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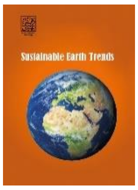

Evaluating the geochemistry of brine springs in the northern Khorram Abad, Iran

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ABSTRACT

For the first time evaluation of the geochemistry of brine springs located in northern Khorram Abad, northern High Zagros zone, was conducted in this study. The springs emerge in carbonate and marl successions of the Lower Miocene Gachsaran Formation, the most important cap rock of oil reservoirs in the Zagros basin, and the carbonate of the middle Cretaceous deposits. A total of 24 samples were taken for XRD analysis, 15 for XRF analysis, and 30 samples for ICP-OES analysis. The results of chemical analysis show that halite is the most important mineral in evaporate deposits, while gypsum, quartz, anhydrite, cristobalite, and dolomite were present at minor levels. Various geochemical evidences such as lower ratio than 1Na/Cl is less than 1 in the brine springs, and the concentration of trace elements B, Na, Cl, K, V, Cr, Mn, Fe, Cu, As, Br, Rb, Sr, Zr, Cs, the indicating their deep-basin origin. These springs have found their way to the earth's surface along the faults in the form of fault springs through the lower (Jurassic) evaporite deposits.

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1. Introduction

Among the common phenomena of nature, springs are notable because of their high usefulness. The essential factors in the production of springs are the source of the water and the rock structure which brings it to the surface. Brine springs are saline water present in the natural environment. They commonly accumulate mineral deposits that are responsive to salinity controls on the mode of mineralization and dominate solution chemistry, which may consist of various combinations of common salt (NaCl), calcium, iodine, lithium, magnesium, potassium, and bromine (Onwuka et al. 2019). Importantly, in some cases, brine resources are the preferred source of metals compared to that of other conventional metallic deposits (Rezaei et al., 2021). For example, salads of the Andean plateau in the central Andes are considered as being the largest lithium deposits on earth, which formed in the drained basins (Steinmetz et al., 2018). Apart from being a source of minerals, brine can also be a source of energy, known as ‘osmotic power’, ‘salinity gradient power’ or ‘blue energy’ (Panagopoulos, 2022). There are many brine springs in the high Zagros zone, especially in its southern parts (e.g., Mirzaei et al., 2016; Rezaei, 2019). Moradi et al. (2023) showed that the occurrence of structural phenomena such as folds, faults, joints and fractures on the one hand and appropriate precipitation on the other hand, have created suitable conditions for the development of karst aquifers and emergence of springs in the high Zagros zone. Intrusion of

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saline water into a karst aquifer can significantly change the quality of karst water (Rezaei et al., 2005).

Bibitalkhone spring with an average annual discharge of 2 m^3/s is one of the important karstic springs in the Southwest of Iran. Kalantari et al. (2016) showed that the dominant water types of Bibitalkhone spring are Cl–Na and HCO_3–Ca for dry (approximately 8 months of year) and rainy (about 4 months of year) periods, respectively. While the host rock of the spring is composed of limestone and dolomite, the dominant type of the spring water doesn't match the lithology of the host aquifer. They suggested one of the most important factors is the input of exotic waters from the neighboring karstic catchment which has circulated within a lithology of high limestone purity. Carbonate formations crop out over about 23% of the Zagros Range of Iran (Raeisi and Kowsar, 1997). Formations overlying and underlying the carbonates are composed of marl and/or ypsum, whereas the interlayers are halite in some parts of the Zagros Mountains. Salt domes are in direct contact with karst formations in some of the anticlines. According to Zarei et al. (2013), the potential sources of salinity in eight small permanent brine springs in the Aghar anticline in, with average total dissolved solids of 326.7 g/L, are the salt diapir, evaporation from shallow water table and deep brines.

Unfortunately, there is no published geochemistry information for them in the northern high Zagros zone. The presence of numerous faults along with carbonate deposits has led to extensive karstification and the emergence of springs. There are brine springs in the northern Khorramabad, northern High Zagros zone, and by pumping the water from these springs into built-in ponds a very large amount of edible salt (more than several thousand tons per year) is harvested after evaporation. According to the residents of the region, salt harvesting dates back more than 500 years, and these springs always contained salty water, and fresh water flowed from these springs only in 1977 and 1978. The host rock of the northern Khorram Abad brine springs is Gachsaran Formation (Miocene age) and Cretaceous limestone. In this research for the first-time elemental geochemistry of the northern Khorramabad brine springs has been evaluated and the source of these evaporate deposits is introduced based on the geochemical characteristics of the salt spring deposits and the stratigraphy of the region.

2. Material and methods

To study brine springs in the northern Khorram Abad, two sections, Sar Namak and Robat Namaki, were measured in the High Zagros Zone. Different fieldwork and laboratory methods were used in this research. During the fieldwork studies, the thickness of the Gachsaran and Cretaceous deposits measured and the stratigraphic column of Gachsaran deposits section was drown. Since several faults were observed among the Cretaceous deposits, it was not possible to prepare a stratigraphic column. For the petrography study, 46 thin sections of the host rocks of the brine spring have been prepared. For classification and texture descriptions, the classification schemes of Dunham (1962) were adopted. After petrography studies, 30 samples of evaporate deposits were selected for geochemical analysis to determine how much of certain elements are in a sample by ICP-OES method. Also, 24 and 15 samples were analyzed by XRD and XRF methods, respectively for mineralogy studies and the determination of major and minor minerals.

2.1. Study area

The Zagros basin is an outstanding orogen running from eastern Turkey to the Makran area (Sembroni et al., 2024). In the Late Cretaceous time, orogenic movement caused Zagros basin on the northeastern margin of the Arabian Plate divided into several zone included the High Zagros, Fars's arc, Izeh, Dezful Embayment (Khuzestan province) and Lorestan Zones, from northeast to south respectively (Fig. 1a). The High Zagros zone is a narrow thrust belt, measuring up to 80 km in width, with a NW–SE trend. It is characterized by high mountains, with a maximum elevation of about 3,000 meters in northwest Zagros, and overthrust anticlines that expose deep-lying sedimentary formations (Khadivi et al., 2012). The High Zagros Zone extends continuously from Main Thrust Zagros to Zagros Fault. The sections studied are located in the northwest Zagros zone. Numerous salt-emission areas exist within the Cretaceous and Miocene outcrop in the northern Khorram Abad in the northern High Zagros zone (Fig.1b). The Cretaceous deposit in the Sar Namak section and consists of 120 m of Marl, limestone, sandy limestone and sandstone. In this stratigraphic section, the marly sediments of this formation include of *Orbitolina* sp and rudist debris.

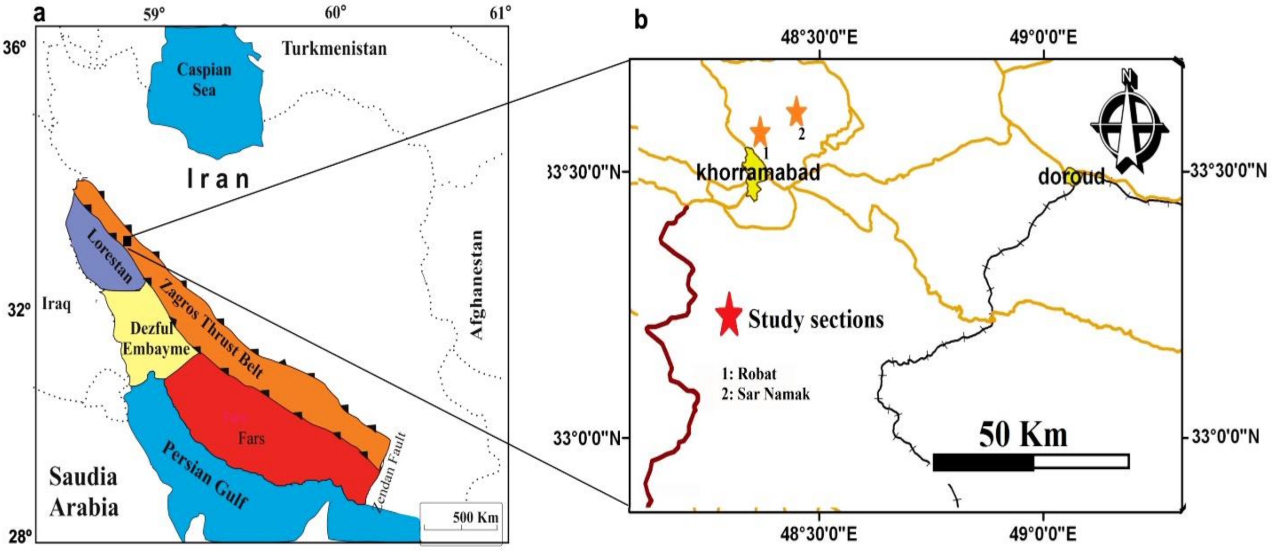

Fig. 1. a) Structhral framework of Zagros Basin (Heydari, 2008); b) location of studied area in the northern Khorram Abad.

The Gachsaran Formation in the Robat Namaki lies conformably on the limestone of the Asmari Formation and its upper boundary is covered. The Asmari Formation in the Robat Namaki section is dated to middle Miocene (Langhian) based on the distribution of the index foraminifera such as *Orbulina* sp. (Roozpeykar et al., 2019). The Gachsaran Formation consists of 146 m limestone, marl, anhydrite and sandstone. Based on the recognized foraminifera, this section is comparable to *Borelis melo- Rotalia-Miogypsina* assemblage zone. The age of the Gachsaran Formation in the studied sections is assigned to the Langhian. The dominant taxa belong to: *Amphistegina* sp., *Borelis melo curdica*, Bryozoair, *Dendritina* sp., *Ditrupa* sp., *Lithothamnium* sp., *Miogypsina* sp., *Neorotalia* sp., Operculina sp. and *Quinqueloqulina* sp. Based on petrography, microfacies and foraminifera content, 3 paleoenvironments, sabkha, lagoon and open marine have been distinguished.

3. Results and discussion

The results of XRD, XRF, ICP and diffraction analysis for the samples from Robat Namaki and Sar Namaki sections are presented in Tables 1, 2, 3, 4, 5, 6 and 7. Samples were analyzed using the XRD determine mineralogical characteristics of evaporates deposits. As illustrated in Table 1; halite was the major mineral, while quartz, anhydrite, gypsum, quartz, dolomite, calcite and cristobalite were the minor ones. The results of chemical analyses indicated that Mg, K, Ba and B did not constitute independent salts. High concentrations of sodium and chloride were observed and confirmed salt purity. The largest source of Cl- in the Earth's crust is the mineral halite in evaporate deposits formed over geologic time by the evaporation of seawater (Zarei, 2013). According to the studies conducted in salt deposits of different ages, the composition of sea water has not changed much since the Cambrian period (Brennan et al., 2004; Li Vigni et al., 2023). The main rare elements in sea salts refer to the relative abundance of some elements in different media. Specifically, the term indicates chemical elements that in natural systems are characterized by concentrations generally lower than 1 mg L^{-1} in solution or 0.1% in solids. The concentration of Li, Zr, Cr, Fe, Cu, Th, Mn and V can reach more than several hundred ppm (Warren, 2006). The amount of Br in seawater is 65 ppm, and as the seawater evaporates, and when halite begins to deposit, this amount increases to about 500 ppm. When the first potassium mineral is deposited, the amount of Br in the brine is around 2300 ppm (Warren, 2006). The amount of Br in the studied evaporates is more than 150 ppm, which indicates the marine origin of the salts in the studied area.

Table 1. Types of minerals that make up the deposits of North Khorram Abad based on XRD analysis. Samples Nos. 2, 9, 10, 16 and 17 are related to table salt deposited in the saltwater springs of the study area.

Samples	**Minerals**	
1	Quartz+ Dolomite+ Calcite	Robat
2	Halite+ Gypsum+ Anhydrite+ Dolomite	
3	Quartz+Gypsum	
4	Gypsum+ Anhydrite+ Cristobalite	
5	Quartze+ Gypsum+ Anhydrite	
6	Gypsum+ Anhydrite+Quartz	
7	Quartz+ Dolomite+ Calcite	
8	Gypsum+ Quartz	
9	Halite+ Gypsum+ Anhydrite+ Dolomite	
10	Halite+ Anhydrite	
11	Calcite+ Dolomite+ Cristobalite	Sar Namak
12	Calcite+ Dolomite+ Cristobalite	
13	Calcite+ Dolomite+ Quartz	
14	Calcite+ Dolomite+ Quartz	
15	Calcite+ Dolomite+ Cristobalite	
16	Halite+ Dolomite+ Gypsum	
17	Halite+ Dolomite+ Gypsum	

Table 2. XRF analysis results of Robat evaporate, values are average weight percent.

	Oxides (w %)									
Samples	**SiO_2**	**Al_2O_3**	**Fe_2O_3**	**Cao**	**K_2O**	**MgO**	**TiO_2**	**MnO**	**P_2O_5**	**BaO**	**SO_3**
R1	30.04	28.78	1.01	18.25	1.24	0.25	0.01	0.02	0.25	1.25	18.9
R2	31.7	28.02	1.2	19.25	0.97	1.41	0.01	0.01	1.28	1.12	15.03
R3	24.68	25.01	0.98	40.10	1.07	0.02	0.02	0.03	0.77	1.02	6.30
R4	27.15	27.15	1.08	26.04	3.45	2.35	0.07	0.71	1.97	1.13	8.90
R5	25.09	21.00	1.04	39.02	3.85	1.02	0.01	0.02	0.85	1.25	6.85
S1	22.25	30.42	1.93	40.26	0.28	2.50	0.05	0.58	0.29	1.20	0.24
S2	18.65	31.45	1.02	42.23	1.28	2.26	0.01	0.01	0.83	1.03	1.23
S3	21.12	32.09	1.14	41.20	0.85	0.93	0.05	0.20	0.23	1.08	1.11
S4	20.84	30.89	1.00	40.89	0.45	1.21	0.01	0.21	0.89	1.11	2.50
S5	14.43	34.59	1.26	42.15	2.14	0.07	0.02	0.01	1.11	1.08	3.14

Table 3. XRD analysis results of Sar Namak evaporate, values are average weight percent.

Samples	**Minerals**
1	Calcite+ Dolomite+ Cristobalite
2	Calcite+ Dolomite+ Cristobalite
3	Calcite+ Dolomite+ Quartz
4	Calcite+ Dolomite+ Quartz
5	Calcite+ Dolomite+ Cristobalite
6	Halite+ Dolomite+ Gypsum
7	Halite+ Dolomite+ Gypsum

Table 4. XRF analysis results of Sar Namak evaporate, values are average weight percent.

	Oxides (w %)									
Samples	**SiO_2**	**Al_2O_3**	**Fe_2O_3**	**Cao**	**K_2O**	**MgO**	**TiO_2**	**MnO**	**P_2O_5**	**BaO**	**SO_3**
S1	22.25	30.42	1.93	40.26	0.28	2.50	0.05	0.58	0.29	1.20	0.24
S2	18.65	31.45	1.02	42.23	1.28	2.26	0.01	0.01	0.83	1.03	1.23
S3	21.12	32.09	1.14	41.20	0.85	0.93	0.05	0.20	0.23	1.08	1.11
S4	20.84	30.89	1.00	40.89	0.45	1.21	0.01	0.21	0.89	1.11	2.50
S5	14.43	34.59	1.26	42.15	2.14	0.07	0.02	0.01	1.11	1.08	3.14

Table 5. The results of ICP analysis of some trace elements of Sar Namak evaporates (values are in ppm). Samples R1 and S1 are related to the salts deposited from the brines.

	Elements (ppm)																
Samples	**As**	**K**	**Rb**	**Y**	**Ni**	**Pb**	**Cr**	**V**	**Br**	**B**	**Zn**	**Cu**	**Sr**	**Na**	**Cl**	**Zr**	**Nb**	**V**
R1	5	420	12	6	12	110	25	15	150	45	112	65	2000	1250	1800	75	3	15
R2	2	210	25	5	30	125	10	25	210	62	62	102	1700	312	362	125	9	25
R3	3	254	10	8	18	950	12	21	168	28	250	85	1500	310	258	248	14	21
R4	9	101	9	2	10	584	7	14	251	41	178	32	752	450	114	158	12	14
R5	7	198	13	1	9	110	21	9	312	78	210	19	970	610	102	236	6	9
S1	5	140	15	4	24	365	10	15	158	98	89	212	652	1120	1870	110	8	15
S2	6	180	21	6	26	215	23	31	89	70	91	67	369	210	240	98	17	31
S3	4	197	18	3	18	89	28	27	215	33	212	98	345	350	110	465	10	27
S4	2	201	14	5	21	146	31	12	111	69	46	115	587	560	212	229	6	12
S5	1	111	9	4	10	851	12	19	124	67	87	189	118	620	310	187	8	19

Table 6. The results of ICP analysis of some trace elements of Sar Namak evaporates.

	Elements (ppm)																
Samples	**As**	**K**	**Rb**	**Y**	**Ni**	**Pb**	**Cr**	**V**	**Br**	**B**	**Zn**	**Cu**	**Sr**	**Na**	**Cl**	**Zr**	**Nb**	**V**
S1	5	140	15	4	24	365	10	15	158	98	89	212	652	1120	1870	110	8	15
S2	6	180	21	6	26	215	23	31	89	70	91	67	369	210	240	98	17	31
S3	4	197	18	3	18	89	28	27	215	33	212	98	345	350	110	465	10	27
S4	2	201	14	5	21	146	31	12	111	69	46	115	587	560	212	229	6	12
S5	1	111	9	4	10	851	12	19	124	67	87	189	118	620	310	187	8	19

Table 7. The results of ICP analysis of some trace elements of North Khorramabad evaporites (values are in ppm). Samples R1 and S1 are related to the salts deposited from the brines.

	Cation and anion (mg/L)								
Samples	**Na^+** mg/L	**K^+** mg/L	**Ca^{2+}** mg/L	**Mg^{2+}** mg/L	**CL^-** mg/L	**F^-** mg/L	**SO_4^{2-}** mg/L	**HCO_3^-** mg/L	**NO_3^-** mg/L
W1	15.21	1.2	108	13.25	103.2	0.58	17.25	208.65	2.9
W2	40.11	0.39	128.96	55.21	98.74	0.28	185	360.14	25.1
W3	9.87	1.14	100.14	20.31	87.36	0.68	25.1	275.14	21.01
W4	98.52	1.12	97.19	19.74	128.21	0.25	55.12	410	2.5
W5	84.01	1.11	98.32	9.4	114.2	0.89	51.21	765	6.3
W6	164000	4.5	2760	641	45000	2.09	40	267000	3.5
W7	19.25	0.36	128.2	25.14	138.02	0.9	56.32	300	5.31
W8	118	0.89	70.36	8.6	118.68	0.56	35.62	345	3.1
W9	61.02	0.34	230.2	2.3	25.63	0.1	55.2	752	3.5
W10	17.25	0.36	111.25	10.25	25.98	0.02	154.02	189.32	3.6
W11	256	0.28	65.32	45.21	100	0.58	68.36	225	12.15
W12	98.02	1.02	115.28	35.21	212.2	0.28	18.58	265.2	9.8
W13	18.36	0.25	56.25	19	25.21	0.36	58.24	369.2	6.3
W14	25.85	0.25	69.58	24.32	189.36	0.25	25.36	30.9.32	3.2
W15	128560	3.2	3652	325	41250	2.02	52	275000	2.4

Chemical composition and quality of groundwater are the result of processes and reactions performed during the formation and density of water in the atmosphere until it appears on the surface based on Todd (1980) definition. In general, groundwater salinity increases with increasing depth and geochemical sequences in groundwater systems are as the shallow carbonates waters change to chloride water in great depth (Todd, 1980). Halite dissolution brines typically have molar Na/Cl ratios of about 1 and deep-basin brines, in contrast, exhibit ratios of less than 1 (Richter and Kreitler, 1986). The results show that water of brine springs in the northern Khorram Abad is enriched in sodium chloride. The results of ICP analysis of the water samples of the studied area showed (Table 5) that the Na/Cl ratios are greater than 1. Therefore, the origin of the salts deposited in the studied area can be attributed to older formations in the area. It is worth mentioning that no evidence of halite has been

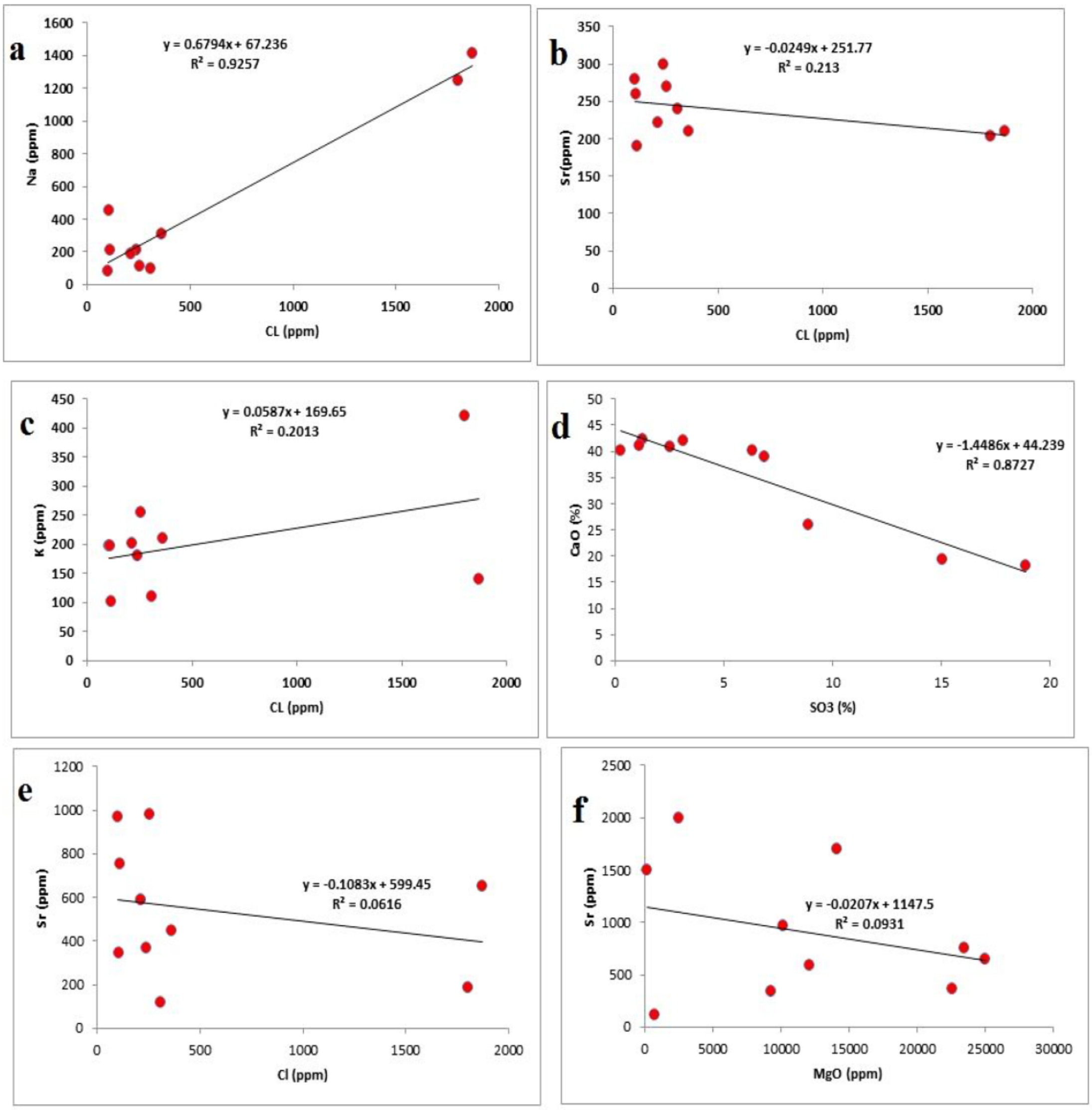

Fig. 2. The ratio of different elements according to geochemical data in Rabat section, a) Cl versus Na; b) Cl versus Sr; c) Positive correlation between Cl and K; d) The graph of changes of CaO compared to SO_3 and their close correlation; e) The ratio of element Sr to Cl and the negative slope of the graph; f) Negative slope of the graph of changes of magnesium against strontium.

seen in Gachsaran Formation, while the Gutnia Formation contains salty deposits (Aoudah et al., 2024). Gotnia Formation that formed under conditions of low sea surface I the late Jurassic age and as a result of the marine recession was contributed to filling the Gotnia basin mainly from evaporite sediments (Sharland et al., 2021). Cl versus sodium and bicarbonate diagrams shows a high correlation between Cl and sodium which indicates of the common origin of the two ions (Fig. 2a). The Na/Cl ratio is less than 1 in which indicates a deep-basin origin. Also, the increasing changes of Sr compared to the decreasing trend of Cl (Fig. 2b) in the studied samples show a good agreement and its high level indicates marine conditions (Warren, 2006). Such a ratio can be observed in Mediterranean deposits after the Messinian event (Boschetti et al., 2013). Potassium-containing salts are found in some marine evaporate sediments and include sylvite (KCl), carnallite ($KMgCl_3$, $6K_2O$) and kainite ($KMgClSo_4$, $3H_2O$) (Warren, 2006). The presence of K with an average of 201.2 ppm in the analyzed samples and the positive correlation between the elements Cl and potassium K (Fig. 3c) are other evidences of the marine origin of the studied salts (Warren, 2006). The correlation between the two elements CaO and SO_3 shows that almost part of the calcium in the environment is left in the form of $CaSO_4$ (Fig. 3d). The regular trend of changes of CaO and SO_3 shows that there is no significant difference between the anhydrites of Gachsaran Formation in terms of composition, and on the other hand, the matching trend of changes of CaO and SO_3 indicates the participation of Ca element in gypsum structure. The amount of strontium element Sr in the studied samples varies between 118 and 2000 ppm (average 899.3 ppm). Based on the studies of Usdowski (1967), the amount of strontium in the first gypsum sediment is about 1400 ppm. When gypsum is converted to anhydrite at 60°C, the amount of strontium remains unchanged. Gypsum receives up to 0.957% and anhydrite up to 1.27% of strontium, and after that, in larger amounts of strontium, this element is easily replaced by Ca^{+2} and precipitates as celestite ($SrSO_4$) (Adabi and Rao, 1991). The decrease in strontium content indicates the formation of secondary gypsum (Usdowski, 1967). The increasing changes of strontium compared to the decreasing trend of chlorine (the negative slope of the graph) show a good match, and the high amount of strontium indicates marine conditions in the sedimentation environment (Fig. 2e). The changes in decrease of magnesium and increase of strontium (Fig. 2f) indicate the marine conditions of the sedimentation basin at the base of Gachsaran Formation. The origin of these salts can be considered as Upper Jurassic sediments (Gutnia Formation) in the region, which have reached the surface of the earth due to faulting. In contrast, brine spring in Khuzestan, south of study area, were sourced from the Gachsaran Formation (Chitsazan et al., 2018). The assessments show that except for Ramshir and Garu brine spring cropping out in Mishan and Asmari Formations, respectively most of other brine springs is located in the evaporative Ghachsaran Formation (Rezaei et al., 2019). It should be noted that Bahroudi and Koyi (2004) have attributed the origin of the evaporations of the Gachsaran Formation to the dissolution and redeposition of the deposits of the Hormuz Formation. Unlike Lorestan and Khorramabad, there are no deposits of the Gotnia Formation in the stratigraphic sequence of Khuzestan.

4. Conclusion

Abundant brine springs have been reported in the High Zagros zone. The present work is the first regional-scale reconnaissance of 2 brine springs located in the northern Khorram Abad, High Zagros zone, Robat and Sar Nasmak brine water. Geologically, Robat brine springs hasemerged in the Lower Miocene Gachsaran Formation. Field observations show that the emergence of springs follows in the lower contact contact of the Gachsaran Formation with Asmari Formation. Sar Namak spring has emerged in the fault zone between limestone of the Cretaceous deposits. According to the results, the dominant mineral of study area was NaCl, which was different from the common types of elements found in Gachsaran Formation. The results of the XRD analysis indicated that halite was the major mineral, while quartz, anhydrite and dolomite were the minor minerals. The ratios of different elements according to geochemical data in Rabat section, e.g., Cl versus Na, Cl versus Sr, Cl versus K, indicate the origin of deep water for the salts in North Khorram Abad. Probably, with reference to the stratigraphic history of the studied area the origin of these salts is the deposits of Gutnia Formation of Late Jurassic age.

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