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Geological and Tectonic Settings Preventing High-Temperature Geothermal Reservoir Development at Mt. Villarrica (Southern Volcanic Zone): Clay Mineralogy and Sulfate-Isotope Geothermometry

Sebastian Held[1], Eva Schill[2], Pablo Sanchez[3], Thomas Neumann[4], Katja Emmerich[5], Diego Morata[3] and Thomas Kohl[1]

[1] Karlsruhe Institute of Technology, Institute of Applied Geosciences, Karlsruhe, Germany

[2] Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal, Karlsruhe,Germany

[3] Universidad de Chile, Andean Geothermal Center of Excellence, Santiago de Chile, Chile

[4] Karlsruhe Institute of Technology, Institute of Mineralogy and Geochemistry, Karlsruhe, Germany

[5] Karlsruhe Institute of Technology, Competence Center for Material Moisture, Karlsruhe, Germany

Sebastian.held@kit.edu

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ABSTRACT

In the vicinity of the volcano Villarrica (South-Central Chile), geothermal manifestations at the surface are evident revealing low-enthalpy geothermal processes in the subsurface. The structural geology of the area, postulated by (Sanchez et al., 2013), set up a first draft of a structural model but also naming the complexity of the structural geology in the area generated by the interaction of two major fault systems and a change in the basement geology. In the first phase of the actual research project, geochemical methods are used to characterize the geothermal system. Besides standard geochemical techniques, clay mineralogy and $\delta^{18}O_{[SO4]}/\delta^{18}O_{[H2O]}$ isotopes are used to determine the characteristics of the geothermal reservoir. The results of these techniques, pointing at a low-enthalpy reservoir in the vicinity of the active Villarrica volcano, are discussed in context of the special tectonic characteristics in the investigation area.

1. INTRODUCTION

Worldwide geothermal resources are classified according to their temperature into low temperature (100–150°C) to ultra-high temperature (>300°C) by Sanyal (2005). Volcanic regions are typically characterized by high temperature resources, where different volcanic rocks, such as rhyolitic and ignimbritic reservoir rocks in Taupo Volcanic Zone, New Zealand (e.g., Graham, 1992; Wood, 1992) or volcanoclastic sediments in Miravalles Geothermal Field, Costa Rica (Mainieri et al., 1985), share high productivity originating from primary or secondary fluid permeability. Exploration of high-temperature systems focuses mainly on determining the cap rock, which seal the systems and prevents geothermal fluid from escaping (Ussher et al., 2000) to guarantee adequate mass flow (Barbier, 2002). Sufficient water recharge is essential for sustainable exploitation (Barbier, 2002; Ellis and Mahon, 1977).

In Chile, the subduction of the Nazca Plate underneath the South American plate yields a volcanic arc (Angermann et al., 1999; Norabuena, 1998) including more than 100 volcanos between 17°S and 54°S (e.g., Stern, 2004). This volcanic chain is interrupted by a volcanic gap between 28°S and 33°S, which is attributed to the Pampean Flat Slab Segment (Barazangi and Isacks, 1976). The Chilean chain can be subdivided into the Central (CVZ, 17–28°S), Southern (SVZ, 33–46°S), and the Austral Volcanic Zones (49–54°S) (Stern, 2004). To the north, the SVZ is limited by the flat slab and to the south it extends to the triple junction between the Nazca, South American, and the Antarctic plates. Recent subduction is slightly oblique to the trench with an angle of N78°E (Somoza, 1998) resulting in a NE–SW maximal horizontal stress in the main cordillera since the Pliocene (Lavenu and Cembrano, 1999; Nakamura, 1977). The N–S to NNE–SSW aligned SVZ includes major stratovolcanoes and minor eruptive centers on top of a crust with decreasing thickness from north to south (Hildreth and Moorbath, 1988; Tassara and Yáñez, 2003). The tectonic environment controls the localization of volcanic edifices to a large extent (Cembrano and Lara, 2009). Associated with this volcanism, high-temperature hot springs, fumaroles, and surface hydrothermal alteration products are often found in the vicinity of suspected or identified high-temperature geothermal reservoirs (Hauser, 1997; Tassi et al., 2010). Examples for geothermal exploration in the SVZ are Planchón-Peteroa and Descabezado Grande (Benavente and Gutierrez, 2011), Sierra Nevada (Munoz et al., 2011), Mariposa (Hickson et al., 2011) and Tinguiririca (Clavero et al., 2011). All these geothermal fields are located between 33°S and 39°S, where the Eocene–Miocene volcano-sedimentary Cura-Mallín Formation occurs underlying the recent volcanoes (Charrier et al., 2007; Jordan et al., 2001; Radic, 2010). The volcano-sedimentary Cura-Mallín Formation was deposited under non-marine conditions in basin environments (Radic, 2010). Two subunits are differentiated: (1) the Guapitrío Formation, a volcanic unit with minor sedimentary deposits, and (2) the Río Pedregoso Unit, a mainly sedimentary association with minor volcanic intercalations (Suarez and Emparan, 1995). Terrestrial sediments were deposited within extensional basins intercalated with lavas and volcanoclastic rocks (Radic, 2010; Suarez and Emparan, 1995).

The first Chilean geothermal power plant in the SVZ is currently built in the vicinity of the Tolhuaca volcano exploiting a high-enthalpy reservoir hosted in Cura-Mallín basement rocks and revealing the typical features such as fumaroles, steam-heated springs with temperatures between 20–60°C and hydrothermal alteration products such as clay minerals and silica sinters (Melosh et al., 2012). About 200 km to the south of Tolhuaca, at Mt. Villarrica, the processes active in the subsurface do not favor the formation of such a high-temperature reservoir even though a magma chamber is present at shallow depth (Hickey-Vargas et al., 1989).

Within this study, we will highlight the differences between high- and low-temperature reservoirs in Sierra Nevada (El Toro side) and Villarrica and discuss the tectonic situations that may be responsible for the absence of a high-enthalpy geothermal reservoir in the vicinity of one of the most active volcanoes of Chile (Ortiz, 2003). In this respect, we have analyzed clay minerals sampled from the surroundings of thermal springs in order to determine the formation of hydrothermal alteration products. We furthermore calculated reservoir temperatures from those springs to exclude the occurrence of hidden high-enthalpy reservoirs as far as possible.

2. GEOLOGICAL SETTING

The geological setting of the Mt. Villarrica area is characterized by three major structural elements of regional significance, the Liquiñe-Ofqui Fault Zone (LOFZ), the Villarrica-Quetrupillán-Lanín lineament, and the change in basement rock (Fig. 1).

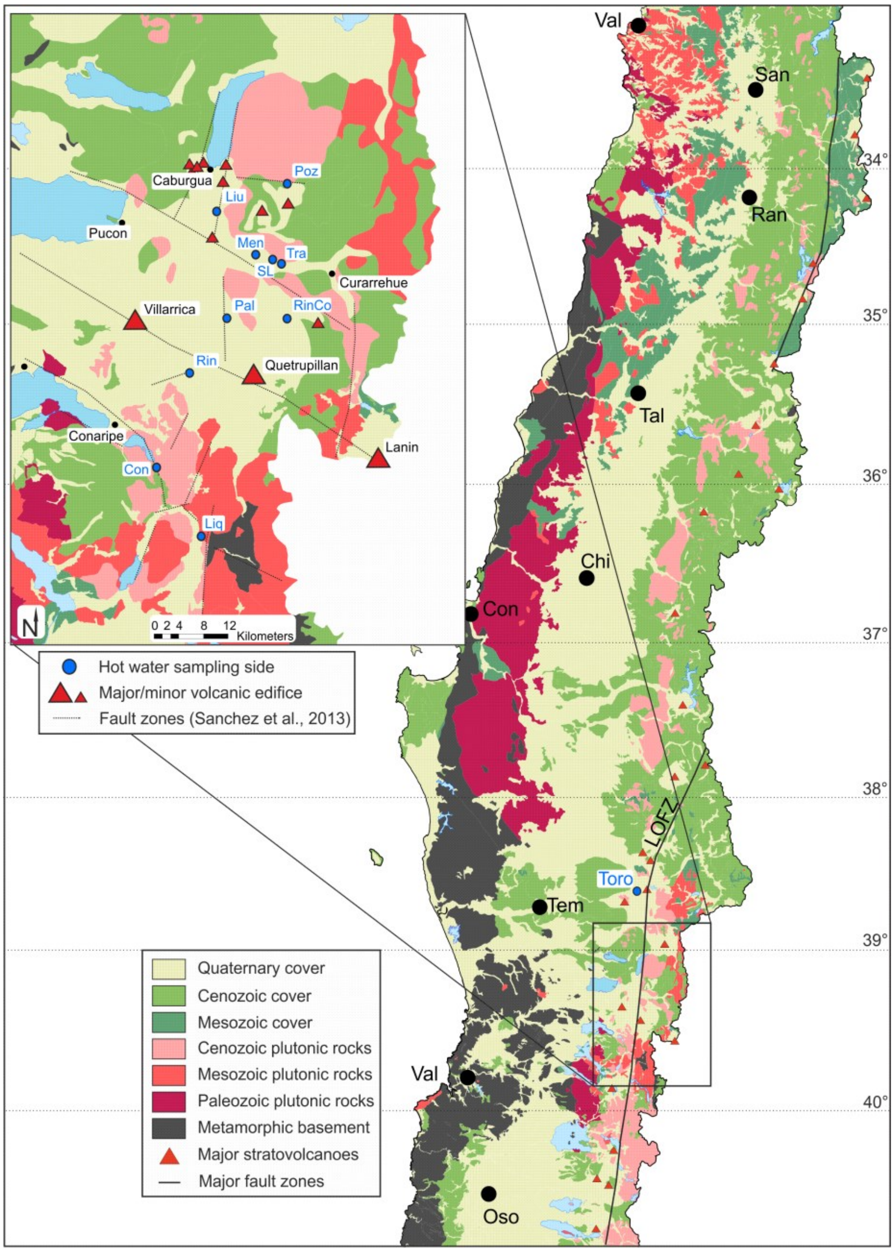

Figure 1: Locations of fluid and clay sampling in the Villarrica and the Sierra Nevada area. The maps is modified after Sanchez et al., (2013), Cembrano and Lara (2009), and 1:1.000.000 scale map of Servicio Nacional de Geología y Minería (SERNAGEOMIN) of Chile

Starting in the vicinity of the triple junction, the LOFZ extends over 1200 km parallel to the volcanic arc (Adriasola et al., 2005; Cembrano et al., 1996; Rosenau et al., 2006) and is associated with dextral strike-slip/dip-slip structures (Cembrano et al., 1996). The LOFZ controls the Cenozoic emplacement of the North Patagonian Batholith (NPB) between 39°S and 54°S, which is interpreted as the root of the volcanic arc (Hervé, 1984; Munizaga et al., 1988; Pankhurst et al., 1999). Granodiorites and tonalites of Cretaceous to Miocene age dominate the complex lithology of the NPB (Herve, 1994; Pankhurst et al., 1999). Besides N-S- to NNE-SSW-aligned faults belonging to LOFZ, subsidiary NE and NW faults are also common in the SVZ. NE aligned fault zones are considered as tension fractures often related to more mafic volcanism (Cembrano and Lara, 2009). NW-trending fault zones, such as the volcanic chain of Villarrica-Quetrupillán-Lanín, are interpreted as pre-Andean deep-seated fault zones created during

Triassic–Jurassic break-up of Gondwana (Rapela and Pankhurst, 1992). Volcanism related to these show a great variety of locally more evolved magmas (Cembrano and Lara, 2009). The research area, located in the vicinity of the Villarrica volcano, is affected by the above described tectonic features. The LOFZ crosscut the area and is displaced by the NW-SE-aligned volcanic lineament (see Fig. 1) probably running from Argentina to the Pacific coast (Lara, 2004). The NPB, occurring extensively south of the volcanic chain, crops out only occasionally as magmatic bodies intruded into or acting as basement of the Cura-Mallín Formation north of the Villarrica volcano. In Chile, the southern expansion of Cura-Mallín basins, characterized as intra-arc basins (Radic, 2010) and sometimes even as pull-apart basins (Herve, 1994), is described only down to 39°S (Radic, 2010). The southern continuation is documented at the eastern slope of the Main Cordillera (Ramos et al., 2014) in Argentina. Gravity data published by Ramos et al. (2014) show an almost continuous gravity low between 37° and 41°S with a negative gravity signature between the Caburgua and Pucón lakes, indicating the occurrence of sedimentary basin structures. Cenozoic volcano-sedimentary Cura-Mallín units occur widespread north of the volcanic chain (see Fig. 1) correlating with gravity anomalies but are almost completely abundant south of it.

In the vicinity of the Villarrica volcano, a limited number of thermal springs occur in direct association with fault zones (see Tab. 1). These are generally of low mineralization (Lara and Moreno, 2004; Sanchez et al., 2013). To the south of the volcano, the hot springs trace the distinct run of the LOFZ. In contrast, thermal springs to the north of the Villarrica-Quetrupillán-Lanín volcanic chain are distributed over an extended area. A high number of faults, with varying orientation, crosscut the northern part often with hot springs discharging at miscellaneous oriented faults mostly between batholithic rocks and Cenozoic basin deposits (Moreno and Lara, 2008). Studies of geothermal fluids from the Villarrica area indicate reservoir temperatures of 120–160°C (Sanchez et al., 2013). Typical volcanic features, such as fumaroles and high-temperature springs, do not occur at the Villarrica volcano.

3. METHODS AND SAMPLING PROCEDURE

10 fluid samples from single thermal springs were sampled to the north and south of the Villarrica-Quetrupillán-Lanín lineament for chemical analysis. For comparison with a site characterized by high-enthalpy reservoir features, El Toro, a sub-boiling spring with steam-heated groundwater and fumarolic activity located on the flank of Sierra Nevada volcano was chosen (Fig. 1). Sampling locations of the El Toro hot springs on the flank of Sierra Nevada is shown in Figure 2. It should be noted that the full fluid chemical analyses including major and trace elements, O/H isotopic, Strontium isotopic, and chlorofluorocarbon analyses will be presented in a forthcoming paper. Furthermore, clays occurring in the vicinity of two springs were analyzed for high-temperature alteration products linked to possible cap layer formation. We would like to point out that except from Coñaripe site (Fig. 2), no clay minerals were found in the outcrops near to the springs in the Villarrica area. Locations of the sampled hot springs are shown in Figure 1. Most of the samples were collected from the Villarrica area.

Figure 2: Left: Sampling location at the El Toro side (Sierra Nevada). In the red and grey parts are sub-boiling springs with 95°C and fumarolic activity. Right: Clay mineral outcrop in the vicinity of the Coñaripe hot spring. The spring is located four meters to the left of the outcrop, with the outcrop continuing until the spring.

Reservoir temperatures, inferred from cation geothermometers for Villarrica sources (Sánchez et al., 2013) are significantly lower than 200°C. As cation geothermometers are vulnerable for dilution by meteoric water or kinematic effects during ascent, in this study, we applied sulfate-isotope geothermometry. The related temperature-dependent oxygen fractionation between water and sulfate species was first described by Hoering and Kennedy (1957), Mizutani and Rafter (1969), and Lloyd (1968). Besides the reduced vulnerability to dilution, its advantage over cation geothermometers is the slow reaction rate at lower temperatures, i.e. during ascent, resulting in fluids representing the fractionation in the deep reservoir (Chiba and Sakai, 1985). The dominant sulfate species at reservoir conditions is governing the fractionation (Sakai, 1977; Chiba and Sakai, 1985; Zeebe, 2010; Boschetti, 2013) and therefore, also the temperature estimate. After Boschetti (2013), at lower temperatures and near neutral pH, SO_4^{2-} is the dominate species resulting in the application of the formulas indicated below in Table 1. For the measurements sulfate was precipitated completely with stoichiometric abundant $BaCl_2$. Measurements of $\delta^{18}O$ were done by isotope-ratio mass spectrometry (IRMS) using a GV Instruments IsoPrime combined with a HTO Pyrolysis from HEKAtech.

To determine the clay mineralogy, samples were grinded to grain size of up to 500 µm with an agate mill and subsequently homogenized. Simultaneous Thermal Analysis (STA) was conducted using a STA 449 C Jupiter from Netzsch connected to a quadrupole mass spectrometer (QMS 403 C, Aeolos, Netzsch). The STA was run between 35 and 1100 °C with a heating rate of 10 K/min. 50 mg sample material were used in a Pt/Rh crucible under a synthetic air/nitrogen atmosphere (50 + 20 ml/min). Powder X-Ray Diffraction (XRD) measurements were done using a Siemens D5000 diffractometer with CuKα radiation. For bulk powder samples the diffractometer varied the 2θ angle between 5 and 80° with a step size of 0.02° and a step time of 3s, whereas the texture samples are measured with an angle between 2 and 35°. Oriented samples were prepared by mixing the sample material with deionized water and pipetted onto a glass slide after ultrasonic treatment. After drying the textured sample were measured starting with an untreated texture sample. Afterwards measurements were continued firstly adding ethylene glycol and afterwards heated up to 375° and 550°C. Settings at the Siemens D5000 diffractometer do not change. Additional Cation Exchange Capacity (CEC) and X-ray Fluorescence (XRF) measurements were conducted to evaluate XRD and STA-MS results but are not presented here. This will be included in a forthcoming paper.

4. RESULTS

Results of STA-MS (Fig. 3) show significant difference between the clays sampled near the El Toro and Coñaripe hot springs. High amounts of swelling 3-layer clay minerals indicated by an endothermic peak in DSC between 123–157°C and the appearance of sulfides, probably pyrite, indicated by an exothermic peak in DSC at 441° and 476°C with corresponding SO_2 peaks in the mass spectrometry, underline the secondary hydrothermal alteration at a high-enthalpy reservoir at El Toro. Further sulfate phases are also detected in the SO_2-MS curve for the El Toro sample. In contrast, Coñaripe samples reveal no indication for hydrothermal alteration minerals. Endothermic peaks in DSC and mass spectrometry rather indicate crystal water. These observations are confirmed by powder XRD analysis (Fig. 4a). The sample only consists of silica and feldspars. The high noise ration points at amorphous material. In contrast, the powder XRD analysis of El Toro sample (Fig. 4b) reveals a much more differentiated mineralogy of the sample. High amount of clay fraction (d(001) > 10 Å) accompanied by feldspars, quartz, sulfides, Gypsum, and probably talc and/or a second sulfate (e.g. Basaluminite) are evident. XRD-texture analyses indicate smectite as the major clay mineral fraction by d(001) = 15.4 Å in the untreated sample (black curve, Fig. 4c) shifted to 17 Å in sample treated with ethylene glycol (red curve, Fig. 4c). Texture preparation confirms the appearance of minor amount of sulfides and sulfates. In conclusion, clay mineralogical analyses indicate secondary hydrothermally formed clay minerals at El Toro hinting at high-temperature processes, whereas the samples from the Coñaripe hot springs where probably formed by low-temperature weathering.

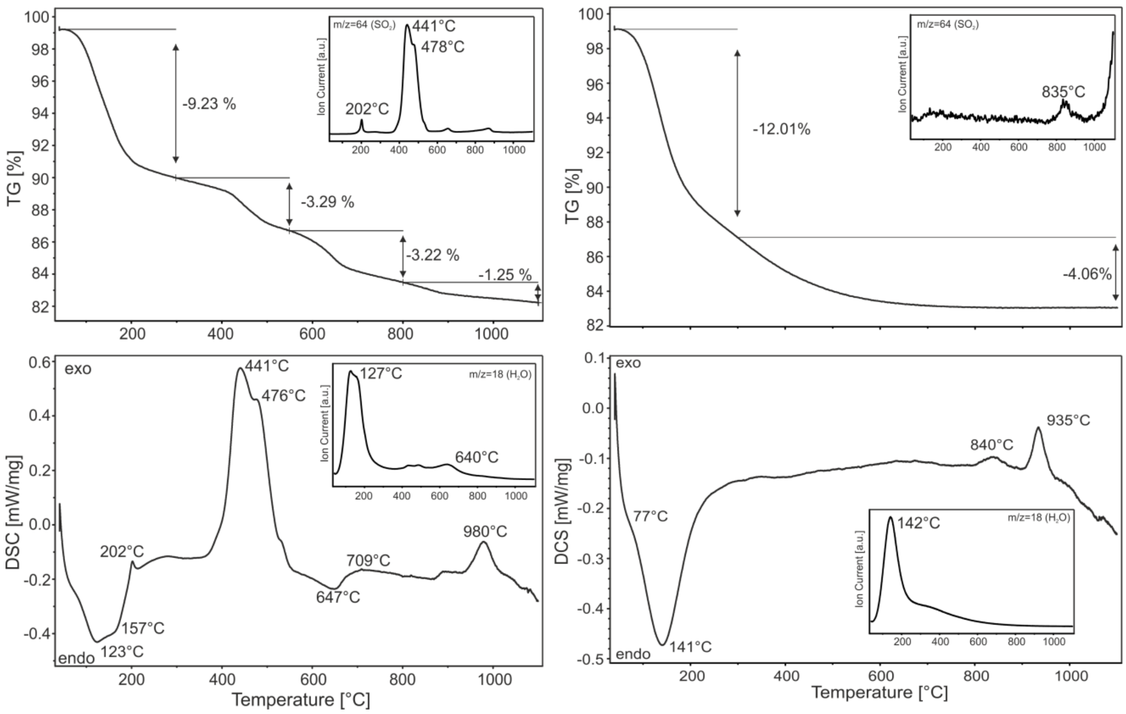

Figure 3: Results from STA-MS analyses of El Toro sample (left) and Coñaripe sample (right). TG = Thermo Gravimetry, DSC= Differential Scanning Calorimetry.

Since clay mineralogy points to high-temperature alteration typical for a cap layer at El Toro site and respective reservoir conditions are exploited there, it is considered as a representative example for high-enthalpy conditions in our study. Sulfate isotope geothermometer for the El Toro site reveals reservoir temperatures of >300°C. It should be noted that in contrast to Villarrica area, at El Toro H_2SO_4 acts as the dominant species. Therefore, calculations after Seal et al., (2000) or Mizutani and Rafter (1969) have to be used, assuming that the origin of the hot fluid is the reservoir and not steam-heated groundwater.

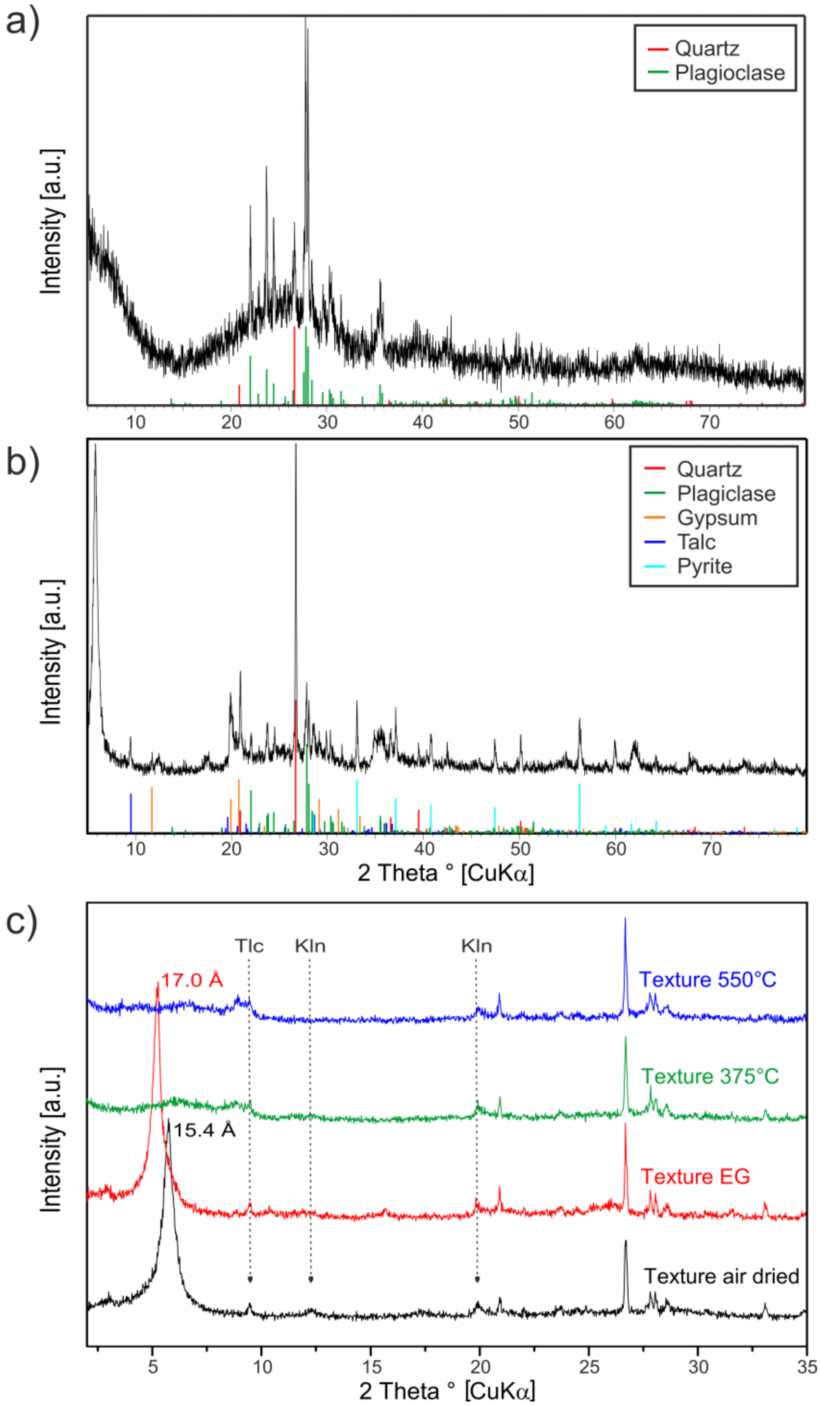

Figure 4: XRD analyses on powder of Coñaripe (top), on powder of El Toro (middle), and texture analysis of El Toro (bottom). Abbreviations: Pyp = Pyrophyllite, Kln = Kaolinite

Sulfate-isotope geothermometry of the sampled springs in the Villarrica area reveals temperatures ranging from 83°C in Liquiñe to 134°C in Coñaripe (see Tab. 1). While cation geothermometers reveal a significantly higher reservoir temperature of 110–150°C for Liquiñe, the reservoir temperature of Coñaripe is within the range of the cation thermometers (see Sanchez et al., 2013 for cation geothermometer data). It should be mentioned here, that we were not allowed to sample the Geometricas springs, which has the highest cation temperatures of 140–180°C (Sánchez et al., 2013). In terms of reservoir temperature no significant differences can be observed between the area north and south of the Villarrica-Quetrupillán-Lanín volcanic chain (different grey shading in Tab. 1). The mean temperature obtained from sulfate-isotope geothermometry in the Villarrica area differs strongly from the high reservoir temperature at the El Toro side.

Table 1: Results from geothermometry using $d^{18}O$ isotopes at SO_4 and H_2O of the Villarrica area. Light grey shaded area indicates springs located to the south of the Villarrica-Quetrupillán-Lanín volcanic chain; dark grey shaded indicates springs located north of this chain. Equations used are described in (Halas and Pluta, 2000) and (Zeebe, 2010) and listed below.

Sample	$\delta^{18}O$ (SO_4) [‰]	$\delta^{18}O$ (H_2O) [‰]	Calculated temperatures after Halas and Pluta (2000)[I] [°C]	Calculated temperatures after Zeebe (2010)[II] [°C]
Rincon	0.33	-9.74	116	117
Liquine	3.78	-9.41	83	87
Coñaripe	0.63	-8.09	134	134
Trancura	0.14	-9.24	125	125
San Luis	0.04	-9.28	126	126
Palguin	0.74	-9.83	110	112
Los Pozones	0.68	-10.2	123	123
Liucura	2.55	-9.07	99	101
Rinconanda	0.56	-11.1	98	101
Menetue	1.55	-9.04	110	112
El Toro	-3.78	-7.14	322[III]	347[IV]

[I] Formulae after Halas and Pluta (2000): $10^3 \ln \alpha(SO_4^{2-} - H_2O)_{empirical} = 2.41 \cdot 10^6/T^2 - 5.77$

[II] Formulae after Zeebe (2010): $10^3 \ln \alpha(SO_4^{2-} - H_2O)_{theoretical} = 2.68 \cdot 10^6/T^2 - 7.45$

[III] Calculated after Seal et al. (2000), [IV] Calculated after Mizutani & Rafter (1969)

5. DISCUSSION

Based on the presented clay mineralogy at Coñaripe, n.b. the spring with the highest reservoir temperature in the Villarrica area (except from Geometricas), the results do not indicate an outcropping cap layer, formed by hydrothermal alteration trough ascending thermal fluids, as often found in high-enthalpy geothermal fields. In addition, there are no hidden high-enthalpy reservoirs indicated by the sulfate-isotope geothermometry in the Villarrica area. In contrast geothermometry at El Toro combined with the appearing cap layer, formed by hydrothermal alteration products, points at an existing high-enthalpy reservoir feeding the hot springs.

Although all sampled springs are located in the vicinity of active volcanoes, it's obvious, that the results indicate different reservoir conditions, over both Villarrica and Sierra Nevada (El Toro) sites. At Mt. Villarrica, even though Hickey-Vargas et al., (1989) has proved the evidence of a shallow magmatic chamber, our results clearly indicate a more Alpine-type, low temperature geothermal system in contrast to the system at Sierra Nevada. Both systems share similar tectonic characteristics (see Tab. 2), which, nevertheless, provoke completely different settings from a geothermal point of view. Structural parameters, which govern the development of geothermal reservoirs in the SVZ, and their impact to the reservoir evolution, have to be investigated systematically using a wide range of different techniques.

In the following section, we will review the geological and structural characteristics of the two areas with respect to the observed geothermal features that affect the reservoir development. As mentioned above, the important tectonic features are:

- LOFZ: The N-S- to NNE-SSW-aligned intra-arc fault zone is the dominant feature in the southern SVZ. The locations of the large number of monogenetic cones and even stratovolcanoes are controlled by this dextral strike-slip fault zone. Interaction with the pre-Andean basement faults can result in horizontal displacement (Hackney et al., 2006) with implications for fluid-flow, sedimentary deposition, and fault behavior. Further north in the vicinity of Sierra Nevada and Tolhuaca volcanoes, the LOFZ bends to a NE-trending lineament, splits in several separate faults and finally fans out next to the Copahue-Callaqui ENE-aligned lineament continuing in the Antinir-Copahue fault zone (Folguera et al., 2004; Potent, 2003).

- NW-SE- or NE-SW-aligned faults: Appearance of the pre-Andean (Triassic–Jurassic) NW-aligned basement fault zones and NE-aligned extensional faults is linked strongly to the evolution of major stratovolcanoes, tectonically affecting magma chemistry due to the orientation of these fault zones (Cembrano and Lara, 2009). Fast magma ascent is simplified on NE-trending extensional fault zones, also favoring vertical fluid flow in extensional fault zones. However, NW-aligned fault zones under compressional/transpressional tectonic stress hinder direct magma ascent often resulting in formation of more evolved magmas (Cembrano and Lara, 2009). Vertical fluid flow is also prevented on NW-oriented faults.

- Basement: Basement rocks consist of granitoids of the NPB or the volcano-sedimentary Cura-Mallín Formation, which is deposited in extensional Eocene–Miocene basins. Basin evolution is probably limited by pre-Andean fault zones acting as margin faults (Folguera et al., 2004; Jordan et al., 2001) with effects on sedimentary development in the direct vicinity of these basement faults. Furthermore, LOFZ lineament has been affected by basement geology with a more distinct run, where it passes the NPB and diversification of the alignment, as it enters areas affected by basin sedimentation. Finally, facial differentiation of the Cura-Mallín Formation (see Geological Setting section) into a more volcanic (Guapitrío Formation) and sedimentary (Río Pedregoso) subunit can also affect the evolution of geothermal reservoirs.

Analyzing the appearance of these features in selected geothermal exploration areas (see Tab. 2), no tectonic feature by itself is responsible for the formation of high-enthalpy reservoirs in the SVZ. Also, combination of different tectonic features cannot explain the appearance of an exploitable resource, because every location is characterized by a variety of significantly different features. For example, basement rocks of Cura-Mallín Formation and granitoids of NPB are present at all locations. A direct link between one basement rock and the formation of high-enthalpy reservoirs cannot be established. The other features show similar ambiguity highlighting the complexity of the geothermal systems in the SVZ. As a result it becomes obvious that the explanation of the evolution of a high-enthalpy reservoir has to be made on a more local level considering the regional distribution of the tectonic features.

Table 2: Tectonic features of the geothermal exploration areas

	LOFZ	NW / NE aligned secondary faults	Basement
Villarrica	N–S-aligned, displaced by NW-aligned pre-Andean fault zone	NW-aligned pre-Andean fault zone with sinistral transpression + NE-aligned fault vents on Villarrica NE-shoulder	NPB, Cura-Mallín starting to the north
Sierra Nevada	NNE-NE-aligned	ENE-aligned fault with sinistral strike-slip movement	NPB, Cura-Mallín probably Guapitrío unit
Tolhuaca	NNE between Tolhuaca and Lonquimay volcano	Lonquimay and Tolhuaca volcanoes as well as fumaroles and hot springs on NW shoulder of Tolhuaca volcano aligned on NW lineament	Cura-Mallín probably Guapitrío unit, partly NPB
Mariposa	Not present	ENE-aligned fault zones in the transition from NSVZ to TSVZ interpreted sometimes as part of a graben system	Cura-Mallín + Granitoids

5. OUTLOOK

On-going geochemical and geophysical measurements will be used to better constrain the reservoir conditions at Mt. Villarrica. Preliminary results from Strontium isotopes indicate two different geothermal systems to the north and south of the Villarrica-Quetrupillán-Lanín volcanic chain. To exclude the dilution as process causing the low estimates of geothermometry, chlorofluorocarbon (CFC) analyses were conducted to determine the amount of dilution of the reservoir fluid by meteoric water. Finally, geochemical alteration experiments, matching the sampled thermal fluids with artificially altered water in equilibrium with possible reservoir rocks, should finally evaluate the hypothesis. Besides, geochemical investigations extensive geophysical surveys were/will be conducted. Gravimetric measurements should determine the extent and thickness of the volcano-sedimentary basin, whereas magnetotelluric investigations should detect fluid flow paths in the subsurface. The local investigations should allow the characterization of the geothermal evolution at Villarrica. Tectonic features hindering the formation of a high-enthalpy reservoir should be determined after intense investigations. Following these, results will be compared to other geothermal exploration areas to validate the hypothesis.

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