blue symbols in the geochemical diagrams.

Table 1. Geographic classification of studied hot springs. VS: Springs related to volcanic systems; LOFS-C: Springs located along LOFS structures in the continental areas; LOFS-I: Springs located along LOFS structures in the intertidal zone.

Geographic group	Hot springs	Temperature [°C]	pH
VS	El Yate	30.2	7.19
	Pichicolo	42.2	7.94
		49.6	7.06
		56.6	8.95
	Porcelana Chico	85.1	8.04
	El Comau	74.8	7.91
	Porcelana Grande	57.3	7.33
LOFS-C	Puyehue	58.2	8.51
	Aguas Calientes	60.8	7.69
	Rupanco	54.4	7.86
	El Callao	55.9	9.5
	Cayetué	50.5	7.64
	El Amarillo	53.8	9.5
		53.1	9.49
LOFS-I	Ralún	40.1	7.25
	Rollizos	29.7	7.82
		28.4	8.04
	Cochamó	25.9	7.73
		24.7	8.02
	Sotomó	30.8	7.94
	Puelo	34.9	8.01
	Llancahué	51.6	7.83
	Cahuelmó	60.9	9.06

4.1.2 Major Elements classification

Using major ions concentrations, it is possible to recognize various clusters in the Giggenbach diagram (Giggenbach, 1991). In the anion diagram (Figure 2), the blue symbols (LOFS-I) fall close to the chloride waters area except for the Cahuelmó waters, while the red symbols are closer to the volcanic waters zone. Puyehue, Aguas Calientes, Rupanco and El Callao are also closer to the volcanic waters zone except for the Porcelana Chico hot spring, which is located in the mature waters area. The Cayetué and

El Amarillo hot springs are plotted close to the steam-heated waters area.

Chlorides are anions are typically known to be chemically conservative constituents encountered in thermal fluids, so plotting cations versus chlorides composition allows groups to be distinguished according to the behavior of local cold surface waters. Based on this relationship, 3 main groups are able to be distinguished:

(a) Group 1: This group consists of the Porcelana Chico, El Comau, Porcelana Grande, Puyehue, Aguas Calientes, Rupanco, Pichicolo and El Yate hot springs. These waters have normal Na concentrations with respect to the homogeneous mixing line (Figure 3) and intermediate concentrations of chlorides (221.7 mg/l average). From these measurements and some cation concentrations, three subgroups are identified (Figure 3):

- Group 1A: Porcelana Chico, El Comau and Porcelana Grande form this group that shows higher Boron and Lithium concentrations and therefore the highest B/Cl ratio of the samples analyzed (Figure 4). Also these samples are located along ALFS structures (Figure 1) and present the highest chloride concentrations of Group 1 (528, 293, 238 mg/l, respectively).

- Group 1B: This subset includes the Puyehue, Aguas Calientes and Rupanco hot springs, which have high to intermediate B/Cl ratios of about 0.01 (Figure 4) and chloride concentrations between 133.97 and 184.96 mg/l. All of these samples are exposed in outcrops along LOFS structures and have a narrow range of temperatures (54.4, 58.2 and 60.8°C, respectively).

- Group 1C: The Pichicolo and El Yate hot springs belong to this group, with higher $Cl/(SO_4+Cl+HCO_3)$ ratios (Figure 2), lower B/Cl ratios (Figure 4) and lower temperatures (Figure 6 and Figure 7) compared to the other subsets (El Yate: 30.2°C; Pichicolo: 42.2-56.6°C).

(b) Group 2: This group is composed of the El Callao, Cayetué, Cahuelmó and El Amarillo hot springs, which have the highest $SO_4/(SO_4+Cl+HCO_3)$ (Figure 2) and Na/Cl ratios (Figure 3) and lowest Cl concentrations (24.59, 17.70, 12.00 and 7.30-7.60 mg/l, respectively).

(c) Group 3: This set is only composed of the LOFS-I hot springs (Ralún, Rollizos, Cochamó, Sotomó, Puelo and Llancahué), which have the highest concentration of chlorides and the lowest surface temperatures (between 24.7 and 40.1°C), with the exception of Llancahué waters that have a temperature of 51.6°C. Moreover, it is important to note that these hot springs have low but varied boron concentrations (Figure 4), with Sotomó waters having similar B/Cl and Br/Cl ratios as estuary and fjords samples (Figure 4 and Figure 5).

4.2 Hydrochemical geothermometry

In this study, three different geothermometers were used to estimate the depth temperatures of analyzed hot spring from Los Lagos District: Na-K-Mg (Giggenbach, 1988), K/Mg (Fournier, 1991); Na/K (Díaz-González *et al.*, 2008) and SiO_2 (Verma and Santoyo, 1997) geothermometers.

The most reliable results were obtained from the SiO_2 geothermometer because the equation has been adapted to systems with temperatures ranging from 20-210°C (Verma and Santoyo, 1997). Also, a plot of SiO_2 and surface temperatures follows a linear trend (Figure 6). In contrast, data from the Na-K-Mg, K/Mg and Na/K geothermometers have a higher dispersion of values that is attributed to a greater influence of water-rock interaction (Ruiz, 2015) as will be discussed below (Figure 7), this wide distribution is visible also in Na vs Cl diagram, (Figure 3a). According to these values, the hot spring that recorded the highest depth temperature with the K/Mg, Na/K and SiO_2 geothermometers is Porcelana Chico (also known as the Porcelana geysers): 398.5, 191.6 and 180.7°C, respectively.

In general, it is also possible to show that the LOFS-I hot springs have the lowest estimated depth temperatures, especially those belonging to Group 3 (Figure 6).

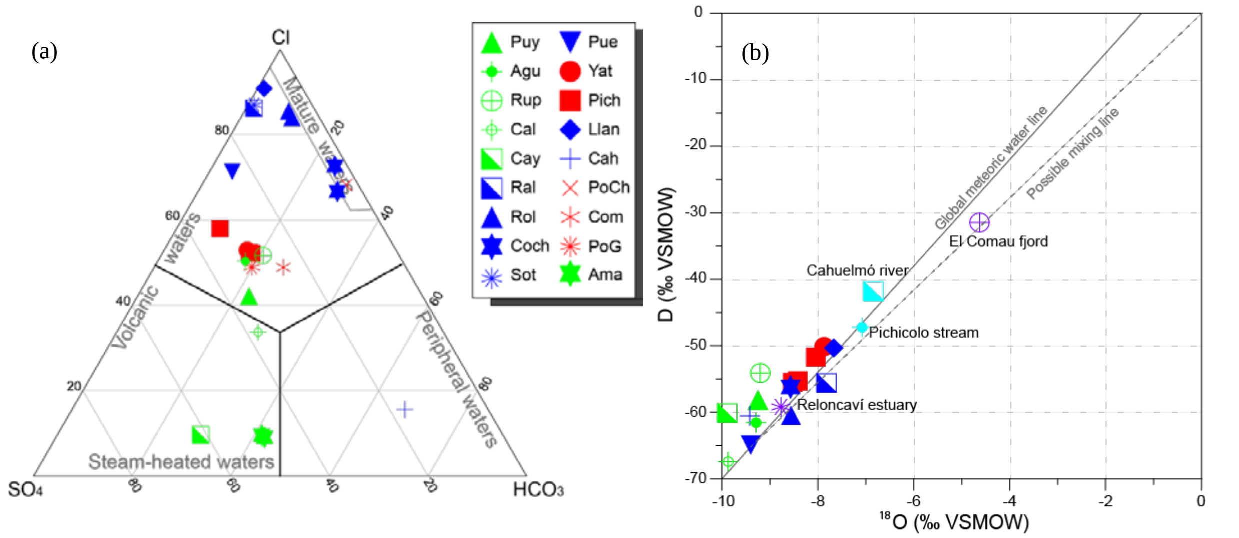

Figure 2: (a) Classification of hot springs from Los Lagos District according to the $Cl-SO_4-HCO_3$ triangular diagram of Giggenbach (1991). (b) Isotopic composition ($\delta^{18}O$ versus δD) of geothermal fluids and their closest cold waters.

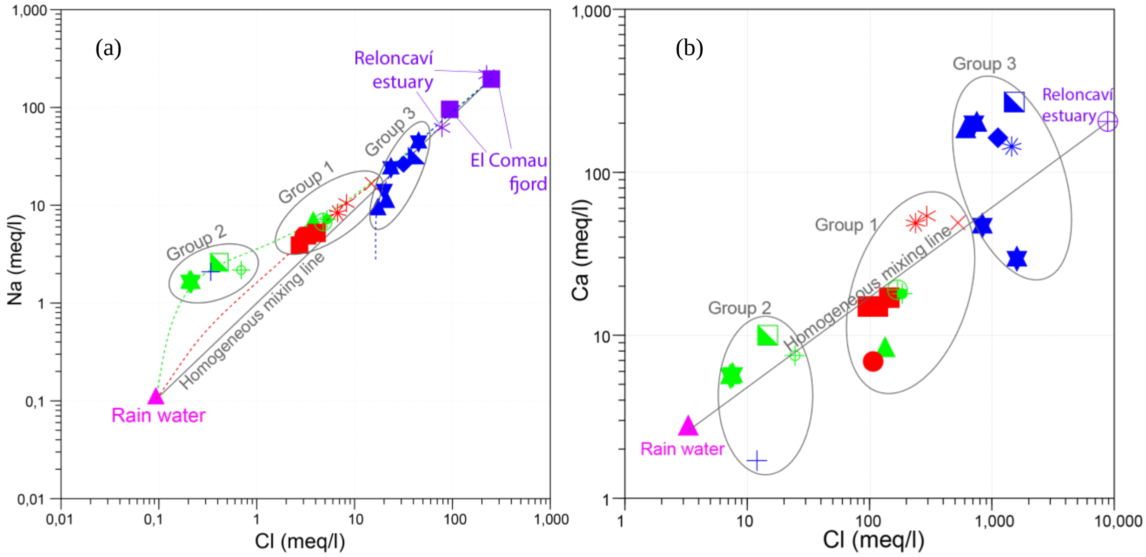

Figure 3: Classification of hot springs waters from Los Lagos District. (a) Binary diagram of Na versus Cl. (b) Binary diagram of Ca versus Cl.

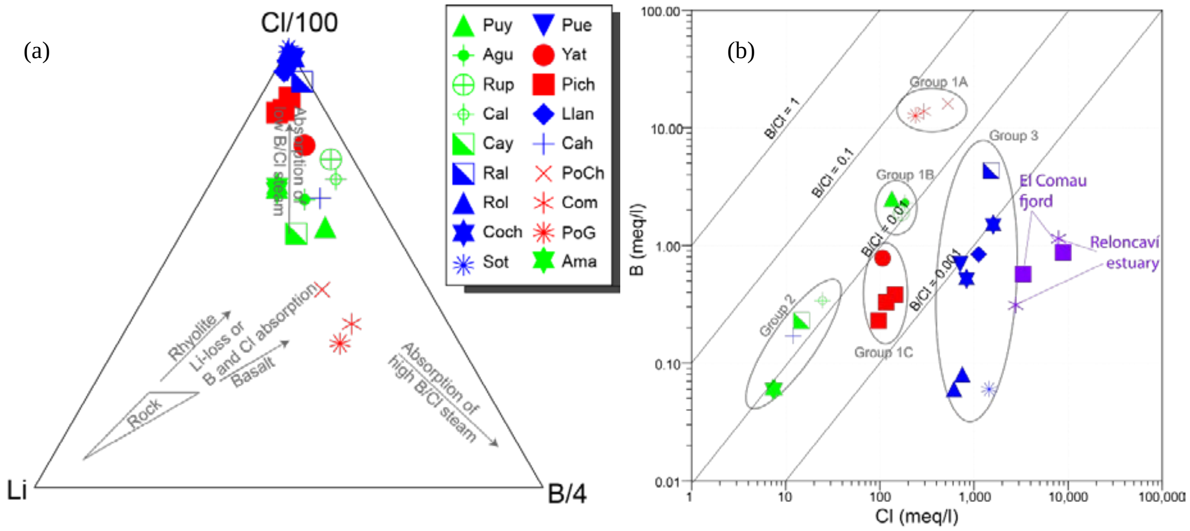

Figure 4: (a) Cl-Li-B ternary diagram (Giggenbach, 1991) showing behavior of hot springs from Los Lagos District. (b) Classification of waters in the binary diagram of B versus Cl.

4.3 Oxygen and hydrogen isotopic characterization

The δD versus $\delta^{18}O$ diagram (Figure 2) plotted with the Global Meteoric Water Line (GMWL) shows that all hot springs have a meteoric water source. Three water samples (Puelo, Rollizos and Ralún) have slightly higher $\delta^{18}O$ values (Ruiz, 2016), which can be from minor $\delta^{18}O$ exchanges between deeply circulating meteoric waters and host rocks in geothermal systems (Truesdell and Hulston, 1980). This exchange could also be associated with higher reservoir temperatures or a small input from seawater. Nevertheless, the samples with these slight $\delta^{18}O$ positive deviations are not high enough and do not correspond to those hot springs with the highest depth temperatures.

In general, samples from the northern part of Los Lagos District exhibit lower $\delta^{18}O$ and δD values (Figure 2), which is believed to be related to reflect the change in latitude.

5. DISCUSSION

5.1 Similarities and differences among hot springs

The sources of thermal waters are mostly meteoric and, in the Rollizos, Cochamó and Sotomó hot springs, also slightly marine, which helps explain the lower B/Cl ratios and seawater chlorides sources (Figure 4 and Figure 5). This likely reflects the existence of a permeable zone, where circulating geothermal waters cross the intertidal zone. However, these hot springs do not have higher $\delta^{18}O$ values plotting a possible mixing line (Figure 2), as would be expected. Therefore, it is possible that geometries of structures can determine the mean flow direction at a local scale disabling the mixing of both fluids, or alternatively isotopic equilibrium could be occurring during fluid ascent.

Moreover, all waters belonging to the LOFS-C group show numerous signs of having undergone processes of water-rock interactions, which are demonstrated not only by the high ratios of Na/Cl and/or Ca/Cl with respect to the other samples, but also by the wide distribution of depth temperatures estimated by the Na/K geothermometer (Figure 7).

Almost all of the hot springs from Los Lagos District show a hydrothermal chloride source, except for the LOFS-I group, which exhibits a scattering of data both in the B versus Cl and Vengosh diagrams (Figure 4 and Figure 5, respectively), allowing to propose an interaction between groundwater and seawater, although the $\delta^{18}O$ values are not high enough.

Thus, it is possible for several groups to be distinguished from the chemical composition of the data set, allowing a primary classification to be made based on geographical location (explained in Part 4.1.1).

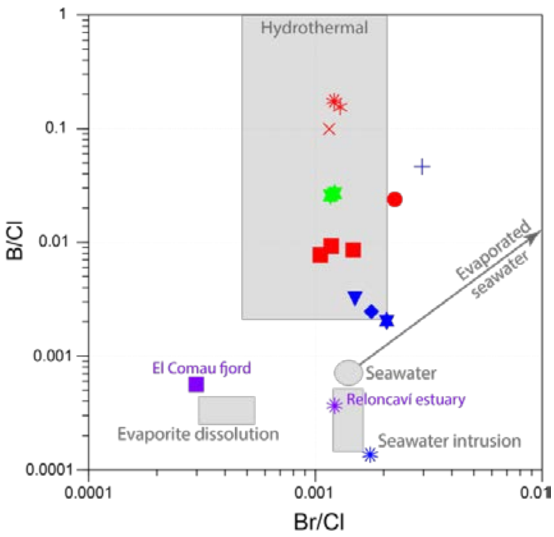

Figure 5: Classification of chlorides sources of hot springs from Los Lagos District according to B/Cl and Br/Cl ratios. Diagram modified from Vengosh (2003).

5.2 Heat and fluid sources

Anion and isotopic compositions of hot springs from Los Lagos District indicate there may be two possible heat sources: one from high heat flux in an intra-arc region associated with deep water flows heated by conduction, and second, from a magmatic source that would generate fluids and subsequent advective heat transport (Sánchez *et al.*, 2013). The group associated with the first heat source is composed of hot springs with low volatile ratios (specifically boron content), high chloride contents (Figure 2 and Figure 4) and low to intermediate surface and depth temperatures (Groups 1B, 1C, 2 and 3). This group includes the Pichicolo, El Yate, Cayetué and El Amarillo hot springs that seem to be related to both types of sources based on the lower B, Cl and B/Cl values and low to intermediate surface and depth temperatures, despite being located close to the area of volcanic and steam heated waters as shown by the Giggenbach diagram (possibly with a volcanic fluids origin). In contrast, the group linked to a magmatic source shows high volatile ratios (indicating a low fluid transport), intermediate chloride and sulfate contents and very high surface and depth temperatures (Group 1A) (Figure 6 and Figure 7).

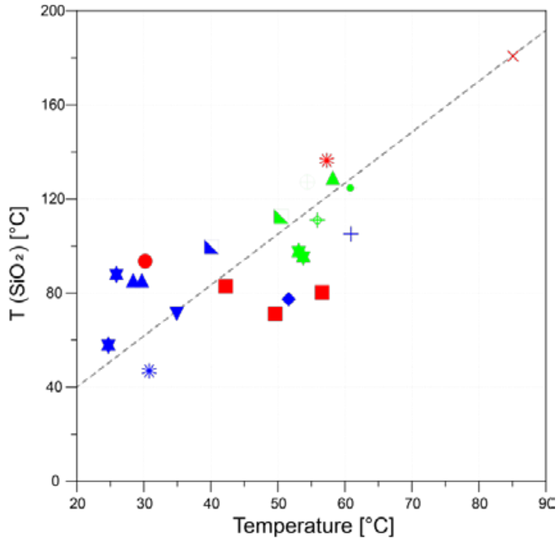

Figure 6: Comparative diagram between temperatures obtained by silica geothermometer (Verma and Santoyo, 1997) and surface measured temperatures.

5.3 Location differences

It is important to note that a general correlation appears to exist between hot springs locations and chemical characteristics, particularly for springs found along the ALFS that exhibit higher surface and depth temperatures and are classified as chloride waters with high volatile contents (Figure 2 and Figure 4). These correlations are not so clear for hot springs located along the LOFS, with only conservative elements being more abundant in this group, indicating that there is a greater transport of geothermal waters.

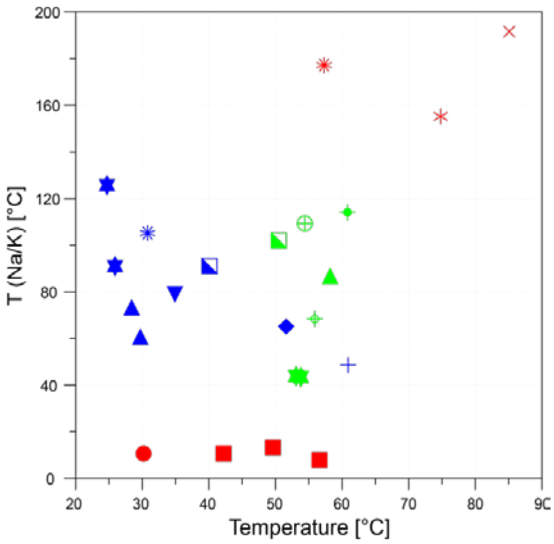

Figure 7: Comparative diagram between temperatures obtained by Na/K geothermometer (Díaz-González *et al.*, 2008) and surface measured temperatures.

The model proposed by Sánchez *et al*. (2013) has been very useful for this study because it has allowed thermal waters to be grouped using both hydrochemical features and location data. This grouping of data has allowed some characteristics of the geothermal fluids transport to be determined, which is primarily vertical through the LOFS and lateral through the ALFS (Figure 8), as shown by low and high volatile rates, respectively, and temperature data. Thus, the two main groups mentioned in section 5.2 can be described broadly and fairly complete in each location group (Figure 8): (1) the hot springs located along LOFS structures are formed by high heat flux in an intra-arc region associated with deep water flows heated by conduction, and (2) the hot springs located along ALFS structures which have a magmatic source of heat and fluids generating heat transport by advection.

6. CONCLUSION

Chemical and isotopic data from hot springs of the Los Lagos District can be divided into two distinct groups relative to the LOFS and ALFS. The first group contains hot waters from deep vertical fluid transport, conductive heat flow and cation exchange that is reflected by the scattering of depth temperatures data estimated by cation geothermometers (Groups 1B, 1C, 2 and 3). The second group brings together hot springs from Group 1A, which represents lower, shallower and lateral fluid transport with a significant influence from magma that is responsible for heat and mass transfer.

The Pichicolo, El Yate, Cayetué and El Amarillo hot springs were considered to be part of the LOFS domain group despite showing a volcanic influence, primarily because of the lower boron and chloride content and lower B/Cl ratio (Giggenbach, 1991; Sánchez *et al.* , 2013), with the possibility of a small localized contribution from volcanic fluids.

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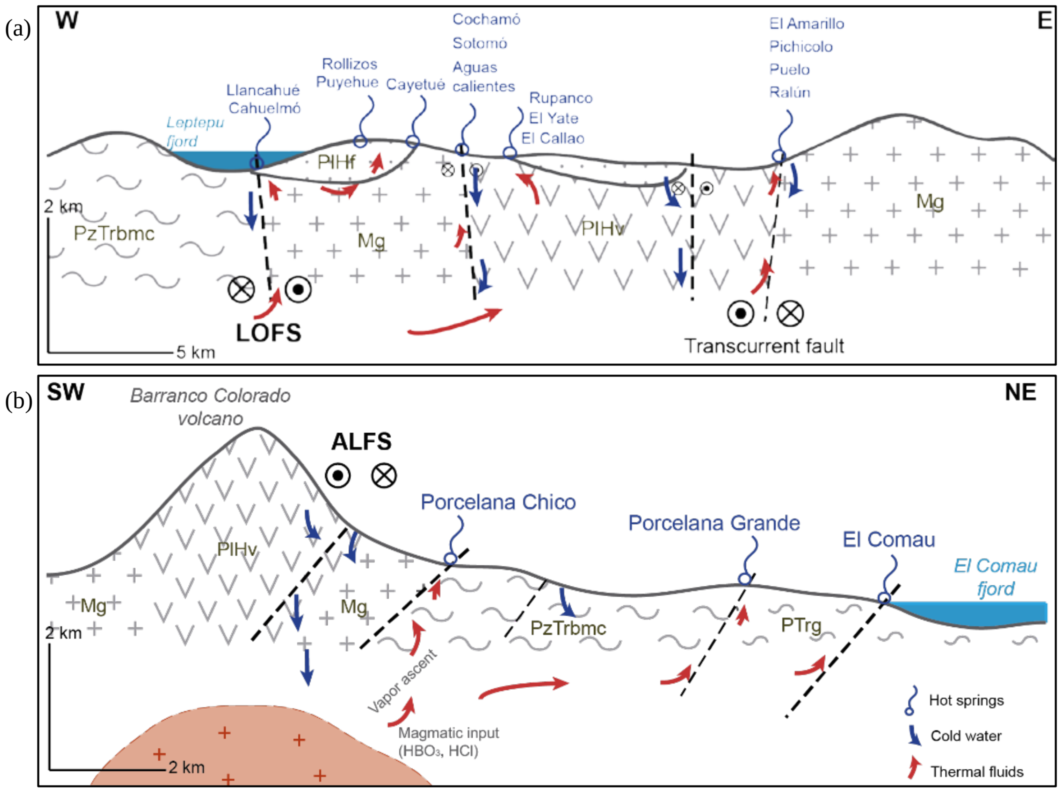

Figure 8: Conceptual model modified from Sánchez *et al.* (2013) showing the geothermal systems for the (a) LOFS domain and (b) ALFS domain. PzTrbmc: Paleozoic and Triassic amphibolite schists and ultramafic rocks; PTrg: Paleozoic and Triassic gabbros; Mg: North Patagonian Batholith granitoids from Miocene; PlHv: Pleistocene-Holocene lavas and pyroclastic deposits; PlHf: Plesitocene-Holocene fluvial deposits.

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