

Hydrogeochemical evolution and mineralization origin in a semi-arid shallow aquifer: a case study of the Barika area in northeast Algeria

Evoluzione idrogeochimica e genesi della mineralizzazione in un acquifero superficiale in contesto semi-arido: Caso studio nell'area di Barika nella parte nord-est dell'Algeria

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ARTICLE INFO

Ricevuto/Received: 20 January 2023

Accettato/Accepted: 8 May 2023

Pubblicato online/Published online:

30 June 2023

Handling Editor:

Nico Dalla Libera

Citation:

Tafrount, A., Drias, T., Chenaf, D., Nafaa, B. (2023). Hydrogeochemical evolution and mineralization origin in a semi-arid shallow aquifer: a case study of the Barika area in northeast Algeria. *Acque Sotteranee - Italian Journal of Groundwater*, 12(2), 19 - 34
<https://doi.org/10.7343/as-2023-624>

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Keywords: *Piper diagram, groundwater, saturation index, mineralization, climate change.*

Parole chiave: Diagramma di Piper, acque sotterranee, indice di saturazione, mineralizzazione, cambiamento climatico.

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Riassunto

Molte regioni africane della parte medio-orientale e settentrionale sono caratterizzate da un clima arido e semi-arido. Per questo motivo, la gestione e l'approvvigionamento dell'acqua potabile sono diventate attività complesse sia per le autorità locali che regionali. L'acquifero presente nei depositi del periodo Mio-Plio Quaternario dell'area di Barika è l'unico reservoir della regione utilizzabile per scopi idropotabili e irrigui. L'obiettivo di questo articolo è quello di identificare l'origine e il processo evolutivo della mineralizzazione delle acque sotterranee avvalendosi degli elementi maggiori come indicatori. Al fine di raggiungere l'obiettivo, nei mesi di giugno 2018 e luglio 2019 sono stati prelevati ed analizzati dei campioni provenienti da pozzi che intercettano l'acquifero indagato. I risultati ottenuti, espressi attraverso l'ausilio dei diagrammi di Gibbs e Piper, correlazioni chimiche e analisi statistiche, hanno permesso di identificare le origini della mineralizzazione delle acque di falda. I processi principali che hanno portato alla mineralizzazione sono imputabili alla dissoluzione di evaporiti, precipitazione di carbonati, scambio cationico e il processo di evapotraspirazione. I risultati delle analisi fisico-chimiche hanno mostrato come queste acque sono principalmente riconducibili alla facies idrochimica "solfato - clorurato - alcalino terrosa", con un leggero cambiamento di facies in alcuni pozzi tra un periodo di campionamento e l'altro. Ciò è imputabile all'interazione delle acque con le formazioni geologiche che compongono il serbatoio e alla scarsità di acqua dovuta al cambiamento climatico.

Abstract

Most Middle Eastern and North African regions are characterized by an arid and semi-arid climate. As such, the drinking water supply and management have become a challenging task for local and regional authorities. The Mio-Plio Quaternary aquifer of the Barika area is the only drinking and irrigation water reservoir in the region. The objective of this paper is to identify the origin and evolution process of the groundwater mineralization of this aquifer using major elements as indicators. To achieve this objective water samples were collected, from several boreholes drilled in the aquifer, in June 2018 and March 2019, and subsequently analyzed. The results obtained in terms of Gibbs plot, Piper, chemical correlation, and statistical analysis of chemical data identified the origins of groundwater mineralization. The dissolution of evaporated minerals, precipitation of carbonates, evapotranspiration, and ion exchange reactions have been identified as the primary processes of mineralization. The results of the physicochemical analysis showed that these waters consisted mainly of chloride, calcium sulfate, and magnesium facies types with a slight change of facies in some boreholes during the two sampling periods. This is due to the interactions with the aquifer geology and to the water scarcity caused by climate change.

Introduction

Water resources play a vital role in our daily lives, serving as a primary source for drinking, irrigation, and industrial activities.

One-third of the world's population uses groundwater for drinking (Sudani, 2018). Today, groundwater supplies approximately 50% of the world's drinking water needs, with some regions relying almost exclusively on groundwater for their water supply. However, it is worth noting that the availability and quality of groundwater vary greatly across regions and can be impacted by factors such as climate change, overuse, pollution, and population growth. Many studies were carried out on groundwater usage and protection (Fehdi et al., 2009; Al-Ahmadi, 2013; Kallel et al., 2018; Hamad et al., 2018; Kumari and Rai, 2020; Nekkoub et al., 2020; Beyaitan Bantin et al., 2020; Hamed et al., 2022).

The accumulated stresses and demands make groundwater management face one of the current global challenges to resilience to water scarcity due to the vital role played by freshwater as a resource for sustaining life and human needs (Chang et al., 2017). Although groundwater reserves are substantial, current climatic conditions often impact their availability, and a changing climate leads to asymmetric and very uneven rainfalls, leaving approximately 40% of the world's population experiencing acute water shortage (Dakoure, 2003; Blinda and Thivet, 2009; Loucks and Van Beek, 2017; Yousefi et al., 2018; Yousif and El-Aassar, 2018; Ncibi et al., 2021; Besser et al., 2021). With the population growth increasing by 3% per year and rapid urbanization and advancements in living standards have resulted in a surge in the demand for water as well. This increase in demand has led to the depletion of groundwater resources, which has been utilized not only for drinking purposes but also for intensified irrigation and industrial activities. Unfortunately, in some instances, the unregulated exploitation of groundwater has caused numerous critical issues worldwide. (Castilla-Rho et al., 2017; Sappa et al., 2019). Therefore, given the fundamental role that water resources play in sustaining ecosystems and supporting human livelihoods, it is essential to evaluate and properly manage groundwater quantity, quality, and demand carefully to ensure its availability for future generations. In light of this critical situation, the scientific community faces a significant challenge to evaluate and protect all available sources of water. (Rouaibia and Djabri, 2017; Rais et al., 2017; Modibo Sidibé et al., 2019).

In Algeria, groundwater is the primary source of water for domestic, industrial, and agricultural use, particularly in the semi-arid southern and eastern regions where water resources are scarce and the aquifers are shallow (Hamad et al., 2018; Drias et al., 2020; Brahmi et al., 2021; Hamed et al., 2022).

The quality of groundwater is controlled by the variations of hydrochemical processes. Thus, this research aims to consider the mineralization processes in the Barika region and to identify its governing actors, where groundwater has always been an important source of drinking water for local people, animals, and for irrigation purposes on over

111,683 ha of agricultural lands (DSA, 2019). It is the first study of its kind in the region, as previous studies have focused on the availability rather than the quality of groundwater.

To achieve this objective, 26 samples collected from the boreholes were analyzed in two periods, June 2018 and March 2019. A combined hydrogeological and geochemical investigation was carried out by inputting data into the PH Redox Equilibrium in Aquifers (PHREEQC) code and analyzing the saturation index, which indicates whether a mineral has a thermodynamic potential for dissolution or precipitation. Hydrochemical facies and mineral dissolution/water was calculated using the DIAGRAMME package software (Fehdi et al., 2009). Multivariate statistical methods, such as Principal Component Analysis (PCA), offer many options for assessing groundwater types in different regions of the world (Saccetti and Timmerman, 2016; Telahigue et al., 2018; Parisi et al., 2023). To interpret the hydrochemical parameters, these methods can be combined with a graphical representation. Gibbs (Gibbs, 1970) proposed the principal natural mechanisms controlling global water chemistry, which can be classified as precipitation dominance, evaporation, precipitation, rock-water interaction, and mixing with seawater (Adimalla et al., 2018; Aouiti et al., 2021; Zhang et al., 2021). Using these methods make it possible to follow the spatial evolution of the hydrochemical parameters and to estimate their mineral origin. Therefore, in order to develop sustainable groundwater management strategies, many studies adopt a combined approach that encompasses both hydraulic and hydrogeochemical analyses. This is because a comprehensive understanding of the hydrochemical properties of groundwater is essential for effective and long-term management of this vital resource (Sappa et al., 2019; Gao et al., 2022). A new improved piezometric map of the shallow aquifer was created in June 2018 using data from a survey of the aquifer's water levels. The groundwater flow is visible on this map, with the direction of water flow naturally occurring in a NE-SW and SE-SW direction. Additionally, the piezometric variations over time show seasonal fluctuations in relation to rainfall variations.

The results of this research are expected to inform current water resources planning efforts and contribute to effective, sustainable natural resource management in the plain of Barika and other similar regions. Ultimately, this research will help promote the long-term health and viability of local ecosystems and communities.

Material and methods

Geographic location and climate

The Barika Plain is situated in the northeastern part of Algeria. In the Barika region which is located in the western part of the Wilaya of Batna, limited to the north by the Wilaya of Sétif, to the west by the Wilaya of M'Sila, to the south by the Wilaya of Biskra. The plain is part of the Hodna watershed and the Barika-Bitam wadi sub-basin (Fig. 1) which covers a surface area of approximately 139,418 km². To the north, the plain is bordered by the Djebel Hodna

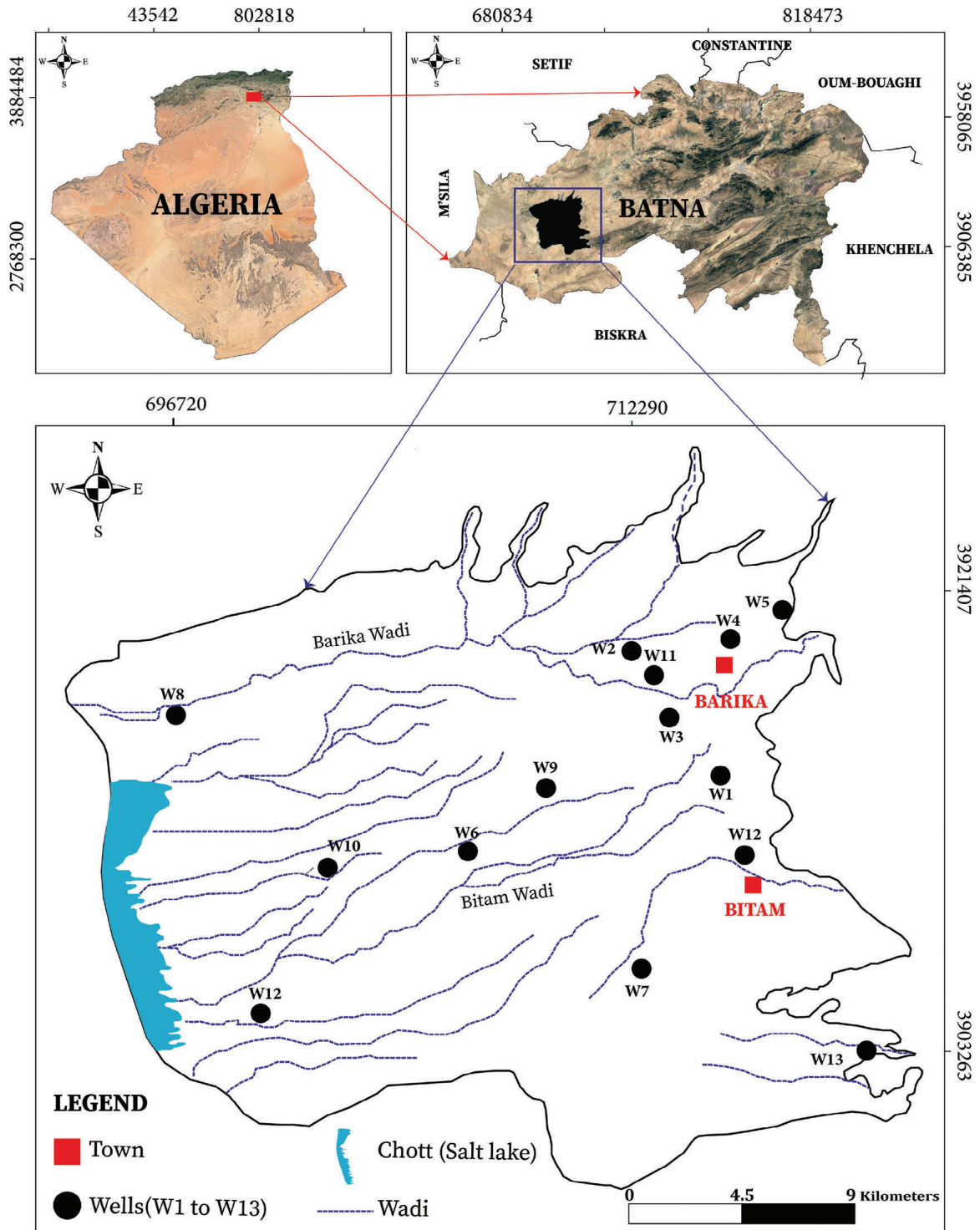


Fig. 1 - Geographical location of the study area of Barika shallow aquifer.

Fig. 1 - Inquadramento geografico dell'area studio dell'acquifero superficiale di Barika.

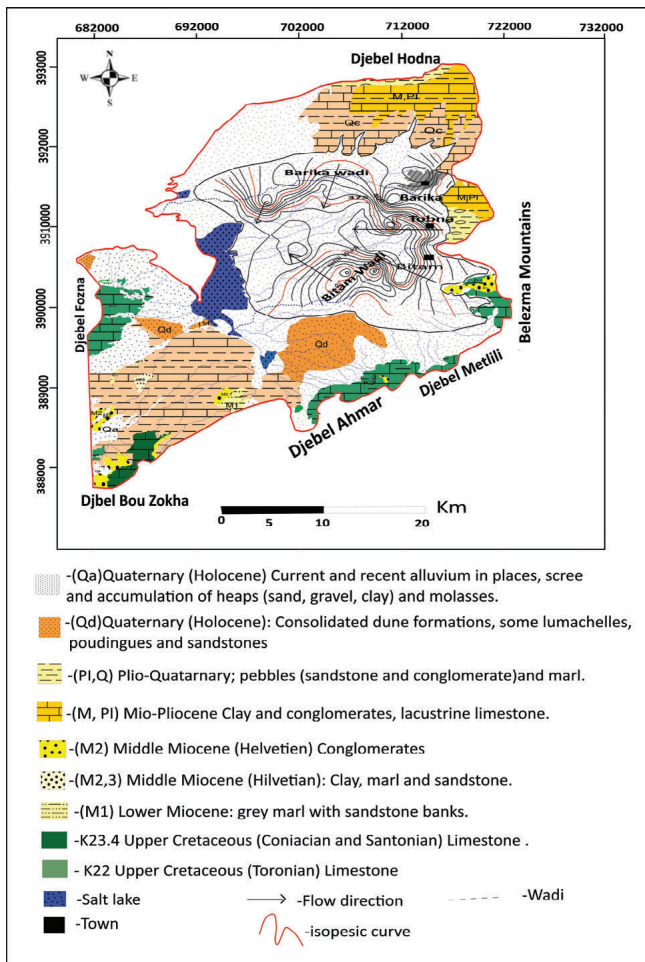


Fig. 3 - Geological map and piezometric curves of the shallow aquifer of Barika area with groundwater flow directions (June 2018).

Fig. 3 - Carta geologica e curve piezometriche dell'acquifero superficiale dell'area di Barika con la direzione di deflusso delle acque sotterranee (Giugno 2018).

it may also receive water inputs from Oued Barika during flood periods and from the Boundary limits through faults in the Cretaceous carbonate formations. The piezometric map (Fig. 3) shows that there are two primary directions of groundwater flow: from the northeastern and southeastern parts of the aquifer towards the west.

Piezometric evolution

Based on the groundwater level measurements obtained from wells W4, W5, W7, W10, and W12 (as depicted in Fig. 4), there has been an average decrease of approximately 6 meters between June 2018 and March 2019. This decline can be attributed to several factors, including the rainfall pattern (as shown in Fig. 5), lateral groundwater recharge/discharge, and intensive exploitation of groundwater resources. Indeed, the agricultural activity in the study area relies heavily on groundwater for irrigation and has significantly contributed to the declining water levels in the absence of sustainable management practices to ensure the preservation of this vital resource.

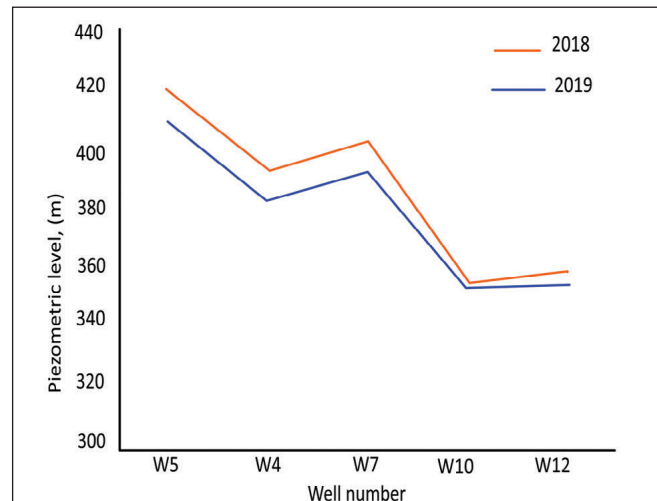


Fig. 4 - Piezometric surface fluctuations in monitored boreholes in the study area of the shallow aquifer of Barika (June 2018).

Fig. 4 - Fluttuazione della superficie piezometrica nei pozzi monitorati nell'area studio dell'acquifero superficiale di Barika (Giugno 2018).

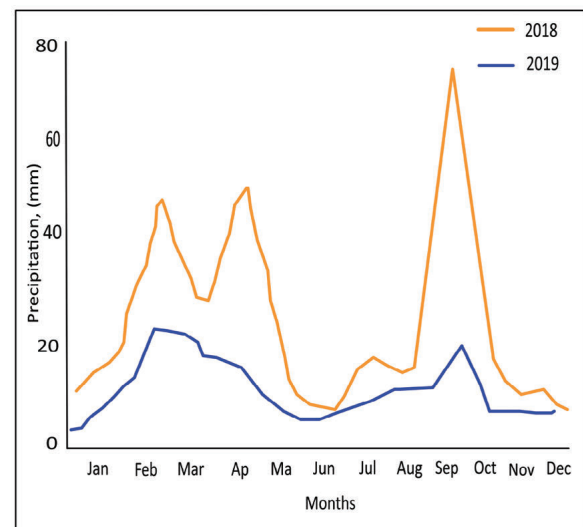


Fig. 5 - Comparing rainfall variation between June 2018 and March 2019.

Fig. 5 - Comparazione della precipitazione tra Giugno 2018 e Marzo 2019.

Sampling and laboratory analyses

To conduct this study, a comprehensive dataset was assembled from various sources, including physico-chemical water parameters and piezometric measurements done in June 2018 (as shown in Fig. 3). In addition, fieldwork was carried out in March 2019, where thirteen sites in the study area, were visited to collect data from the same wells as in June 2018. During fieldwork, all water samples were collected and temporarily stored in a portable cooler at 4 degrees Celsius before being transferred to a refrigerator for storage until analysis. The samples were analyzed for major cations (Na⁺, K⁺, Ca²⁺, and Mg²⁺); and anions (Cl⁻, SO₄²⁻, HCO₃⁻, NO₃⁻) in the analytical laboratory using standard methods recommended by the J. Rodier 8th edition and ISO 5667. The chloride contents were determined using the Mohr method (ISO 9297 - NA 6917, ISO 5667-3: 2003), while sulfates were determined by UV/Visible spectrophotometry and nitrates by the sodium salicylate method (Rodier, Standard Afnor

T90-012). The pH of the water was determined using an electrometric method, and sodium (Na^+) and potassium (K^+) were analyzed by atomic absorption spectroscopy (AAS), while bicarbonate (HCO_3^-) was measured using the volumetric method. The accuracy of the chemical analysis was verified by calculating the ion-balance errors, which were always less than 10% (Cidu et al., 2009), as recommended by Subramani et al. (2005). The results obtained from the different analytical techniques were analyzed using a combination of arithmetic techniques and statistical methods. To better visualize and characterize the physicochemical data of the water, we used two complementary approaches, namely Piper diagrams and Phreeque by DIAGRAMME. Phreeque is a module within the DIAGRAMME software package that uses principal component analysis (PCA) to mathematically describe the hydrochemical characteristics of water samples. Specifically, Phreeque uses a variant of PCA called non-negative matrix factorization (NNMF) to identify the underlying chemical processes that give rise to the observed hydrochemical variability. The output of Phreeque is a set of linear equations that describe the contribution of different chemical processes (such as mineral dissolution or ion exchange) to the overall hydrochemical characteristics of the water. The investigation of the water mineralization, water pooling and the identification of the causes of these clusters of water science were performed by the mathematical description of water using PCA and MS Excel Statistics (XLSTAT-pro2018) software as in similar studies (Zouita et al., 2018; Gouaidia et al., 2017).

Results and discussions

Hydrogeochemical facies and mechanisms of controlling groundwater chemistry

To better understand the hydrochemical evolution of groundwater, analytical data collected from groundwater samples can be plotted on a Piper Trilinear diagram, which displays the major cations and anions. This approach was originally proposed by Piper in 1944. By analyzing the data using the Piper diagram (Fig. 6), it is possible to identify an almost homogeneous chemical facies of water in the study area between the two periods: chloride and calcium sulfate and magnesium. This indicates a mix of fresh water from a carbonatic environment and brackish water, likely linked by evaporitic deposits. This spatial homogeneity can be explained by the phenomenon of Base Exchange, which occurs when water and clays exchange sodium and potassium for calcium and magnesium. Another possible explanation is inverse ionic exchange. The dilution of water during its journey to the Chott is also an important mechanism to consider. To ensure the waters become evaporative, it is necessary to comment on the points that have changed positions (W5, W7, W4, W12, W13), as these changes are directly related to the formation of gypsum (a Triassic mineral).

One of the facies tends to carbonate (W7) as of June 2018. This evolution of facies is likely due to the exchange of bases, caused by the proximity to Bitham wadi. However, it tends to return to its original facies after dilution by

rainwater. The examination of major element contents reveals that mineralization is governed by calcium, sodium, and magnesium for cations and sulfates for anions. The arid and semi-arid climate can also affect the chemical composition by concentrating the ions during evaporation. To analyze the chemical composition of groundwater, to detect any chemical changes or chemical alterations, and differentiate natural or meteoric characteristics of groundwater, groundwater chemistry tests were conducted and the results were analyzed. These test results revealed that calcium, sodium, and magnesium were the dominant cations, while sulfates were the main anions responsible for mineralization. These test results were converted from ions to equivalent weights and plotted to establish correlations that could help to identify potential sources of groundwater chemistry (Nazzal et al., 2014). The Gibbs diagram, which is widely used to determine the relationship between lithological characteristics of an aquifer and its water composition (Krishna Kumar et al., 2015), was also used in this study, and they are calculated by the following Eq. 1 and 2:

$$\text{Gibbs ratio I (for anions)} = \frac{\text{Cl}^-}{\text{Cl}^- + \text{HCO}_3^-} \quad (1)$$

$$\text{Gibbs ratio II (for cations)} = \frac{(\text{Na}^+ + \text{K}^+)}{(\text{Na}^+ + \text{K}^+ + \text{Ca}^{2+})} \quad (2)$$

Whereas the ionic concentrations are presented in meq/L.

The Gibbs diagram depicts five processes, including: precipitation dominance, evaporation, precipitation, rock-water interaction, and mixing with seawater, that control the overall groundwater chemistry (Gibbs, 1970; Sadooni and Al Awadi, 2009). The analysis of the samples using the Gibbs diagram revealed that the predominant samples fall into the evaporation and precipitation dominance fields of the diagram (Fig. 7). This suggests that evaporation is a dominant factor in increasing the mineralization of groundwater. This is not surprising, as evaporation can increase the concentration of ions formed by chemical weathering of rock, leading to a higher degree of mineralization.

Groundwater mineralization processes

Elementary statistics

A coefficient of variation (CV) of less than 50% indicates homogeneity in the amplitude of the observed contents between the minimum and maximum values (Tab. 1), indicating that the observed values are close to the calculated average. However, variables such as Mg^{2+} , Na^+ , Cl^- , and SO_4^{2-} have a CV greater than 50%, indicating a large deviation from the average of all observations, which can be attributed to the evaporitic soil and the period. The variables Ca^{2+} and HCO_3^- show a CV lower than 50% whereas K^+ and NO_3^- show a CV close to 50%, indicating a certain degree of heterogeneity. During the March period, the CV% was lower than 50% for all the elements except for Na, indicating a small amplitude between the minimum and maximum value and a fairly homogeneous spatial distribution of the ions.

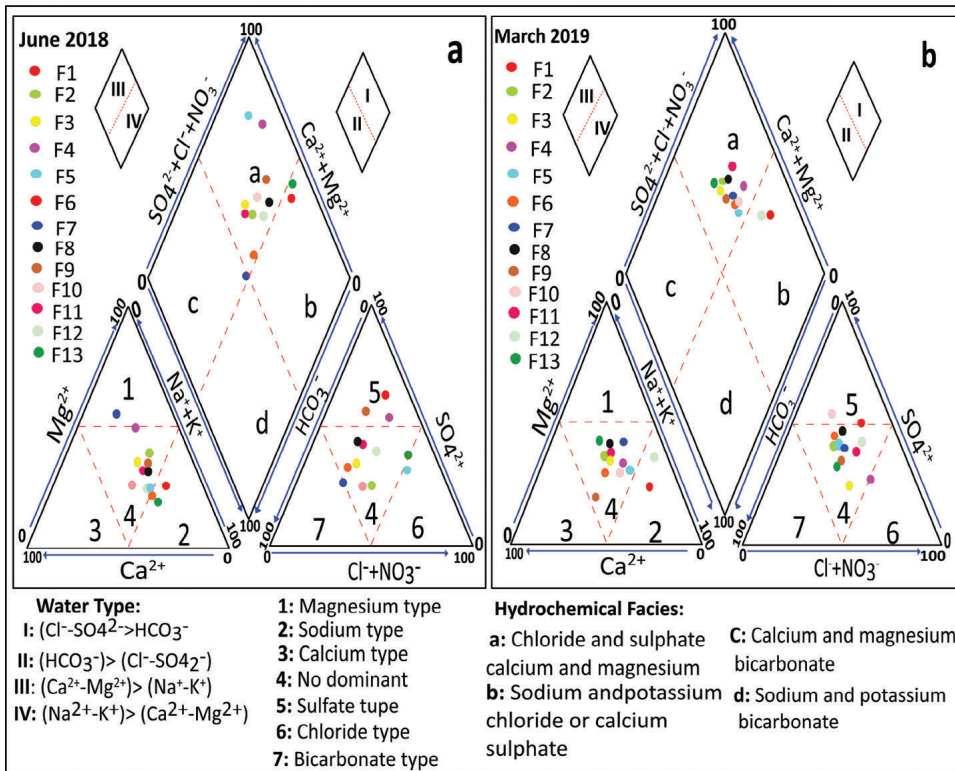


Fig. 6 - Plot of the collected water samples on Piper's diagram.

Fig. 6 - Diagramma di Piper per i campioni prelevati.

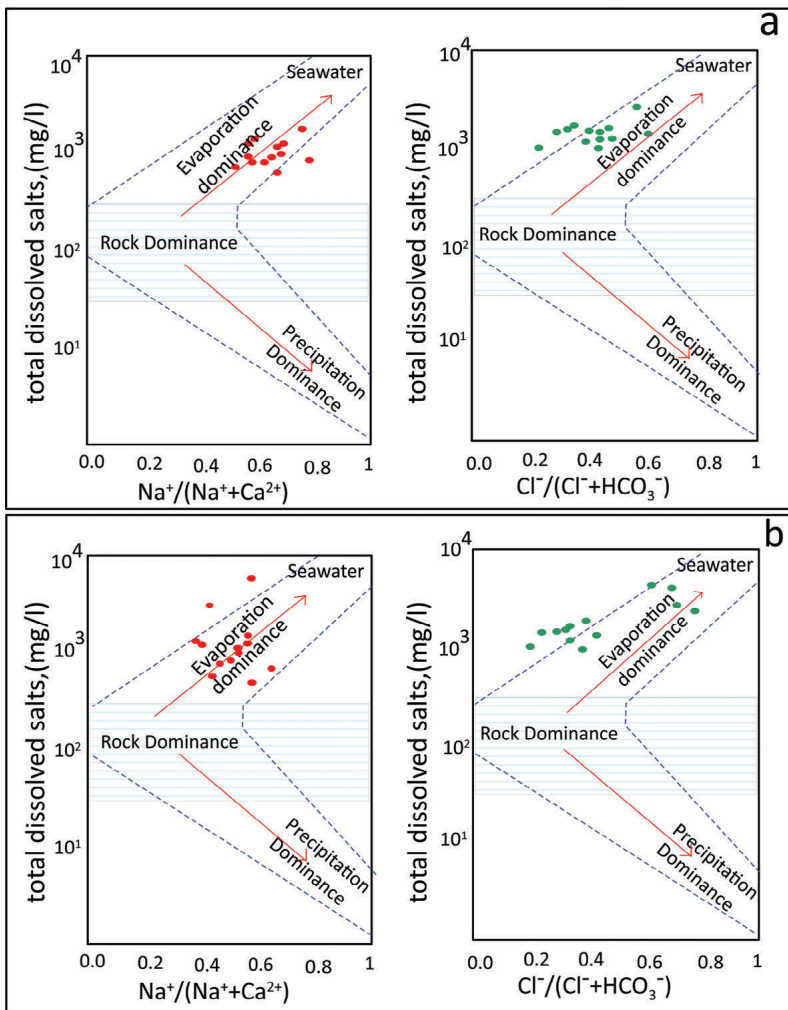


Fig. 7 - Plot of the collected water samples on Gibbs's diagram.

Fig. 7 - Diagramma di Gibbs per i campioni prelevati.

Tab. 1 - Summary of the descriptive statistics for the measured parameters.

Tab. 1 - Riassunto della statistica descrittiva per i parametri misurati.

Variables	June 2018					March 2019				
	Min	Max	Mean	SD	CV%	Min	Max	Mean	SD	CV%
Ca ²⁺ (mg/L)	62	228.32	130.71	56.25	130.71	16.67	150.03	108.29	33.47	30.91
Mg ²⁺ (mg/L)	34	275.22	100.30	80.32	100.30	51.30	115.96	88.86	22.82	25.68
Na ⁺ (mg/L)	112	576	208.46	134.04	208.46	114.55	560.52	197	119.50	60.66
K ⁺ (mg/L)	2	10	5.28	2.74	5.28	2.78	5.83	4.39	0.95	21.63
HCO ₃ ⁻ (mg/L)	183	398.52	310.44	59.77	310.44	168.09	380.91	327.52	57.62	17.59
Cl ⁻ (mg/L)	78	600	266.68	188.73	266.68	92.10	429.18	225.54	97.04	43.03
SO ₄ ²⁻ (mg/L)	150	1250	467.95	360.40	467.95	206.48	814.14	398.76	145.79	36.56
NO ₃ ⁻ (mg/L)	2	40	20.40	11.42	20.40	7.58	33	16.91	7.46	44.14
EC (us/cm)	1300	34	2056.23	820.20	2056.23	1420	3340	1858.53	560.31	30.15
PH	7	8.10	7.52	0.35	7.52	6.83	7.79	7.20	0.38	5.34

Correlation matrix and data analyses

The correlation matrix is a statistical tool for exploring the relationships between different variables (Li et al., 2019). In this study, we used Pearson correlation matrices for both sampling periods of June 2018 (Tab. 2) and March 2019 (Tab. 3) to identify the chemical parameters that affect water

mineralization in the Barika region. Our results show that during the summer period, the key parameters are Ca²⁺, Mg²⁺, Na⁺, SO₄²⁻, and Cl⁻ (Tab. 2), while during the winter period, they are Na⁺, SO₄²⁻, and Cl⁻ (Tab. 3). We found that Ca²⁺, Mg²⁺, Na⁺, SO₄²⁻, Cl⁻, and electrical conductivity (EC) were strongly correlated ($0.9 < r^2 > 0.7$).

Tab. 2 - The correlation between major ionic parameters in water samples collected in June 2018.

Tab. 2 - Correlazione tra gli ioni maggiori rilevati nei campioni prelevati a Giugno 2018.

Variables	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	HCO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻	CE
Ca ²⁺	1								
Mg ²⁺	0,832	1							
Na ⁺	0,517	0,249	1						
K ⁺	0,499	0,515	0,352	1					
HCO ₃ ⁻	-0,321	0,040	-0,374	0,186	1				
Cl ⁻	0,945	0,780	0,612	0,258	-0,356	1			
SO ₄ ²⁻	0,708	0,668	0,779	0,518	-0,277	0,637	1		
NO ₃ ⁻	0,223	0,177	0,268	0,764	0,054	-0,018	0,308	1	
EC	0,921	0,727	0,754	0,535	-0,271	0,884	0,839	0,264	1

Tab. 3 - Investigating the correlation between major ionic parameters in water samples collected in March 2019.

Tab. 3 - Analisi della correlazione tra gli ioni maggiori rilevati nei campioni prelevati a Marzo 2019.

Variables	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	HCO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻	CE
Ca ²⁺	1								
Mg ²⁺	0,604	1							
Na ⁺	0,269	0,419	1						
K ⁺	0,377	0,420	0,515	1					
HCO ₃ ⁻	0,627	0,337	0,127	0,498	1				
Cl ⁻	0,428	0,644	0,745	0,533	0,176	1			
SO ₄ ²⁻	0,251	0,283	0,810	0,488	0,082	0,362	1		
NO ₃ ⁻	0,037	-0,266	-0,186	-0,124	-0,183	-0,216	0,045	1	
EC	0,152	0,211	0,902	0,501	0,050	0,647	0,654	-0,211	1

The significant links between various elements found in the Barika region can probably be attributed to their common origins. Sulfate-type waters, calcium, and magnesium chlorides, for example, all have similar origins, which is confirmed by the grouping of these elements with EC on the F1 axis (Fig. 8). The mineralization of waters in the Barika region appears to be mainly governed by three mechanisms: the residence time in the aquifer, high evaporation due to the semi-arid climate, and ion exchange. There was a significant positive correlation between Ca^{2+} and Mg^{2+} (0.8 - 0.6) in the two periods, June 2018 and March 2019, respectively. This suggests that Ca^{2+} and Mg^{2+} have common origins in carbonate and evaporitic sources. In addition, high correlations were observed between magnesium and chloride ($r^2 = 0.78$) as well as between magnesium and sulfate ($r^2 = 0.66$) in June 2018 and between magnesium and chloride ($r^2 = 0.64$) in March 2019. This suggests that a part of the mineralization may also be linked to the dissolution of MgSO_4 and MgCl_2 . Sodium (Na) is found to be positively associated with Cl^- and SO_4^{2-} , with Na-Cl at the rate of 0.50 and Na- SO_4^- at the rate of 0.77 in June 2018, and 0.7 and 0.8, in March 2019, respectively (refer to Tab. 2 and 3). This suggests that sodic-plagioclase weathering is likely to be a crucial source of excess Na^+ concentration in the study area, in addition to halite dissolution. The weak association observed between calcium and sulfate indicates that carbonate dissolution and cation exchange processes are the primary sources of excess Ca^{2+} levels, with the exception of gypsum dissolution. In March, a good correlation between Ca^{2+} and SO_4^{2-} ($r^2 = 0.7$) suggests possible dissolution of sulfate minerals, such as gypsum, with a possible reverse effect of the calcite precipitation process known as dedolomitization. HCO_3^- is found to have a weak to very weak negative correlation with Ca^{2+} (-0.3) and Mg^{2+} (0.3), indicating that the source of these ions may not be carbonate mineral dissolution. In contrast, in the winter period, there is a good association between HCO_3^- and Ca^{2+} ($r^2 = 0.6$), which might be due to ion exchange processes operating in the aquifer. The very weak correlation observed between nitrates (NO_3^-) and chlorides (Cl^-) in both periods suggests that nitrates have a superficial origin.

Principal Component Analysis (PCA)

Principal Component Analysis (PCA) is a statistical technique used to study and graphically illustrate the relationships between variables and observations in large datasets (Mouassa et al., 2020; Mitryasova and Pohrebennyk, 2020; Gueddari et al., 2022). PCA was performed using the program XLSTAT 2018 to identify the most important factors controlling groundwater geochemistry. The dataset included 11 variables (HCO_3^- , SO_4^{2-} , Cl^- , NO_3^- , Ca^{2+} , Mg^{2+} , Na^+ , K^+ , pH, T, and EC) which were analyzed on 26 samples collected during the two campaigns of June 2018 and March 2019. The results confirmed the source, different mechanisms and most important factors that characterize the mineralization of the groundwater. The first three components extracted (F1, F2, and F3) in June 2018 and March 2019

represented 79.07% and 74.93% of the total variance in the dataset, respectively (Fig. 8, Tab. 4 and 5). The first factor (F1), which accounted for 46.97% and 46.45% of the total variance, respectively, included Ca^{2+} , Mg^{2+} , SO_4^- , Na^+ , Cl^- , and EC. This factor represents the most important process controlling water chemistry and appears as an axis of global water mineralization (Tab. 4 and 5), related to evaporitic elements. The chemical composition of groundwater is influenced by various natural and anthropogenic factors. Cation exchange and natural weathering reactions by water-soil/rock interactions are some of the natural processes that control groundwater chemistry. In the summer period, the second principal component (F2), which explains 18.56% of the total variance, is characterized by high concentrations of K^+ and NO_3^- (0.62 and 0.75, respectively). This suggests that groundwater is contaminated by anthropogenic activities, mainly associated with agricultural activity. In contrast, the third principal component (F3) explains only 13.54% of the variance and has a moderate positive connection with Mg^{2+} (0.53) and HCO_3^- (0.53), indicating that it is due to the dissolution of carbonate rocks (Tab. 5).

In the wet season (March 2019), the second principal component (F2) explains 19.30% of the total variance and is characterized by high concentrations of Ca^{2+} (0.59) and HCO_3^- (0.79), which suggests that groundwater is influenced by the dissolution of carbonate rocks representing the water coming from the recharge areas. These two factors (F1 and F2) show that the groundwater in the region follows two different processes in the dissolution of minerals, namely that of carbonates and that of evaporates. The third principal component (F3) explains only 10.18% of the variance and is dominated by NO_3^- (0.9), which is linked to anthropogenic activities. The reversal of the factors F2 and F3 between the two periods can be explained by the increase in water requirement during summer months, which leads to a higher density of irrigation and the use of chemical fertilizers. This results in a higher percentage of pollution during summer, with the pollution factor (F2) becoming more dominant than (F3) in June 2019. Overall, the results suggest that groundwater quality in the region is influenced by both natural and anthropogenic factors, which should be considered when designing strategies for groundwater management and protection.

Origin of the main major elements

To highlight the different mechanisms leading to the mineralization of groundwater, the ionic correlation diagrams produced by the molar concentration of the main elements was used. The Na^+ vs. Cl^- (Fig. 9a) identified the mechanism of water salinity acquisition in arid and semi-arid areas (Dixon and Chiswell, 1992; Jalali, 2009). In natural waters, the presence of Na^+ and Cl^- comes largely from the dissolution of halite (NaCl) found in Triassic formations (Capaccioni et al., 2005). The Bivariate Na^+ vs. Cl^- in Figure 9a shows a linear distribution of the data with medium to high correlation coefficients, in the order of 0.5 and 0.76, respectively, in June 2018 and March 2019, indicating the origin of the elements

Tab. 4 - Factors of water samples collected in June 2018.

Tab. 4 - Fattori dei campioni d'acqua prelevati in Giugno 2018.

(A)	F1	F2	F3
Ca ²⁺	0,930	-0,214	0,101
Mg ²⁺	0,778	-0,254	0,552
Na ⁺	0,741	0,141	-0,548
K ⁺	0,642	0,626	0,325
HCO ₃ ⁻	-0,290	0,342	0,534
Cl ⁻	0,848	-0,443	0,012
SO ₄ ²⁻	0,880	-0,033	-0,057
NO ₃ ⁻	0,393	0,756	0,186
EC	0,972	-0,094	-0,092
PH	0,190	0,708	-0,089

Tab. 5 - Factors of water samples collected in March 2019.

Tab. 5 - Fattori dei campioni d'acqua prelevati in Marzo 2019.

(B)	F1	F2	F3
Ca ²⁺	0,548	0,593	0,372
Mg ²⁺	0,664	0,444	-0,006
Na ⁺	0,910	-0,283	-0,091
K ⁺	0,729	0,168	0,175
HCO ₃ ⁻	0,358	0,791	0,084
Cl ⁻	0,839	-0,058	-0,071
SO ₄ ²⁻	0,733	-0,272	0,182
NO ₃ ⁻	-0,241	-0,217	0,908
EC	0,807	-0,429	-0,135
PH	0,392	-0,726	0,203

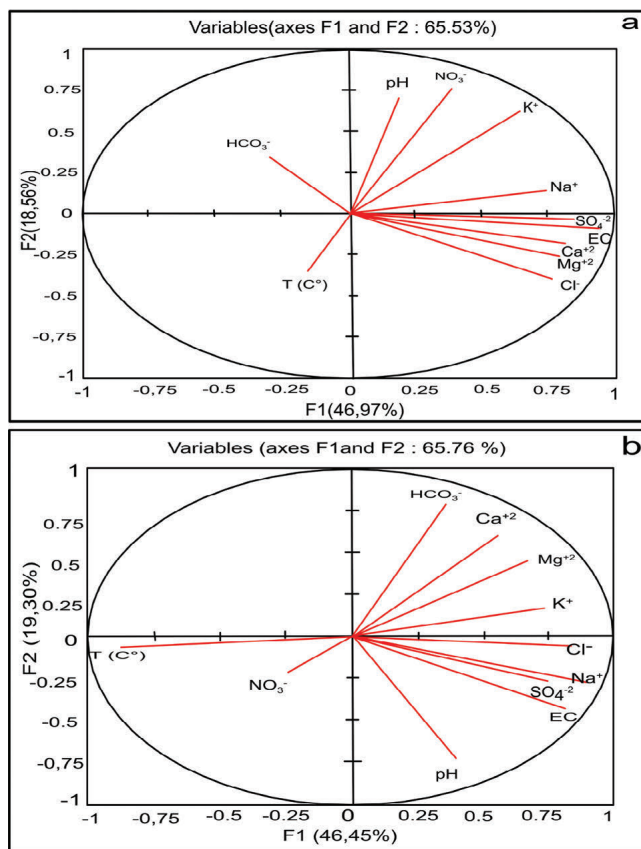


Fig. 8 - Correlating hydrochemical data from shallow aquifers of Barika: collected in Jun 2018 and March 2019.

Fig. 8 - Correlazione tra i dati idrochimici presi dall'acquifero superficiale di Barika: acquisiti in Giugno 2018 e Marzo 2019.

Na⁺ and Cl⁻ is largely due to dissolution of the halite (Djabri et al., 2008). This dissolution is, however, confirmed by the negative saturation indexes of groundwater with respect to halite which indicates an under-saturation state (Ben Moussa et al., 2009). During the winter period, some water points exhibit a higher sodium content compared to the expected slope of 1. This excess of sodium ions cannot be explained solely by the presence of halite, indicating the existence of another source for the sodium ions. One possible explanation for this increase is the phenomenon of direct ion exchange,

which occurs when water interacts with clay minerals. In this process, calcium ions are fixed by the clay minerals, leading to the release of two sodium ions into the water. This hypothesis is supported by the analysis of two samples from each period. On the other hand, there is a net enrichment in chlorides of 85% in a sample. This could be attributed to the anthropogenic sources, dissolution of other evaporate minerals (MgCl₂, CaCl₂), or cation exchange by reverse ion exchange (Na⁺ reduction) (Barzegar et al., 2019). In the second plot Ca²⁺ vs HCO₃⁻ (Fig. 9b), calcium is present in excess compared to bicarbonates. Furthermore, the Ca²⁺ vs SO₄²⁻ + HCO₃⁻ diagram (Fig. 9c) shows that the majority of points are shifted to the HCO₃⁻ + SO₄²⁻ pole, indicating a double origin for calcium, with one origin hydrolysis of secondary carbonates (aragonite, calcite), this reflects two other chemical facies, one bicarbonate-sodic, and the other bicarbonate-magnesium, which were particularly produced in June. (Fig. 9d), The findings show that silicate weathering dominates the dissolution of gypsum and anhydrite. Further in the plot for HCO₃⁻ + SO₄²⁻ vs Ca²⁺ + Mg²⁺ the majority of samples fall below the 1:1 aquiline line (Fig. 9e) indicating the dominance of alkaline earth over bicarbonate, signifying the process of silicate weathering in the study area. Generally, the couple Mg²⁺ vs Cl⁻ shows an average correlation between the two elements (r² = 0.6 in June and 0.4 in March) (Fig. 9f) and thus shows that the cation Mg²⁺ contributes to the salinity of groundwater in the summer period. While the excess of these ions in some samples can be attributed to the phenomenon of dolomitization, which would result from the irreversible dissolution of sulfates along with the precipitation of calcite, the deficit of these ions in some wells is explained by the fixing of magnesium ions by the process of base exchange. (Djamaï et al., 2011). For the contribution to Ca²⁺/Mg²⁺ (Fig. 9g), Hounslow suggests that there are three possibilities such as in the first case, the dissolution of gypsum (Mg²⁺/Ca²⁺ = 0.5), the second alteration of dolomitic limestone (Mg²⁺/Ca²⁺ < 0.5), and the third, dissolution of dolomite and calcite precipitation (Mg²⁺/Ca²⁺ > 0.5) (Hounslow, 2018). In the graph HCO₃⁻/(Cl⁻+SO₄²⁻) vs EC, the relationship shows the global origin of salinity. Indeed, this diagram (Fig. 9h) shows

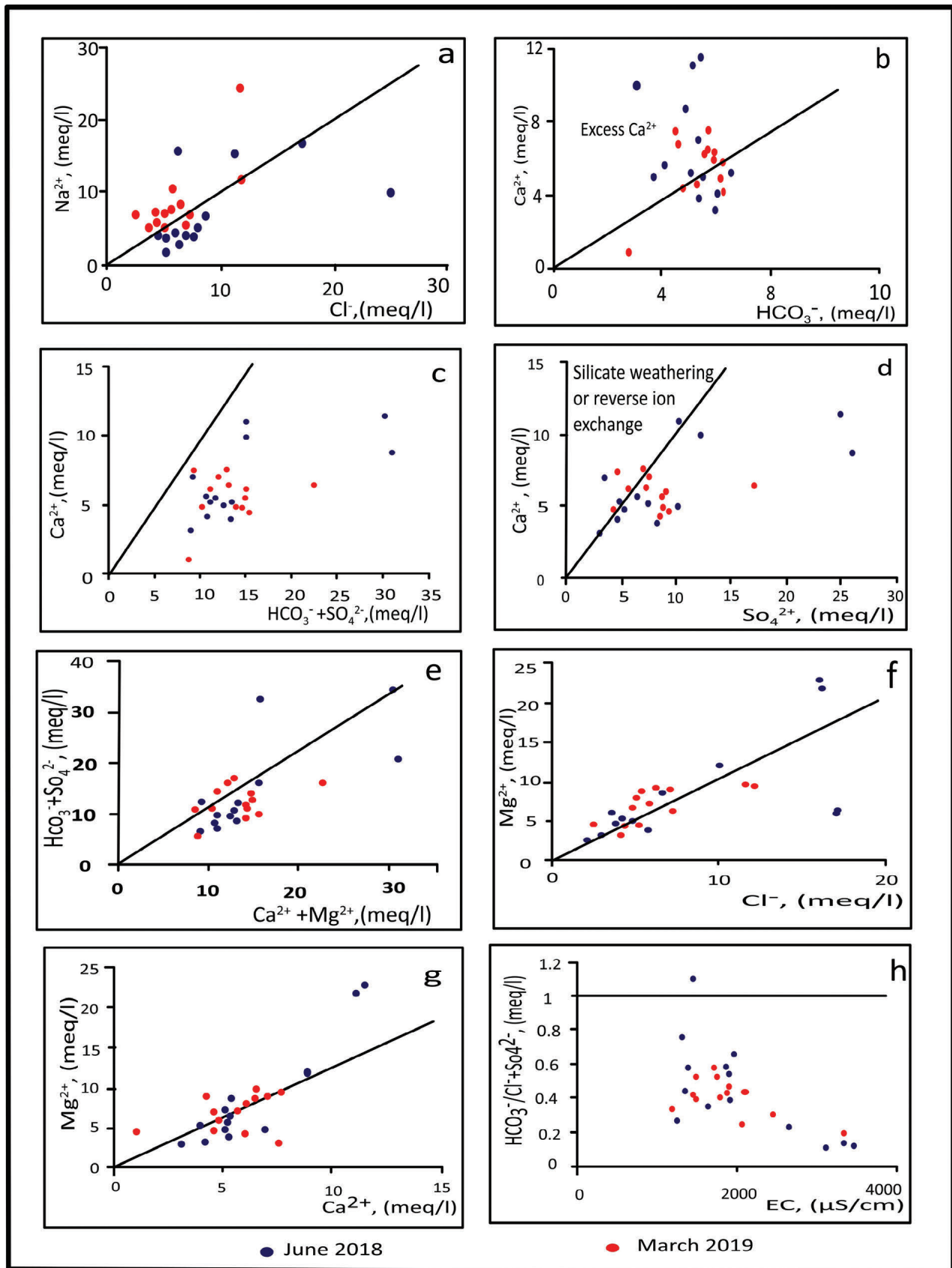


Fig. 9 - Plots of a) Na^+ versus Cl^- , b) Ca^{2+} versus HCO_3^- ; c) Ca^{2+} versus $\text{HCO}_3^- + \text{SO}_4^{2-}$; d) Ca^{2+} versus SO_4^{2-} ; e) $\text{HCO}_3^- + \text{SO}_4^{2-}$ versus $\text{Ca}^{2+} + \text{Mg}^{2+}$; f) Mg^{2+} versus $\text{Cl}^-/\text{Mg}^{2+}$; g) Mg^{2+} versus Ca^{2+} ; h) $\text{HCO}_3^-/(\text{Cl}^- + \text{SO}_4^{2-})$ versus EC.

Fig. 9 - Grafico di a) Na^+ versus Cl^- , b) Ca^{2+} versus HCO_3^- ; c) Ca^{2+} versus $\text{HCO}_3^- + \text{SO}_4^{2-}$; d) Ca^{2+} versus SO_4^{2-} ; e) $\text{HCO}_3^- + \text{SO}_4^{2-}$ versus $\text{Ca}^{2+} + \text{Mg}^{2+}$; f) Mg^{2+} versus $\text{Cl}^-/\text{Mg}^{2+}$; g) Mg^{2+} versus Ca^{2+} ; h) $\text{HCO}_3^-/(\text{Cl}^- + \text{SO}_4^{2-})$ versus EC.

one pole that can be individualized by its salinity where the most predominant ions are sulfate, chloride, and sulfuric at a rate of 99% in the summer period and 100% in the winter period. Additionally, this ratio increases as the salinity rises, and the salinity rises in the direction of groundwater flow. The infiltration of rainwater through the carbonate formations in the eastern part of the study area, (Metlili mount) at the edges, allows the dissolution of Upper Cretaceous limestone. During the flow of groundwater, water allows the dissolution of evaporated minerals, gypsum, and/or anhydrite, of the Mio-Plio-Quaternary age (the plain of Barika). Thus, we have interpreted the sample located at the carbonate pole.

Geochemical modeling

The saturation index (SI), as described by Cidu et al. (2009) and Raj and Shaji (2017), is a useful tool for characterizing the state of soluble minerals in water. This index can be used to distinguish between different phases of hydrochemical evolution and to identify the key geochemical reactions that control water chemistry (Mouassa et al., 2020). When the SI values are close to zero, an equilibrium state can be assumed, indicating a balance between the dissolution and precipitation of minerals in the water.

When water comes into contact with a mineral, the concentration of the solution can increase to a point where it reaches a maximum under certain physicochemical conditions. The SI (Eq. 3) is a measure of the solution's ability to dissolve a given mineral. A positive SI indicates that the solution has dissolved all of the mineral and cannot dissolve any more of it. Conversely, a negative SI shows that the groundwater is under-saturated with a specific mineral, meaning that more of the mineral can dissolve into the water. The SI of a mineral can be calculated using the following equation (Appelo and Postma, 2004).

$$\text{Saturation Index (SI)} = \log\left(\frac{IAP}{K_T}\right) \quad (3)$$

With K_T the equilibrium constant of mineral dissolution and IAP the ionic product in a mineral dissolution reaction. In this study, the open source USGS geochemical modeling software PHREEQC (PH Redox Equilibrium in Aqueous Systems) is used to calculate saturation indices for several minerals, including halite, gypsum, anhydrite, calcite, and aragonite. PHREEQC can be used to calculate the chemical speciation and solubility of minerals in water as well as to simulate chemical reactions in groundwater systems. It is able to model a wide range of natural and industrial processes, such as water-rock interactions, mineral precipitation and dissolution, acid-base reactions, and gas exchange between water and air. It is a widely used software in the fields of hydrogeology, environmental geochemistry, and geochemical modeling.

The results obtained are shown in Figure 10, which indicates that most of the solutions were under-saturated with respect to gypsum and anhydrite. This suggests that these evaporate minerals may have been in contact with the water

for a relatively long time and are therefore more prone to dissolution. On the other hand, halite was found to be largely under-saturated, which means that it can dissolve more easily in water and contribute to an increase in mineralization. The majority of samples were close to zero, indicating that they were in the equilibrium zone for carbonaceous minerals such as calcite and aragonite). The higher equilibrium constant (K) of evaporated minerals allows them to remain in water at high concentrations, which could explain our findings.

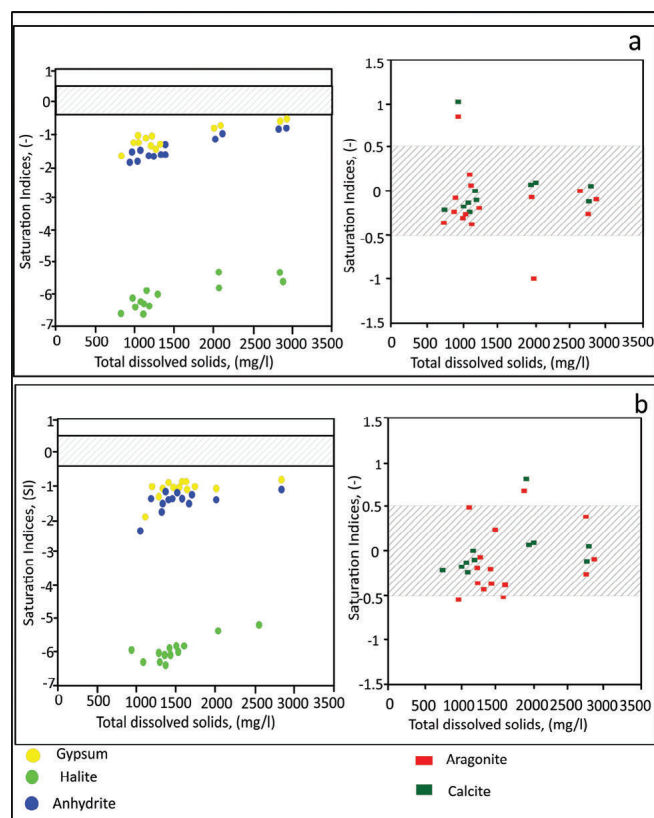


Fig. 10 - Saturation indices for carbonate and evaporate minerals using PHREEQC and Total Dissolved Solids, TDS data (a) June 2018 and (b) March 2019).

Fig. 10 - Indici di saturazione per carbonati e minerali evaporitici ottenuti con PHREEQC e valore dei solidi sospesi totali (a) Giugno 2018 e (b). Marzo 2019).

Evolution of mineralization vs groundwater direction

According to the mineralization distribution map (Fig. 11), mineralization decreases along groundwater flow paths from recharge to discharge areas, with high values found in the eastern part of the study area. Higher conductivity values characterize north-eastern wells, with an average EC of 3460 $\mu\text{S}/\text{cm}$ (June 2018) and 3340 $\mu\text{S}/\text{cm}$ (March 2019) and decrease to the south and southwest. These high conductivity and mineralization values suggest that natural processes, such as the presence of evaporates, mainly influence water-rock interactions, while other interactions involving carbonates, silicates, and clays also contribute to the geochemistry of groundwater in the plain. This is demonstrated by the concentrations of chemical elements that significantly contribute to mineralization, especially SO_4^{2-} and Cl^- , with values up to 1250 mg/L – 419 mg/L in June 2018 and

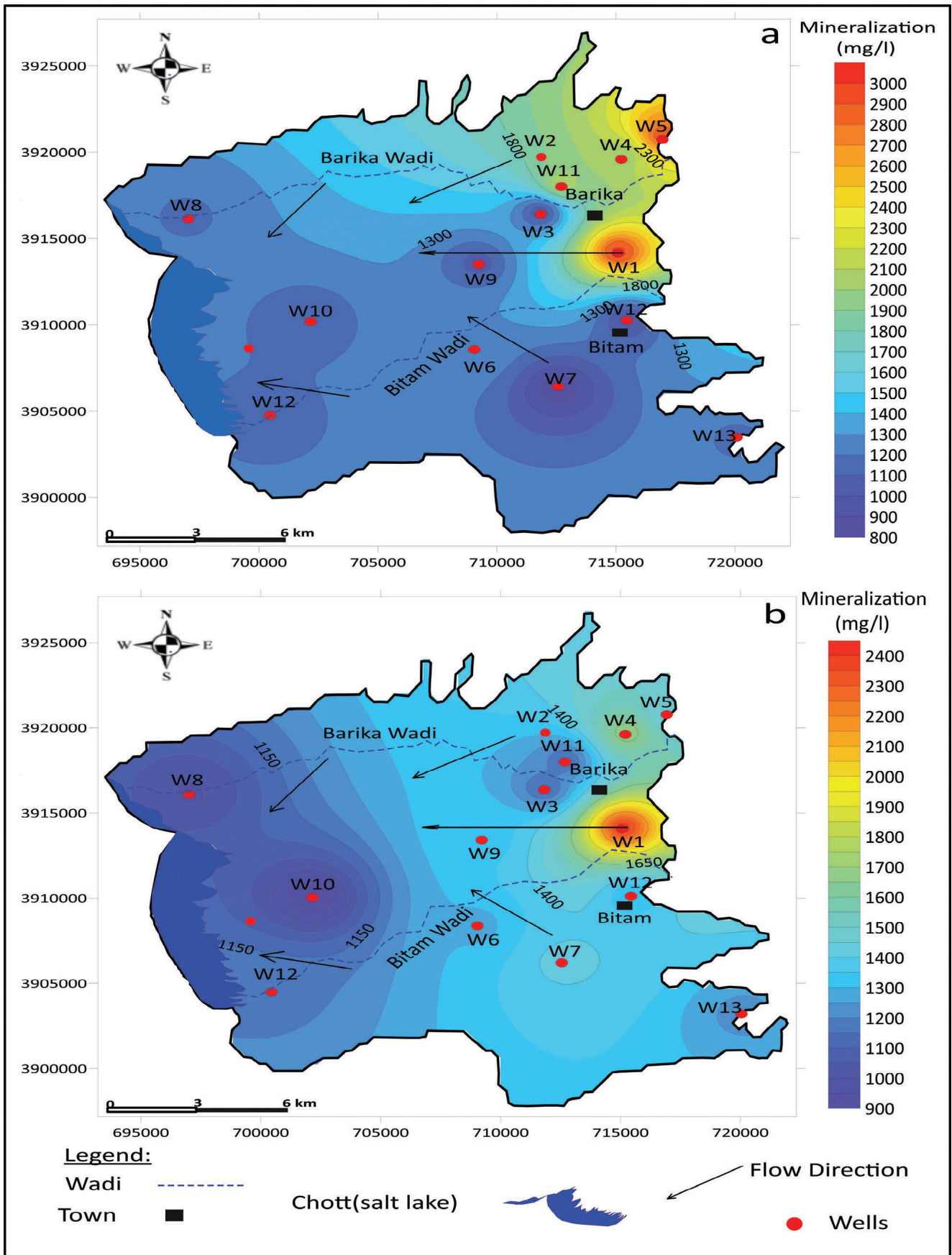


Fig. 11 - Evolution of mineralization: a study of June 2018 and March 2019 data.

Fig. 11 - Evoluzione della mineralizzazione: studio dei dati del periodo di Giugno 2018 e Marzo 2019.

814.14 mg/L – 419 mg/L in March 2019 in the eastern part of the region. Magnesium decreases westward. The active ingredient found in feeding areas, HCO_3^- , has a value of 317 mg/L – 364 mg/L in both periods (June and March), which is due to the slow residence time of groundwater flowing from the carbonate formations that border the plain to the aquifer. As the groundwater passes through heterogeneous materials such as silt, gravel, sand, and evaporates, bicarbonate undergoes dilution, and ion exchange reactions occur, leading to a decrease in mineralization in the direction of flow.

Conclusion

In conclusion, this study represents the first comprehensive investigation of its kind into the mineralization process and chemical behavior of groundwater in the Barika region of north-eastern Algeria. The findings provide valuable insights into the mineralization process and chemical behavior of its groundwater. The results reveal that groundwater in the region has high mineralization levels, particularly during the dry season, and that certain elements such as SO_4^{2-} and Cl^- are present in slightly higher concentrations during this time. The hydrochemical analysis indicates that natural processes, such as water-rock interactions, precipitation of carbonate minerals, and cation exchange reactions, contribute to groundwater mineralization, with the dissolution of evaporates playing a significant role. The study also demonstrates significant connections between electrical conductivity and various chemical elements, highlighting the importance of sulfates, calcium, chlorides, sodium, and magnesium in mineralization acquisition. These findings have important implications for policymakers in developing effective water management strategies in response to increasing water demands for various purposes.

Further research is recommended to confirm the general mineralization process mechanism and groundwater quality in the Barika region. This study contributes to a better understanding of the sources of water quality variability in the region and can serve as a useful guide for developing more effective management strategies for groundwater resources. As global climate change continues to impact the hydrological cycle, it is crucial to monitor and understand how these changes may affect groundwater chemistry in the future, potentially altering the suitability of groundwater for various uses. Therefore, ongoing research and monitoring efforts will be crucial for managing and protecting this vital resource in the Barika region and beyond.

Acknowledgments

The authors thank the Algerian Ministry of Higher Education and Research (MHER) for the Scholarship awarded to the first author. This article was the result of a scientific collaboration between the Department of Geology of the University Mustapha Ben Boulaid Batna 2 (Algeria) and the Department of Civil Engineering of the RMC of Canada (Kingston, Ontario). Finally, the authors would like to acknowledge the constructive suggestions and review comments on an earlier version of this manuscript by two anonymous reviewers.

Funding source

Tafrount Assia is a PhD candidate and received a scholarship from the Algerian Ministry of Higher Education and Research.

Competing interest

The authors declare no competing interest.

Author contributions

The material preparation, data collection, and analysis were conducted by Author 1, Mrs. A. Tafrount with guidance and support from both authors 2 and 3, Profs. T. Drias and D. Chenaf who are the PhD supervisor and co-supervisor, respectively, of the first author. The continuous improvement of the paper's writing, editing, and structure was carried out collaboratively among the three first authors. The fourth author, Prof. N. Brinis, contributed to the hydrogeochemical interpretation of the test results. All authors have read and approved the published manuscript.

All authors have read and approved the published manuscript.

Additional information

Supplementary information is available for this paper at <https://doi.org/10.7343/as-2023-624>

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