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Journal of Stratigraphy and Sedimentology Researches

E-ISSN: 2423-8007

Vol. 41, Issue 4, No. 101, 2025, 43-57

Received: 13.09.2025

Accepted: 14.10.2025

Research Paper

Lithological influence on water quality evolution: A case study of the Zuzan Plain, northeast Iran

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Abstract:

This study investigates the influence of lithological characteristics on the evolution of water quality in the Zuzan Plain, Northeast Iran, a region characterized by limited water resources. A total of 23 water samples were collected from various sources across the area. Physical parameters, including acidity (between 7.2 and 8.7), dissolved solids (1180–7415 mg/l), electrical conductivity (ranging from 2100 to 12,210 microsiemens/cm), and salinity, were measured on-site using an Extech model multimeter. Water hardness (100–1960 mg/lCaCO₃) was analyzed at the laboratory. Statistics software was used to analyze the correlations between elements and their relationship with the region's lithology. Water facies were identified using RockWorks 16 software. The Piper and Stiff diagrams revealed that most samples exhibited sodic facies with chloride type, except for two samples, which displayed sodic facies with sulfate type. Furthermore, the Gibbs diagram was employed to distinguish the dominant hydrogeochemical processes, indicating that both evaporation and rock-water interactions strongly influence the groundwater composition. To better understand spatial controls, a groundwater flow direction map was prepared, clarifying the distribution of water facies in relation to regional hydrogeology. Cluster analysis and principal component analysis of the water samples revealed two main components influencing the source variations. The presence of acidic igneous units, limestone, sandstone, conglomerate, siltstone, and shale in the region suggests their significant role in controlling the chemical composition of the water. Limestone, due to its higher solubility, is identified as the primary regulator of water quality in the Zuzan Plain, while igneous and sedimentary rocks exert secondary influences.

Keywords: Water resources, lithology, sedimentation, Zuzan plain, Iran

Introduction

Water plays a crucial role from both a health and economic perspective. Many health problems in developing countries are attributed to the lack of access to clean drinking water. As the cornerstone of sustainable development, human health is directly linked to the availability of safe drinking water; without it, the well-being of society cannot be achieved. Economically, water is essential for industrial activities and agricultural productivity. Therefore, beyond the quantity of available water, the quality and pollution levels of water

resources are critical factors that impact water supply. Assessing the quality and pollution levels of surface and groundwater, particularly in areas where groundwater is used for drinking, is of paramount importance. Water quality is influenced by the ions it contains, which are primarily controlled by the region's lithology and geological development (Naseem et al. 2010; Deocampo and Jones 2014; Cuoco et al. 2015; Chitsazan et al. 2018; Geris et al. 2022; Ahmed and Singh 2022; Azari and Tabari, 2024).

In this regard, monitoring and zoning water resources

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Javanbakht M. Moradian F. Raftari Sh. and Pourjavad H. (2025). Lithological influence on water quality evolution: A case study of the Zuzan Plain, northeast Iran. *Journal of Stratigraphy and Sedimentology Researches*, 41(4):43-57. <https://doi.org/10.22108/jssr.2025.146633.1321>



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<https://doi.org/10.22108/jssr.2025.146633.1321>

should be considered essential components of planning. The quality of water is impacted by various environmental and anthropogenic factors (Rabah et al. 2011; Tabari and Eilbeigy 2017; Ramya and Elango 2022). The spatial distribution of water quality is closely related to the regional structure, and analyzing how these variables evolve over consecutive years helps clarify the processes driving changes (Koponen et al. 2002; Zhao et al. 2017; Nemati et al. 2023).

The understanding of water quality and its evolution is critical for proper water resource management and future planning. Various methods and models have been proposed to estimate water quality parameters (Banerjee and Sikdar 2022). The chemical composition of water in closed basins is primarily influenced by the lithology of the rocks and sediments exposed to weathering processes such as evaporation, gypsum dissolution, and silicate weathering (Hardie and Eugster 1970; Jones et al. 1977; Hardie et al. 1978; Eugster and Jones 1979; Eugster 1980; Deacampo and Jones 2014; Rezende-Filho et al. 2015; Li et al. 2023).

In particular, evaporation processes increase the concentration of solutes in water bodies, often leading to higher salinity levels. Gypsum dissolution refers to the process where gypsum (CaSO_4) dissolves in water, contributing sulfate ions (SO_4^{2-}) and influencing water chemistry. Silicate weathering involves the breakdown of silicate minerals, which releases ions like calcium (Ca^{2+}) and bicarbonates (HCO_3^-) into water, affecting its pH and hardness.

Ensuring the regulation of pollutant concentrations in water resources is crucial for protecting public health. This often requires extensive chemical testing of water sources (Pulliainen et al. 2000, 2001; Nanbakhsh 2003). Water quality is also affected by river flow patterns (Qiao et al. 2018), with evolution interactions between river systems and surrounding geological formations further influencing water chemistry (Gipperth and Elmgren 2005; Mishra et al. 2024).

In the Zuzan Plain, providing adequate drinking water has become increasingly challenging due to the growing demand from the industrial sector, particularly iron processing in the Sangan region. This creates a critical need to evaluate the quality of water resources in the Zuzan Plain and assess how water quality changes over time. Despite the significance of this issue, a research gap remains regarding the interplay between lithological characteristics and the evolution of water quality in the region. This study aims to bridge this gap by conducting a detailed investigation of water quality in the Zuzan Plain and analyzing the processes that drive changes in water quality over time. By highlighting the relationship between lithology and water quality evolution, this research seeks to provide valuable insights into the factors affecting water resource management and sustainability in the region.

The study's primary objectives include assessing the current quality of water resources in the Zuzan Plain, analyzing the impact of lithological features on the evolution of water quality, identifying key factors that affect changes in water quality over time, and, based on the study's findings, offering recommendations for effective water resource management and conservation strategies.

Geological setting Study Area

The Zuzan region is located in the southeast of Razavi

Khorasan Province, between longitude $59^\circ 30'$ to $60^\circ 00'$ east and latitude $34^\circ 00'$ to $34^\circ 30'$ north (Figure 1). Based on Iran's climatic conditions, Zuzan falls in the desert and semi-desert region, characterized by cold, dry winters with minimal precipitation and hot summers with strong winds and dust. This area experiences limited rainfall throughout the year, typically restricted to the winter and early spring seasons, with total annual rainfall not exceeding 300 mm. Topographically, the central parts of the region consist of relatively high altitudes that decrease towards the edges, forming relatively flat plains in the eastern and western parts.

Geology of the region

Understanding the geology of closed basins is essential for comprehending the chemistry of water in its initial phase. Both surface and groundwater serve as sources of ions that ultimately form salts deposited within the basin. Acidic waters induce the chemical weathering of surface rocks in the catchment area, and this weathering process varies depending on the types of rocks present, resulting in water with different cations and anions (Erfanian et al. 2020; Poaty Plante et al. 2021). The geology of Zuzan consists of rock formations ranging from the Paleozoic to Quaternary, with notable stratigraphic gaps.

To summarize, the geological map (Figure 1) indicates that the rock masses around this plain can be divided into two zones. The first zone comprises units extending from the north to the east, while the second zone includes units starting from the north and extending to the west and south. The investigation reveals two primary lithological units in the eastern to northeastern parts: sedimentary deposits such as limestone, dolomite, conglomerate, sandstone, and shale, which account for about 50% of the units, and volcanic units, including dacite, andesite, and basalt, comprising 26% of the region's rock units.

In the western to southern areas, three main lithological units are identified, covering the largest area: medium to basic igneous units (andesite and basalt), clastic units containing fragments of these rocks (sandstone and conglomerate), which make up about 51.5% of the area, and acidic igneous units (granite and microgranite), covering about 15.5%. Additionally, there are carbonate, detrital, and evaporite sedimentary deposits occupying 7% of the area.

Impact of human activities on water quality in the Zuzan region

In addition to natural and lithological factors, human activities also play a significant role in altering the water quality of the Zuzan Plain. One of the most prominent anthropogenic influences in the region is the large-scale iron ore extraction and processing in the Sangan area, which has expanded considerably in recent years. Mining operations, the discharge of industrial waste, the use of chemicals during ore beneficiation, and increased water withdrawal for industrial purposes can contribute to elevated concentrations of specific ions—such as heavy metals, sulfates, and chlorides—in both surface and groundwater. To minimize the potential confounding effects of these activities, this study aimed to select sampling points away from direct industrial zones or to compare samples from lithologically similar areas at varying distances from human activities (Tavakoli and Ghanbari 2023).



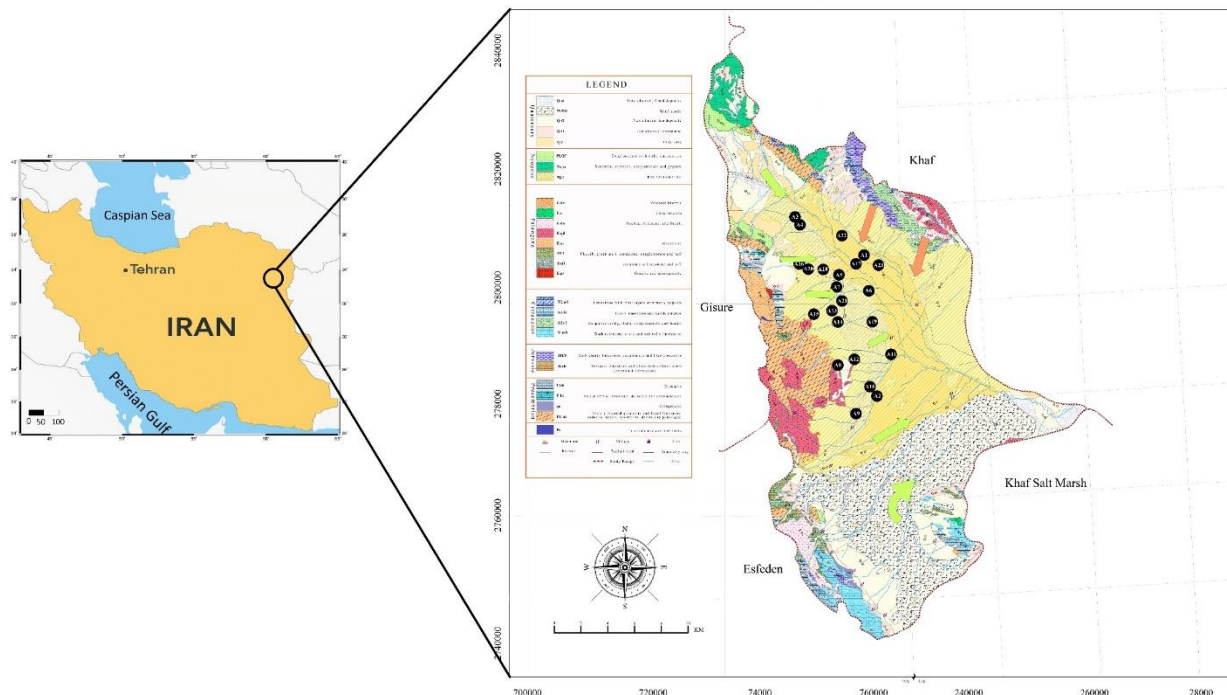


Fig 1- Geographical location of Zuzan Plain in northeast Iran, sampled points in the area, and water flow direction

Material & Methods

Initially, a geological map of the area was created to identify access routes and suitable sampling locations. After pinpointing the sampling points on the map, 23 samples were collected from wells and aqueducts in the study area to assess the quality of the water sources (Fig 1). It should be noted that the sources available for sampling are wells and aqueducts in the region. Therefore, 23 wells or aqueducts were available for sampling (containing water). In this study, all water samples were collected during the same season (Spring 2023) to minimize the impact of seasonal fluctuations on water quality. Conducting the sampling within a uniform time frame (during a week and between 10:00 and 14:00 each day) ensured that variations in temperature, precipitation, and evaporation had minimal influence on the results, allowing for a more accurate attribution of the observed hydrochemical patterns to the region's lithological and geological characteristics. Consequently, the variations in ion composition across the samples are primarily related to geological factors rather than seasonal effects. The coordinates of each sampling site were recorded using GPS. Before collecting the samples, the sampling bottles were thoroughly rinsed with site water, and two samples were collected from each site. Physical parameters such as acidity, dissolved solids, electrical conductivity, and salinity were measured at the sampling site

using an Extech multimeter. The samples were then transported to the environmental laboratory at Islamic Azad University, Mashhad, where hardness was analyzed. Other parameters were determined at the Zarazma laboratory in Tehran. It should be noted that in order to maintain the standard of the measured parameters, the water sampling instructions of Iranian publication number 274 were used, which attempted to take two containers of each water sample, reduce the pH of one sample with acid to minimize changes in elements, even heavy metals, and then transport both samples to the laboratory at a temperature of less than 3 degrees. SPSS 22 software was used to analyze the correlation between the elements and their relationship to the region's lithology, while RockWorks 16 software was used to determine the water facies of the area (Table 1). Future studies involving multi-seasonal or long-term datasets could more accurately assess temporal evolution in water quality. In addition to standard sampling and laboratory analyses, a groundwater flow direction map was prepared using well data and topographic information. This map was critical for visualizing groundwater movement and interpreting the spatial evolution of hydrochemical facies. Moreover, the Gibbs diagram was applied to evaluate the relative roles of precipitation, evaporation, and rock weathering in controlling groundwater chemistry.

Table 1- Physical parameters of the sampled points

sample	X	Y	PH	T.D.S (mg/l)	EC (μ s/cm)	TH (mg/lCaCo3)	Ca ⁺² (mg/l)	Na ⁺ (mg/l)	Mg ⁺² (mg/l)	K ⁺ (mg/l)	HCO ₃ ⁻ (mg/l)	CO ₃ ⁻² (mg/l)	Cl ⁻ (mg/l)	SO ₄ ⁻⁴ (mg/l)
A1	767430	3808025	7/9	6678	10600	1120	10	82	12.4	4.6	3.2	0	80	26.5
A2	767418	3780156	7/7	5405	8580	920	9.4	67	9	2.9	2.7	0	52	33.8
A3	755424	3816496	8/1	6073/2	9640	1360	13	66	14.2	4.4	2.8	0	69	26.2
A4	755942	3814875	8	5556/6	8820	900	10	67.9	8	3.2	2.1	0	56	31.3
A5	764029	3805083	8	1341/9	2130	180	1.5	18.2	2.1	0	5.6	0	7.5	9.8
A6	770026	3801623	8	6898/5	10950	1040	13.5	84	7.3	6.7	3.4	0	78	30.6
A7	764600	3803876	8/2	3679/2	5840	560	6.3	46.4	4.9	1.7	3	0.3	33.5	22.7
A8	759560	3789189	8/5	4195/8	6660	500	4.2	54.8	5.8	2.2	4	0.5	43.5	19.2
A9	762486	3781308	7/7	6766/2	10740	1440	13.6	74	15.2	6.2	3	0	80	27
A10	767706	3781713	7/9	4473	7100	740	10.7	54.2	4.1	3.1	2.5	0	40	29.8
A11	773830	3789553	8/3	2986/2	4740	380	4.5	40	3.1	0.4	3	0.3	30.6	14.3
A12	763469	3788745	8	7415/1	11770	1960	25.3	73	13.9	6.3	2	0	84	33
A13	763141	3798970	8/4	2375/1	3770	260	2.8	33.5	2.4	0	3.4	0.4	24	11
A14	764199	3796933	8/7	1323	2100	100	1.1	19.6	0.9	0	4.7	0.6	10	6.4
A15	753829	3797390	8/2	2847/6	4520	640	6	32.5	6.8	0.5	2.3	0.2	32.5	11
A16	755500	3807950	8/3	4830	9680	680	5.2	80	8.4	3.2	2.3	0.4	75	19.1
A17	766600	3807950	8/4	6100	12210	620	5.6	108	6.8	3.6	1.8	0.8	102.5	20.4
A18	760450	3806400	8/2	4450	8210	600	5.6	70	6.4	3	2.9	0	62.60	20
A19	769150	3796000	7/8	1180	2950	155	1.8	26.4	1.3	0	5.1	0	12.50	11.9
A20	757350	3807000	8/4	1650	3010	190	1.8	26.5	2	0	2.6	1.2	16.5	10.1
A21	764250	3799800	8/6	1700	3400	115	0.6	31.7	1.7	0	3.8	1.6	9.2	19.4
A22	764000	3812800	8/1	3280	6550	960	8.7	46	10.5	0.8	3/3	0	42.5	20.8
A23	770300	3806350	7/2	4930	9860	1260	8	72	17.2	4.8	3/6	0	71	28

Results & Discussion

The Piper plot was used to determine the water type (Figure 2). The investigations show that the samples from the Zuzan Plain predominantly fall into the chloride type and sodic facies. In this classification, waters are divided into three types of magnesia, calcium, and sodic facies based on cations, and three types of bicarbonate, sulfate, and chloride based on anions (Table 2). Examination of the cations and anions and their comparison in the diagram (Piper 1944) indicated that all samples display a sodium-bearing facies and a chloride type, except for samples A5 and A21, which exhibit a sodium-bearing facies and a sulfate type and are almost in the middle of the plain. A closer examination of samples A5 and A21, which differ from the majority of samples by exhibiting a sulfate-sodic facies instead of a chloride-sodic type, suggests

that this deviation is likely due to specific lithological features and localized hydrogeological conditions. Both samples are located in the central part of the Zuzan Plain, an area characterized by the presence of evaporitic formations such as gypsum (CaSO₄) and soft sedimentary rocks like marl and shale. The dissolution of gypsum-rich layers contributes to elevated sulfate concentrations in groundwater, thus shifting the facies toward the sulfate type. Additionally, local hydrogeological factors—such as reduced groundwater flow or prolonged water-rock interaction time—may further intensify these processes. Consequently, the combined effect of lithology and site-specific hydrogeological conditions appears to be the most plausible explanation for the observed deviation in facies in these two samples.



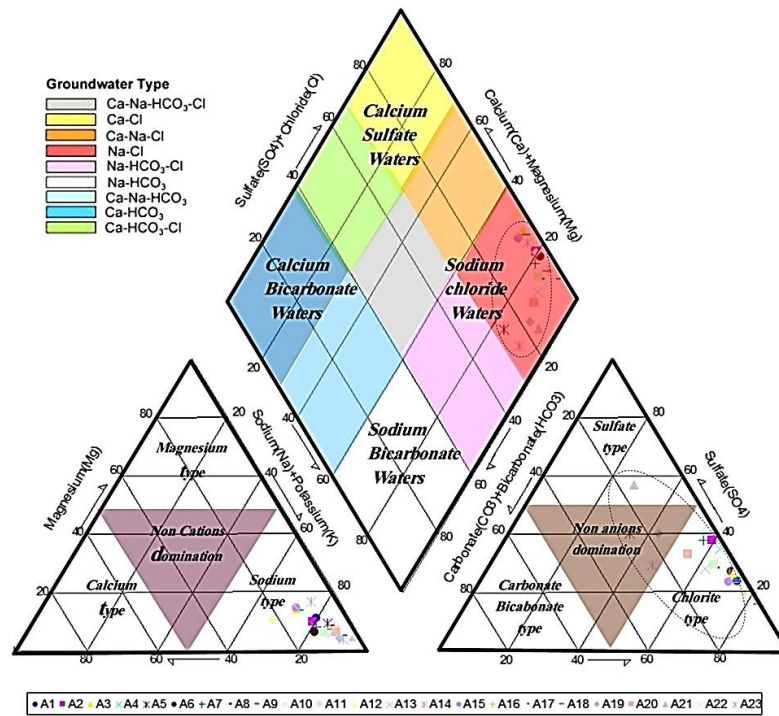


Fig 2- Piper diagram of the collected samples of the studied area (Piper 1944)

Table 2- Type and facies of water in the Zuzan region

Sample number	Concentration of anion	Concentration of cations	Water type	Water facies	Type and water	Type and face development
A1	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A2	Cl > SO4 > HCO3	Na+K > Ca > Mg	chloride	sodic	chloride sodic	Transitional development
A3	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A4	Cl > SO4 > HCO3	Na+K > Ca > Mg	chloride	sodic	chloride sodic	Transitional development
A5	SO4 > Cl > HCO3	Na+K > Mg > Ca	sulfate	sodic	sulfate sodic	Transitional development
A6	Cl > SO4 > HCO3	Na+K > Ca > Mg	chloride	sodic	chloride sodic	Transitional development
A7	Cl > SO4 > HCO3	Na+K > Ca > Mg	chloride	sodic	chloride sodic	Transitional development
A8	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A9	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A10	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A11	Cl > SO4 > HCO3	Na+K > Ca > Mg	chloride	sodic	chloride sodic	Transitional development
A12	Cl > SO4 > HCO3	Na+K > Ca > Mg	chloride	sodic	chloride sodic	Transitional development
A13	Cl > SO4 > HCO3	Na+K > Ca > Mg	chloride	sodic	chloride sodic	Transitional development
A14	Cl > SO4 > HCO3	Na+K > Ca > Mg	chloride	sodic	chloride sodic	Transitional development
A15	Cl > SO4 > HCO3	Na+K > Ca > Mg	chloride	sodic	chloride sodic	Transitional development
A16	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A17	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A18	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A19	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A20	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A21	SO4 > Cl > HCO3	Na+K > Mg > Ca	sulfate	sodic	sulfate sodic	Transitional development
A22	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development
A23	Cl > SO4 > HCO3	Na+K > Mg > Ca	chloride	sodic	chloride sodic	Transitional development

Composition and hydrogeochemical changes of water in the Zuzan region

The chemical composition of water depends on the salts

present in the incoming water, in the atmospheric water, and in the chemical weathering reactions (Eugster and Jones 1979; Eugster 1980). The initial deposition of relatively insoluble

minerals, such as alkaline earth carbonates (low magnesium calcite, high magnesium calcite, and aragonite) and gypsum, is the basic stage that controls the development of brine (Hardie and Eugster 1970; Warren 2010; Thivya et al. 2013).

According to the studies carried out, the electrical conductivity of the waters in the Zuzan region ranges from 2100 to 12,210 microsiemens/cm, while the pH value of the samples varies between 7.2 and 8.7. The waters of the studied area are rich in sodium and chloride ions. Based on the results regarding the anions and cations in the samples, the most abundant anions in the waters of the region are $Cl^- > SO_4^{2-} > HCO_3^- > CO_3^{2-}$, while the most abundant cations are $Na^+ > Ca^{2+} > Mg^{2+} > K^+$.

The processes responsible for the evolution of water entering the concentrated aquifers in a closed basin include continuous evaporation, simultaneous recirculation with the deposition of previously formed minerals, diagenetic reactions, exchange with pore fluids, sulfate generation, and ion exchange (Drever and Smith 1978; Devito et al. 2000).

The quality of water resources is related to the type of rock units around the study area and their interaction with the available water. The type of geological formations around the catchment area is considered one of the most important factors in the development of water resources. Chemical weathering of different rocks produces different cations and anions as they react with the incoming water, resulting in variations in the original solution composition (Eugster and Jones 1979; Eugster 1980; Deocampo and Jones 2014). Rocks and sediments in the Zuzan Plain include sediments (7.5%) and sedimentary rocks (51.5%), igneous rocks (15.5%), and a few metamorphic rocks (0.6%). Based on the research of Deocampo and Jones (2014), it appears that the water entering the basin has certain characteristics related to each of these rock types (Fig 3). Small changes in the weather over different periods of time affect surface water and groundwater, altering the flow of incoming water and subsequently influencing the groundwater level, which, in turn, causes changes in the chemical composition of the water.

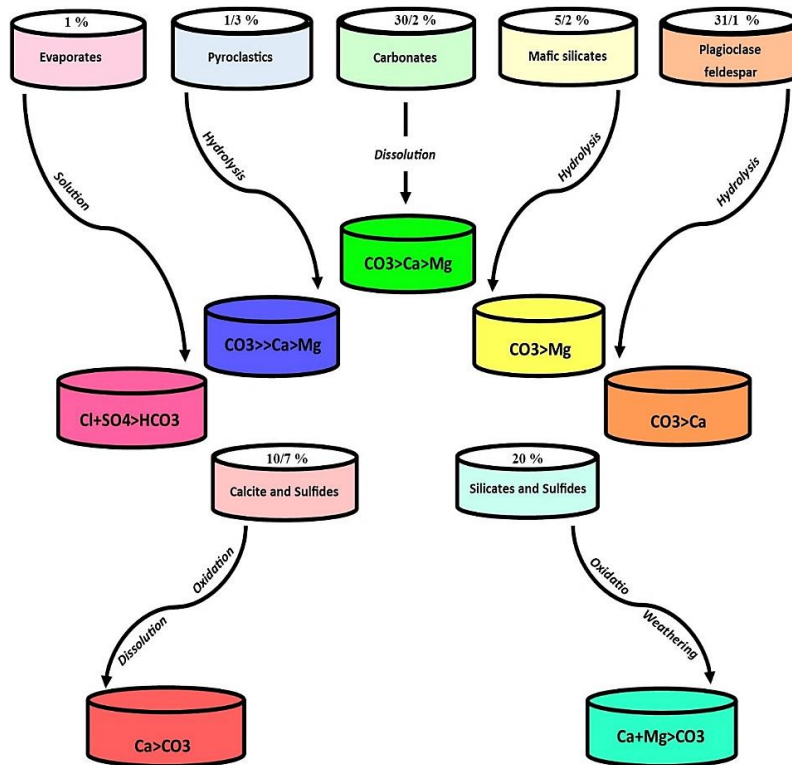


Fig 3- The relationship between the origin and composition of saline water, adapted from Deocampo and Jones (2014). The percentage of rock units and the trends in the study area are marked with different colors. It should be noted that first, the percentage of each unit was determined in the GIS software environment, and then, with the help of rock geochemistry data (Geological Survey of Iran), the percentage of each unit in this division was determined. Then, the information obtained from petrological studies and thin rock sections (Raftari 2017) determined the percentage of each unit.

Based on the effects of the predominant formations in the region on groundwater, it can be concluded that acidic igneous units, limestone, sandstone, conglomerate, siltstone, and shale are the most abundant units in the region and therefore control

the chemical composition of the water. However, since the solubility of limestone is greater, these rocks can be considered the primary controllers of the chemical composition of the water resources in the Zuzan Plain. To further explore the

relationships among major ions and their controlling factors, Principal Component Analysis (PCA) and Cluster Analysis (CA) were applied (Rezaei and Naderi 2024). The PCA identified two principal components: PC1, which included Ca, Mg, K, CO₃, and HCO₃, reflected the influence of carbonate lithology; and PC2, associated with Na and Cl, represented the impact of evaporation and weathering of igneous rocks. The dendrogram resulting from CA also revealed two major clusters, corroborating the PCA results. These findings reinforce the strong dependence of water chemistry on regional lithological variations (Fig 4a). The diagram in Figure (4b) shows that from the perspective of the collected samples, they are divided into two main distinct clusters, Cluster 1 includes samples such as A5, A13, A14, A19, A20, A21 - and samples such as A7, A8, A11, A15, A18, A22 - waters influenced by alluvial and evaporation, and Cluster 2 includes A1, A2, A3, A4, A6, A9, A10, A12, A16, A17, A23 - more evolved waters influenced by water-rock interaction.

Alluvial layers (such as silt, sand, marl, and other fine-grained sediments) play an important role in determining groundwater quality. These layers can affect the chemical composition of water through various geochemical processes. Alluvial layers usually contain minerals that are capable of ion exchange with water. This process can cause changes in the concentration of ions such as Na⁺, Cl⁻, and Ca²⁺. Especially in areas with clay alluvial deposits, these layers can increase the concentration of sodium and chloride in the water, which leads to increased salinity and water hardness. Alluvial layers can play an important role in evaporation processes. In areas with high evaporation, these layers can concentrate ions such as SO₄²⁻ and Cl⁻, increasing water hardness. In areas where evaporation is excessive, alluvial layers can play an important role in enriching water with evaporative ions.

The Gibbs diagram (Fig 5) was used to investigate the origin of the elements. Most of the points are located in the Rock Dominance zone. This shows that the ionic composition of the waters is mainly influenced by the dissolution of the rocks of the water-bearing formation (such as carbonates, silicates, or evaporates). That is, the reaction process of water with the minerals of the bedrock is the main factor determining the chemical composition of the water. A number of samples are located on the border of Rock and Evaporation Dominance. This could indicate that in parts of the region, in addition to the dissolution of minerals, the evaporation process also plays a role. Therefore, considering that the dominant elemental composition is in the sodium chloride range, it can be stated that the dissolution of rocks and the evolutionary process on the one hand and the evaporation process, especially in the central and lower parts of the plain, have probably been influential. These reasons confirm the two main separated

clusters (Fig 4).

Evolutionary geochemical pathway of the water resources of the Zuzan Plain

The processes that cause the evolution of incoming water in a region and its transformation into saline water include continuous evaporation, recirculation concurrent with the deposition of previously formed minerals, diagenetic reactions, exchange with pore fluids, sulfate regeneration, and ion exchange (Warren 2006, 2010). The initial deposition of relatively insoluble minerals, such as alkaline earth carbonates (low magnesium calcite, permagnesium calcite, and aragonite) and gypsum, is a crucial step in controlling the development of water salinization (Hardie and Eugster 1970). Many studies have been conducted on the evolution of saline water, including the works of Garless and Mackenzie (1967), Eugster and Jones (1979), Fayazi, Lak and Nakhaei (2007), Tchamako et al. (2013), and Erfanian et al. (2020). According to the studies of Hardie and Eugster (1970) on the evolution of saline water, it has been shown that three main types of inflowing water influence the chemical evolution of water in closed basins and play a role in the determination of saline water.

To investigate the evolutionary process of the saline waters of the Zuzan Plain, the following results were obtained based on the analysis of the samples and their comparison with the studies of Hardy and Eugster (1970). These samples follow processes I, II, and III. According to Deocampo and Jones (2014), most of the water resources in the region follow process I, while a smaller percentage undergo processes II and III. To comprehensively analyze the diagram of Hardy and Eugster (1970) (Fig 6), the type of existing water resources should first be determined so that the past development process and future trend of these resources can be identified. To determine the type of water (saline water), the amounts of cations and anions were first calculated in milliequivalents per liter. Then, the molar percentage of cations and anions was calculated separately, so that the sum of the cations is 100% and the sum of the anions is also 100%. Ions whose quantities are less than 5% are not considered in the designation. Ions between 5% and 25% are written in brackets, and values above 25% are considered (Hardie and Eugster 1970).

According to the results, the predominant type in the waters of the Zuzan Plain is Na-Cl-SO₄, which is the result of the influence of geological units and the evolutionary process. For anions, there are eight samples of Cl-SO₄ type, five samples of Cl-(SO₄) type, one sample of Cl-SO₄-HCO₃ type, and nine samples of Cl-SO₄-(HCO₃) type. For cations, there are also 17 samples of Na-(Mg)-(Ca) type, two samples of Na-(Mg)-(Ca)-(K) type, two samples of Na-(Ca) type, and two samples of Na-(Mg) type (Table 3).

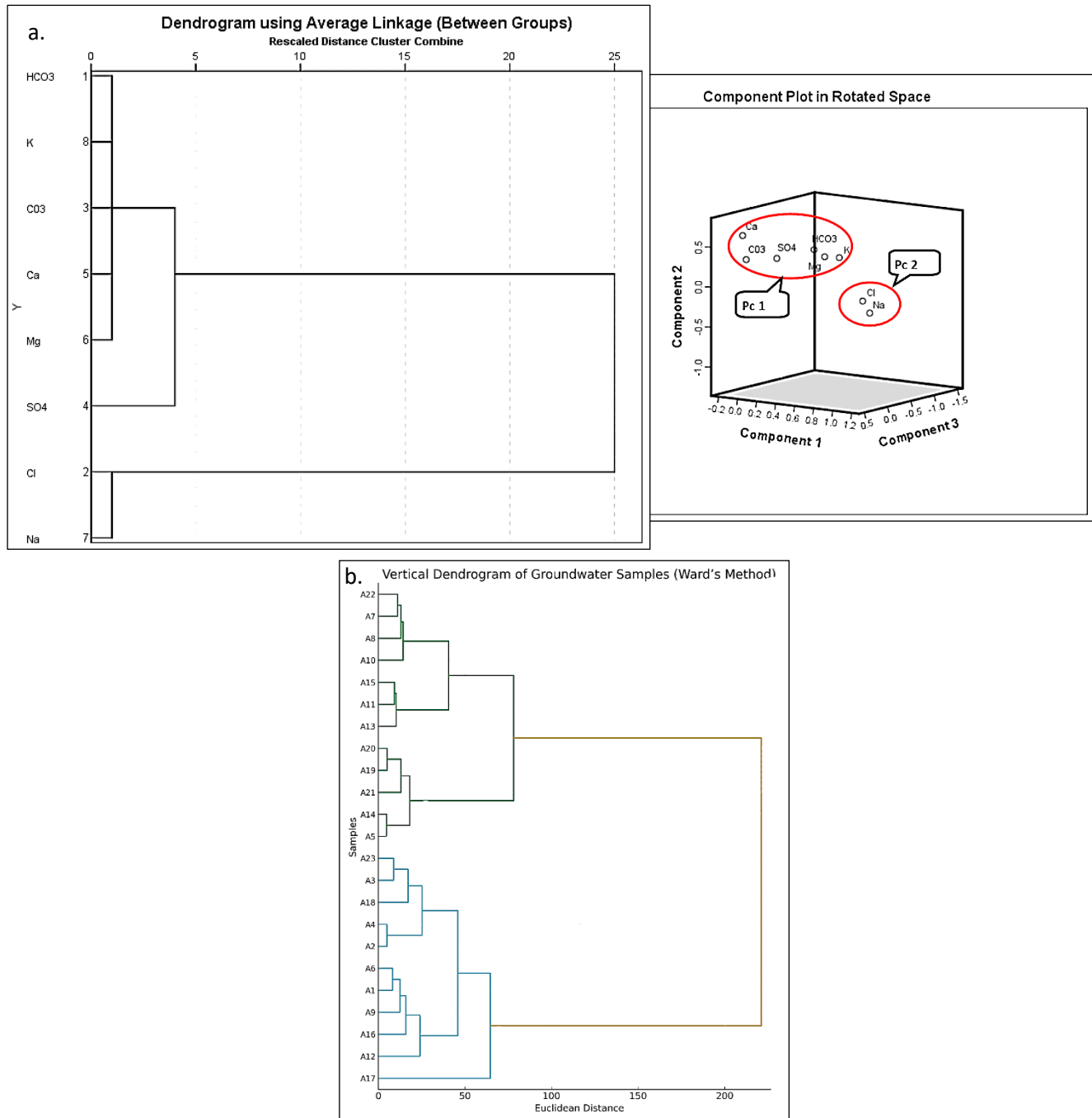


Fig 4- Hierarchical cluster dendrogram of major ions (Mg, Ca, K, CO₃, HCO₃, Cl, Na) in the water samples of the Zuzan Plain. The diagram reveals two primary clusters: one dominated by carbonate-related ions and another by salinity-related ions, indicating distinct lithological influences (a.b.).

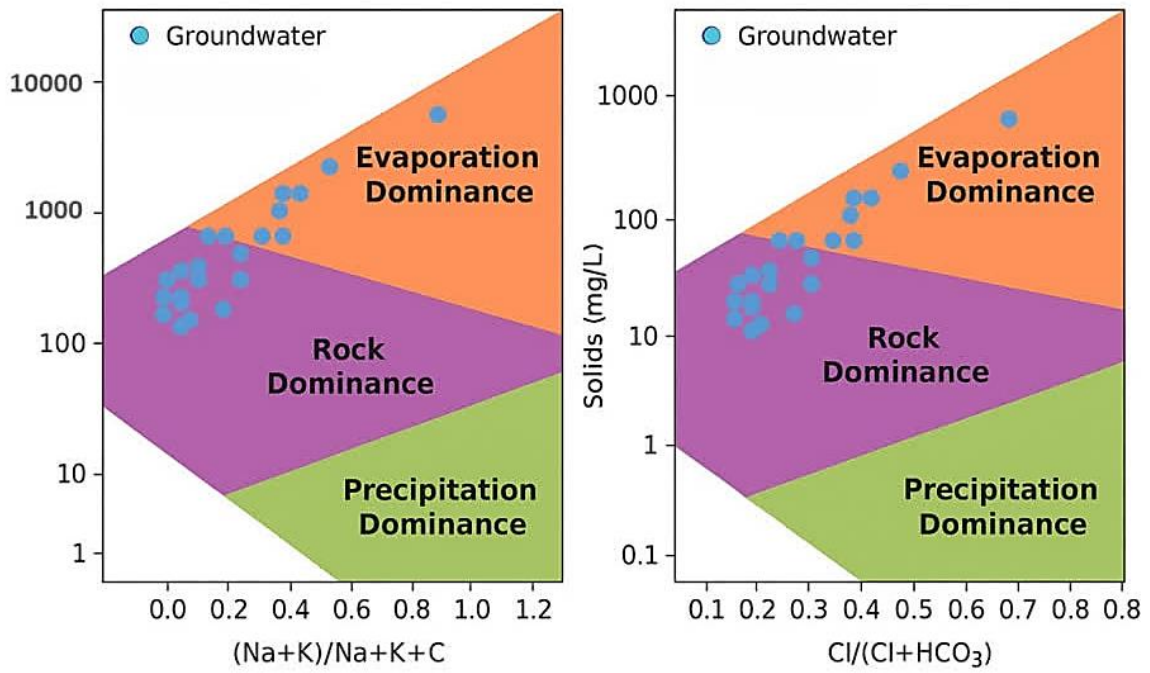


Fig 5- Gibbs diagram in relation to the origin of elements in the study area

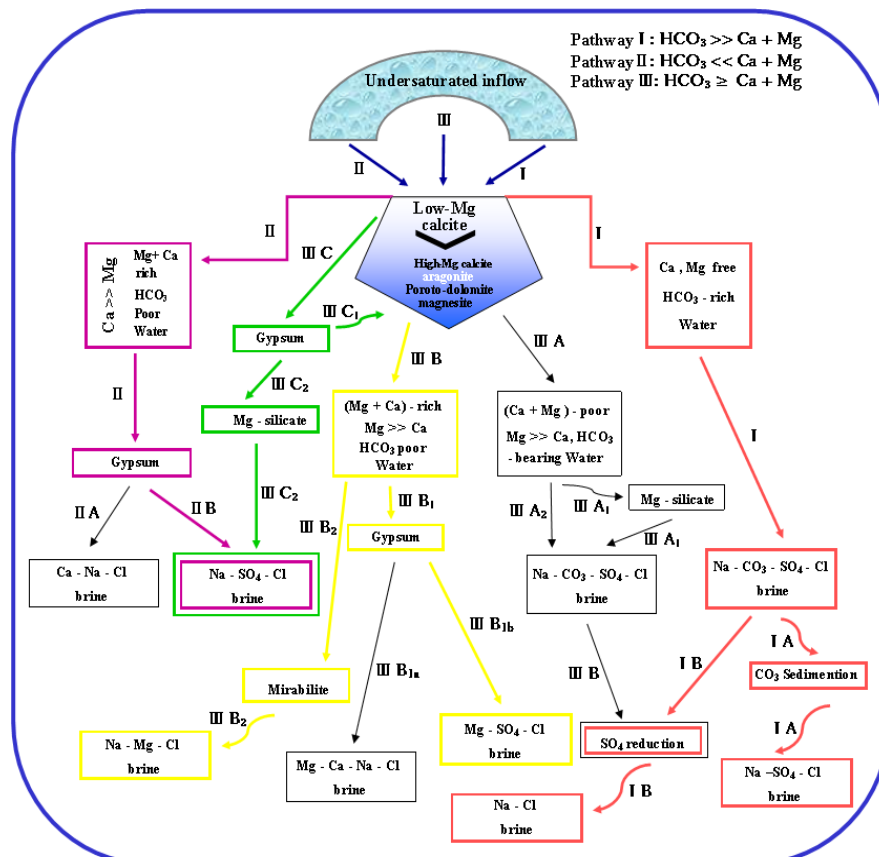


Fig 6- Diagram of the development of saline water (Hardie & Eugster 1978; Warren 2006)

Table 3- Water type of the studied area in terms of anions and cations

Sample	SO ₄ ²⁻	CO ₃ ²⁻	Cl ⁻	HCO ₃ ⁻	Anion A.	Type	Ca ²⁺	K ⁺	Na ⁺	Mg ²⁺	Cation A.	Type
A1	26.5	0	80	3.2	109.7	CL-(SO ₄)	10	4.6	82	12.4	109	Na-(Mg)-(Ca)
A2	33.8	0	52	2.7	88.5	CL-SO ₄	9.4	2.9	67	9	88.3	Na-(Mg)-(Ca)
A3	26.2	0	69	2.8	98	CL-SO ₄	13	4.4	66	14.2	97.6	Na-(Mg)-(Ca)
A4	31.3	0	56	2.1	89.4	CL-SO ₄	10	3.2	67.9	8	89.1	Na-(Mg)-(Ca)
A5	8.9	0	7.5	5.6	22	CL-SO ₄ -HCO ₃	1.5	0	18.2	2.1	21.8	Na-(Mg)-(Ca)
A6	30.6	0	78	3.4	112	CL-SO ₄	13.5	6.7	84	7.3	111.5	Na-(Mg)-(Ca)
A7	22.7	0.3	33.5	3	59.5	CL-SO ₄ -(HCO ₃)	6.3	1.7	46.4	4.9	59.3	Na-(Mg)-(Ca)
A8	19.2	0.5	43.5	4	67.2	CL-SO ₄ -(HCO ₃)	4.2	2.2	54.8	5.8	67	Na-(Mg)-(Ca)
A9	27	0	80	3	110	CL-(SO ₄)	13.6	6.2	74	15.2	109	Na-(Mg)-(Ca)-(K)
A10	29.8	0	40	2.5	72.3	CL-SO ₄	10.7	3.1	54.2	4.1	72.1	Na-(Mg)-(Ca)
A11	14.3	0.3	30.6	3	48.2	CL-SO ₄ -(HCO ₃)	4.5	0.4	40	3.1	48	Na-(Mg)-(Ca)
A12	33	0	84	2	119	CL-SO ₄	25.3	6.3	73	13.9	118.5	Na-(Mg)-(Ca)-(K)
A13	11	0.4	24	3.4	38.8	CL-SO ₄ -(HCO ₃)	2.8	0	33.5	2.4	38.7	Na-(Mg)-(Ca)
A14	6.4	0.6	10	4.7	21.7	CL-SO ₄ -(HCO ₃)	1.1	0	19.6	0.9	21.6	Na-(Ca)
A15	11	0.2	32.5	2.3	46	CL-(SO ₄)-(HCO ₃)	6	0.5	32.5	6.8	45.8	Na-(Mg)-(Ca)
A16	19.1	0.4	75	2.3	96.8	CL-(SO ₄)	5.2	3.2	80	8.4	96.8	Na-(Mg)-(Ca)
A17	20.4	0.8	102.5	1.8	125.5	CL-(SO ₄)	5.6	3.6	108	6.8	124	Na-(Mg)
A18	20	0	62.6	2.9	85.5	CL-(SO ₄)	5.6	3	70	6.4	85	Na-(Mg)-(Ca)
A19	11.9	0	12.5	5.1	29.5	CL-SO ₄ -(HCO ₃)	1.8	0	26.4	1.3	29.5	Na-(Ca)
A20	10.1	1.2	16.5	2.6	30.4	CL-SO ₄ -(HCO ₃)	1.8	0	26.5	2	30.3	Na-(Mg)-(Ca)
A21	19.4	1.6	9.2	3.8	34	CL-SO ₄ -(HCO ₃)	0.6	0	31.7	1.7	34	Na-(Mg)
A22	20.8	0	42.5	3.3	66.6	CL-SO ₄	8.7	0.8	46	10.5	66	Na-(Mg)-(Ca)
A23	28	0	71	3.6	102.2	CL-SO ₄	8	4.8	72	17.2	102	Na-(Mg)-(Ca)

According to Figure 7, most of the water sources in the Zuzan Plain follow process I, in which initially saline waters of the Na-CO₃-SO₄-Cl type are formed and then, by the separation of carbonate ions IA or the reduction of sulfate IB, brines rich in sodium and chlorine with sulfate or by the decrease in sulfate without this anion are produced. However, studies show that aqueous sediments resulting from the oxidation and dissolution of calcite, sulfite, and silicates in the igneous or magmatic rocks of the region (composition HCO₃ >> Ca+Mg), undergo process II. After the precipitation of

gypsum minerals (Fig 8), saline waters of the Na-SO₄-Cl type are formed. A small percentage of the rocks of the region (gravels, igneous rocks, and magmatic rocks) have caused the evolution process III (HCO₃>Ca+Mg) in the water sources of the Zuzan Plain. Depending on the type of brine in the area, either process IIIC, which activated process IIB after precipitation of gypsum minerals to form brines of the Na-SO₄-Cl type, or process IIIB and subprocesses IIIB₁ or IIIB₂ occurred.

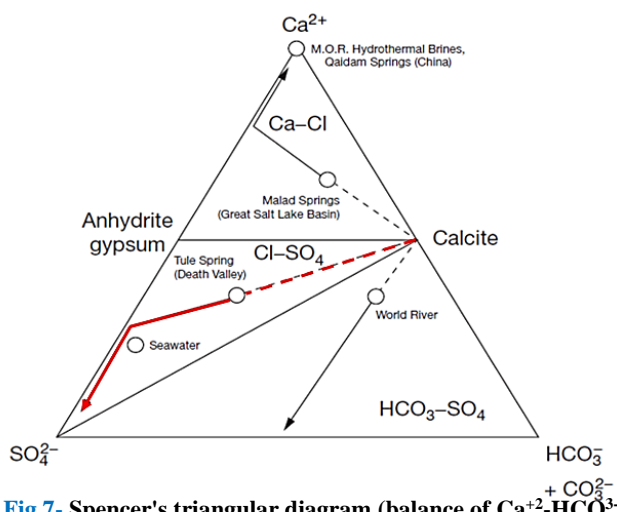


Fig 7- Spencer's triangular diagram (balance of Ca²⁺-HCO₃⁻+CO₃²⁻-SO₄²⁻ ions and development path of saline waters in the study area) (Spencer et al. 1984)



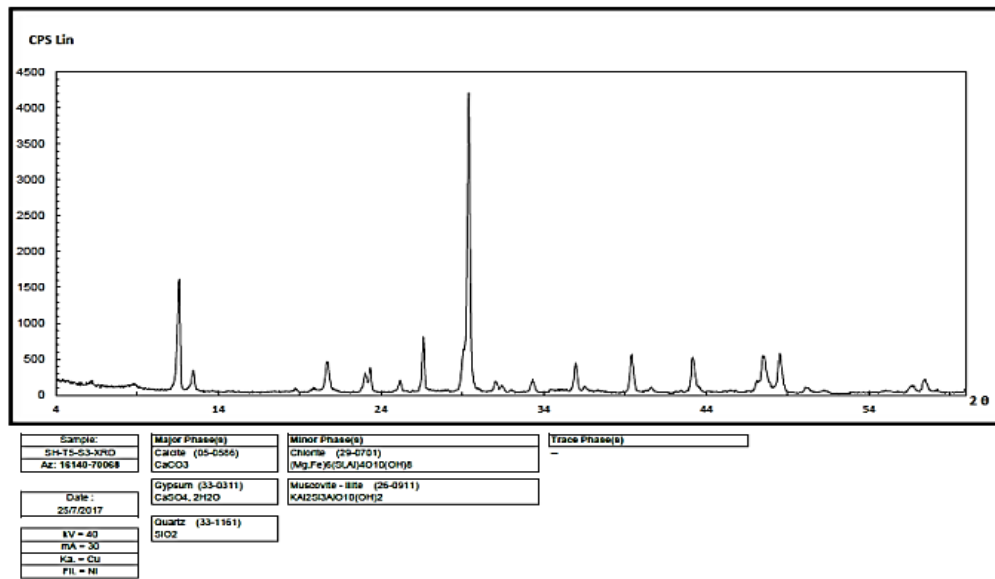


Fig 8- X-ray diffraction analysis of the salt in sample 22, showing the deposition of calcite and gypsum in the area

To better understand the evolution process of saline waters in the Zuzan region, three diagrams for anions and cations ($\text{Ca}^{2+}\text{-HCO}_3^- \text{-CO}_3^{2-}\text{-SO}_4^{2-}$), based on the Spencer diagram (Figure 7), were used (Eugster and Jones 1979; Spencer et al. 1984; Spencer 1985a; Spencer 1985b). The ions Ca^{2+} , SO_4^{2-} , and $\text{HCO}_3^- + \text{CO}_3^{2-}$, which are located in the corners of this diagram, are in equilibrium with each other (Table 4). This diagram divides water into the main phases $\text{HCO}_3^- \text{-SO}_4^{2-}$, $\text{Ca}^{2+}\text{-Cl}^-$, and $\text{Cl}^- \text{-SO}_4^{2-}$. The $\text{HCO}_3^- \text{-SO}_4^{2-}$ phase includes the average chemical composition of the world's rivers, the $\text{Ca}^{2+}\text{-Cl}^-$ phase includes thermal springs, and the $\text{Cl}^- \text{-SO}_4^{2-}$ phase includes seawater.

Plotting the average samples of the studied area on the diagram reveals that the saline waters in this area are of the $\text{Cl}^- \text{-SO}_4^{2-}$ type and undergo this process. As a result, they move towards the SO_4^{2-} pole, indicating that after the sedimentation of carbonates (calcite), the concentration of Ca^{2+} and HCO_3^- ions in the waters of the region gradually decreases, and the development process progresses towards the sedimentation of sulfates. The minerals in the region, especially gypsum, confirm this trend (Figure 8). Therefore, the waters of the Zuzan region are considered $\text{Cl}^- \text{-SO}_4^{2-}$.

Table 4- Average anion and cation of the waters of the studied area

TDS	K^+	Na^+	Mg^{2+}	Ca^{2+}	SO_4^{2-}	Cl^-	HCO_3^-	CO_3^{2-}	Average elements
96134/4	57/6	1273/7	164/4	169/2	481/4	1112/4	73/1	6/3	Groundwater

Geochemical evolutionary processes: Clarification of Types I, II, and III

To better interpret the hydrogeochemical development in the Zuzan Plain, three primary evolutionary processes—labeled as Process I, II, and III based on the model by Hardie and Eugster (1970)—are briefly described below, along with region-specific examples:

Process I: Evaporation and carbonate-sulfate precipitation: Continuous evaporation leads to the precipitation of carbonate minerals (e.g., calcite, aragonite), followed by gypsum, resulting in increasingly saline Na-Cl-rich waters.

In the Zuzan Plain most water samples (e.g., A1, A6) follow this process, where high chloride and moderate sulfate levels dominate, as evidenced by mineral deposition patterns and the Spencer diagram (Figure 7).

Process II: Gypsum dissolution and volcanic lithology influence: This pathway is driven by the dissolution of gypsum and oxidation of sulfur-bearing minerals, influenced by volcanic rock interactions. The resulting water chemistry tends

to be Na- SO_4 -Cl type.

In the Zuzan Plain samples A5 and A21, located near gypsum-bearing zones and acidic volcanic units, reflect this process with dominant sulfate concentrations.

Process III: Silicate weathering and ion exchange: The weathering of silicate rocks such as granite releases sodium and bicarbonate into the water. Ion exchange with clays may further enhance sodium levels.

In western and southern Zuzan, where granite and microgranite units are present, samples such as A19 and A20 exhibit geochemical signatures consistent with this process.

Comparison of water quality in different geological zones of the Zuzan Plain

A comparative analysis of water quality between the eastern and western parts of the Zuzan Plain reveals significant differences in chemical composition, which are closely linked to the lithological characteristics of each region. In the east zone, which is predominantly composed of sedimentary rocks such as limestone, dolomite, shale, and conglomerate, higher

concentrations of bicarbonate (HCO_3^-) and calcium (Ca^{2+}) ions are observed. This is attributed to the greater solubility of carbonate rocks, which contributes to increased water hardness. In contrast, the western part of the plain, characterized by igneous rocks (both acidic and basic types such as granite, andesite, and basalt), shows elevated levels of sodium (Na^+) and chloride (Cl^-) ions, indicating the influence of silicate weathering and stronger evaporation processes. Furthermore, electrical conductivity (EC) and total dissolved solids (TDS) are generally higher in the western samples, reflecting greater mineralization and concentration of dissolved salts. These findings demonstrate that the lithological composition of different areas plays a significant role in determining the hydrochemical characteristics of groundwater in the Zuzan Plain. Although the lithological units of the Zuzan Plain—such as acidic igneous rocks, limestone, dolomite, shale, sandstone, and gypsum—have been broadly categorized, a more precise correlation between individual formations and specific water samples enhances the clarity of interpretation. Samples A5 and A21, exhibiting a sulfate-sodic facies, are located near gypsum-rich evaporitic layers, which likely contribute to elevated sulfate concentrations.

Samples A19 and A20, taken from the western sector of the plain characterized by granite and microgranite units, show high sodium and bicarbonate content—consistent with silicate weathering and ion exchange processes.

Samples A3, A4, and A9, from the eastern zone dominated by limestone and dolomite, have higher levels of calcium and bicarbonate, reflecting carbonate dissolution.

Based on the information provided and the groundwater flow direction map (Figure 1), it can be stated that the main movement of water in the Zuzan Plain is from the northeastern highlands to the southwestern lowlands. This pattern is influenced by the topographic slope and lithological conditions of the region. Along the flow path, the chemical quality of water changes due to continuous interaction with rocks and also mixing with alluvial sediments. The correspondence of the distribution of chemical facies with the flow direction indicates that both lithology and flow processing processes play a fundamental role in the formation of groundwater quality. In the downstream parts, especially in the central and southwestern regions of the plain, the salinity and hardness of water have increased, which is due to the increase in water retention time and the stronger effect of evaporation and rock-water reactions.

Origin of the sodium-chloride facies and its origin

According to Table 2, the dominant facies of the groundwater of the Zuzan Plain is sodium-chloride (Na-Cl). However, field and geochemical evidence show that this pattern does not have a fixed origin, but is the result of the overlap of several processes and spatial heterogeneity in the basin:

Dissolution of halite and evaporate salts (direct terraneous origin of chloride): The presence of evaporate deposits (halite/evaporates) in the central parts, and the increase in conductivity (EC) along with the simultaneous increase in Na and Cl with a molar ratio of $\text{Na}/\text{Cl} \approx 1$ is consistent with direct dissolution of halite. Such behavior is more pronounced in samples from the end areas of the flow path (downstream).

Evaporation and concentration along the flow path: In arid plain climates, evaporation from the water/soil surface

increases TDS and EC and shifts samples towards the sodium-chloride corner of Piper diagrams and the “rock/evaporation” fields of the Gibbs diagram. This mechanism intensifies the Na-Cl pattern, regardless of the initial source of the ions, and therefore the “appearance of Na-Cl” does not necessarily mean a fixed source.

Cation Ion Exchange in Fine-Grained Alluvium: The process of exchanging Na^+ with $\text{Ca}^{2+}/\text{Mg}^{2+}$ on the surface of clays can cause water to become sodium-rich without producing excess chloride. In this case, $\text{Na}/\text{Cl} > 1$ (in milliequivalents) and equilibrium indices such as $\frac{(\text{Ca}+\text{Mg})-(\text{HCO}_3+\text{SO}_4)}{(\text{Ca}+\text{Mg})-(\text{HCO}_3+\text{SO}_4)}$ versus $\frac{(\text{Na}-\text{Cl})}{(\text{Na}-\text{Cl})}$ tend towards ion exchange gradients. This means that even when the final facies is reported as Na-Cl, some of the sodium does not necessarily come from halite chloride.

Contribution of upstream igneous/silicate rocks: Weathering of plagioclases and sodic feldspars in marginal igneous units can produce sodium and bicarbonate; if further evaporation or mixing with chloride-rich waters occurs, the Na-Cl facies is formed, while some of the Na is of silicate origin. This is consistent with the components extracted from multivariate analyses (PCA/cluster), which show two origins: “carbonate dissolution” and “evaporative/Na-Cl concentration with igneous input”.

Evidence of local contrast in non-chlorid facies: Core samples A5 and A21A5 and A21A5 with Na- SO_4 highlight the role of gypsum/anhydrite and increased residence time; these cases prove that the origin of water chemistry is not uniform across the plain and is sensitive to local lithology and flow direction.

Comparison with similar arid and semi-arid regions

To assess the generalizability of the findings from the Zuzan Plain, comparisons were made with studies conducted in other arid and semi-arid regions of Iran and beyond. In several comparable areas—such as the Maharlou Basin in southern Iran (Fayazi et al. 2007), the Qorveh Plain in western Iran (Rezaei and Naderi 2024), and the Yazd-Ardakan region (Thivya et al. 2013)—the predominant hydrochemical facies were similarly of sodic-chloride type, aligning with the results of this study. International studies from other semi-arid areas, including the South Bengal Basin in India (Banerjee and Sikdar 2022) and parts of East Africa (Kebede and Ayenew 2022), also confirmed that lithological units—particularly limestone and volcanic rocks—significantly influence groundwater chemistry. This is consistent with observations in the Zuzan Plain, where the dissolution of limestone and weathering of igneous rocks play key roles in shaping water quality (Figure 9). However, some localized features make the Zuzan Plain unique. For example, samples A5 and A21, which exhibit a sulfate facies rather than the dominant chloride type, are located in areas with overlapping acidic igneous and carbonate lithologies. Additionally, anthropogenic influences such as industrial iron ore processing in the nearby Sangan area may contribute to water quality variations—factors not commonly reported in the comparison regions. Therefore, while the hydrogeochemical evolution in Zuzan shares common traits with other arid regions, the specific combination of lithology and human activity appears to impart distinctive characteristics to the region’s water resources.

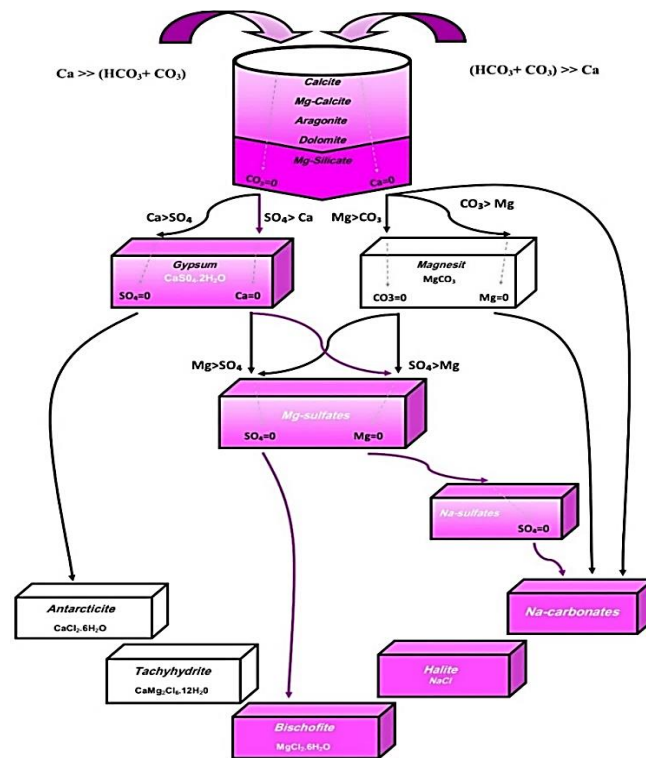


Fig 9- Diagram of the evolution of saline water originating from non-marine water (Hardie & Eugster 1970; Warren 2006)

Conclusions

Based on the analysis of the Piper diagram for cations and anions in the Zuzan water, it is evident that the majority of samples exhibit a sodic facies with chloride-type composition. Notably, exceptions are observed in samples A5 and A21, which display a sodic facies with a sulfate-type composition and are located centrally within the plain. This composition reflects the predominant influence of various geological formations in the region, notably acidic igneous units, limestone, sandstone, conglomerate, siltstone, and shale. Among these formations, limestone stands out due to its higher solubility, consequently exerting significant control over the chemical composition of the water resources in the Zuzan plain.

Further examination reveals distinct characteristics in the water related to the geological formations, such as the relative presence of chloride, sulfate, bicarbonate, and carbonate ions. The saltwater evolution diagram illustrates that the samples generally follow processes I, II, and III, with a prevalence of trend I and fewer occurrences of trends II and III. The dominant water type in the Zuzan Plain emerges as Na-Cl-SO₄, as confirmed by both the Spencer diagram for Cl-SO₄ and the diagram for Mg²⁺-HCO₃⁻+CO₃²⁻-SO₄²⁻.

Moreover, the evolution diagrams depict the sequential deposition of minerals, with carbonate minerals precipitating initially, followed by sulfate minerals, particularly due to the high concentration of Cl in the region's samples. The deposition process further elucidates the evolution towards the sedimentation of sodium sulfate minerals and sodium carbonate, with the potential deposition of halite upon water condensation. Conversely, if magnesium sulfate minerals form

while the waters are depleted of SO₄, the sedimentation process would lead to the formation of bischofite mineral. These findings underscore the complex interplay between geological formations and water chemistry in the Zuzan region, providing valuable insights for understanding and managing water resources in the area.

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