University of Tehran Press

Pollution

Print ISSN: 2383-451X
Online ISSN: 2383-4501

https://jpoll.ut.ac.ir/

Physical and Geochemical Characteristics of the Typical Spring's Mineral Water in the NW of Iran, Case studies Lighvan and Toptapan Springs Mineral Water

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Article Info

Article type:
Research Article

Article history:
Received: 09.06.2022
Revised: 24.07.2022
Accepted: 29.10.2022

Keywords:
Geochemistry
environmental
Springs mineral water
Eastern Azarbaijan
Iran

ABSTRACT

Lighvan hot spring and Toptapan mineral spring are located in the Eastern Azarbaijan, NW of Iran. The host rocks of Lighvan hot spring are dacite, andesite and Quaternary volcanic tuffs. Their main rock forming minerals are quartz, plagioclase, biotite and rarely amphibole. The host rocks of Toptapan mineral water spring are Cretaceous and Jurassic sandstone, shales and carbonate sedimentary rocks. Their main rock forming minerals are quartz, calcite, dolomite and clays. Due to the deposition of mineral water springs, travertine is the main Quaternary sediments around the springs. Water samples were collected from Toptapan mineral spring and Lighvan hot spring in July (dry season). The sampling method was according to standard methods for geochemical analysis. Field parameters such as PH, temperature, and EC were measured in situ, and samples were analyzed by ICP-OEC and ICP-MS in the laboratory of the Geological Survey of Iran. The measuring data showed that pH varies between 6.1 to 6.4. The surface temperature varies from 20.1˚C to 32.8˚C. The concentration of anions and cations in the Piper diagram show calcic bicarbonate type for Toptapan mineral spring and sodic bicarbonate type for Lighvan hot spring respectively. According to Lunglier – Ludwig diagram, the dissolution of carbonate and silicate minerals is the most important factor in increasing calcic cation. The Cl-Li-B diagram shows that the dissolution of sodic minerals and clays and ionic exchange are also the most important factors for increasing sodium in these springs. These data are in agreement to the host rocks, their mineralogy and their chemical composition. Based on the Ca-Mg-K geothermometer diagram, the geothermal reservoir temperature for Lighvan hot spring is 95-100 ˚C with a depth of about 2Km and for Toptapan mineral spring is 65-85 ˚C with a depth of less than 1Km. Also, high concentrations of chlorine show a deep geothermal primary reservoir in the Lighvan hot spring. These geochemical data show that these cold and hot springs are not polluted and not harmful for environmental point of views.

Cite this article: Yazdi, M., Mohammadi, F., Navi, P., & Behzadi, M. (2023). *Physical and Geochemical Characteristics of the Typical Spring's Mineral Water in the NW of Iran, Case studies Lighvan and Toptapan Springs Mineral Water*. Pollution, 9(1): 169-182.
http//doi.org/10.22059/poll.2022.344213.1498

DOI: http//doi.org/10.22059/poll.2022.344213.1498

INTRODUCTION

One of the most important geothermal resources of Iran is located in the Eastern Azerbaijan region, around the Sahand- Sabalan volcanoes, NW of Iran (Nabavi, 1976, Mehdipour Ghazi and Moazzen, 2015). These geothermal sources are located in the Urmia-Dokhtar plutonic-

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volcanic geotectonic zone of Iran (Figure 1). These geothermal resources were created due to the tectonic forces and plutonic heat sources in the western and southwestern part of the Azarbaijan region, especially is related to the Tabriz major fault and Sahand - Sabalan old volcanoes (Yousefi et al., 2010, Mohammadi, 2016).

The present paper is the result of our research on two of the most important springs in this region. We used Lighvan hot spring and Toptapan cold mineral spring to compare the geochemical features and genesis of these two different springs. Lighvan hot spring is located in 65 km east of Urmia Lake and 30 km southeast of Tabriz city. Toptapan cold mineral spring is located at about 25 km east of Urmia Lake and 45 km SW of Tabriz city (Figure 1). These areas are cold, rainy-snow, with long winter climate and mild climate and short summers. The annual average rainfall is 288 mm and annual average temperature is 12.5 °C (Mohammadi, 2016). The host rocks of Lighvan hot spring are Sahand volcano igneous rocks, dacite, andesite and Quaternary volcanic tuffs (Figure 2). Their main rock forming minerals are quartz, plagioclase, biotite and rarely amphibole. The host rocks of Toptapan mineral water spring are Cretaceous and Jurassic carbonate sedimentary rocks. Their main rock forming minerals are calcite, dolomite and clays. Due to the deposition of mineral water springs, travertine sediments are seen around the spring (Figure 3) which is a sign of the activity of these springs. Relocation of spring can be due to fault activities at different times.

In order to analyze the structure of the study area and understand the nature of the layers and surrounding subsurface structures of these springs, two cross sections have been drawn perpendicular to the trend of the structures (Figures 2 and 3). The cross section of Lighvan area has been drawn in a length of 6Km with NE-SW trend (Figure 2). The BB´ section show that the main basement rocks of the area are Neogene tuffs, andesite, dacite-andesite and lahar. There is a normal fault with the NW-SE trend that passes from the south and southwest of the hot spring. Atmospheric water seems to be entered to the ground through these faults and fractures and after being heated by heat engine of Sahand volcano (Figure 2). The cross section of the Toptapan area has been drawn with a length 9 km with NE-SW trend (Figure 3). The AA´ section show that the main basement rocks of the area are dolomite, dolomitic limestone (Triassic), sandstone, shales

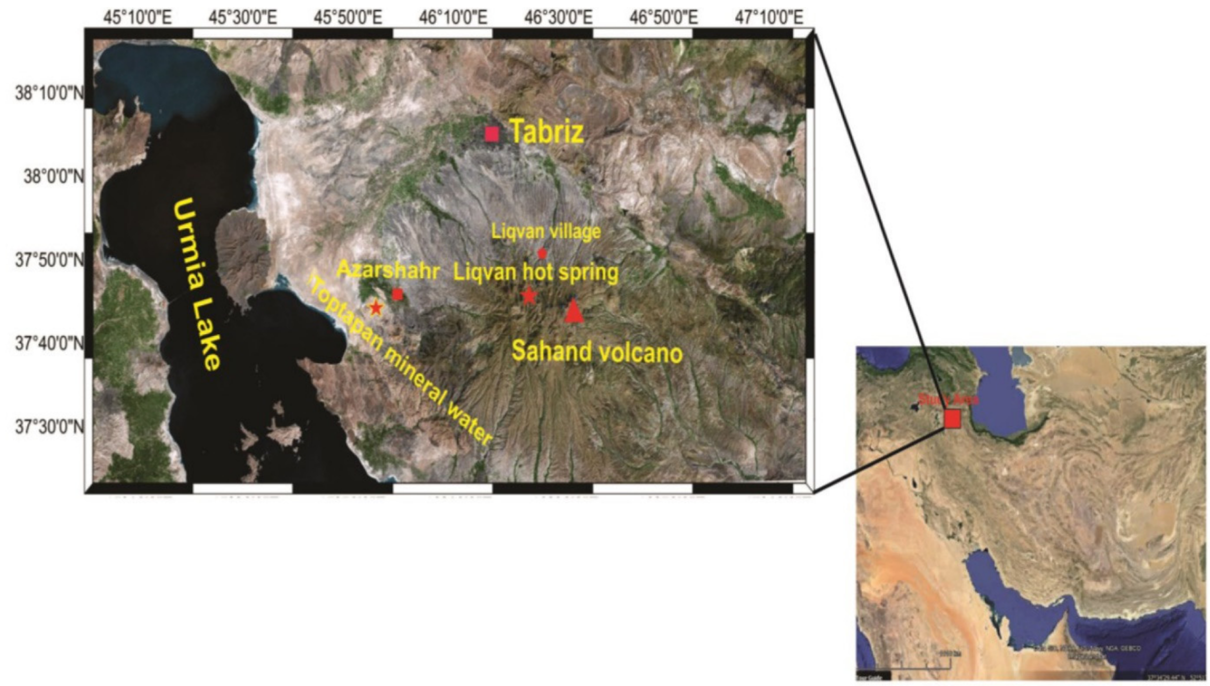

Fig. 1. The geographical location of Lighvan hot springs and Toptapan cold mineral spring

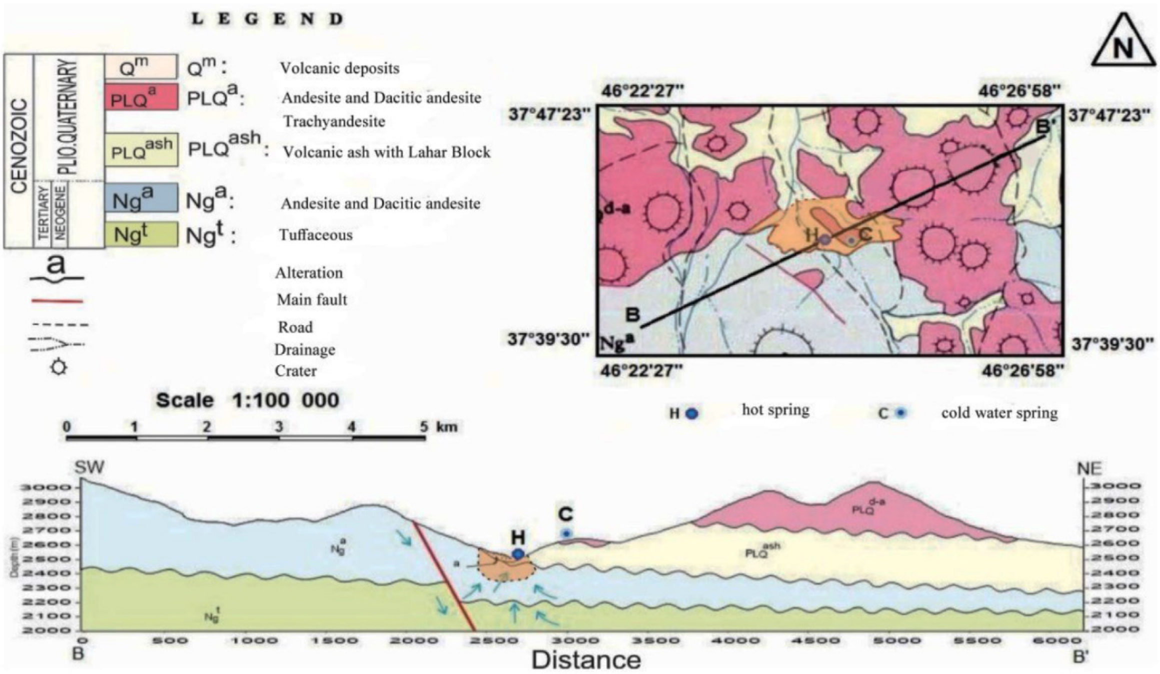

Fig. 2. Geological map (adapted from the geological map of Osko 1:100000) and structural section of Lighvan region (Khodabandeh et al., Fazel. 1995).

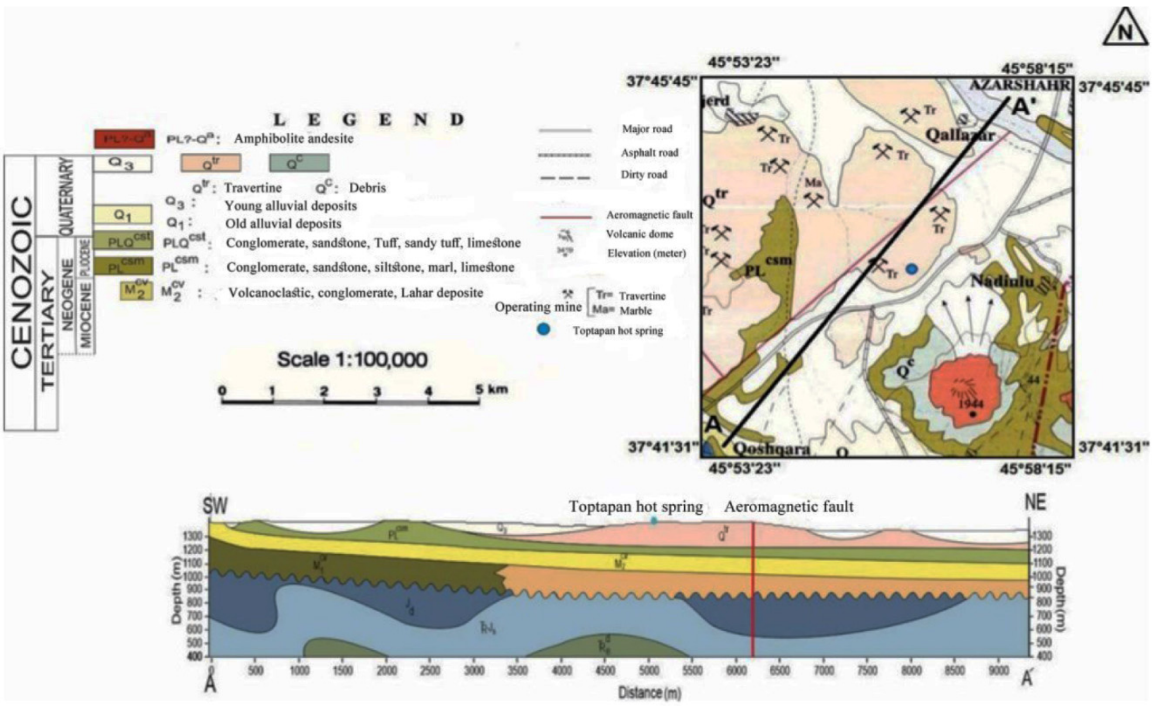

Fig. 3. Geological map (derived from the geological 1:100000 map of Azarshahr) and structural section of Toptapan area (Gadirzadeh et al.,2002)

with interlayers of limestone (Jurassic Shemshak Formation), gray to white limestones (Middle Jurassic Delichay Formation), Miocene volcanic breccia, conglomerate, sandstone, siltstone, marl, Pliocene limestone and Quaternary travertines (Shahrabi,1994, Khodabandeh et al, 1995, Gadirzadeh et al., 2002, Mohajjel and Taghipour 2014).

MATERIALS AND METHODS

A total of 8 water samples were collected from Lighvan hot spring, Lighvan cold water and Toptapan mineral water springs (Table 1). To minimize dilution of the hot-springs water with infiltrated meteoric water and to investigate the trace element concentrations in dry season, samples were collected during the summer (dry period). Some in situ measurements such as pH, electrical conductivity (EC), and temperature were conducted during sampling. All samples were collected as two filtered batches into 250-ml polyethylene containers (Moghimi,2006). A few drops of nitric acid were added to the containers to prevent metal deposition and adsorption to the container walls (Yazdi, 2003). The samples were stored in a cool, dark place until they reached the laboratory. The samples were analyzed by using a standard method suggested by the American Public Health Association (APHA, 2020) in the Geological Survey of Iran (GSI) laboratory. The ICP-OES and ICP-MS method was applied to analyze major and trace elements and cations measured by flame photometry. Sulfate concentration was measured by spectrophotometry. Chloride and bicarbonate concentrations were measured by titration methods. The TDS values were also analyzed. The term TDS (Total Dissolved Solids) describes the inorganic salts and colloids present in solution in water (Nik Peyman et al., 2019). The geographical coordinates, physical and chemical parameters of these springs are presented in Tables 1 and 2. Petrographic studies were done by polarized microscope in laboratory of Faculty of Earth Sciences, Shahid Beheshti University. Also, we analyzed some representative samples by X-ray diffraction (XRD) method in the Geological Survey of Iran (GSI) laboratory.

RESULTS AND DISCUSSION

Hydrogeochemical results

In order to investigate the error rate and accuracy of the analysis, the error percentage of the ion balance was calculated using the following formula (Yazdi, 2013). Error= Σ Cations – Σ Anions/ Σ Cations + Σ Anions × 100% Our calculations show that the difference between cations and anions in milliequivalents per liter (meq/l) is 5% for Lighvan hot springs and for Toptapan and cold-water springs are 9% and 11% respectively. The ionic balance indicates the accuracy of the chemical test in the hot springs. AquaChem 2011.1 software were used for data processing and drawing of Piper diagram (Piper, 1944). The results show that water of Lighvan hot spring enriched in sodium chloride, and the water of Toptapan mineral water springs and Lighvan cold spring enriched in calcium bicarbonate (Figure 4). According to petrographic studies and rock samples analysis by X-ray diffraction (XRD) method, it seems that the high amount of bicarbonate in Lighvan hot springs compared to Toptapan mineral water spring is due to Ca-plagioclase alteration of igneous rocks.

The mineral saturation index is an indicator which used to clarify the tendency of water to precipitate or dissolve minerals. Saturation index (SI) is obtained from comparing the chemical

Table 1. Geographical coordinates and some physicochemical parameters of studied springs

parameter	Toptapan mineral spring (Td)	Lighvan cold spring (Nsd)	Liqvan hot spring (Ld1)	Liqvan hot spring (Ld2)
longitude	″60΄56 ˚45	″26΄24 ˚46	″8/22΄24 ˚46	″8/22΄24 ˚46
latitude	″38΄43˚37	″21΄46˚37	″5/24΄46˚37	″6/24΄46˚37
Temperature (˚C)	20.1	6.2	32.8	31.6
pH	6.17	8	6.43	6.25
EC (μs/cm)	4700	70	7000	7500
TDS (mg/l)	2300	70	4710	4770

Table 2. Concentration of main anions and cations and some chemical elements (ppm)

spring	Hot water 2	Hot water 1	Cold water	Toptapan
type	Calcium Bicarbonate	Calcium Bicarbonate	Calcium Bicarbonate	Calcium Bicarbonate
HCO^{3-}	2000	1781	45	1708
CO_3^{-2}	0.3	60	0.3	0.3
SO_4^{-2}	654.17	698.33	5	213.3
Cl^-	1064	1064	2.5	142
Ca	171.76	136	12.2	211
K	69.16	71.75	2.83	33.5
Mg	57.47	58.3	2.42	94.3
Na	1224	1224	5.2	218
Si	20	20	18.1	16.7
Sr	4.7	4.5	0.1	1.67
Li	4.2	4.3	0.18	0.3
As	2.5	2.4	0.002	1.1
B	16.5	15.3	0.009	2.74
Cs	0.547	0.553	0.00018	0.496
%Error	5	5	11	9

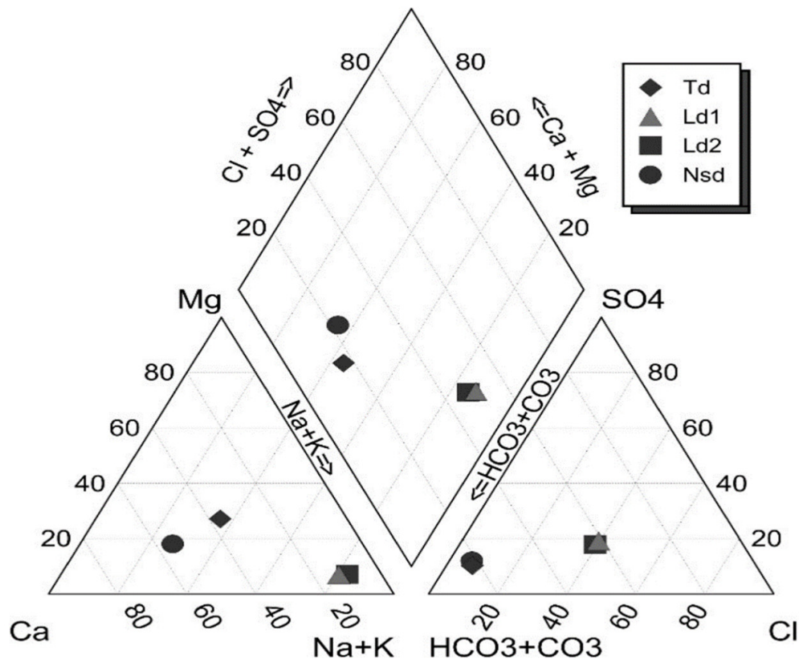

Fig. 4. Type and facies of Liqvan hot springs and Toptapan cold water springs in Piper diagram (% meq) (Piper, 1944).

activity of soluble ions (IAP: Ion Activity Product) with their dissolution rate and it can be calculated through the following formula (Langmuir,1997): $SI = \log (IAP / Ksp)$

The saturation index for different minerals is used to evaluate the balance between water and minerals, which the change in water saturation in relation to different minerals determines the geochemical reactions that control water chemistry (Jalali, 2006). If the saturation index (SI) is less than zero, the water is below saturation mode and the solution has more solubility. If SI is equal to zero, there is a balance between the solid and solution phases. If the SI quantity is more than zero and less than one, the water is on the verge of saturation, and if the SI is more than one, the water is supersaturated (Langelier and Ludwig, 1942, Deutsch, 1997, Merkel et al., 2008).

The saturation index of calcite mineral for cold spring water samples was negative. But this index for hot spring was positive. Also, despite the high concentration of sulfate and chloride in hot springs and mineral springs, the saturation index of springs is negative compared to gypsum, anhydrite, and halite minerals (Table 3). This indicates that the groundwater of the area is under the saturation rate with these minerals. It seems that these minerals can be more dissolved in the groundwater of the area and increase the salinity of groundwater. Also, saturation rate of the CO_2 gas is low. This means that more CO_2 can be dissolved and increase the acidity of the groundwater of the area.

Chlorine versus sodium and bicarbonate diagrams show a high correlation between chlorine and sodium which is indicating of the common origin of the two ions (Figure 5). The Na/Cl

Table 3. Saturation Index for Lighvan hot springs and Toptapan cold water (calculated in Phreeqc software,2021)

Spring	$SI_{Calcite}$	$SI_{Aragonite}$	$SI_{Anhydrite}$	SI_{Gypsum}	SI_{Halite}	SI_{CO2}
Td	0.13	-0.02	-1.39	-1.15	-6.14	-0.58
Nsd	-1.70	-2.61	-3.62	-3.37	-9.37	-2.67
Ld1	0.37	0.23	-1.12	-0.93	-4.60	-0.64
Ld_2	0.23	0.09	-1.24	-1.05	-4.60	-0.51

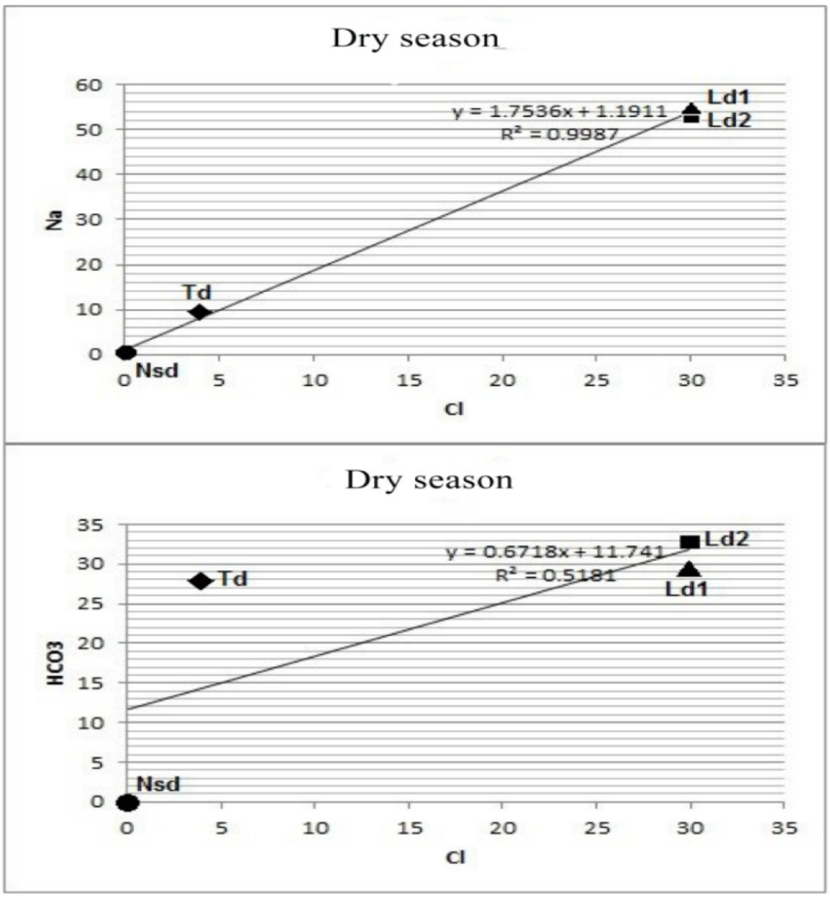

Fig. 5. Chlorine versus sodium and bicarbonate (meq/l) for Lighvan hot springs and Toptapan cold water springs.

ratio is more than 1 in hot springs which indicates the ion exchange in these waters. Higher concentrations of sodium compared to chlorine indicate that normal ion exchange in these waters (Shamohammadi, 2015). On the other hand, chlorine shows low correlation to bicarbonate. The Ca/Cl average ratio in Lighvan hot springs is lower than Lighvan cold springs. In cold springs this ratio is 3/27 and in hot and mineral springs is 0.13 and 1.37 respectively. The Na versus Cl is used to determine the source of sodium in the studied springs (Figure 6). The Na/Cl average ratio is more than 1 which seems to be natural origin for Na. It may be to the decomposition of sodium-containing clays such as montmorillonite (Boslik 2011, Zorratipour et al., 2021).

One of the diagrams that show the type of water-soluble minerals, is the Langlier-Ludwig diagram (Langelier. and Ludwig. 1942, Jeelani et al., 2011). This diagram is based on the concentration of bicarbonate, sulfate, chloride, calcium, magnesium, sodium, and potassium ions. The three main parameters identified in this diagram include gypsum/anhydrite dissolution, carbonate and silicate dissolution. The results of our data showed that the dissolution of carbonate and silicate minerals has an important role in the chemical composition of the studied water samples (Figure 7).

The most important process that affects the composition of geothermal fluids, is the dissolution of primary minerals and the deposition of secondary minerals (Esmaeili-Vardanjani, 2015). The dissolution process increases the concentration of constituents such as B, Br, Cl, and other constituents in aquifer fluids, therefore the composition of geothermal water is very different. The best way to determine the geothermal water type is to use the Cl-HCO_3-SO_4 ternary plot proposed by Giggenbach, 1991. The composition of the hot springs and their location in the plot and also their proximity to the top of HCO_3 in this diagram shows that the composition of geothermal waters in the region is of the bicarbonate type (Figure 8). The water-soluble minerals diagram show that the dissolution of carbonate and silicate is the most important factor in the chemical composition of these spring. According to petrographic studies and rock samples analysis by X-ray diffraction (XRD) method, it seems that the high amount of bicarbonate in Lighvan hot springs compared to Toptapan mineral water spring is due to Ca-plagioclase alteration of igneous rocks. These data are in agreement to the host rocks, their mineralogy and their chemical composition.

The HCO_3/Cl average ratio in Lighvan cold water spring is 18 and in Toptapan mineral water

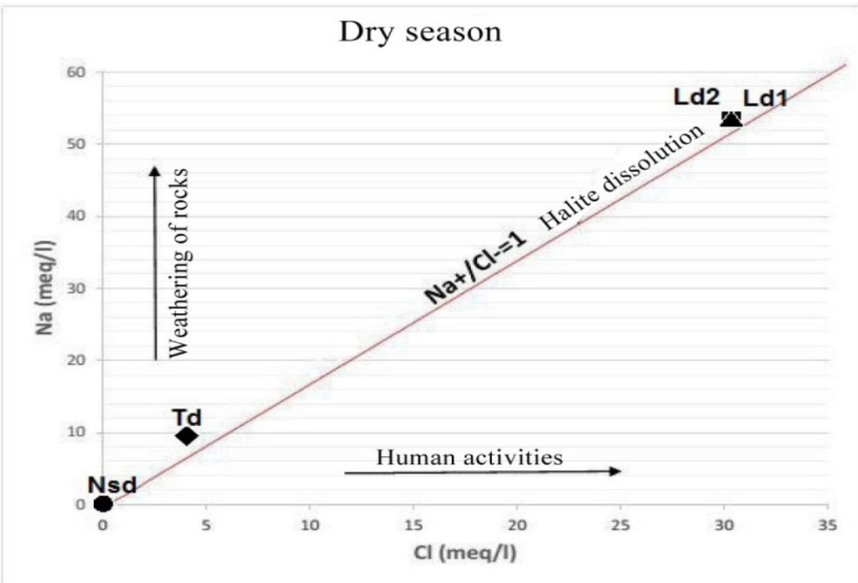

Fig. 6. Sodium vs. chlorine for Lighvan hot springs and Toptapan cold water springs (Hem,1989)

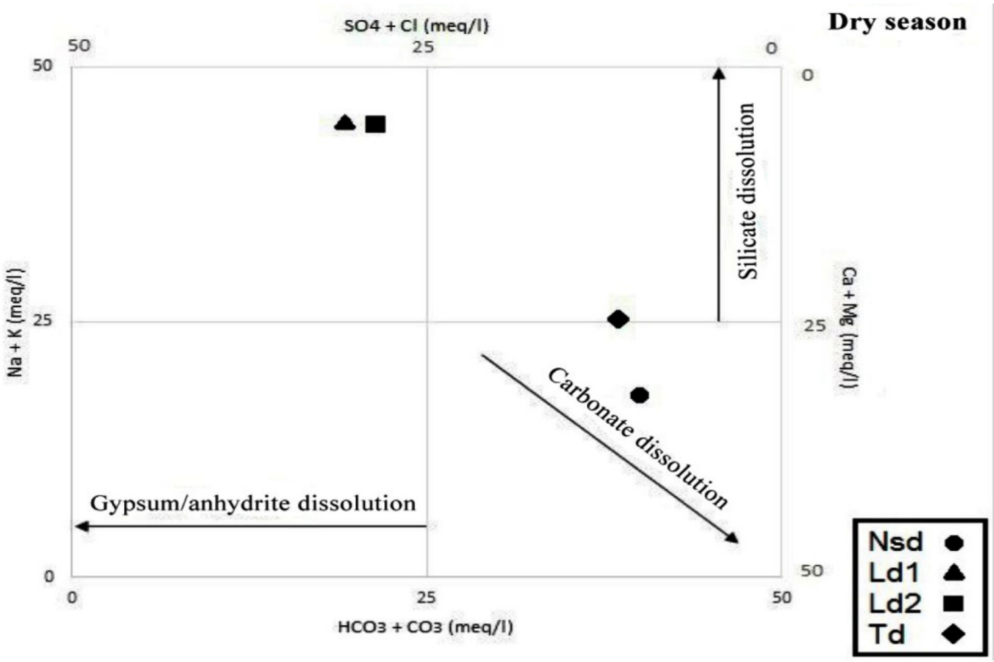

Fig. 7. Langlier-Ludwig diagram for Lighvan hot spring and Toptapan cold water spring (Merkel and Planer-Friedrich, 2008).

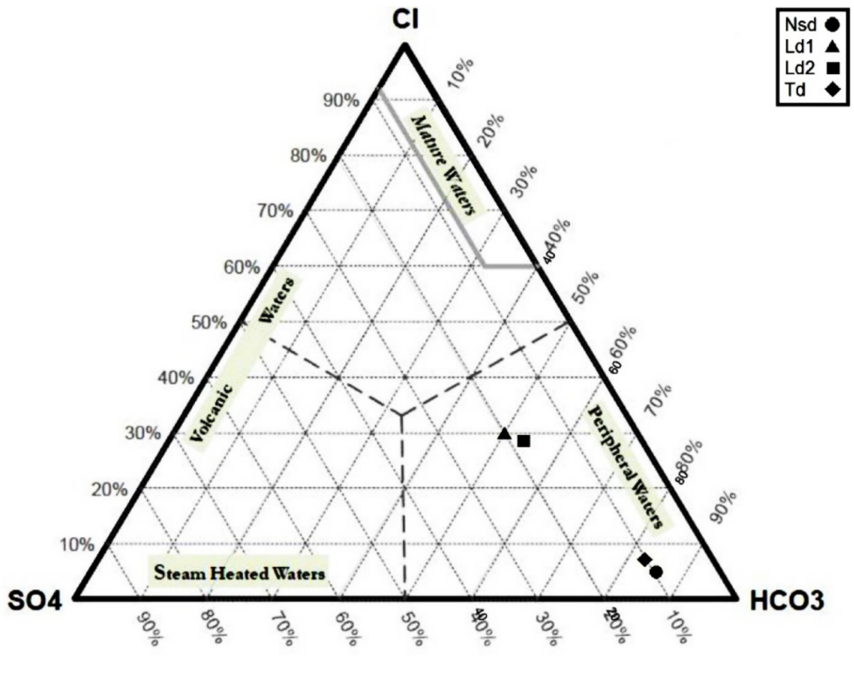

Fig. 8. Cl-HCO_3-SO_4 plot (meq/l) showing the geothermal water type of these springs (Giggenbach, 1991).

spring is 12 and in Lighvan hot springs is 1.77. Therefore, the high value of this ratio in cold spring water is a reflection of the short-term flow and fast water cycle while its low value in warm waters indicates that these waters have circulated underground for a longer period of time and the water cycle has occurred in depth (Han et al., 2010). The Li is an alkaline metal that is not affected by secondary processes and therefore it is used as a trace element to evaluate the

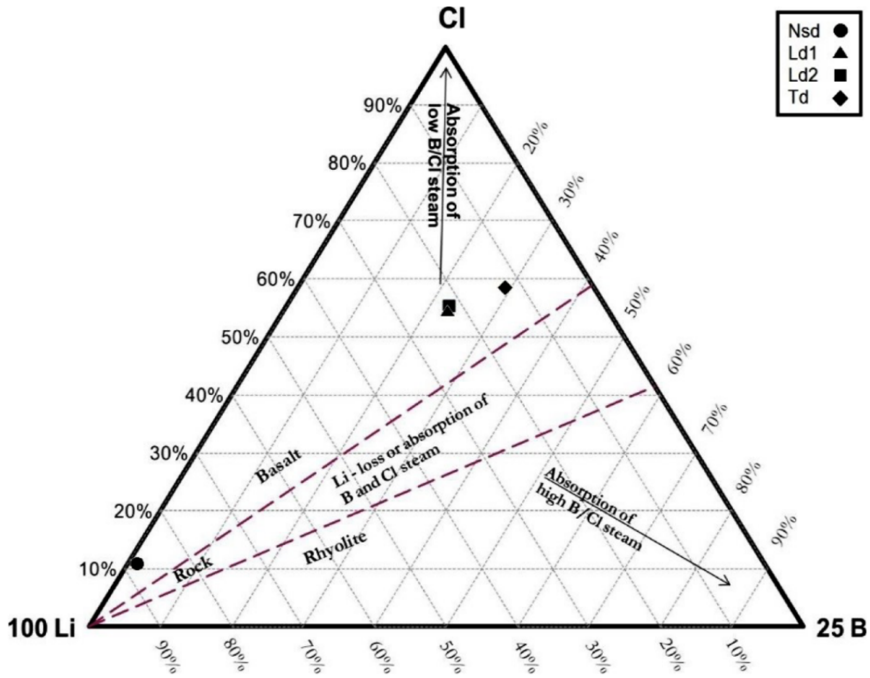

Fig. 9. Source of geothermal water for Lighvan hot springs and Toptapan cold water (meq/l) (Muhwezi, 2009)

possible origin of Cl and B as well as to determine the dissolution process of rocks (Muhwezi, 2009). The Cl-Li-B ternary plot is usually used when the chlorine concentration in these samples is more than 10% (Figure 9). According to this diagram, these hot springs samples are located near the top of chlorine ratio which shows that these waters originate from an old geothermal system. The high amount of chlorine in spring water indicates the original depth of the geothermal reservoir (O'Brien, 2010). Changes in the Cl to B ratio in a geothermal system can be due to fluid origin, lithological changes in the fluid passage and adsorption of boron by clays, or reactions with basement rocks such as basalts and andesites (Giggenbach, 1988). The amount of boron (B) is a reflection of the degree of maturity of the geothermal system, because evaporation during heating and rising causes the release of elements such as B, As, Sb, and Hg, so old heating systems can be depleted of these elements (Kipng'ok and Kanda, 2011).

Stable elements such as Rb, Cs, and Cl are used for detecting of heavy elements, fluids, and rock origin since they are not affected by dilution and boiling processes. The Cl-Rb-Cs triangular diagram shows that the hot and cold springs are located near the rock dissolution zone (Figure 10). This suggests that these springs are probably due to the result of the dissolution of magmatic gases in groundwater, which ultimately led to the iso-chemical dissolution of rocks. Li/Cs average ratio in Lighvan hot springs is 7.8. This ratio is close to the ratio of intermediate acidic rocks which, given the low chlorine content and no severe depletion of B, may have been the source of hot springs and mineral springs at moderate depths (Giggenbach and Glover, 1992).

Geo-thermometry

Geochemical data of the chemical decomposition of spring water can also be used to estimate subsurface temperature (Yazdi et al., 2018). Many reactions in the geothermal system are temperature-dependent so that at low temperatures their kinetic energy is very slow and at high temperatures it is high. Therefore, their equilibrium properties are still maintained in the ground surface and cooling conditions. For this reason, subsurface heat can be calculated from the concentration and chemical composition of surface geothermal solutions (Yazdi et al., 2016). By using the Na-K-Mg diagram (Figure 11), the temperature of the geothermal reservoir

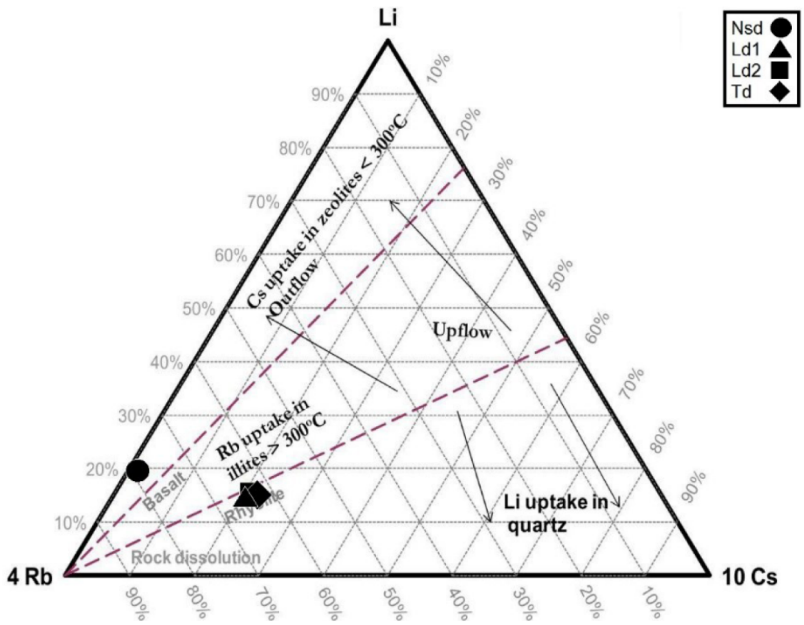

Fig. 10. Cl-Rb-Cs (meq/l) diagram for Lighvan hot springs and Toptapan cold water spring (Kipng'ok and Kanda, 2011).

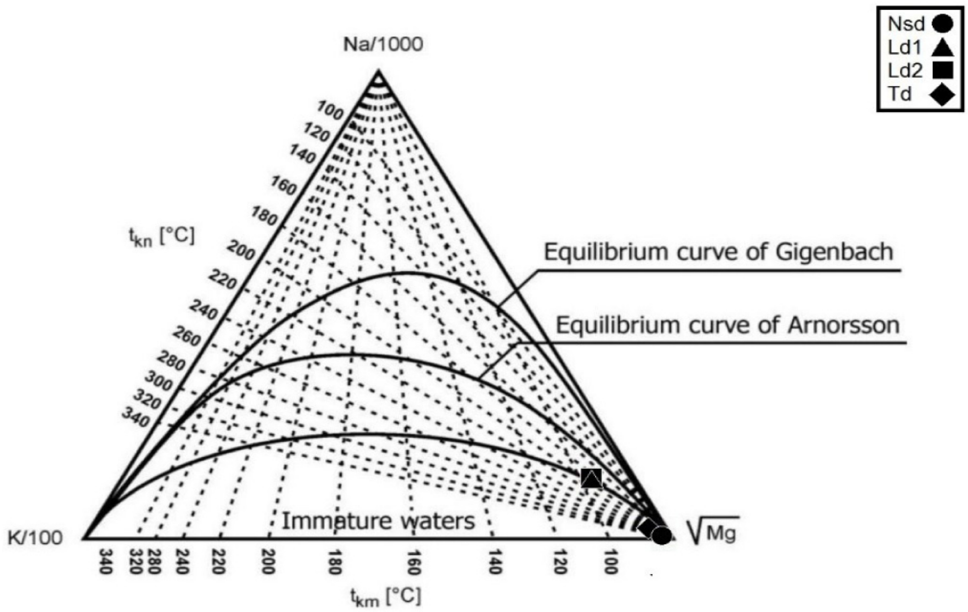

Fig. 11. Determination of geothermal water reservoir temperature for Lighvan hot springs and Toptapan cold water (meq/l) (Giggenbach, 1988).

associated with the springs can be estimated (Giggenbach, 1991). In hot waters with a deep origin, the concentration of Mg ions is much lower than Ca (Nik Peyman et al., 2019).

The result of chemical geothermometers is reliable when there is a balance between water and rock while the movement of the fluid to the surface and mixing with surface water gives unreliable results. The ternary plot of Na/1000 - K/100 - $\sqrt{Mg}$ can be used to measure the reservoir temperature of geothermal that has reached equilibrium with the host rock (Giggenbach, 1988). Figures 11 and 12 show that none of the samples of Lighvan hot springs have a chemical water-

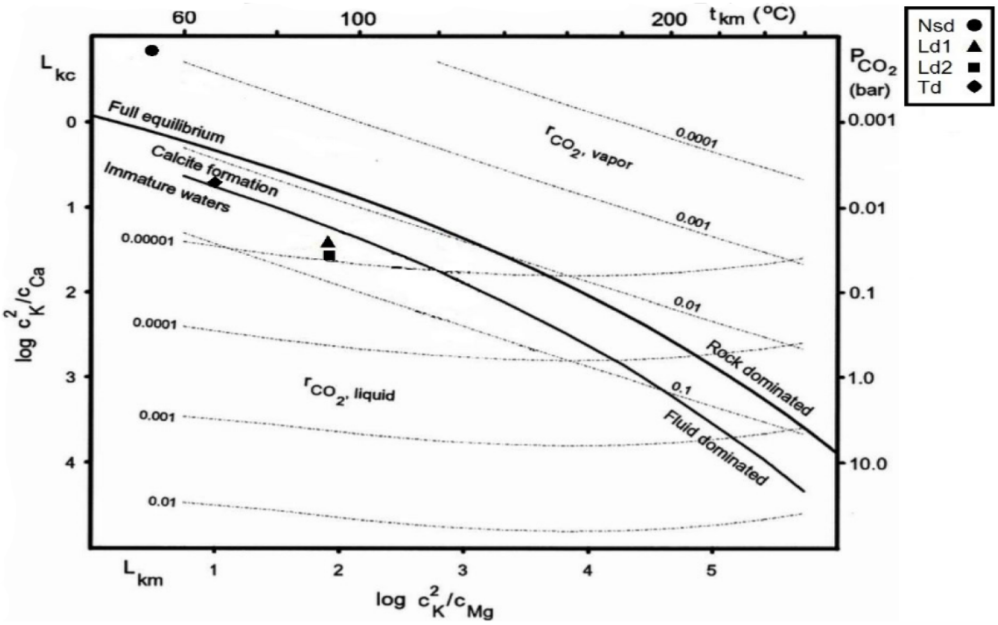

Fig. 12. Thermometer diagram of Ca-Mg-K (meq/l) for the studied springs (O'Brien, 2010)

rock balance and are located in the area of immature (shallow) waters. The calculated temperature based on the linear trend shows the samples of Lighvan hot springs are about 95-100 ° C and in the depth of 2 km approximately. These diagrams also show that the samples of Toptapan cold water spring are in equilibrium with calcite and are in the range of mature waters. The estimated temperature of the geothermal reservoir of Toptapan mineral water spring is about 65 to 85 °C at a depth of less than 1 Km close to the surface.

Spring formation process

The hypothetical model for the formation of Lighvan hot springs can be presented in such a way that atmospheric water has penetrated deep into the earth along the main and minor faults and permeable rocks of the region. These waters are heated by the geothermal slope and volcanic rocks and the solubility of their surrounding rocks is also increased. Then they possibly move upward in an ascending path along faults and fractures, mixing with water from the volcanic and igneous activity and its chemical composition changing during the heat losing. During of the hot water movement, host rocks are dissolving and soluble elements are increasing.

The heat factor in the depths can be attributed to the presence of volcanic activity. The reason for the heat of hot springs can be attributed to thermal anomalies caused by the geothermal gradient and Sahand volcano. The hypothetical model and formation of many springs in this region, especially in Toptapan mineral water springs can be explained. All travertine deposits in the region are located on the direction's major faults and minor faults. The major and minor faults are the suitable channel to penetrate surface water into the underground. Water heating in the depth can be due to the result of geothermal gradient. Also, intrusions related to Sahand volcano and active tectonics are increasing the temperature of these waters. Faults and intersecting fractures and permeable rocks are the best way for circulation of the hydrothermal fluids and increases their solubility in reaction with host rocks. Therefore, waters, especially hot springs, move upwards along these faults due to their lifting force. The thermometer diagram of Ca-Mg-K shows that the temperature of the heating reservoir of Lighvan hot spring is 95 to 100 ° C and for Toptapan mineral water spring is about 65 to 85°C (Figures 11-12). When water ascends along the host rocks, it loses its temperature. As a result, more gases and volatiles are dissolved in the water. The shallow cold-water mixing can also be effective in the process of dissolution of carbonate rocks. The ascent of water to the upper parts and the decreasing of hydrostatic pressure causes the release of gases, including CO_2, and the saturation of water with calcite. It is resulting deposition of calcium carbonate such as travertines.

CONCLUSIONS

This research shows that there is geochemistry, environmental features, geo-thermometry and genesis differences between Lighvan hot spring, Lighvan cold water and Toptapan mineral water springs. The geochemical data show that Lighvan hot springs are sodium bicarbonate type. Toptapan mineral water and Lighvan cold water are of calcium bicarbonate type.

The water-soluble minerals diagram show that the dissolution of carbonate and silicate is the most important factor in the chemical composition of these spring. According to petrographic studies and rock samples analysis by X-ray diffraction (XRD) method, it seems that the high amount of bicarbonate in Lighvan hot springs compared to Toptapan mineral water spring is due to Ca-plagioclase alteration of igneous rocks. These data are in agreement to the host rocks, their mineralogy and their chemical composition. The Cl-Rb-Cs ternary plot diagram shows that hot and cold springs are located in the rock dissolution zone. This suggests that these springs may have been affected by the dissolution of magmatic gases in groundwater. Eventually, this process caused the iso-chemical dissolution of the host rocks.

The thermometer diagram of Ca-Mg-K shows that the temperature of the heating reservoir of Lighvan hot spring is 95 to 100 ° C and for Toptapan mineral water spring is about 65 to 85 ° C. The high concentration of chloride in this hot spring indicates the depth of the primary geothermal reservoir. Also, geochemical data show that Lighvan hot springs have a chemical water-rock balance and are located in the area of immature (shallow) waters. The geochemical data confirms that the Toptapan cold water spring are in equilibrium with calcite and are in the range of mature waters. These geochemical data show that these cold and hot springs are not polluted and not harmful for environmental point of views.

ACKNOWLEDGMENT

The present research did not receive any financial support.

GRANT SUPPORT DETAILS

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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