The Chemical Composition of Geothermal Hot Springs as an Indicator of Various Geologic Sub-surfaces in Iceland and Serbia

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ABSTRACT

Geothermal hot springs are frequently located along tectonic boundaries proving environmentally friendly energy source to fulfill energy demand. Generally, geothermal activity emerges near divergent tectonic plate boundaries as in the Icelandic case or in orogeny geological settings as in the Serbian case. The chemical composition of a hot spring is a signature of the sub-surface geology and its chemical composition. The sub-surface geology and composition of sources in Iceland have been studied intensively using geophysical methods and exploration drilling as required for estimating the geothermal potential of a source. Hence, the comparison of the chemical composition of geothermal sources can help us provide more detailed information prior to subsurface exploration. In order to assess the suitability of geothermal sources we analyzed and compared the chemical composition in Icelandic and Serbian geothermal sources. For this purpose, samples were collected from various hot springs in Iceland and in Serbia and analyzed using ion chromatography (IC) to determine ion concentrations and using inductively coupled plasma-optical emission spectrometry (ICP - OES) and inductively coupled plasma mass spectrometry (ICP - MS) to determine other elements concentrations. The results reveal that chemical composition reflects differences between locations of sampling or host rock composition. In general, sodium (Na+) is the dominating ion in all samples. The data obtained from the chemical analysis from sources located near to the ocean in Iceland indicate a high concentration of sodium (Na+), potassium (K+) and chloride (Cl-) ions, revealing an inflow of seawater. The dominant elements in Icelandic geothermal hot springs are iron (Fe), strontium (Sr), boron (B), barium (Ba) and aluminium (Al) The data from the chemical analysis of the samples obtained from sources in Serbia, however, shows a high concentration of sodium (Na+), calcium (Ca2+) and sulfate (SO42-). The dominant elements are boron, strontium, and arsenic. These results reveal that Serbian geothermal sources are characterized by very hard water and accordingly have to be used with precaution. Further analysis of the temporal evolution of the chemical composition is advisable before sub-surface exploration.

1. INTRODUCTION

Geothermal hot springs are located in various tectonic environments proving environmentally friendly energy source to fulfill energy demand (Barbier, 2002). Natural geothermal hot springs exist around the world, in particular in, Chile, Hungary, Iceland, Israel, Japan, New Zealand, Romania, Fiji, Canada and the United States(Ferguson et al., 2011). The high-temperature hot springs are found next to tectonic faults while the low and medium temperature hot spring are usually located further away from volcanoes and tectonic fractures. In order to use geothermal energy for electricity production, temperatures have to be above 150°C to enable adequate Carnot efficiency (Polettini, et al., 2015). The worldwide geothermal energy capacity accounts for 6300 MW-electric of the energy production, whereof 28% are located in the USA (Fridleifsson et al., 1994; Holm, et al., 2010; Moeck, 2014; Romitti, 2015).

The feasibility of electricity production from geothermal sources depends also on the chemical composition of the geothermal water. Geothermal hot springs contain various concentrations of dissolved chemical compounds. The amount and nature of dissolved compounds are impacted by temperature, pressure, and local sub-surface geology. Geothermal fluids move through rocks and chemically react with the rocks which usually have a complex chemical composition. Typically, geothermal sources are mineralized and dominated by sodium (Na+), calcium (Ca2+), potassium (K+), chloride (Cl-), silica (SiO2), sulfate (SO42-) and bicarbonate (HCO3-). Minor components include a wide range of elements with mercury (Hg), fluorine (F), boron (B) and arsenic (As). Dissolved gasses usually involve carbon dioxide (CO2), hydrogen sulfide (H2S), ammonia (NH3) and methane (CH4) (Ármannsson, 2016; Sano, et al., 1985).

Settings of tectonic plates have a crucial impact on the geothermal activity. Geologic and geothermal activity is usually located along moving tectonic plates. A particularly interesting case is Iceland, located along the divergent tectonic plate boundaries of the North American Plate and the Eurasian plate. The Mid-Atlantic ridge disparts the island from the Reykjanes ridge on the southwest to the Kolbeinsey ridge on the northeast. Accordingly, the tectonic movement of the two plates results in intense volcanic, seismic and high-temperature geothermal activity (Jacoby, et al., 2007; Thordarson, et al., 2007). Consequently, the primary energy production in Iceland stems to 80% from geothermal energy (Björnsson, 2010). The primary use of geothermal energy is for district heating, greenhouse heating, and swimming pools. Geothermal energy accounts also for about 25% of the electricity production in Iceland (National Energy Authority, 2014).

In contrast, Serbia is located at the Eurasian tectonic plate and it is a part of Alpine orogeny geological settings (Schmid, et al., 2008). The territory of Serbia is divided into four geotectonic plate units: Dinarides, Serbian-Macedonian massif, Charpato-Balkanides, and Pannonian basin. The settings and composition of these geotectonic units are characterized by many low and middle-temperature

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geothermal sources with temperatures rarely exceeding 150°C. Accordingly, no geothermal electricity production exists in Serbia. However, about 158MW the geothermal energy for heating, pools and other thermal usages are generated in Serbia. Nevertheless, the potential of the total geothermal capacity is much greater (Martinovic, et al., 2010). Most of the geothermal hot springs are used in balneology, for swimming pools and spa centers, only a small number of sources are used for district heating and in agriculture.

The most common problem of the usage of the geothermal hot springs has been related to the chemical composition of the springs. Mineralogical composition of hot springs can be quite high and contain minerals which can cause scaling and corrosion of the installation used for the extraction of the water.

This paper gives an overview of the chemical composition of geothermal hot-springs in Iceland and Serbia. For this purpose water samples from geothermal hot springs were collected from nine locations in Iceland and five locations in Serbia. The chemical composition of all samples was analyzed using ion chromatography (IC) to determine anion and cation concentrations, inductively coupled plasma optical emission spectrometry (ICP-OES) and inductively coupled plasma mass spectrometry (ICP-MS) to determine the concentrations of other elements. The paper concludes by characterizing geothermal spring and highlighting the difference between geothermal hot springs in the two countries.

2. STUDY SITE

Between December 2017 and January 2019 water samples from geothermal hot springs were collected from nine locations in Iceland and five locations in Serbia. A description of sampling locations including main characteristics is listed in Table 1 and located on an overview map in Figure 1.

2.1 Sampling locations in Iceland

In Iceland, samples were collected from 9 natural geothermal sources. In the following we provide a short description of each location:

- The Geysir geothermal field lies in a shallow valley named Haukadalur located in south-west Iceland, about 80 km from the capital, Reykjavik and 50 km from the western seashore. While thermal activity in a form of warm springs is distributed along the tectonic Reykjanes ridge, most of the geothermal activities occur in within the vicinity of the great Geysir (sample 1 and 2), Iceland's most famous spring. The average temperature of the hot springs is around 200°C at 850m depth (Ármannsson, 2016). Geothermal hot springs in the Geysir area are of different types. In the western and southern part of the area, some of the geothermal hot springs are constantly boiling and emit hot water rising from deep sources to the surface (sample 3). In the northern part of the area are fumaroles, a type of geothermal activity where only steam and gas are emitted from the source (sample 4). The southern part of the area is characterized by mud pots. Mud pots are fumaroles that are boiling up through the surface or the groundwater. Geothermal sources from the Geysir area are primarily touristic attraction, only a few sources are used for district heating by local hotels.
- 2) Solheimar Eco Village is one of the first Eco villages in the world located in the southern part of Iceland85 km from Reykjavik. Solheimar is a sustainable community known for its ecological atmosphere where about 100 people live and work together. The village is an educational center about environmental issues and hosts exhibitions for sustainable buildings. Only a few geothermal sources (sample 5 and 6) are located in this area with a temperature around 90°C. Geothermal energy from Solheimar is used for district heating, hot water and a small pool in the village.
- 3) Hveragerði geothermal area is located in the southern part of Iceland, 50 km east of Reykjavik. The surrounding area is characterized by a high geothermal activity from the adjacent Hengill central volcano system. In the Hveragerði area, geothermal manifestations consist of the fumaroles located in the north and hot springs (samples 7 and 8) most abundant in the southern part of the village. The maximum sub-surface temperature of the Hveragerði geothermal systems is around 260°C. Hot springs in this area are used for greenhouses heating, district heating, swimming pools and industrial processes such as retreading of car tires and wool washing (Ragnarsson, 2013).
- 4) Reykjadaluris a valley few kilometers north of the town Hveragerði (see the previous location). The valley is abundant in numerous hot springs and mud pots and the surface water runoff is drained by a tributary hot river (sample 9) of River Varmá, which eventually flows through Hveragerði. The Reykjadalur valley is also a part of the Hengill central volcano system. Geothermal hot springs from this area are a popular touristic attraction.
- 5) Hveradalir is a geothermal hot spring (sample I10) in the southern part of the Hengill central volcano system, at the foot of the Hellisheiði lava plateau. The Hellisheiði Power Station was built here in 2008 providing hot water and electricity to the Reykjavik capital area. The Hellisheiði Power Station is the third-largest geothermal power plant in the world with an installed capacity of 303 MWe (Ragnarsson, 2013). Although the last eruption from the Hengill volcano system occurred approximately 2000 years ago, the area is abundant in geothermal activity with numerous hot springs and mud pots.
- Krýsuvík is a part of the high-temperature geothermal area located in the Reykjanes Peninsula in southwest Iceland, 35 km west of Reykjavík. It emerges along the fissure zone of Mid-Atlantic ridge which crosses Iceland. The area is abundant in geothermal activity in a form of fumaroles, mud pots and hot springs (sample I10, I11, and I12). The soil is colored white, yellow, red and green, due to the high content of sulfate salts. The highest temperature of the sources in this area is around 100°C and sources are used as a tourist attraction.
- The Reykjanes power plant is located in the most western part of the Reykjanes peninsula in southwestern Iceland. The plant generates 100 MWe, using geothermal fluids from boreholes reaching temperatures up to 320°C. The area is part of the Reykjanes volcano system which last erupted in 1240. The Reykjanes power plant is cooled with seawater, making it a unique power plant using saltwater for cooling purposes. Cooling water is discharged into the sea at the western end of the peninsula (sample I13). Some natural geothermal springs in the vicinity of the plant have created a lagoon in the vicinity of the power plant (sample I14).

- 8) The Blue Lagoon is a geothermal spa in south-western Iceland located in the lava fields north of the town Grindavík. The geothermal water in the spa comes from the Svartsengi geothermal power station, a 74 MW power plant, using 100°C geothermal fluid for electricity production. The power plant outlet is characterized by silicate mixtures which have proven to have positive effects on treating psoriasis (Olafsson, 1994). Since 1999 the water from the power plant outlet (sample I15) is used by the adjacent Blue lagoon spa for recreation and rehabilitation.
- 9) Deildartunguhver is the largest natural geothermal hot spring in Iceland, delivering about 180 l/s and 100°C hot water (sample I16). It is located in Reykholtsdalur about 100 km northeast of Reykjavik. The spring is used for hot water supply to the towns Borgarnes and Akranes.

2.2 Sampling locations in Serbia

Water samples of geothermal hot springs were collected from five different geothermal sources in Serbia located primarily in the southern part of Serbia. Below is a description of the sampling locations:

- Niška spa is a natural spa located in the south-east part of Serbia, 10 km away from the city of Niš and 255 km away from the capital, Belgrade. Niška spa is a famous touristic place and rehabilitation area known since the Roman Empire. Due to geothermal activity in the area numerous hot springs (sample S1) and mineral mud can be found. The chemical composition of the waters from this area is attributed to a high natural curative effect. Some springs contain radioactive "bigar" rocks and the radioactive gas Radon. Bigar is a soft, hollow sedimentary rock and it is formed by precipitation from waters containing a high amount of dissolved calcium-carbonate. The radioactive bigar that exists in Niška spa is a very rare and special exception but here the deposits are abundant in high concentration. The main curative factor of bigar is rheumatic diseases. The temperature of the waters is around 37 °C and it is used only for therapeutical purposes. Nevertheless, there is a high potential for using geothermal sources for district heating.
- Vranjska spa is a natural spa located in the southern part of Serbia, 350 km from Belgrade in the vicinity of the city Vranje. The area is remarkably abundant in geothermal energy in the form of hot springs (samples S2, S3, and S4). Vranjska spa is the hottest geothermal hot spring in Serbia with discharge temperatures of 96 °C. The water has a high curacy effect due to the high concentration of hydrogen sulfide (H2S) and it is used for various medical treatments. The latest research concluded that geothermal energy from Vranjska spa could potentially support a power generation of up to 20 MWe while only 10% is currently in use (Tulinius, et al., 2015).
- 3) Sijarinska spa is a spa town in southern Serbia, 330 km from Belgrade. The area is well known for geothermal energy. There are eighteen geothermal hot springs distributed along the area with temperatures between 32 °C to 72 °C which contains various chemical compositions (samples S5, S6, S7, and S8). There is also a radioactive sulfurous mud and a hot water geyser whose water pillar reaches the high of 8 m. The waters from this area are used for various medical treatments.
- Lukovska spa is located in the southern part of Serbia, 300 km from Belgrade on the slopes of Kopaonik mountain. Nevertheless, Lukovska spa is the spa with the highest altitude in Serbia (681 masl). The spa is well-known for its numerous geothermal hot springs (samples S9, S10, S11, S12, S13, and S14). The temperatures of waters vary between 36°C to 90 °C. The chemical composition of the geothermal hot springs from this area as well as the amount of minerals makes Lukovska spa one of the wealthiest spas in Serbia. The use of geothermal energy in this area is well-known since the time of the Roman Empire. This is confirmed by archeological discoveries of an over 2000-year-old water pipes system. Nowadays, waters from this area are used for district heating of local hotels and the main usage is in medical treatments.
- 5) Jošanička spa is a spa town located in the southern part of Serbia, 250 km from Belgrade. Jošanička spa is the second-highest spa in Serbia with 550 masl. There are five geothermal sources in this area (samples S15 and S16) whose temperature is between 36 °C and 78 °C. The main usage of the waters from this area is in balneology due to the various chemical composition of geothermal hot springs.

In particular, a high concentration of Fe was measured in the sample from Krýsuvík with a concentration of 8982 μ g/l. Also, a high concentration of Fe was measured in the samples from Hveradalir with a concentration of 5223 μ g/l and Solheimar Eco Village with a concentration of 1341 μ g/l. Sr, B, and Ba are dominant elements in the samples from the Reykjanes power plant outlet and the Blue Lagoon. In the sample from the Reykjanes power plant measured concentration of strontium is 7545 μ g/l, boron 6880 μ g/l and barium 7077 μ g/l. In the sample from the Blue Lagoon, Sr was measured in a concentration of 4299 μ g/l, B in a concentration of 5479 μ g/l and Ba reached a concentration of 1250 μ g/l. The high concentrations of Al were measured in the samples from Solheimar Eco Village, with a concentration of 4340 μ g/l and from Krýsuvík with the concentration of 3813 μ g/l. Manganese, arsenic, rubidium, and molybdenum were measured in lower concentration with an average concentration below 1000 μ g/l.

4.2 Results from Serbia

Results of ionic composition from geothermal hot springs in Serbia reveal that the dominant ion is Na+ with a concentration of up to 1.151 g/l. Results of Ca2+ and SO42- reveal lower concentrations of 0.147 g/l and 0.406 g/l, respectively. Significantly lower concentrations were found for K+, Mg2+, Cl- and F- with average concentration ranging between 0.011 g/l and 0.084 g/l. Total mineralogical composition of all analyzed samples from Serbia revealed the highest concentration in samples collected in Sijarinska and Vranjska spa with the concentrations of 1.406 g/l and 0.911 g/l, respectively. The lowest mineralogical composition was found in the geothermal water from Niška spa with a concentration of 0.129 g/l.

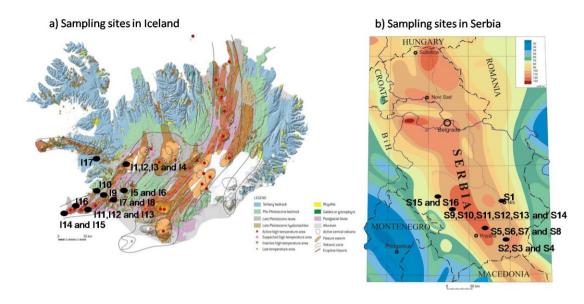


Figure 1: Overview of all sampling locations: a) sampling locations in Iceland, b) sampling locations in Serbia. Maps modified from Jóhannesson and Saemundsson (1999) and Milivojević (2010).

Table 1: Overview of the sampling locations in Iceland and Serbia

Area	Location	Type	Usage	Temperature [°C]	Sample name				
Sampling locations in Iceland									
Árnessýsla	Geysir	Natural spring	Tourist attraction	>100	I1, I2, I3				
					I4				
	Solheimar Eco Village	Borehole outlet	Heating	90	I5				
			Tap water		I6				
	Hveragerði	Natural spring	Agriculture	150	I7				
		Borehole outlet	Heating		I8				
	Reykjadalur	Natural spring	Tourist attraction	>100	19				
Hengill	Hveradalir	Natural spring	No particular use	>100	I10				
The Reykjanes	Krýsuvík	Natural spring	Tourist attraction	100	I11, I12, I13				
	Reykjanes power plant	Plant outlet	Electricity production	~320	I14				
	(RPP)	Lagoon	Natural and some plant outlet	~320	I15				
	The Blue lagoon	Plant outlet	Rehabilitation center		I16				
Reykholtsdalur	Deildartunguhver	Natural spring	Heating	>100	I17				

Sampling locations in Serbia								
Nišava district	Niška spa	Natural spring	Rehabilitation center	37	S1			
Pčinja district	Vranjska spa	Natural spring	Rehabilitation center	96	S2, S3			
		Borehole outlet	Heating	96	S4			
Jablanica district	Sijarinska spa	Natural spring	Rehabilitation center	70	S5, S6, S7			
					S8			
Toplica district	Lukovska spa	Natural spring	Rehabilitation center	36 – 90	S9, S10, S11,			
					S12, S13, S14			
Raška district	Jošanička spa	Natural spring	Rehabilitation center	36 - 78	S15, S16			

3. METHODS

In order to characterize the chemical composition of geothermal hot springs, water samples were collected from nine locations in Iceland (August to December 2018) and five locations in Serbia (December 2017 and June 2018).

For extracting water samples from hot geothermal springs, a metal container was used to fill up the sampling bottles. The metal container was first washed with acid and distilled water and rinsed three times with sampling fluid before sampling. Sampling bottles consisted of high-density PVC with a volume of 50 ml were cleaned by rinsing with nitric acid and deionized water. Before sampling the bottles were rinsed three times with the sample fluid and filled with geothermal water to the top. The cap was tightened and samples were kept in a refrigerator at a temperature of 4°C until the analysis in the laboratory.

In order to analyze ionic composition in water samples a Dionex ICS-900cation chromatographer was used to determine the concentration of sodium (Na+), potassium (K+), magnesium (Mg2+) and calcium (Ca2+) and a Dionex ICS-900 anion chromatographer to determine the concentration of fluoride (F-), chloride (Cl-) and sulfate (SO42-). An inductively coupled plasma optical emission spectrometer, Thermo Scientific ICP-OES iCAP 6500 Duo was used to analyze the concentration of aluminum (Al), boron (B), barium (Ba), iron (Fe) and strontium (Sr). An Inductively coupled plasma mass spectrometer, Thermo Scientific ICP-MS iCAP Q was used to determine the concentration of manganese (Mn), arsenic (As), rubidium (Rb) and molybdenum (Mo). Further elements were also analyzed but are not discussed here due to their low concentrations in the samples.

The accuracy of methods was verified using standard solutions for each method. The uncertainty of measurements was obtained by accurate standardization, blank measurements, and duplicate samples measurements.

Concentrations of the ionic compositions are expressed in g/l while the concentrations of the elements are expressed in µg/l.

4. RESULTS

Cations including Na+, K+, Mg2+, Ca2+, and anions including F-, Cl- and SO42- are naturally present in groundwater and soil. Ion concentrations found in all collected water samples from Iceland and Serbia are illustrated in Figure 2. In general, Na+ is the dominating ion in all samples. With the exception of samples collected from the Svartsengi power plant outlet and the Blue lagoon, the total mineralogical composition reveals similar concentrations, with average concentrations amounting up to 0.416g/l in Iceland and 1.406 g/l in Serbia.

Concentrations of analyzed elements including aluminium (Al), boron (B), barium (Ba), iron (Fe), strontium (Sr), manganese (Mn), arsenic (As), rubidium (Rb) and molybdenum (Mo) from the samples in Iceland and Serbia are illustrated in Figure 3.

4.1 Results from Iceland

In Iceland Na+, K+ and Ca2+were the dominating ions, with concentration reaching over 14 mg/l for Na+, 2 g/l for K+, and Ca2+. Significantly lower were average concentrations of Mg2+ and Cl- with concentrations ranging between 0,001 g/l and 0,004 g/l. The least abundant ions were F- and SO42-with average concentrations below 0,007 g/l.

ICP Analysis

Depending on the location, the dominant elements in Icelandic geothermal hot springs are iron (Fe), strontium (Sr), boron (B), barium (Ba) and aluminum (Al).

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ICP Analysis

In Serbian geothermal hot springs, the dominant elements are boron, strontium, and arsenic. Boron was found at all locations in fairly high concentrations between 141 μ g/l for Lukovska spa and 7531 μ g/l for Sijarinska spa. Strontium is the second highest abundant element in Serbian geothermal hot springs with concentrations between 77 μ g/l for Jošanička spa and 1238 μ g/l for Lukovska spa. A high amount of arsenic with a concentration of 1575 μ g/l was measured in the sample from Lukovska spa.

Other elements including aluminum, barium, iron, manganese, rubidium, and molybdenum were found in lower concentrations with the average concentration below $400 \mu g/l$.

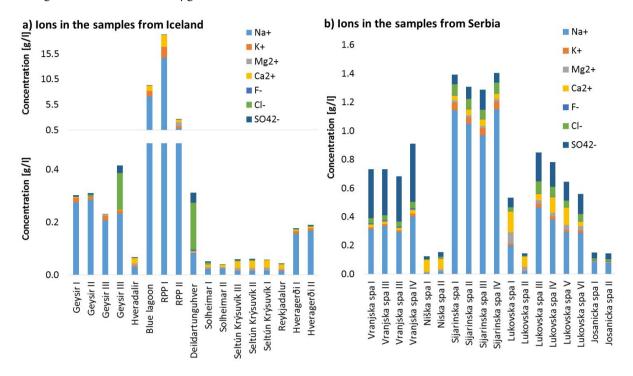


Figure 2: Ion concentrations of samples collected in a) Iceland and b) Serbia.

5 DISCUSSION

Based on the presented results the most dominant ions in all samples were sodium and sulfate and the dominant elements boron and strontium. The interaction of fluids and rocks at a high temperature could be a source of sodium and sulfate explaining their high concentration in all samples.

The highest concentration of total measured parameters was found in the samples from the Blue Lagoon and the Reykjanes power plant in Iceland, and samples from Sijarinska and Lukovska spa in Serbia.

Water samples from hot springs in Iceland reveal sodium as the dominating anion, which can be attributed to seawater content in some samples. This is especially the case for samples taken from the Reykjanes power plant outlet, where seawater is used for cooling purposes.

In Iceland iron was highly abundant in many samples, especially in waters from Krýsuvík. Furthermore, in Solheimar and Krýsuvík aluminum was found in relatively high concentrations.

In Serbia, the relative ratio of sulfate was higher than in most samples from Iceland. This could possibly be caused by dissolving and weathering rocks in sub-surface at a high temperature.

Boron is the most abundant element in the samples collected in Serbia. Furthermore, strontium and arsenic are highly abundant in Serbian samples which could explain the usage of these waters in various medical treatments.

6 CONCLUSIONS

In this paper water samples from various geothermal sources in Iceland and Serbia were collected and analyzed for their chemical composition. In total 32 water samples from 14 locations were analyzed for their anion, cation and other elements content. Based on the presented results we conclude the following:

- The dominating ion in all samples was sodium. While Sodium concentrations were highest at two locations in Iceland, it is probable that these two water samples contained seawater from the adjacent Atlantic. In most other samples sodium concentrations were slightly higher in samples from Serbia.
- Sulfate concentrations were relatively high in samples collected in Serbia.

• Iron was highly abundant in the sample of the Icelandic location Krýsuvík while boron was highly abundant in most samples from Serbia.

Based on these conclusions it seems that the chemical composition of geothermal water in Serbia has a high potential for thermal energy, therapeutical and recreational purposes and other commercial uses similar to the Icelandic case. Nevertheless, Serbia is currently using only small amounts of the geothermal sources at few locations. The most common use of geothermal energy in Serbia is balneotherapy, sport, and recreation. Since geothermal energy is a renewable energy source that can be utilized in a sustainable way, it would be wasteful not to use it.

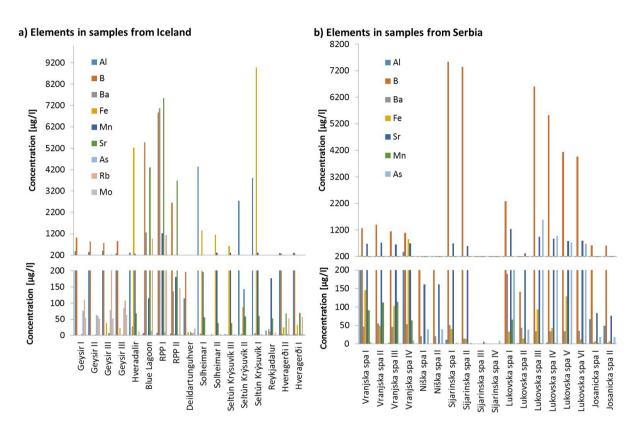


Figure 3: Concentration of dominant elements in samples collected in a) Iceland and b) Serbia.

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